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## Light-Driven Dynamics in Hybrid Nanostructures

### From Ultrafast Photoemission Studies to Functional Optical Systems

Zaiats, Nelia

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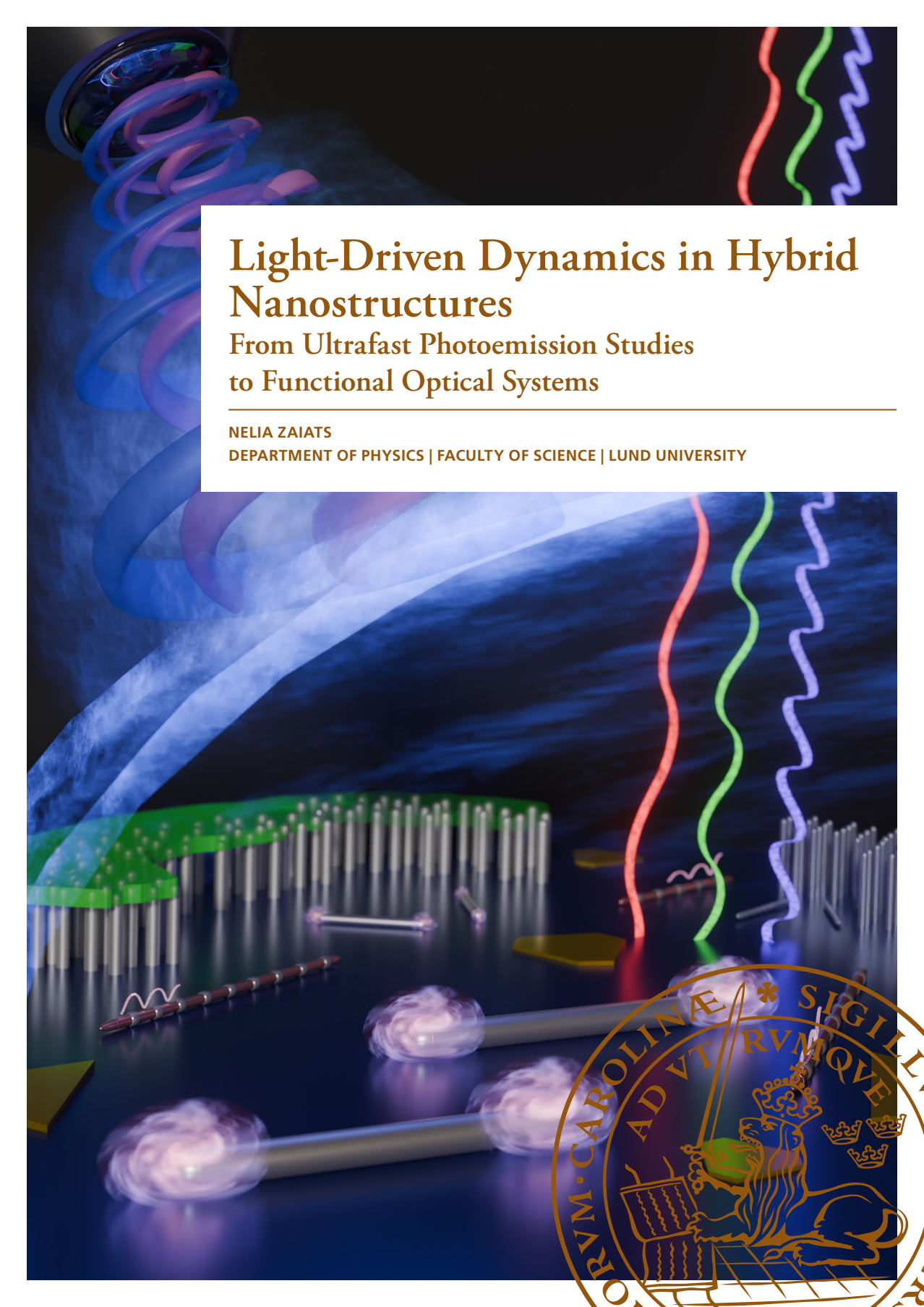
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# Light-Driven Dynamics in Hybrid Nanostructures

From Ultrafast Photoemission Studies  
to Functional Optical Systems

NELIA ZAIATS

DEPARTMENT OF PHYSICS | FACULTY OF SCIENCE | LUND UNIVERSITY





# Light-Driven Dynamics in Hybrid Nanostructures

From Ultrafast Photoemission Studies  
to Functional Optical Systems

by Nelia Zaiats



**LUND**  
UNIVERSITY

DOCTORAL DISSERTATION

Thesis for the degree of Doctor of Philosophy (PhD)

Thesis advisors: Anders Mikkelsen, Rainer Timm, Jan Vogelsang

Faculty opponent: Zhi-Heng Loh

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in the Rydberg lecture hall (Rydbergsalen) at the Department of Physics on Friday, the 11<sup>th</sup> of  
September 2026 at 09:15.

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| <b>Abstract</b><br>Understanding and controlling light-driven electronic processes at the nanometer scale is important for the continued development of next-generation optoelectronic devices, energy-efficient information processing, and emerging neuromorphic systems. This thesis investigates light-matter interactions in nanoscale systems across a broad range of timescales, from femtosecond plasmon dynamics to molecular memory persisting for hours or days.<br>The first part of this work is focused on fundamental studies of ultrafast electronic processes using time-resolved photoemission electron microscopy (TR-PEEM). Direct visualization of plasmonic near fields in silver nanowires excited by few-cycle short-wavelength infrared (SWIR) pulses reveals strong spatially localized field enhancement and demonstrates the impact of nanoscale geometrical variations on plasmonic behavior. The extension of this concept is further explored in semiconductor-metal nanowire systems, highlighting the role of composition and structure in determining carrier excitation and propagation dynamics. The experimental observations are supported by electromagnetic simulations and an interpretation of photoemission processes.<br>Beyond these fundamental studies, this work also explores hybrid nanowire-molecule systems aimed at neuromorphic device functionality. For this, donor-acceptor Stenhouse adduct (DASA) dyes are integrated with semiconductor nanowire devices, combining reversible molecular switching with electronic readout. In these hybrid devices, optical writing and electrical reading are largely decoupled, enabling dynamic control of the device response. Furthermore, the concept is extended to wavelength-selective multi-dye systems, demonstrating the possibility of independently addressable optical channels within a single device architecture. The resulting devices exhibit memory, history-dependent behavior, and tunable temporal dynamics that resemble one of the key aspects of neuromorphic functionality.<br>By bridging fundamental studies of ultrafast nanoscale processes with the development of functional hybrid devices, this thesis explores how light can be used to probe electronic processes on their natural timescales and how experimental techniques and materials can be combined to advance light-controlled electronics and information processing. |                                |                                                                     |  |
| <b>Key words</b><br>Photoemission electron microscopy (PEEM), ultrafast dynamics, plasmonics, nanowire, hybrid nanostructure, Donor-acceptor Stenhouse adducts (DASA)                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |                                |                                                                     |  |
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# Light-Driven Dynamics in Hybrid Nanostructures

From Ultrafast Photoemission Studies  
to Functional Optical Systems

Nelia Zaiats



**LUND**  
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**Cover illustration front:** Conceptual illustration of light exciting diverse nanoscale systems, where ultrafast electron dynamics in nanostructures and adaptive optical switching in nanowire–DASA hybrid devices are converted into measurable electrical signals. Cover artwork by Ulrike Küst, created for this thesis in collaboration with the author.

**Cover illustration back:** “The real voyage of discovery consists not in seeking new landscapes, but in having new eyes.” - Marcel Proust

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*Dedicated to my loved ones,  
who help me see a little further.*



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# Popular summary in English

## *Seeing the invisible*

When you look at a solar panel, a computer chip, or even a piece of metal, you are seeing only the surface of a much smaller world. Beneath what our eyes can observe, electrons move through materials and interact with light, shaping many of the technologies we rely on every day.

The difficulty of studying electrons begins with their extraordinarily small size. To appreciate just how small this world is, it helps to compare it with the vast range of scales found in nature. The outer reaches of the Solar System extend to roughly  $10^{15}$  meters – about 1,000,000,000,000,000 meters. At the opposite extreme, an atomic nucleus is approximately  $10^{-15}$  meters in size – just 0.000000000000001 meters. In a sense, the distance from everyday human scales down to the atomic nucleus is comparable to the distance from everyday scales up to the outer Solar System. It is tempting to think that studying electrons is like being an astronaut in reverse: instead of journeying across vast distances into outer space, we venture inward toward a universe of astonishingly small dimensions.

The microscopic world is also strange in another way. Electrons are particles, but unlike anything we encounter in everyday life, they also exhibit wave-like behavior. Rather than always following a well-defined path, they can spread through space and interact in ways that challenge our intuition. Understanding how electrons move and exchange energy with light is essential for improving many of the technologies that surround us.

However, the challenge is not only the size, but also the speed.

A femtosecond ( $10^{-15}$  seconds) is one millionth of a billionth of a second, and it is related to one second in the same way that one second is to approximately 30 million years. Light travels at nearly  $3 \times 10^8$  meters per second, and when it interacts with a material, it can transfer energy to electrons almost instantaneously. As a result, electrons can absorb this energy, redistribute it, and change their state on timescales far shorter than anything we encounter in everyday life. In the time of a single heartbeat, countless electronic processes can take place inside a material. Events happening this quickly cannot be captured by any conventional camera. Instead, we can use ultrafast laser light to “take a photo” of electrons.

In the fundamental part of this thesis, a technique called *time-resolved photoemission electron microscopy* (TR-PEEM) is used to observe how electrons respond to light. The method works by using a laser to eject electrons from a material and then forming an image from these emitted electrons. By exciting a nanostructure with one laser pulse and probing it with a second pulse arriving at a controlled time later, it becomes possible

to reconstruct a movie of electronic processes unfolding on femtosecond timescales.

Using this approach, the thesis investigates metallic and semiconductor nanostructures, including nanowires that are thousands of times thinner than a human hair. The measurements with TR-PEEM allow us to observe where electrons are emitted in these nanostructures, how they move, and how their distribution evolves over time. By imaging this light-driven electron dynamics, this thesis provides new insight into how optical energy is concentrated and converted into electronic motion at the nanoscale.

### *Learning from nature*

Sometimes, the best ideas do not come only from physics and engineering; they can also be found in nature itself. Throughout history, engineers and scientists have borrowed ideas from biological systems that have been refined through millions of years of evolution. Airplane wings, for example, were inspired by birds, while high-speed trains in Japan were redesigned based on the streamlined shape of a kingfisher's beak to reduce noise and improve efficiency. Even modern robots increasingly imitate the movement of animals to navigate complex environments more efficiently.

The same idea can be applied to information processing. The human brain performs calculations, recognizes patterns, and learns from experience while consuming only about 20 W of power – less energy than a small light bulb. Even much simpler nervous systems, such as those of insects, can navigate, communicate, and adapt to changing environments with remarkable efficiency. These biological systems do not work like conventional computers; instead, the connections between neurons, called synapses, continuously adapt depending on previous signals. Inspired by these principles, researchers have begun developing neuromorphic technologies that are designed to mimic certain aspects of how the brain processes information. Rather than separating computation and memory into different components, these devices can adapt their response and, in some cases, even learn from previous inputs.

One possible route toward such technologies is explored in parts of this thesis through the combination of light-sensitive molecules and semiconductor nanowires. These molecules, known as DASA molecules, change their structure and color when illuminated by specific wavelengths of light. They do not immediately return to their original state but instead recover gradually over time. In the devices studied in this thesis, the molecules are deposited directly onto semiconductor nanowires, creating a hybrid system in which light and electronics work together. Light is used to modify the state of the molecules, while the nanowire converts the resulting optical change into an electrical signal that can be measured. In this way, information can be written optically and read out electrically. By combining molecules with different optical responses, it also becomes possible to create multiple independent channels within a single device, enabling more complex forms of information storage and processing. As a result, the response of the

device depends not only on the light applied at a given moment but also on the history of previous illumination, resembling the memory behavior of biological synapses.

By integrating photoswitchable DASA molecules with semiconductor nanowires, this thesis demonstrates how light can be used to create memory-like and adaptive behavior in simple nanoscale devices. While these devices are far simpler than a biological brain, they illustrate how principles developed by nature over millions of years can inspire entirely new approaches to information processing. Instead of making computers only faster, future technologies may become smarter by incorporating memory, adaptation, and learning directly into the materials themselves.

*So what is it all for?*

At its core, this thesis explores how light can be used both to reveal and to control processes occurring far beyond the limits of human perception.

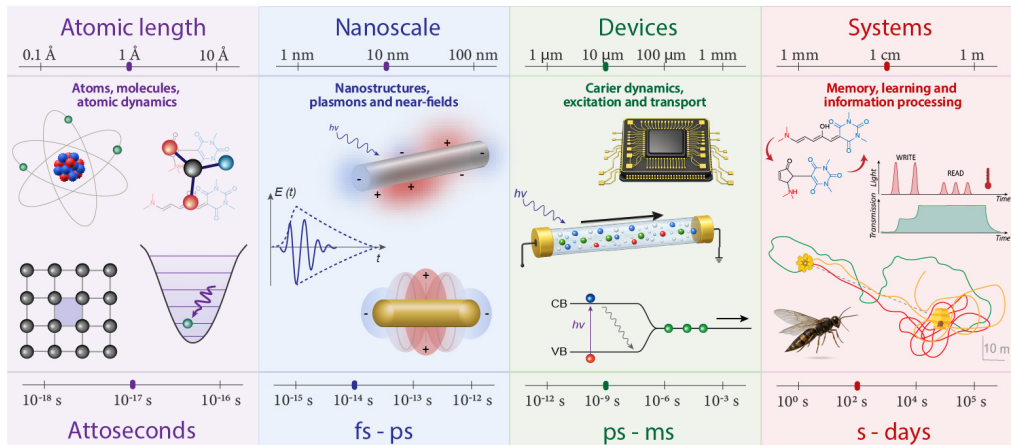


**Figure 1:** Schematic overview of the length and time scales relevant to this thesis, ranging from atomic and molecular electron dynamics through ultrafast plasmonic phenomena in nanostructures to carrier transport, memory, and fundamental physical and information-processing processes in functional optoelectronic and neuromorphic systems.

Figure 1 illustrates the broad range of length and time scales covered in this thesis. Although the investigated systems differ in their physical realization, they are united by a common theme: understanding how light interacts with matter across a vast range of length and timescales.

The studies presented here span more than fifteen orders of magnitude in time, from electron dynamics unfolding within femtoseconds to molecular memory effects persisting for seconds, minutes, or longer. On the shortest timescales, ultrafast photoemission electron microscopy makes it possible to visualize how optical energy is concentrated

within metallic and semiconductor nanostructures and converted into electronic motion. On much longer timescales, photoswitchable DASA molecules integrated with semiconductor nanowires demonstrate how light can modify material properties, store information, and create adaptive responses reminiscent of biological synapses.

Together, these studies show how the interaction between light and matter can lead to both fundamental discoveries and new technological possibilities. Insights gained from ultrafast electron dynamics may help guide the development of future optoelectronic devices, while nanowires combined with molecules provide a platform for adaptive sensing, optical memory, and neuromorphic information processing.

# Populärvetenskaplig sammanfattning på Svenska

## *Att se det osynliga*

När du tittar på en solpanel, ett datorchip eller till och med en bit metall ser du bara ytan av en mycket mindre värld. Under ytan döljer det sig en värld där elektroner rör sig genom material och interagerar med ljus, vilket utgör grunden för många av de teknologier vi förlitar oss på varje dag.

Svårigheten med att studera elektroner börjar med deras extraordinärt mikroskopiska storlek. För att förstå hur liten denna värld verkligen är hjälper det att jämföra den med det enorma spann av skalor som finns i naturen. De yttersta delarna av solsystemet sträcker sig till ungefär  $10^{15}$  meter – alltså 1 000 000 000 000 000 meter. I den motsatta extremen har en atomkärna en diameter på ungefär  $10^{-15}$  meter – endast 0.000 000 000 000 001 meter. På så sätt är avståndet från vardagliga mänskliga skalor ner till atomkärnan jämförbart med avståndet från vardagliga skalor upp till solsystemets yttersta delar. Det är frestande att tänka att studerandet av elektroner är som att vara en astronaut i omvänd riktning: istället för att färdas över enorma avstånd ut i rymden färdas vi inåt mot ett universum av häpnadsväckande små dimensioner.

Den mikroskopiska världen är också märklig på ett annat sätt. Elektroner är partiklar, men till skillnad från allt vi möter i vardagen uppvisar de också vågliknande beteende. Istället för att alltid följa en väldefinierad bana kan de sprida sig genom rummet och interagera på ett sätt som utmanar vår intuition. Att förstå hur elektroner rör sig och utbyter energi med ljus är avgörande för att förbättra många av de teknologier som omger oss.

Utmaningen handlar dock inte bara om storleken, utan också om hastigheten.

En femtosekund ( $10^{-15}$  sekunder) är en miljondel av en miljarddels sekund, och den förhåller sig till en sekund på samma sätt som en sekund förhåller sig till ungefär 30 miljoner år. Ljus färdas med nästan  $3 \times 10^8$  meter per sekund, och när det interagerar med ett material kan det överföra energi till elektroner nästan omedelbart. Som ett resultat kan elektroner absorbera denna energi, omfördela den och ändra sitt tillstånd på tidsskalor som är långt kortare än något vi möter i vardagen. Under tiden för ett enda hjärtslag kan otaliga elektroniska processer äga rum inuti ett material. Händelser som sker så snabbt kan inte fångas av någon konventionell kamera. Istället kan vi använda ultrasnabba laserpulser för att “ta ett fotografi” av elektroner.

I den grundläggande delen av denna avhandling används en teknik som kallas *tidsupplöst fotoemissionselektronmikroskopi* (TR-PEEM) för att observera hur elektroner reagerar på ljus. Metoden fungerar genom att använda en laser för att slå ut elektroner från ett material och sedan skapa en bild av dessa emitterade elektroner. Genom att excitera en

nanostruktur med en laserpuls och undersöka den med en andra puls som anländer vid en kontrollerad tidpunkt senare blir det möjligt att rekonstruera en film av elektroniska processer som utvecklas på femtosekundsskalan.

Med hjälp av detta tillvägagångssätt undersöker avhandlingen metalliska och halvledande nanostrukturer, inklusive nanotrådar som är tusentals gånger tunnare än ett mänskligt hårstrå. Mätningarna med TR-PEEM gör det möjligt att observera var elektroner emitteras i dessa nanostrukturer, hur de rör sig och hur deras fördelning utvecklas över tid. Genom att avbilda denna ljusdrivna elektrondynamik ger avhandlingen nya insikter i hur optisk energi koncentreras och omvandlas till elektronisk rörelse på nanoskalan.

### *Att lära av naturen*

Ibland kommer de bästa idéerna inte från fysik och ingenjörsvetenskap, utan från naturen. Genom historien har ingenjörer och forskare lånat idéer från biologiska system som har förfinats genom miljontals år av evolution. Flygplansvingar inspirerades till exempel av fåglar, medan höghastighetståg i Japan omdesignades utifrån den strömlinjeformade formen hos en kungsfiskares näbb för att minska buller och förbättra effektiviteten. Till och med moderna robotar efterliknar i allt större utsträckning djurs rörelser för att navigera mer effektivt i komplexa miljöer.

Samma idé kan tillämpas på informationsbehandling. Den mänskliga hjärnan utför beräkningar, känner igen mönster och lär sig av erfarenhet samtidigt som den förbrukar endast omkring 20 W effekt – mindre energi än en liten glödlampa. Även mycket enklare nervsystem, såsom insekters, kan navigera, kommunicera och anpassa sig till föränderliga miljöer med anmärkningsvärd effektivitet. Dessa biologiska system fungerar inte som konventionella datorer; istället anpassas kopplingarna mellan neuroner, kallade synapser, kontinuerligt beroende på tidigare signaler. Inspirerade av dessa principer har forskare börjat utveckla neuromorfa teknologier som är utformade för att efterlikna vissa aspekter av hur hjärnan bearbetar information. Istället för att separera beräkning och minne i olika komponenter kan dessa enheter anpassa sitt svar och i vissa fall till och med lära sig av tidigare insignaler.

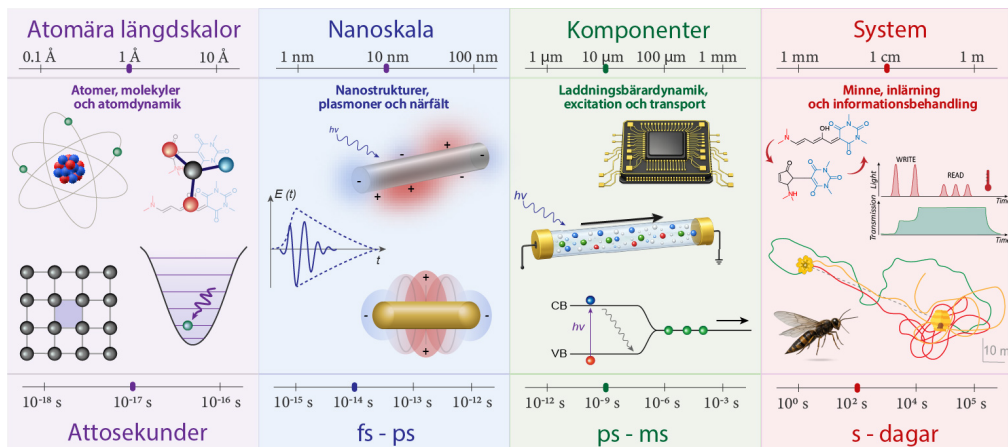
En möjlig väg mot sådana teknologier utforskas i delar av denna avhandling genom kombinationen av ljuskänsliga molekyler och halvledarnanotrådar. Dessa molekyler, kända som DASA-molekyler, ändrar sin struktur och färg när de belyses med specifika ljusvåglängder. De återgår inte omedelbart till sitt ursprungliga tillstånd utan återhämtar sig gradvis över tid. I de enheter som studeras i denna avhandling depone-ras molekylerna direkt på halvledarnanotrådar, vilket skapar ett hybridssystem där ljus och elektronik samverkar. Ljus används för att förändra molekylernas tillstånd, medan nanotråden omvandlar den resulterande optiska förändringen till en elektrisk signal som kan mätas. På detta sätt kan information skrivas optiskt och läsas ut elektriskt. Genom

att kombinera molekyler med olika optiska responser blir det också möjligt att skapa flera oberoende kanaler inom en enda enhet, vilket möjliggör mer komplexa former av informationslagring och informationsbehandling. Som ett resultat beror enhetens respons inte bara på det ljus som appliceras vid ett givet ögonblick utan också på historiken av tidigare belysning, vilket påminner om minnesbeteendet hos biologiska synapser.

Genom att integrera fotoswitchbara DASA-molekyler med halvledarnanotrådar visar avhandlingen hur ljus kan användas för att skapa minnesliknande och adaptivt beteende i enkla enheter på nanoskalan. Även om dessa enheter är mycket enklare än en biologisk hjärna illustrerar de hur principer som utvecklats av naturen under miljontals år kan inspirera helt nya sätt att behandla information. Istället för att bara göra datorer snabbare kan framtida teknologier bli smartare genom att integrera minne, anpassning och inlärning direkt i själva materialen.

*Så vad är allt detta till för?*

I grunden undersöker denna avhandling hur ljus kan användas både för att avslöja och för att kontrollera processer som sker långt bortom gränserna för mänsklig perception.



**Figur 1:** Schematisk översikt över de längd- och tidsskalor som är relevanta för denna avhandling, från atomär och molekylär elektrondynamik, via ultrasnabba plasmoniska fenomen i nanostrukturer, till laddningsbärartransport, minne samt grundläggande fysik och informationsbearbetning i funktionella optoelektroniska och neuromorfa system.

Figur 1 illustrerar det breda spektrum av längd- och tidsskalor som behandlas i denna avhandling. Även om de undersökta systemen skiljer sig åt i sin fysiska realisering förenas de av ett gemensamt tema: att förstå hur ljus växelverkar med materia över ett mycket brett spann av längd- och tidsskalor.

Studierna som presenteras här spänner över mer än femton storleksordningar i tid, från

elektrondynamik som utspelar sig inom femtosekunder till molekylära minnes effekter som kvarstår i sekunder, minuter eller längre. På de kortaste tidsskalorna gör ultrasnabb fotoemissionselektronmikroskopi det möjligt att visualisera hur optisk energi koncentreras inom metalliska och halvledande nanostrukturer och omvandlas till elektronisk rörelse. På betydligt längre tidsskalor visar fotoswitchbara DASA-molekyler integrerade med halvledarnanotrådar hur ljus kan förändra materialegenskaper, lagra information och skapa adaptiva responser som påminner om biologiska synapser.

Tillsammans visar dessa studier hur interaktionen mellan ljus och materia kan leda till både grundläggande upptäckter och nya teknologiska möjligheter. Insikter från ultrasnabb elektrondynamik kan bidra till utvecklingen av framtida optoelektroniska enheter, medan nanotrådar kombinerade med molekyler erbjuder en plattform för adaptiv avkänning, optiskt minne och neuromorf informationsbehandling.

# Науково-популярний огляд Українською

## *Побачити невидиме*

Коли ви дивитеся на сонячну панель, комп'ютерний чип або навіть шматок металу, ви бачите лише поверхню значно меншого світу. Поза тим, що можуть спостерігати наші очі, електрони рухаються крізь матеріали та взаємодіють зі світлом, формуючи багато технологічних явищ, на які ми щодня покладаємося.

Складність вивчення електронів починається з їхнього надзвичайно малого розміру. Щоб зрозуміти, наскільки малим є цей світ, корисно порівняти його з величезним діапазоном масштабів, які існують у природі. Зовнішні межі Сонячної системи простягаються приблизно до  $10^{15}$  метрів – близько 1 000 000 000 000 000 метрів. На протилежному кінці шкали атомне ядро має розмір приблизно  $10^{-15}$  метра – лише 0.000 000 000 000 001 метра. У певному сенсі відстань від звичних людських масштабів до атомного ядра можна порівняти з відстанню від звичних масштабів до зовнішніх меж Сонячної системи. Напрощується цікава аналогія: вивчення електронів схоже на космічну подорож навпаки – замість того щоб долати величезні відстані у відкритому космосі, ми прямуємо до всесвіту дивовижно малих масштабів.

Мікроскопічний світ є дивним і з іншої причини. Електрони є частинками матерії, але, на відміну від будь-чого, що ми зустрічаємо в повсякденному житті, вони також можуть поводитися подібно до хвиль. Замість того щоб завжди рухатися чітко визначеною траєкторією, вони можуть поширюватися в просторі та взаємодіяти способами, які суперечать нашій інтуїції. Дослідження того, як електрони рухаються та обмінюються енергією зі світлом, допомагає вдосконаленню багатьох технологій, що нас оточують.

Однак виклик полягає не лише в розмірах, а й у швидкості.

Фемтосекунда ( $10^{-15}$  секунди) – це одна мільйонна від однієї мільярдної частки секунди, і вона співвідноситься з однією секундою так само, як одна секунда співвідноситься приблизно з 30 мільйонами років. Світло поширюється зі швидкістю майже  $3 \times 10^8$  метрів за секунду, і коли воно взаємодіє з матеріалом, воно може майже миттєво передавати енергію електронам. У результаті електрони можуть поглинати цю енергію, перерозподіляти її та змінювати свій стан за надзвичайно короткий час – значно швидше, ніж будь-які процеси, які ми спостерігаємо в повсякденному житті. За час одного серцебиття всередині матеріалу можуть відбутися незлічені процеси за участю електронів. Події, що відбуваються так швидко, неможливо зафіксувати жодною звичайною камерою. Натомість ми можемо використовувати надшвидке лазерне світло, щоб “сфотографувати” електронів.

У фундаментальній частині цієї дисертації використовується метод часово-роздільної фотоemisійної електронної мікроскопії, або TR-PEEM, який дає змогу спостерігати за тим, як електрони реагують на світло. Метод працює шляхом використання лазера для вибивання електронів із матеріалу та подальшого формування зображення з цих випромінених електронів. Спрямовуючи на наноструктуру один лазерний імпульс і досліджуючи її другим імпульсом, що надходить через точно контрольований проміжок часу, можна відтворити своєрідний фільм електронних процесів, які розгортаються протягом фемтосекунд.

За допомогою цього підходу в дисертації досліджуються металеві та напівпровідникові наноструктури, включаючи нанодрти, які в тисячі разів тонші за людську волосину. Вимірювання за допомогою TR-PEEM дозволяють спостерігати, де саме електрони випромінюються в цих наноструктурах, як вони рухаються та як змінюється їхній розподіл із часом. Завдяки візуалізації цієї керованої світлом динаміки електронів ця дисертація надає нове розуміння того, як оптична енергія концентрується та перетворюється на електронний рух на наномасштабі.

### *Вчитися у природи*

Іноді найкращі ідеї приходять не лише з фізики та інженерії – їх також можна знайти в самій природі. Протягом історії інженери та науковці запозичували ідеї з біологічних систем, які вдосконалювалися мільйонами років еволюції. Наприклад, крила літаків були натхненні птахами, тоді як високошвидкісні поїзди в Японії були перепроєктовані за зразком дзьоба зимородка, щоб зменшити шум і підвищити ефективність. Навіть сучасні роботи дедалі частіше наслідують рухи тварин для ефективнішого пересування в складних середовищах.

Ту саму ідею можна застосувати до обробки інформації. Людський мозок виконує обчислення, розпізнає закономірності та навчається на досвіді, споживаючи лише близько 20 Вт потужності – менше енергії, ніж лампочка. Навіть значно простіші нервові системи, наприклад у комах, можуть орієнтуватися, спілкуватися та адаптуватися до змінних умов із вражаючою ефективністю. Ці біологічні системи працюють не так, як звичайні комп'ютери; натомість зв'язки між нейронами, які називаються синапсами, безперервно адаптуються залежно від попередніх сигналів. Натхненні цими принципами, дослідники почали розробляти нейроморфні технології, призначені для імітації певних аспектів того, як мозок обробляє інформацію. Замість розділення обчислень і пам'яті на різні компоненти такі пристрої можуть адаптувати свою реакцію, а в деяких випадках навіть навчатися на попередніх вхідних сигналах.

Один із можливих шляхів до таких технологій досліджується в окремих частинах цієї дисертації через поєднання світлочутливих молекул і напівпровідникових нанодротів. Ці молекули, відомі як молекули DASA, змінюють свою структуру та

колір під дією світла певних довжин хвиль. Вони не повертаються одразу до свого початкового стану, а поступово відновлюються з часом. У пристроях, досліджених у цій дисертації, молекули безпосередньо наносяться на напівпровідникові нанодроти, створюючи гібридну систему, у якій світло та електроніка працюють разом. Світло використовується для зміни стану молекул, тоді як нанодріт перетворює отриману оптичну зміну на електричний сигнал, який можна виміряти. Таким чином інформація може записуватися оптично та зчитуватися електрично. Поєднуючи молекули з різними оптичними відгуками, також стає можливим створення кількох незалежних каналів в одному пристрої, що дозволяє реалізувати складніші форми зберігання та обробки інформації. У результаті реакція пристрою залежить не лише від світла, прикладеного в певний момент, але й від історії попереднього освітлення, нагадуючи поведінку пам'яті біологічних синапсів.

Інтегруючи фотоперемикні молекули DASA з напівпровідниковими нанодротами, дисертація демонструє, як світло можна використовувати для створення поведінки, подібної до пам'яті та адаптації, у простих пристроях наномасштабу. Хоча ці пристрої набагато простіші за біологічний мозок, вони показують, як принципи, вироблені природою протягом мільйонів років, можуть надихати абсолютно нові підходи до обробки інформації. Замість того щоб робити комп'ютери лише швидшими, майбутні технології можуть стати розумнішими завдяки інтеграції пам'яті, адаптації та навчання безпосередньо в самі матеріали.

*То для чого все це?*

У своїй основі ця дисертація досліджує, як світло можна використовувати як для виявлення, так і для керування процесами, що відбуваються далеко за межами людського сприйняття.

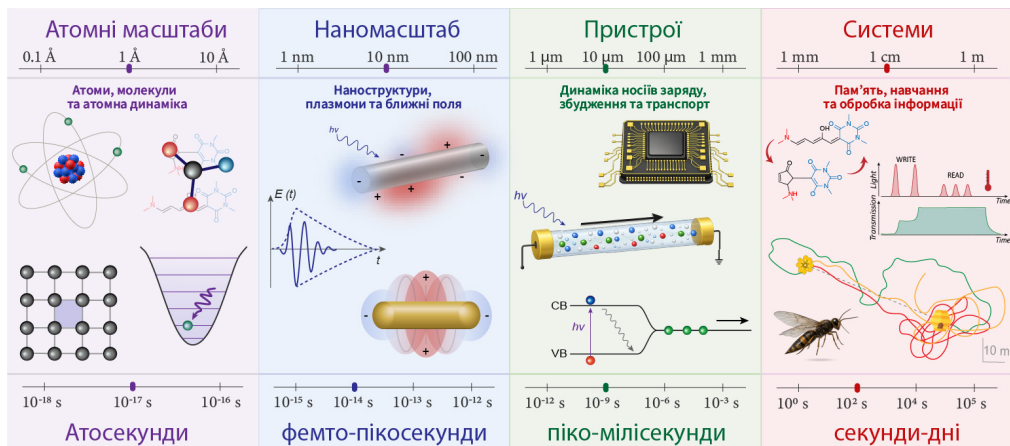


**Рис. 1:** Схематичний огляд масштабів довжини та часу, що є релевантними для цієї дисертації: від електронної динаміки на атомному та молекулярному рівнях, через надшвидкі плазмонні явища в наноструктурах, до транспорту носіїв заряду, пам'яті, а також фундаментальних фізичних процесів і обробки інформації у функціональних оптоелектронних та нейроморфних системах.

Рисунок 1 ілюструє широкий діапазон масштабів довжини та часу, охоплених у цій дисертації. Хоча досліджувані системи відрізняються своєю фізичною реалізацією, їх об'єднує спільна тема: розуміння того, як світло взаємодіє з речовиною в надзвичайно широкому діапазоні просторових і часових масштабів.

Дослідження, представлені тут, охоплюють понад п'ятнадцять порядків величини за часом – від електронної динаміки, що розгортається протягом фемтосекунд, до молекулярних ефектів пам'яті, які зберігаються протягом секунд, хвилин або довше. На найкоротших часових масштабах надшвидка фотоемісійна електронна мікроскопія дозволяє візуалізувати, як оптична енергія концентрується в металевих і напівпровідникових наноструктурах та перетворюється на електронний рух. На значно довгих часових масштабах фоточутливі молекули DASA, інтегровані з напівпровідниками, демонструють, як світло може змінювати властивості матеріалів, зберігати інформацію та створювати адаптивні реакції, що нагадують біологічні синапси.

Разом ці дослідження показують, як взаємодія світла і матерії може призводити як до фундаментальних відкриттів, так і до нових технологічних можливостей. Знання, отримані завдяки дослідженню надшвидкої електронної динаміки, можуть допомогти спрямувати розробку майбутніх оптоелектронних пристроїв, тоді як надроти в поєднанні з молекулами створюють платформу для адаптивного сенсорного аналізу, оптичної пам'яті та нейроморфної обробки інформації.



# List of publications

Publications included in this thesis:

- I **Ultrafast near-field dynamics in silver nanowires driven by few-cycle short-wave infrared pulses**  
**Nelia Zaiats\***, Lukas Wittenbecher, Ivan Sytceвич, Anne-Lise Viotti, Chen Guo, Chandni Babu, Xiaolu Zhuo, Luis M. Liz-Marzán, Eduardo J. C. Dias, F. Javier García de Abajo, Jan Vogelsang, Anne L’Huillier, Cord Arnold, Anders Mikkelsen  
*ACS Photonics* 2026, 13(8), 2248–2259
- II **Ultrafast dynamics of InAs nanowires with epitaxial Al-rings studied by TR-PEEM**  
Mahesh Saini, Saurabh Bokar, Thomas K. Jensen, **Nelia Zaiats**, Joachim E. Sestoft, Jesper Nygård, Donatas Zigmantas, Anders Mikkelsen  
*Manuscript in writing*
- III **Integrating molecular photoswitch memory with nanoscale optoelectronics for neuromorphic computing**  
David Alcer\*, **Nelia Zaiats\***, Thomas K. Jensen\*, Abbey M. Philip, Evripidis Gkani- as, Nils Ceberg, Abhijit Das, Vidar Flodgren, Stanley Heinze, Magnus T. Borgström, Barbara Webb, Bo W. Laursen, Anders Mikkelsen  
\*These authors contributed equally to the work  
*Communications Materials* 6, 11 (2025)
- IV **All-optical molecular synapses with wavelength-selectivity on III-V nanowire devices**  
**Nelia Zaiats\***, Xinyu He\*, Thomas K. Jensen, Abbey M. Philip, Bo W. Laursen, Anders Mikkelsen  
\*These authors contributed equally to the work  
*Manuscript in writing*

Publications not included in this thesis:

- V **Ultrafast photoemission from InP nanowires mediated by local optical field enhancement**  
Nedjma Ouahioune, Mahesh Saini, Emil Östberg, Zhen Zhao, Thomas K. Jensen, Caroline Juliano, Nelia Zaiats, Mathieu Gisselbrecht, Chen Guo, Anne-Lise Viotti, Cord L. Arnold, Claudio Verdozzi, Anders Mikkelsen  
*Manuscript in writing*
- VI **Ion migration and redox reactions in axial heterojunction perovskite  $\text{CsPb}(\text{Br}_{1-x}\text{Cl}_x)_3$  nanowire devices revealed by operando nanofocused X-ray photoelectron spectroscopy**  
Yen-Po Liu, Nils Lamers, Zhaojun Zhang, Nelia Zaiats, Anders Mikkelsen, Jesper Wallentin, Regina Dittmann, Rainer Timm  
*ACS Nano* 2024, 18(51), 34763–34775



# Abbreviations

|           |                                                    |
|-----------|----------------------------------------------------|
| AFM       | Atomic Force Microscopy                            |
| Ag        | Silver                                             |
| Al        | Aluminum                                           |
| ARPES     | Angle-Resolved Photoemission Electron Spectroscopy |
| Au        | Gold                                               |
| CB        | Conduction Band                                    |
| CEP       | Carrier-Envelope Phase                             |
| DASA      | Donor–Acceptor Stenhouse Adduct                    |
| DLD       | Delay-Line Detector                                |
| EBL       | Electron Beam Lithography                          |
| EQE       | External Quantum Efficiency                        |
| $E_F$     | Fermi Energy                                       |
| $E_{VAC}$ | Vacuum Level                                       |
| FDTD      | Finite-Difference Time-Domain                      |
| FWHM      | Full Width at Half Maximum                         |
| HHG       | High Harmonic Generation                           |
| IAC       | Interferometric Autocorrelation                    |
| InAs      | Indium Arsenide                                    |
| InP       | Indium Phosphide                                   |
| IR        | Infrared                                           |
| ITO       | Indium Tin Oxide                                   |
| LSP       | Localized Surface Plasmon                          |
| MCP       | Microchannel Plate                                 |
| MOVPE     | Metal–Organic Vapor Phase Epitaxy                  |
| NIR       | Near Infrared                                      |
| NOPA      | Noncollinear Optical Parametric Amplifier          |
| OPA       | Optical Parametric Amplifier                       |
| PAMS      | Poly( $\alpha$ -methylstyrene)                     |
| PEEM      | Photoemission Electron Microscopy                  |
| SEM       | Scanning Electron Microscopy                       |
| SHG       | Second Harmonic Generation                         |
| Si        | Silicon                                            |
| $SiO_2$   | Silicon Dioxide                                    |
| SPP       | Surface Plasmon Polariton                          |
| STM       | Scanning Tunneling Microscopy                      |
| SWIR      | Short-Wavelength Infrared                          |
| TEM       | Transmission Electron Microscopy                   |
| THG       | Third Harmonic Generation                          |
| ToF-PEEM  | Time-of-Flight Photoemission Electron Microscopy   |
| TR-PEEM   | Time-Resolved Photoemission Electron Microscopy    |
| UHV       | Ultra-High Vacuum                                  |
| UV        | Ultraviolet                                        |
| VB        | Valence Band                                       |
| XUV       | Extreme Ultraviolet                                |
| ZnO       | Zinc Oxide                                         |
| III–V     | Group III–V Semiconductors                         |



# Introduction

## 1.1 Technological motivation and the drive toward nanoscale systems

Electrons are fundamental carriers of charge and energy in matter, determining the electrical, optical, and chemical properties of materials and forming the basis of modern electronics. Understanding and controlling electron behavior in solids has enabled applications ranging from semiconductors and integrated circuits, to modern computing, communication, and energy conversion. Yet the continuous downscaling of devices to nanometer scales over the last decades is now reaching its limits in terms of energy dissipation, speed, and control over material properties.

For optoelectronic devices, such as solar cells and photodetectors, ultrafast carrier relaxation and recombination can also further reduce efficiency. At length scales approaching atomic dimensions, surface and interface effects become increasingly important, and quantum phenomena increasingly govern material behavior beyond the limits of bulk properties.

These challenges have led to the development of new device architectures and a deeper understanding of electron dynamics at the nanometer scale. For example, nanowire-based photodetectors and solar cells offer efficient light absorption and carrier collection<sup>1,2</sup>, and plasmonic nanostructures based on noble metals enable subwavelength confinement and enhancement of electromagnetic fields for sensing and nanoscale optics<sup>3,4</sup>.

At the same time, beyond improvements to existing technologies, there has also been growing interest in alternative models. One such approach is neuromorphic computing, a brain-inspired computing paradigm that designs hardware and software to mimic the neural and synaptic behavior of biological nervous systems. Neuromorphic computing aims to achieve information processing through novel adaptive, non-linear and history-dependent responses of the hardware. In this concept, insect brains, which achieve complex navigation and sensory processing with minimal energy, have directly motivated hardware implementation ranging from mixed-signal neuromorphic chips<sup>5</sup> to nanophotonic neurons based on optical networks<sup>6-8</sup>.

These challenges motivate the work presented in this thesis, which explores how light can be used both to investigate and to control processes in functional materials. The studies range from fundamental light-driven dynamics in nanoscale structures to hybrid molecular systems capable of adaptive and memory-like behavior.

## 1.2 The role of light as a probe and excitation tool in nanoscale research

Light provides a powerful tool for probing and manipulating electronic dynamics in matter, covering a broad range of energy and length scales that enable the efficient excitation and probing of electronic states, drive interband transitions, launch plasmons, and induce photoemission. The interaction of light with matter is one of the central objectives in modern physics. The observation of atomic spectra and the development of quantum mechanics established the quantized nature of electronic states in matter, and the invention of the laser revolutionized the study and control of light-matter interactions. More recently, experimental techniques such as ultrafast spectroscopy have enabled quantitative studies of carrier relaxation in semiconductors and energy transfer in molecular and biological systems in real time. Furthermore, optical techniques are non-invasive, making them particularly valuable for studying nanoscale systems where direct probing may perturb or even destroy the system.

A central challenge in nanoscale science is that many relevant processes occur simultaneously on nanometre length scales and femtosecond time scales. Conventional optical techniques provide excellent temporal resolution but are diffraction limited in space, whereas electron microscopy offers nanometre spatial resolution but is often insensitive to ultrafast dynamics. Among the experimental techniques available to probe ultrafast phenomena, photoemission electron microscopy (PEEM) stands out for its ability to combine high spatial and temporal resolution. By imaging electrons emitted from a material under optical excitation, PEEM achieves nanometer-scale spatial resolution while preserving femtosecond resolution<sup>9–11</sup>.

Beyond its role as a probe, light also provides a means to control and manipulate matter<sup>12–15</sup>. Time-resolved photoemission electron microscopy (TR-PEEM) combines both capabilities by imaging photoelectrons emitted from a surface following optical excitation. As a result, TR-PEEM using ultrafast light sources sheds light on the excitation dynamics of carriers, plasmons, and local optical fields<sup>10–12</sup>. For example, interferometric three-photon PEEM using 7 fs pulses has mapped plasmon-mode beating and coupling in complex nanostructures, demonstrating temporal control of near-field distributions<sup>16</sup>. Similar experiments on bowtie nanoantennas have visualized evolving localized surface plasmon modes and shown that orthogonally polarized pulses can phase-control optical near fields<sup>15</sup>. Extensions to attosecond extreme-ultraviolet (XUV) pulses combined with carrier-envelope-phase tagging have pushed PEEM into the attosecond regime, enabling direct probing of electron dynamics and plasmonic fields at metal surfaces<sup>17</sup>. These studies illustrate how time-resolved PEEM provides a unique window into the excitation dynamics of charge carriers, plasmons, and local optical fields, revealing the fundamental processes that govern nanoscale systems.

## 1.3 Developments in hybrid nanostructures

Advances in nanofabrication have permitted the generation of individual nanostructures with desired optical and electronic properties. For instance, semiconductor nanowires are efficient at absorbing light and transporting charge carriers, making them attractive for optoelectronic applications such as photodetectors and solar cells<sup>1,2</sup>. Their geometry combines a high optical confinement factor with a sensitivity to surface effects, making them useful for nanoscale material studies. Metallic plasmonic nanostructures, meanwhile, can strongly localize and

enhance electromagnetic fields, thereby increasing the efficiency of optical excitation, emission, and other light-driven processes<sup>18,19</sup>. These capabilities make plasmonic systems well suited for investigating ultrafast phenomena and a wide range of optical processes at the nanoscale.

Beyond individual materials, hybrid nanostructures that combine different material classes have emerged as a powerful platform for tailoring optical and electronic properties. Integration of plasmonic metals with semiconductors, where localized electromagnetic field enhancement provided by the metal can be combined with efficient carrier generation and transport in the semiconductor, offers new opportunities for controlling dynamics at the nanoscale.

More recently, hybrid systems that integrate molecular functionality with nanostructures have also attracted considerable attention, allowing additional control over material properties on the nanometer scale. Photochromic molecules such as donor–acceptor Stenhouse adducts (DASAs) undergo reversible changes in their optical properties upon illumination, enabling photoswitchable control over light absorption<sup>20,21</sup>.

Beyond providing optical functionality, hybrid nanowire–molecule systems are also attractive for neuromorphic computing, where device operation is inspired by biological concepts. Biological information processing is based on networks of interconnected neurons that communicate through synapses. Neurons receive and integrate signals from many neighboring neurons before generating an electrical output, and synapses regulate the strength of communication between neurons. Synaptic transmission can be modified by previous activity (synaptic plasticity), and in this way biological neural networks are able to learn and store information. Reproducing these adaptive functions is one of the central goals of neuromorphic computing.

When integrated with nanowire optoelectronics, DASAs can act as synaptic weights that exhibit reversible wavelength switching, thermal reset, and memory dynamics spanning timescales from sub-seconds to days<sup>21</sup>. In hybrid nanowire–molecule systems, this effect can be used to decouple the optical control of a molecular layer from the electrical readout of the nanowire, where the molecular layer modulates the light available to the nanowire, which in turn generates an electrical signal dependent on the incident illumination. The decoupling of “writing” (optical control) and “reading” (electrical response) introduces a level of functional complexity not present in conventional devices.

By incorporating molecules with different spectral and temporal characteristics, wavelength- and time-dependent responses, as well as switching, can be realized. These hybrid systems provide a platform for exploring new device concepts that combine optical control, electronic readout, and dynamic functionality<sup>22</sup>. Many of these concepts are inspired by biological neural systems, which achieve robust sensing, memory, and decision-making while consuming orders of magnitude less energy than modern computing hardware. In particular, insect brains perform complex tasks such as navigation, visual processing, and learning using only a small number of neurons and limited power. These capabilities motivate the development of neuromorphic hardware that reproduces selected biological functionalities using physical devices rather than conventional digital computation.

Despite their promise, hybrid nanostructures that combine photonic, plasmonic, molecular, or biological components remain challenging because their optical responses often occur on different length scales, time scales, and wavelength ranges<sup>3,23–25</sup>. Nonetheless, integrating different material classes can combine functionalities that are difficult to achieve within a single platform, enabling enhanced light–matter interactions, dynamic optical control, and adaptive device behaviour. As a result, hybrid systems have emerged as promising candidates for next-generation sensing, optoelectronic, and neuromorphic technologies<sup>23,26,27</sup>.

## 1.4 Structure of the thesis

This thesis investigates light-driven electronic processes in nanoscale systems, combining fundamental studies with device-oriented applications. It covers both the underlying physical principles and experimental techniques required to understand and control electron dynamics at the nanoscale.

Chapter 2 sketches the theoretical background relevant for the thesis, starting from the electronic band structure and extending to properties and fabrication of nanostructures. Thereafter, the light and its interaction with the electronic structures are discussed, focusing on the light properties relevant for this work and the potential of light as an experimental tool.

The experimental methods used in this thesis are presented in Chapter 3. First, the basic characterization of optical, electrical, and structural characteristics is explained. The main experimental technique, PEEM is introduced. This includes a description of the basic working principle, the principle of the time-resolved photoemission electron microscope and possible alternative modes. Sample preparation and limitations of the technique are also covered.

Main results obtained with the time-resolved PEEM technique on silver nanowires are presented in chapter 4, in which the experimental setup, the data analysis, as well as the plasmonic and electronic dynamics are introduced and compared with simulations. Furthermore, the chapter presents the study of semiconductor-metal nanowire systems.

Chapter 5 considers device-based implementation, in particular for hybrid devices based on semiconductor nanowires functionalized with photochromic molecules, specifically DASAs. It reviews these devices in terms of optically controlled devices and neuromorphic computing.

Lastly, Chapter 6 summarizes the findings of the thesis and presents suggestions for future work.

# Concepts and methods

Electrons are fundamental constituents of matter that determine many of the electronic, optical, and chemical properties of materials<sup>28,29</sup>. Studying the electronic properties of materials therefore remains an active area of research, as it provides insight into fundamental processes such as charge transport, chemical bonding, and light-matter interaction.

This section introduces the key physical concepts required to describe electronic structure, light-matter interaction, and nanoscale systems relevant to this work. The interaction of light with matter and the resulting electronic excitation and emission processes are discussed. In addition, the basic principles underlying one-, two-, and three-dimensional nanoscale structures are introduced, together with key concepts in nanostructure fabrication<sup>28,30</sup>.

In this thesis, particular focus is placed on III-V semiconductors such as indium arsenide (InAs), which exhibit a direct band gap and high electron mobility, making them highly suitable for optoelectronic and ultrafast studies<sup>30-32</sup>. In addition, plasmonic metals such as silver are employed to generate localized electromagnetic fields<sup>19,33</sup>, while organic photochromic molecules, in particular donor-acceptor Stenhouse adducts (DASAs), provide tunable optical absorption and, when integrated with semiconductor devices, enable coupling between optical excitation and electrical readout<sup>20,21</sup>. These hybrid material platforms combine semiconductor physics, plasmonics, and molecular functionality, providing new opportunities for controlling light-matter interactions and realizing adaptive optoelectronic devices<sup>21,22,34</sup>.

## 2.1 Materials and their physical properties

All solids consist of atoms, which in turn are made up of subatomic particles: the atomic nuclei with neutrons and protons, with electrons bound to the nuclei in discrete quantum states. While the nuclei constitute almost all of the mass, it is the electrons that primarily determine how atoms bond to each other, forming materials, and how those materials respond to external electric fields and light<sup>28,35</sup>.

In a single atom, electrons occupy well-defined energy states on their orbitals around the nucleus bound by its positive charge<sup>36,37</sup>. When atoms form a solid, the overlap of their electronic wavefunctions leads to the formation of states giving rise to energy bands<sup>35,38</sup>. The resulting electronic band structure with forbidden energy regions determines many of the electrical and optical properties of the material.

At thermal equilibrium, electrons occupy the available energy states, filling the lowest-energy states first. The highest occupied energy at absolute zero temperature is defined as the Fermi

energy ( $E_F$ )<sup>35,39</sup>. At finite temperature, the Fermi level defines the energy at which the probability of occupation is 50%.

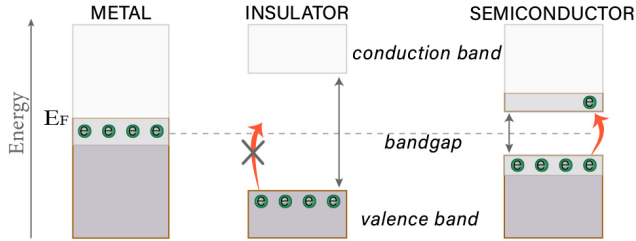


**Figure 2.1:** Schematic of band structures for metals, insulators, and semiconductors. Highlighted are the relative positions of the valence band, conduction band, and bandgap<sup>28,35</sup>.

A simplified representation of the electronic band structure for metals, semiconductors, and insulators is shown in Figure 2.1. In semiconductors and insulators, the valence band is separated from the conduction band by a bandgap. Above the conduction band lies the vacuum level, which represents the energy ( $E_{VAC}$ ) an electron must have to escape from the material into the vacuum. The energy difference between the Fermi level and the vacuum defines the work function of the material. Additionally, another important value is the binding energy, which describes how far an electronic state lies below the Fermi level and indicates how strongly the electron is bound within the material. Together, work function and binding energy determine the total energy needed for an electron to escape the material and, as a result, be imaged with PEEM.

The band arrangement shown in Figure 2.1 provides a simple picture that helps to distinguish different classes of materials<sup>28,35,40</sup>. In metals, the highest occupied band is partially filled, or the Fermi level lies within a band, allowing electrons to move freely and giving rise to electrical conductivity. In insulators, electron motion is restricted because occupied and empty states are separated by a large energy gap. Between these two extremes lie semiconductors, where a smaller bandgap allows a finite number of electrons to be thermally excited into the conduction band. In addition, impurities or dopants introduce shallow energy levels near the band edges, further increasing the number of charge carriers.

Those materials classes are described in a bit more detail further, as differences in band structure determine how materials respond to applied electrical and optical excitation, forming the basis for modern electronic devices.

### 2.1.1 Metals

Metallic bonding is collective rather than directional, meaning that the valence electrons are not localized between specific pairs of atoms but are shared throughout the crystal. This delocalization of electrons gives rise to characteristic metallic properties such as electrical conductivity.

This picture is often described as a “sea of electrons”. The positively charged atomic cores (nuclei with tightly bound inner electrons) form a periodic background, while the valence

electrons are delocalized, shared among atoms. A fraction of these electrons occupy partly filled states at the Fermi level and behave as free charge carriers<sup>33,41</sup>. The free electrons can then move through the material under an applied electric field. This collective electronic behavior will also be important later when discussing plasmonic excitations.

From the band-structure perspective, metals are distinguished by the absence of a bandgap at the Fermi energy. This situation is often described in textbooks in two equivalent ways: either the highest occupied band is only partially filled, or several bands overlap in energy around the Fermi level<sup>28,35</sup>. In both cases, there are available empty electronic states at energies close to the Fermi level. It is therefore appropriate to describe metals as having a sequence of energy bands, with lower-energy bands fully filled and higher-energy bands partially occupied, and the Fermi energy in a metal lies inside a band. At absolute zero temperature, states below the Fermi level are occupied, and states above are empty, but at finite temperatures the occupation changes gradually around the Fermi level. Because empty states exist arbitrarily close to the Fermi level, electrons can be accelerated by an external electric field, giving rise to electrical conduction.

In this work, metallic nanostructures (e.g. silver) are used to support plasmonic excitations, which couple strongly to an incident optical field due to the collective oscillation of conduction electrons (plasmons). The resulting near-field enhancement drives nonlinear photoemission processes that can be directly visualized with PEEM.

### 2.1.2 Insulators

Insulators, unlike metals, are materials in which electronic motion is strongly restricted by their electronic band structure. In an insulator, a large bandgap separates the valence and conduction bands. This energy gap is typically several eV wide, making thermal excitation of electrons across the gap negligible at room temperature.

This can be better understood by first considering the metal, for example aluminum, in which each atom contributes valence electrons to form bonds, producing a partially filled energy band with available empty states that allow electrons to move freely<sup>28,35</sup>. Oxygen, on the other hand, has atoms that strongly attract valence electrons from metals, forming stable, tight bonds<sup>42</sup>. So when aluminum combines with oxygen to form  $\text{Al}_2\text{O}_3$ , valence electrons become localized in bonding states formed between Al and O atoms<sup>42</sup>. Those bonds fully occupy low-energy bonding states, at the same time opening a large bandgap and promoting insulating behavior. For this reason, random oxidation caused by exposure to air is often undesirable in electronic applications and difficult to avoid.

Typical solid insulators are first of all oxides such as silicon dioxide ( $\text{SiO}_2$ ), aluminum oxide ( $\text{Al}_2\text{O}_3$ ), and magnesium oxide ( $\text{MgO}$ ), which play an important role in modern electronics<sup>39,42</sup> by supplying electrical insulation and controlling electric fields and charge transport at interfaces.

### 2.1.3 Semiconductors: doping and the p-n junction

As discussed earlier, semiconductors are materials that have electrical properties between metals and insulators, consisting of a valence band, conduction band, and a bandgap in

the range of approximately 0.5–3 eV<sup>43,44</sup>. However, some materials exhibit significantly larger bandgaps and are often referred to as wide-bandgap semiconductors<sup>45,46</sup>, such as GaN (~3.4 eV)<sup>45,47</sup>. In these materials, thermal excitation of electrons across the bandgap at room temperature is negligible, and electrical conductivity is therefore primarily achieved through doping that is described in more detail later.

When electrons are promoted to the conduction band, they leave behind positively charged holes in the valence band. The electrical conductivity of semiconductors depends on both electron and hole transport and can be expressed as

$$\sigma = q(n\mu_n + p\mu_p), \quad (1)$$

where  $n$  and  $p$  are the electron and hole concentrations, and  $\mu_n$  and  $\mu_p$  are their respective mobilities<sup>28,48</sup>.

These carrier concentrations depend on how easily electrons can cross the bandgap, and thermal energy also plays a role in this process. As temperature increases, a larger fraction of electrons can access higher-energy states, leading to an increase in conductivity. This reflects the temperature-dependent occupation of electronic states near the band edges.

### *Doping and charge carrier control*

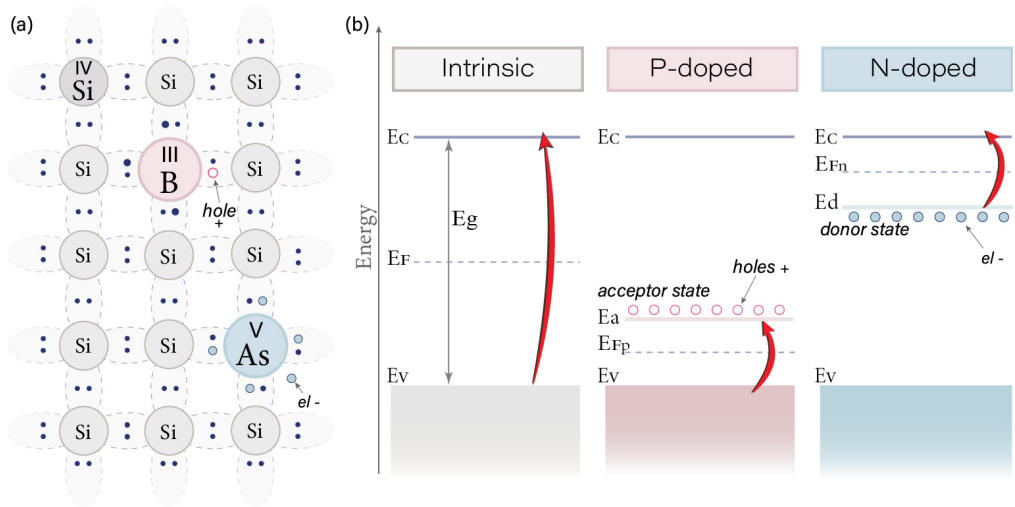


**Figure 2.2:** Doping and charge carrier control in semiconductors. (a) Intrinsic Si atoms with four valence electrons, doping with group III (B) and group V (As) atoms, creating holes and electrons as majority carriers. (b) Energy band diagrams of intrinsic, p-doped, and n-doped semiconductors showing acceptor and donor states and the corresponding shift of the Fermi level.

While intrinsic conductivity is limited, the electrical properties of semiconductors can be modified very effectively through doping. In other words, by introducing atoms of another material, the carrier concentration, Fermi level position, and conductivity can be engineered. In a

broader sense, in materials with large bandgaps, doping can introduce additional localized states within the bandgap, which may contribute to electrical conductivity if these states are sufficiently close to the band edges.

A basic example is silicon, which belongs to group IV of the periodic table<sup>28,42</sup>, meaning that its atoms have four valence electrons. When silicon is doped with a group III element, for example boron, that replaces a silicon atom, one covalent bond is missing an electron. This creates an unfilled bonding state and, as a result, an acceptor level slightly above the valence band. An electron from the valence band can occupy this acceptor level, leaving a mobile hole behind. As a result, holes become the majority charge carriers and the material becomes p-type (p for positive). On the contrary, if silicon is doped with a group V element, such as arsenic, an extra valence electron is introduced. This electron occupies a donor level close to the conduction band and can be thermally excited into it<sup>39,48</sup>. The material then becomes n-type (for negative), with electrons as the majority carriers. The schematic representation of this process is illustrated in Figure 2.2.

### *PN junction and diode operation*

Doping shifts the Fermi level toward the valence band in p-type material and toward the conduction band in n-type material, while the bandgap itself remains the same. When those p-type and n-type regions are brought together, a p–n junction is then formed. In this configuration, electrons diffuse from the n-side to the p-side and holes diffuse in the opposite direction. Near the interface, electrons and holes recombine. This creates a region without mobile charge carriers, known as the depletion region<sup>39,49</sup>. The remaining ionized donor and acceptor atoms create an internal electric field that opposes further diffusion.

The p–n junction is the basis of the diode, the simplest type of electronic component. The electrical characteristics of a p–n junction, relating the current flowing through the junction to the applied voltage, are described by the diode equation<sup>28,39</sup>:

$$I = I_0 \left( e^{\frac{eV}{n k_B T}} - 1 \right), \quad (2)$$

where  $I_0$  is the reverse saturation current,  $V$  is the applied voltage,  $k_B$  is Boltzmann’s constant,  $T$  is the temperature, and  $n$  is the ideality factor accounting for recombination and other non-ideal effects<sup>39,49</sup>.

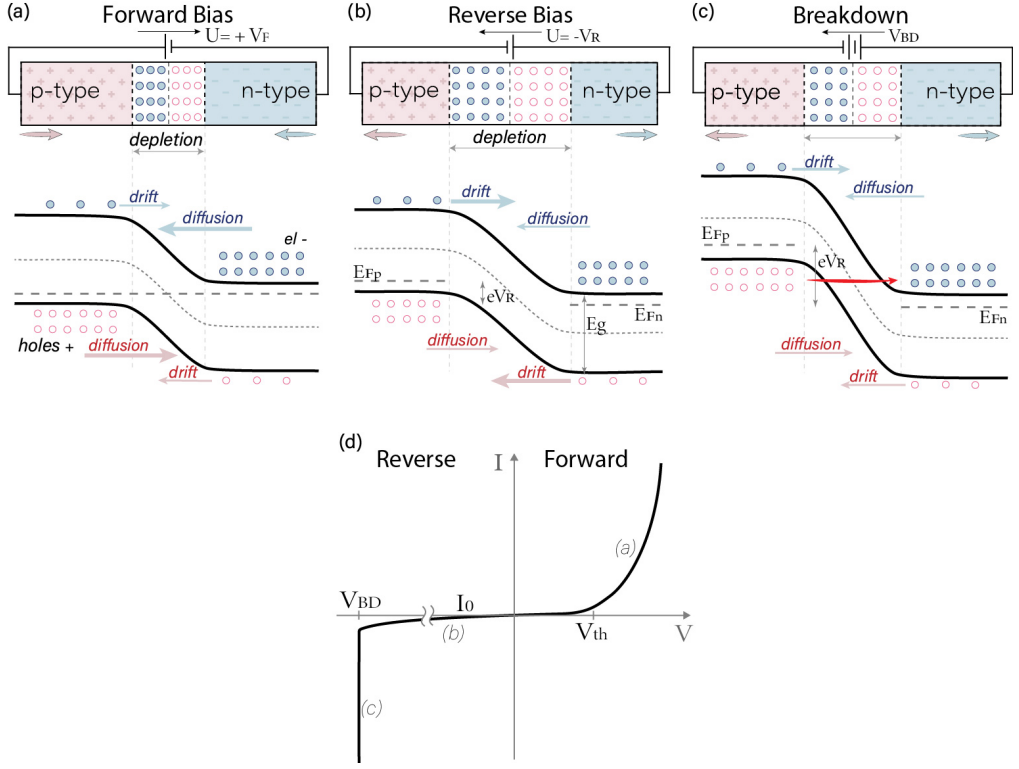


**Figure 2.3:** Schematic of a p-n junction operation under different bias conditions. (a) Forward bias, where the potential barrier is reduced and the depletion region narrows, allowing carriers to diffuse across the junction and recombine. (b) Reverse bias: the potential barrier increases and the depletion region widens, suppressing carrier transport and leaving only a small reverse drift current. (c) Breakdown regime: a strong electric field leads to carrier multiplication through tunneling or impact ionization. (d) Corresponding current–voltage characteristic of the diode showing exponential current increase in forward bias, a small reverse saturation current, and breakdown at  $V_{BD}$ .

When there is no external voltage applied, the diffusion of charge carriers across the junction is balanced by the internal electric field in the depletion region, so current won't flow. Applying an external voltage disturbs this balance and leads to charge accumulation on both sides and diode operation.

When a forward bias is applied (Figure 2.3a), the p-side is connected to a positive potential and the n-side to a negative potential. This reduces the built-in potential barrier and enables charge carriers to be injected across the junction, leading to an increased carrier concentration near the interface. In this case, the applied voltage reduces the potential barrier and narrows the depletion region. As a result, charge carriers can cross the junction more easily and recombine near the interface. This leads to an exponential increase in current with applied voltage, as described by the diode equation.

This recombination can occur via radiative recombination, in which an electron from the conduction band recombines with a hole in the valence band, emitting a photon, which is the

basis of LEDs<sup>50–52</sup>. Or non-radiative recombination<sup>53,54</sup> occurring through defect states or phonon interaction with energy released as heat.

In contrast, when a reverse bias is applied (Figure 2.3b), meaning that the p-side is connected to a negative potential and the n-side to a positive potential, the applied electric field causes an increase in the potential barrier and widening of the depletion region, as majority carriers are pulled away from the junction, while thermally generated minority carriers drift across the depletion region. It suppresses the transport of the carriers across the junction<sup>39,49</sup>. Only a minor reverse current,  $I_0$ , remains from thermally generated minority carriers, which can be swept across the depletion region by the electric field.

This behavior of the reverse-biased junction is closely related to the solar cell’s working principle<sup>55,56</sup>. When light with a photon energy greater than the bandgap is absorbed, it can excite electrons from the valence band to the conduction band, creating electron–hole pairs. In the depletion region, the built-in electric field separates carriers, driving electrons to the n-side and holes to the p-side, thereby producing the measurable photocurrent. Under short-circuit conditions, when the applied voltage is zero, the generated carriers can still flow through the circuit, creating short-circuit current.

If the reverse bias is increased further (Figure 2.3c), the electric field in the depletion region becomes strong enough to cause breakdown. Depending on the junction properties, this occurs either through tunneling (Zener breakdown)<sup>57</sup> or through impact ionization (avalanche breakdown)<sup>56,58</sup>, leading to a sharp increase in reverse current. This behavior is reflected in the diode current–voltage characteristic shown in Figure 2.3d.

### Bandgap control

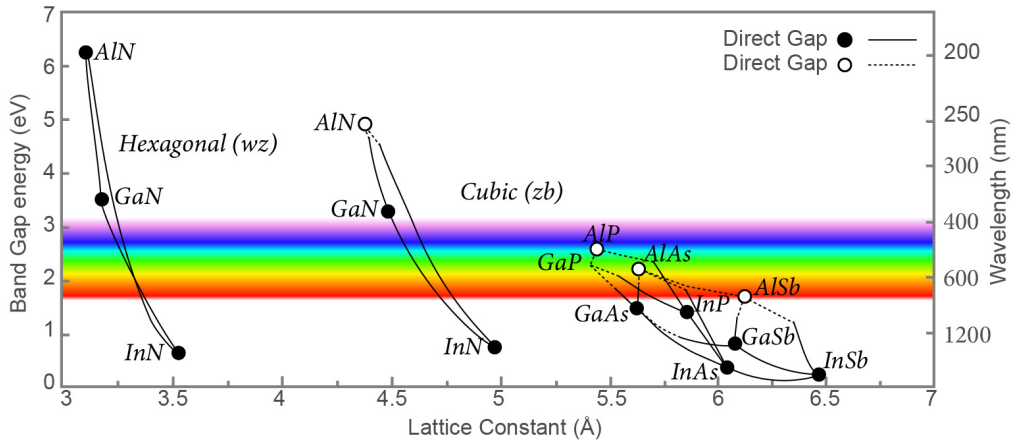


**Figure 2.4:** Bandgap energies of common III–V semiconductors as a function of lattice constant, adapted from<sup>31,45</sup>. The colored region marks the visible spectrum, illustrating how bandgap engineering allows tuning of emission wavelength for optoelectronic devices such as LEDs and lasers.

The bandgap of a semiconductor is determined by its material composition and crystal structure. The lattice constant together with chemistry, orbital energies, and crystal structure defines the bandgap (Figure 2.4). Two processes that are also mentioned earlier in more detail

are particularly important. The first is the generation of electron–hole pairs as a result of photon absorption, which forms the basis of photodetectors and solar cells. The second is radiative electron–hole recombination, which is used in light-emitting diodes and lasers.

The energy of the emitted photon is determined by the semiconductor bandgap:

$$E_g = h\nu = \frac{hc}{\lambda}, \quad (3)$$

where  $h$  is Planck’s constant,  $\nu$ ,  $c$ , and  $\lambda$  are the photon frequency, speed of light, and photon wavelength, respectively. As a result, the color of the emitted light is linked to the bandgap energy. Smaller bandgaps correspond to lower-energy photons (red or infrared light), and larger bandgaps correspond to higher-energy photons (blue or UV light). For visible light emission, bandgaps in the range of approximately 2–3 eV are required.

As discussed above, the bandgap also determines optical absorption. Photons with energies equal to or larger than the bandgap can be absorbed, while lower-energy photons are transmitted. For example, silicon, with a bandgap of about 1.12 eV<sup>59,60</sup>, absorbs a large part of the visible spectrum, and therefore to the human eye a silicon wafer appears dark. In contrast, wide-bandgap materials such as gallium nitride (GaN, with a bandgap of approximately 3.4 eV) are transparent in the visible spectrum<sup>31,47</sup>.

Depending on the band structure, the efficiency of optical transitions varies significantly. In indirect bandgap semiconductors, such as silicon, optical transitions require additional phonons to resolve the momentum mismatch between the nearest opposite energy levels, reducing efficiency. As a result, silicon is widely used in electronics and solar cells, but not as an efficient light emitter. For light-emitting applications, direct bandgap semiconductors, such as gallium arsenide (GaAs), gallium nitride (GaN), and indium phosphide (InP), are preferred<sup>30,31,52,61,62</sup>, as they allow electrons and holes to recombine directly without phonon assistance.

The electronic and optical properties discussed above ultimately originate from the atomic arrangement of the semiconductor. Therefore, it is important to consider the crystal structure of these materials.

### *Crystal structure*

Semiconductors used in electronic and optoelectronic devices crystallize in well-defined lattice structures. Silicon, for example, adopts a diamond cubic structure<sup>35,42</sup>, where each atom forms covalent bonds with four nearest neighbors, as also illustrated in Figure 2.2. Compound semiconductors such as GaAs, GaN or InAs typically form zinc blende (ZB) or wurtzite (WZ) structures<sup>63–65</sup>, which are closely related but consist of two different atomic elements occupying the crystal lattice.

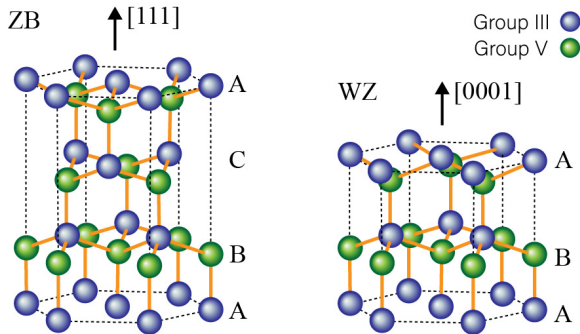


**Figure 2.5:** Zinc blende (ZB) and wurtzite (WZ) crystal structures for III–V semiconductors. The two phases share tetrahedral orientation but differ in symmetry and stacking sequence (ABC for ZB along (111), AB for WZ along (0001))<sup>66</sup>.

Both ZB and WZ structures imaged in Figure 2.5 consist of tetrahedrally coordinated atoms, where each atom is bonded to four nearest neighbors<sup>67</sup>. The main difference lies in the crystal symmetry. In zinc blende, the crystal has a cubic symmetry with atoms arranged in a face-centered cubic lattice, stacked in an ABC sequence along the (111) direction, while wurtzite exhibits a hexagonal symmetry with an AB stacking sequence along the (0001) direction<sup>66</sup>.

Although the local bonding geometry is similar, the difference in crystal symmetry leads to distinct electronic properties. For instance, the bandgap of InP varies between the ZB and WZ phases by approximately 80–90 meV<sup>68–70</sup>. In addition, the lack of inversion symmetry in the wurtzite structure gives rise to spontaneous polarization and piezoelectric effects, which can strongly influence carrier separation and recombination<sup>71,72</sup>. In contrast, zinc blende structures, which are centrosymmetric, typically do not exhibit such internal fields. At the nanoscale, for instance within nanowires, both crystal phases can be manufactured within a single structure by selective control of growth parameters<sup>12,73</sup>, allowing tuning of the electronic properties.

In nanowire geometries, such as those studied in this work, both zinc blende and wurtzite phases can coexist within a single structure. This phase mixing provides an additional degree of freedom in tuning the electronic and optical properties, influencing carrier behavior. As a result, crystal structure engineering is an important parameter to consider when interpreting the experimental observations presented in this thesis.

## 2.2 Nanostructures and fabrication principles

The reduction of dimensionality and nanoscale structuring of materials changes the coupling of light and matter, altering electromagnetic local field confinement, as explored in this work using plasmonic nanostructures<sup>74</sup> and semiconductor nanowires. Nanostructuring provides additional control by shaping electromagnetic fields, confining electronic motion, and introducing interfaces, which are central to the light-driven dynamics in nanowires and hybrid nanostructures presented in this thesis.

Crystal structure, as discussed earlier, is one important factor influencing excitation processes. For example, in the study by Wittenbecher *et al.*<sup>12</sup>, clear differences in excitation between the WZ and ZB crystal structures are observed. The crystal structure determines the electronic band dispersion and symmetry, which influences carrier scattering and excitation pathways, which, in turn, influence carrier dynamics and excitation efficiency.

Geometrical confinement determines the localization and resonance conditions of surface plasmons, especially in the presence of sharp edges, curvatures, and small gaps between metallic surfaces<sup>33,75</sup>. In these cases, the electromagnetic fields are confined within small volumes, which directly influences nonlinear excitation processes and emission efficiency.

### 2.2.1 Surfaces and thin films (2D)

Thin films are the simplest examples of dimensional reduction. Such quasi-two-dimensional systems retain a bulk-like electronic structure, but due to their small thickness, surface and interface effects become more prominent<sup>76,77</sup>. Thin films<sup>11,78,79</sup> and flakes<sup>79,80</sup> can serve as model systems for studying plasmon propagation, near-field enhancement, and ultrafast carrier dynamics, which are directly visualized in PEEM experiments. Because PEEM detects photoelectrons that escape from only the near-surface region, thin films are particularly suitable for connecting surface structure, interfaces, and photoemission response<sup>10,12</sup>.

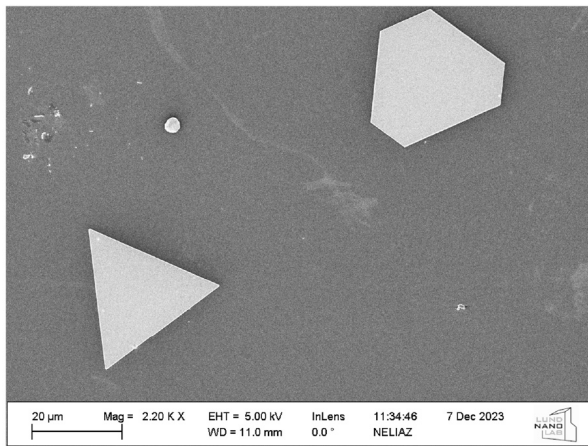


**Figure 2.6:** Gold nanoflake as an example of a two-dimensional plasmonic structure recorded with SEM.

Very similar to thin films are monolayered heterostructures in which several layers of crystals are stacked on top of each other. Examples include metallic nanoflakes, which combine lateral extension with nanoscale thickness. In this case, the excitation can occur at one edge and can propagate across a flat surface, allowing plasmonic and carrier dynamics to be spatially resolved using PEEM<sup>81,82</sup> (Figure 2.6).

Thin films are commonly fabricated using physical vapor deposition (PVD) techniques<sup>83</sup>. PVD enables precise control of film thickness and allows the deposition of smooth, homogeneous

layers. In this process, a source material, for example gold, is heated under high vacuum until it evaporates. The evaporated atoms travel ballistically through the vacuum and condense onto a cooler substrate, forming a thin film. The mean free path of the evaporated atoms is larger than the dimensions of the chamber under high vacuum, so scattering in the gas phase is negligible, and the atoms are deposited in a directional manner with the monitored film thickness. By varying the deposition rate, substrate temperature, and vacuum, the morphology and quality of the film (grain size, surface roughness, and film continuity) can be tuned<sup>84,85</sup>.

To introduce lateral structuring, create hybrid nanostructures, or enable electrical connection, thin films can be combined with lithographic patterning. Lithography enables the definition of nanoscale features on continuous films, allowing spatial control over both the material composition and the geometry of the structured surface<sup>86,87</sup>. In a typical lithography process, a resist is applied to a substrate and then selectively exposed to light in photolithography or to a focused electron beam in electron-beam lithography (EBL), resulting in a chemical modification of the resist at the exposed regions.

Once the desired pattern is formed in the resist, additional manufacturing steps, such as metal deposition or etching, are performed. In this case, metal is deposited on both the exposed areas and the resist pattern. Lift-off consists of removing the resist, thereby leaving the metal deposited directly on the substrate, which allows the fabrication of fine, well-structured metal components and patterns with good spatial accuracy.

Lithography techniques can be divided into fast photolithography for large-area patterning through a mask, and slower but higher-resolution techniques such as EBL, where a focused electron beam is used to directly write patterns into the resist. In contrast to photolithography, EBL does not require a mask and allows higher spatial resolution, as the pattern is defined by scanning the beam point-by-point<sup>87</sup>. However, this makes the process significantly slower. Despite this limitation, EBL enables the fabrication of nanoscale features with high precision, which is essential for defining individual nanowire devices and hybrid nanostructures studied in this work.

## 2.2.2 Nanowires (1D)

Perhaps the most used nanostructures in this work are nanowires, which represent one-dimensional systems with a high aspect ratio. In this thesis, semiconductor and metal nanowires, particularly silver<sup>88,89</sup> and indium arsenide, are used as model systems to investigate light-driven carrier dynamics. Their one-dimensional geometry leads to strong polarization-dependent absorption and enhanced optical confinement<sup>90–92</sup>. This makes them highly suitable for studying light-driven carrier dynamics.

Furthermore, semiconductor nanowires provide efficient absorption of incident light, which can be directly converted into an electrical signal<sup>93</sup>. These nanowires (in our case (Ga)InP) provide a versatile platform for integrating additional functional elements, such as molecular layers, enabling hybrid systems where optical signals can be modulated before being converted into electrical responses by nanowires.

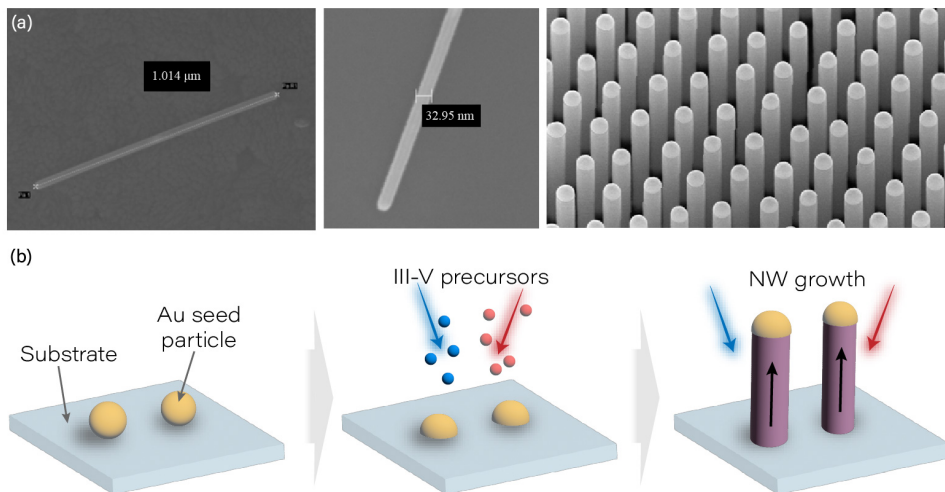


**Figure 2.7:** Nanowires as model one-dimensional structures and their growth mechanism. (a) Electron microscopy images of silver nanowires and InP arrays<sup>93</sup>. (b) Schematic of nanowire growth via metal-organic vapor phase epitaxy (MOVPE). Gold seed particles deposited on a substrate collect decomposed III-V precursors, and supersaturation of the catalyst particle leads to nanowire growth along a crystallographic direction<sup>94,95</sup>.

Nanowires are very robust structures, which can be of different lengths and diameters, freestanding<sup>89</sup> or embedded on a two-dimensional surface<sup>96</sup>, as part of an array or as single structures, made of different materials<sup>88,97</sup>, including III-V semiconductors<sup>12,93</sup> and metals<sup>88,89</sup>. Due to their elongated shape and high surface-to-volume ratio, nanowires exhibit polarization-dependent light absorption, and surface states and band bending strongly affect carrier dynamics and photoemission.

Nanowires can be grown in different ways (top-down by etching, through masks, or chemically, as for silver nanowires in Figure 2.7a). Among these, metal-organic vapor phase epitaxy (MOVPE), illustrated in more detail in Figure 2.7b, is the most commonly used. It provides excellent control over nanowire diameter, length, and composition. By adjusting precursor flow, pressure and temperature, it is possible to control the crystal structure and even switch between wurtzite and zinc blende phases within the same nanowire<sup>73</sup>.

First, preparation for the growth requires lithography. This is used to define the position of catalyst particles, most commonly gold, which act as seeds for nanowire growth. Lithography can be performed using electron-beam lithography (EBL), where a resist layer is patterned with high spatial resolution, followed by metal deposition and lift-off to create well-defined arrays of catalyst particles<sup>87</sup>. The size of these particles determines the nanowire diameter, while their position defines the spatial arrangement of the array.

On the next step, metal-organic precursors containing the required elements for the nanowire are introduced into a heated reactor chamber. There, the precursors decompose at elevated temperatures and the constituent atoms dissolve into the liquid catalyst particle. Once the catalyst becomes supersaturated with the precursor material, crystal growth begins at the

interface between the catalyst particle and the substrate. The nanowire then elongates along a specific crystallographic direction through the continuous supply of precursor material<sup>73,98</sup>. The length of the nanowire can be controlled by adjusting the growth time, while its composition can be tuned by varying the precursor flow rates.

For electrical measurements, additional processing steps are required. After growth, the gold catalyst particle can either be removed or incorporated into the device, depending on the intended design, and the nanowires can be coated with dielectric or conductive layers to enable electrical contacting. Electrical contacts are then typically fabricated using lithography followed by metal deposition and lift-off. Finally, wire bonding or probe contacting is used to connect the fabricated device to external measurement electronics.

For PEEM measurements, isolated and well-defined nanowires are required. Since as-grown nanowires are often arranged in dense arrays, additional processing is necessary to separate or transfer individual structures onto suitable substrates. The simplest approach is mechanical transfer, in which nanowires are dispersed onto a substrate using a dry transfer technique. Although this method is straightforward and fast, the resulting nanowire positions and orientations are random.

A more controlled placement method is achieved using micromanipulation techniques, where individual nanowires are picked up using a sharp probe or micromanipulator tip and positioned at predefined locations on a new substrate. This approach enables deterministic placement and precise control of nanowire orientation. Depending on the experimental setup, the manipulation can be performed under an optical microscope or inside a scanning electron microscope (SEM). Although this method provides excellent control over the final device geometry, it is considerably more time-consuming than simple mechanical transfer.

Another approach consists of prefabricating the receiving substrate with alignment structures, such as trenches or guiding patterns, that facilitate nanowire positioning during transfer. By sweeping nanowires across the patterned surface, individual nanowires can become aligned and trapped within these structures, providing a compromise between deterministic placement and fabrication throughput<sup>96</sup>.

In some cases, nanowires are supplied in liquid suspension, as is common for chemically synthesized silver nanowires used in PEEM measurements. For single-nanowire studies, the suspension is typically diluted to reduce the nanowire density. A clean substrate is then brought into contact with a droplet of the diluted suspension for a short period, followed by rinsing and drying. This procedure provides a simple and reliable method for depositing isolated nanowires, although the final distribution remains largely random and can only be influenced to a limited extent by parameters such as substrate inclination during drying.

In addition to standard growth and lithographic patterning of thin films, nanowires and molecules, advanced fabrication techniques such as atomic layer deposition<sup>99</sup>, directed assembly of nanowires<sup>96,100</sup>, integration with embedded waveguide materials<sup>93</sup>, and functionalization with photochromic layers enable increasingly complex nanophotonic architectures.

### 2.2.3 Molecular systems as 0D absorbers and device elements

As part of this work, more complex hybrid structures are made using additional molecular patterning. In particular, DASA dyes are then used as representative molecular absorbers.

While they provide strong, tunable optical responses, the direct electrical readout of molecular switching is limited. That is why a good strategy is to incorporate photochromic molecules into semiconductor nanowires that convert optical signals into electrical signals.

Unlike nanowires with their diameters on the order of tens to hundreds of nanometers, DASA molecules are only a few nanometers ( $\sim 1\text{--}2\text{ nm}$ ) long. This large difference in scale allows the molecular layer to be distributed around and between nanowires, forming a nanoscale coating that interacts with light at the molecular level before the light hits the nanostructures. In this hybrid device system, nanowires act as efficient light-absorbing structures that couple to the transmitted light by DASAs and convert it into measurable photocurrents.

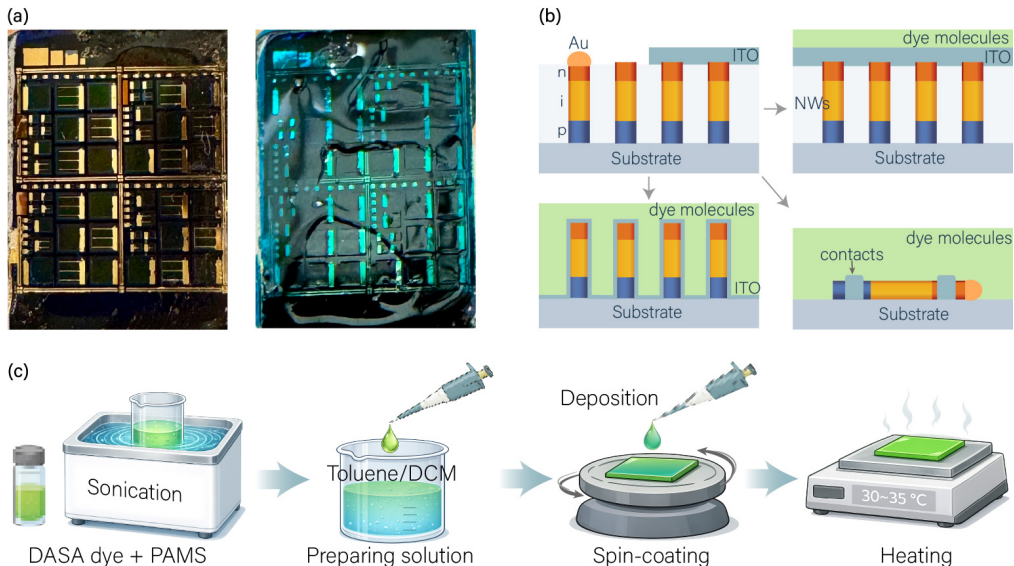


**Figure 2.8:** Schematic illustration of hybrid nanowire-molecule devices. (a) DASA dye sample preparation process involving sonication of dye-based solution, spin coating and heating. (b) Semiconductor nanowires coated with a molecular layer of photochromic DASA dyes incorporated in the polymer. (c) Alternative dye molecules depositions on top, around nanowires or in separate single-nanowire devices.

Due to their direct bandgap, high carrier mobility, and strong optical confinement, nanowires efficiently absorb light and generate photocurrent<sup>93</sup>. The molecular layer acts as an optically programmable filter that controls the amount of light reaching the nanowires. In the colored open form, DASA molecules strongly absorb visible light, reducing the optical flux transmitted to the nanowires and therefore decreasing the generated photocurrent. When exposed to wavelength-selective light, the molecules bleach into the closed form<sup>20</sup>, and the higher amount of light passing through the nanowires increases the photocurrent. Thus, the layer of molecules can be used like an optically tunable filter, while the layer of semiconductor nanowires can be used like a converter of an optical signal into an electrical signal, which can be integrated together in the form of arrays or even single nanowires (Figure 2.8a–b). This separation of optical control and electronic readout is a key concept explored in parts of this thesis.

Molecular layers are prepared using solution-based methods such as drop-casting or spin-coating. In this work, DASA dyes in solid form are dissolved in organic solvents (toluene and

dichloromethane) and incorporated into a polymer matrix (poly( $\alpha$ -methylstyrene), PAMS) to ensure distribution and separation of the molecules and to define how easily the molecules can change shape. After sonication to ensure a homogeneous solution is obtained, it is pipetted using a micropipette onto a clean substrate or a prefabricated nanowire device (Figure 2.8c).

More uniform films of controllable thickness can be produced using spin-coating techniques, in which the sample is rotated during deposition to control film thickness. The spin speed was optimized to about 2000 rpm to provide a suitable thickness for the experiments. Alternatively, films can be deposited by simple drop-casting, resulting in thicker films, although with less control over uniformity.

After deposition, these films are often annealed at moderate temperatures (typically 30–35 °C) for 5 minutes to remove residual solvent and stabilize molecules. Care must be taken, as heating above this temperature will usually result in evaporation of solvent and instability of the film<sup>20,21,101</sup>. The samples are stored in the dark to avoid inadvertent photo-switching before the measurements are made, allowing reproducible layer formation with defined optical properties on nanowire-based devices.

## 2.3 Key characteristics of light

While the previous subsections focused on the role of electrons in determining material properties, light, in the form of photons, provides the means to excite, probe, and control these electronic dynamics. Its interaction with materials depends critically on photon energy  $E$  (or equivalently wavelength  $\lambda$ ), temporal structure characterized by the pulse duration  $\tau$ , and the electric field strength  $E_0$ .

In this thesis, different spectral regions are employed to access distinct physical processes. SWIR femtosecond pulses are used to drive strong-field and multiphoton excitation in nanostructures, visible light is used to control molecular switching in DASA systems, and UV excitation is used to induce photoemission for PEEM imaging.

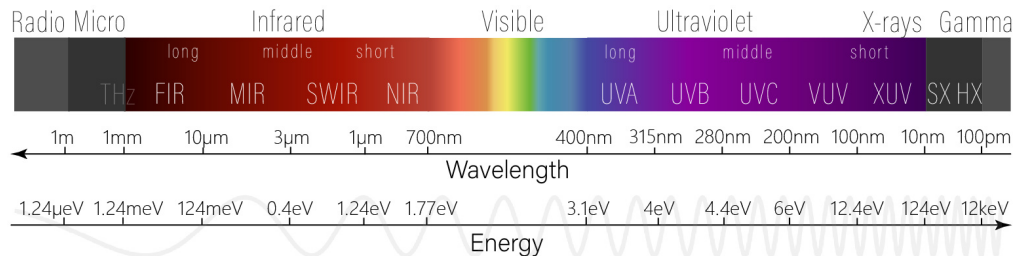


**Figure 2.9:** Electromagnetic spectrum showing the relationship between wavelength and photon energy. Wavelength decreases from left to right while photon energy and frequency increase.

The wavelength  $\lambda$  and frequency  $\nu$  of light are related through  $c = \lambda\nu$  in vacuum. In the material, the speed of light is affected by the refractive index  $n$ :

$$\frac{c}{n} = \lambda\nu. \quad (4)$$

From a quantum mechanics perspective, light can also be described as a flow of photons, each carrying energy given by

$$E = h\nu, \quad (5)$$

where  $h$  is Planck's constant. This relation establishes a connection between the wave-like description of light (frequency) and its particle-like behavior (discrete energy transfer), as shown in Figure 2.9. Infrared and terahertz radiation typically couple to low-energy excitations such as phonons<sup>102–104</sup>. Visible and near-infrared light typically drives interband transitions<sup>105,106</sup>, leading to absorptions of light and photocurrent generation, and can efficiently excite localized surface plasmons in metallic nanostructures. UV and extreme-UV photons have high energy that helps to probe electronic properties by direct photoemission<sup>107–109</sup>, which are commonly provided by a synchrotron radiation source or can also be realized through high-harmonic generation (HHG), which converts intense ultrafast IR laser pulses into extreme-UV radiation.

### 2.3.1 Ultrafast light

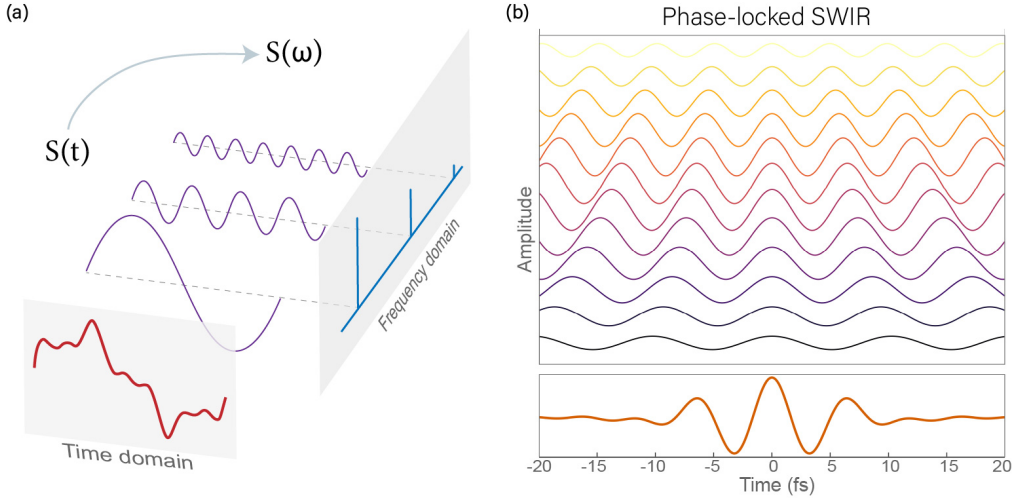


**Figure 2.10:** Time-frequency representation of an ultrafast pulse. (a) Illustration of the Fourier-transform relationship between the time-domain electric field and its frequency-domain spectrum. (b) Time-dependent electric field of 12 different monochromatic waves with wavelengths in the SWIR range (top) and the sum of all the waves, corresponding to an ultrafast pulse with about 2 optical cycles.

Because the temporal and spectral characteristics of the field are related to each other through Fourier transforms<sup>110</sup>, which connects the time and frequency domains of the electromagnetic signal, the bandwidth  $\Delta\omega$  and pulse duration  $\Delta t$  are also dependent on each other.

In Figure 2.10a, the time- and energy-domain representations of an ultrashort pulse are shown, illustrating that the generation of a few-cycle pulse requires a large wavelength range. Besides, Figure 2.10b shows how ultrashort pulses in the SWIR range form from locked modes that interfere in time. In the extreme case of few-cycle pulses, such as a two-cycle pulse, the pulse duration approaches only a few oscillations of the electric field itself<sup>111</sup>. These pulses therefore require a very broad spectral bandwidth, and in this regime the carrier-envelope phase (CEP), defined as the phase offset between the peak of the pulse envelope and underlying electric field oscillations, becomes important, as it affects the instantaneous electric-field waveform even though the pulse envelope remains the same<sup>112,113</sup>. The electric field of such pulses is strongly time-localized, resulting in very high peak fields even at moderate pulse energies.

The ultrafast laser pulses considered here are used to access highly nonequilibrium electronic states and to induce strong-field or multiphoton photoemission processes, realized with few-cycle laser pulses, which are central to the PEEM experiments presented in this thesis.

### 2.3.2 Photoemission

When light interacts with electrons, the electromagnetic field transfers energy to electrons, causing photoemission, in which an electron gains sufficient energy to escape from a surface into the vacuum.

To leave the material, the electron must overcome the surface barrier, which is the potential energy barrier at the material-vacuum interface, defined by the difference between the Fermi level and the vacuum level (work function). Photoemission can occur through several mechanisms and depends on photon energy, pulse intensity, and electric field strength.

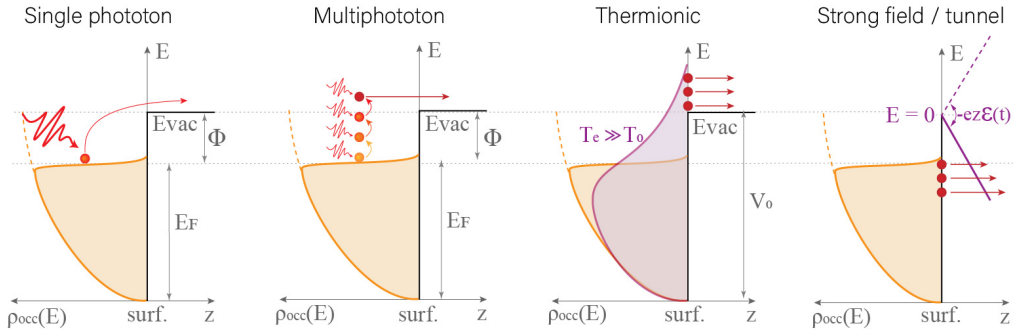


**Figure 2.11:** Schematic overview of photoemission mechanisms. From left to right: single-photon photoemission, multiphoton photoemission, thermally assisted emission, and strong-field photoemission<sup>114,115</sup>.  $\rho_{occ}$  represents the density of states,  $z$  the vacuum distance away from the metal surface. The barrier height  $V_0$ , the Fermi level  $E_F$ , and the work function  $\Phi$ .

Photoemission regimes, shown in Figure 2.11, include multiphoton photoemission, thermally assisted emission, and strong-field tunneling emission<sup>114,115</sup>. In direct one-photon photoemission, an electron absorbs one single photon. If the photon energy is greater than the work function, the electron can directly overcome the surface barrier and be emitted into the vacuum.

Under strong optical excitation, the electronic system can also be driven out of equilibrium, leading to heating of the electron distribution and the population of higher-energy states, from which electrons can overcome the surface barrier. As a result, some electrons reach energies above the emission threshold and escape into the vacuum.

In the case of very strong electric fields, the surface potential itself becomes distorted by the optical field. As a result, the barrier width is reduced, and electrons can tunnel through it even if their energy is below the nominal work function<sup>116</sup>. This strong-field or tunneling regime is typically associated with intense, ultrafast laser pulses and represents a transition from photon-driven to field-driven emission<sup>117</sup>.

Depending on the excitation conditions, multiple photoemission mechanisms can contribute simultaneously, and the observed signal reflects a combination of these processes, as well as material-specific properties such as band structure, surface states, and local-field enhancement.

### 2.3.3 Excitation of surface plasmons

Since, as described earlier, metals have a high density of free electrons, when an external field is applied, they can move over short distances on fast time scales. Because electrons are negatively charged, they are pushed in the direction opposite to the applied field. In this way the external field induces local collective oscillations of groups of electrons, leading to plasmonic excitation<sup>41,118</sup> such as localized and propagating surface plasmon polaritons (LSPPs and SPPs). Such excitations lead to strong near-field enhancement, which is especially important for applications in nanoscale optics and ultrafast photoemission experiments<sup>9,118–121</sup>.

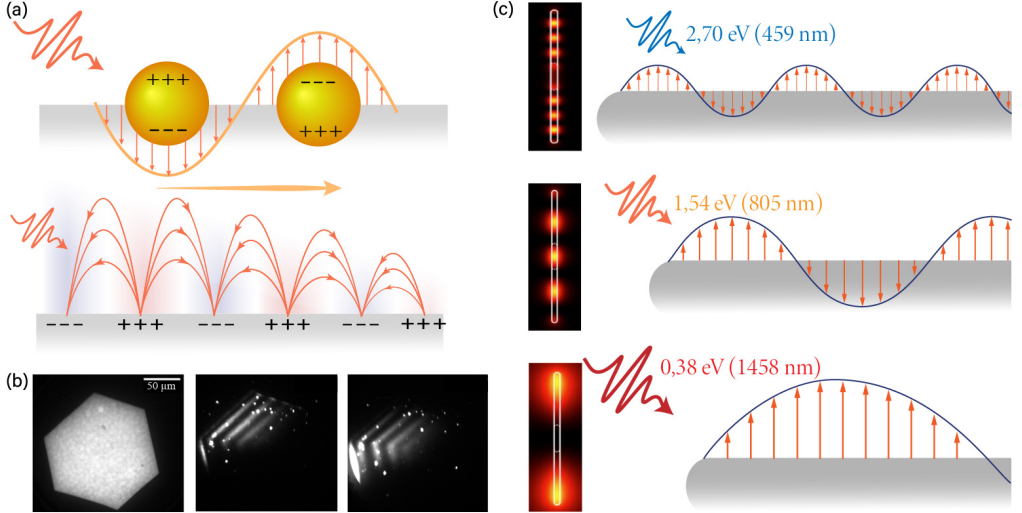


**Figure 2.12:** Excitation of surface plasmons in metallic nanostructures. (a) Collective oscillation of conduction electrons driven by an external electromagnetic field. Bottom: example images of a metallic nanoflake, recorded with PEEM, illustrating spatially resolved plasmon excitation on a flat surface under UV and IR illumination. (b) Size-dependent plasmon resonances showing how the excitation wavelength and nanoparticle geometry determine the resonance energy, with strongly enhanced near-fields localized at the surface<sup>122,123</sup>.

Plasmons are generated when the external electric field drives collective oscillations of conduction electrons. Because the positive ion background is fixed, it pulls electrons back, leading to a wave-like collective movement of the groups of electrons (Figure 2.12a). In a 1-D small nanoparticle, electrons just oscillate back and forth, and this motion creates regions of charge accumulation on the sides of the particle.

For larger dimensions, such as metallic nanoflakes, a similar oscillation propagates on the surface, with the electric field tightly confined near the metal–dielectric boundary in the form of SPPs. Such modes can exist only when the optical response of the metal, described by its dielectric function, differs sufficiently from the surrounding dielectric. Since the dielectric function is wavelength dependent, the excitation and confinement of SPPs are also strongly wavelength dependent<sup>124</sup>. Those strong near-fields are evanescent and decay exponentially away from the interface, with a characteristic decay length typically ranging from tens to hundreds of nanometers depending on the wavelength and dielectric environment<sup>124</sup>. To capture those fields, advanced techniques with nanometer spatial and femtosecond temporal resolutions are needed, making PEEM an ideal tool<sup>12,125</sup>. One of the most common plasmonic metals is gold, as shown in Figure 2.12b, where spatial patterns of plasmon excitation are observed on a nanoflake under IR.

Different photon energies lead to different spatial field distributions, ranging from localized excitation to more extended field patterns. This geometrical dependence becomes even more pronounced in one-dimensional systems such as nanowires, where the excitation is guided along the wire axis<sup>126</sup>. As shown in Figure 2.12c, plasmonic excitation in nanowires is strongly influenced by their length, diameter, and orientation, leading to characteristic spatial intensity distributions that can be resolved using PEEM.

Since plasmonic fields depend on the surface geometry, their properties are also influenced by the crystal lattices<sup>12,127</sup>. Different crystal lattices give rise to distinct band dispersions and anisotropies, making the lattice and nanostructure symmetries another important factor in determining the conduction and optical response.

### 2.3.4 Light interaction with molecules

When light interacts with a molecule, photons can be absorbed if their energy matches the difference between two molecular energy levels, promoting an electron from the ground state to an excited state and triggering a variety of photophysical or photochemical processes<sup>128–130</sup>. In many molecules, this excitation involves a redistribution of the electron density, which changes how electrons are shared across the molecule. This process modifies the forces between atoms and can boost structural changes. In some molecular systems, optical excitation can induce structural rearrangements that modify the molecular geometry and, consequently, its optical properties.

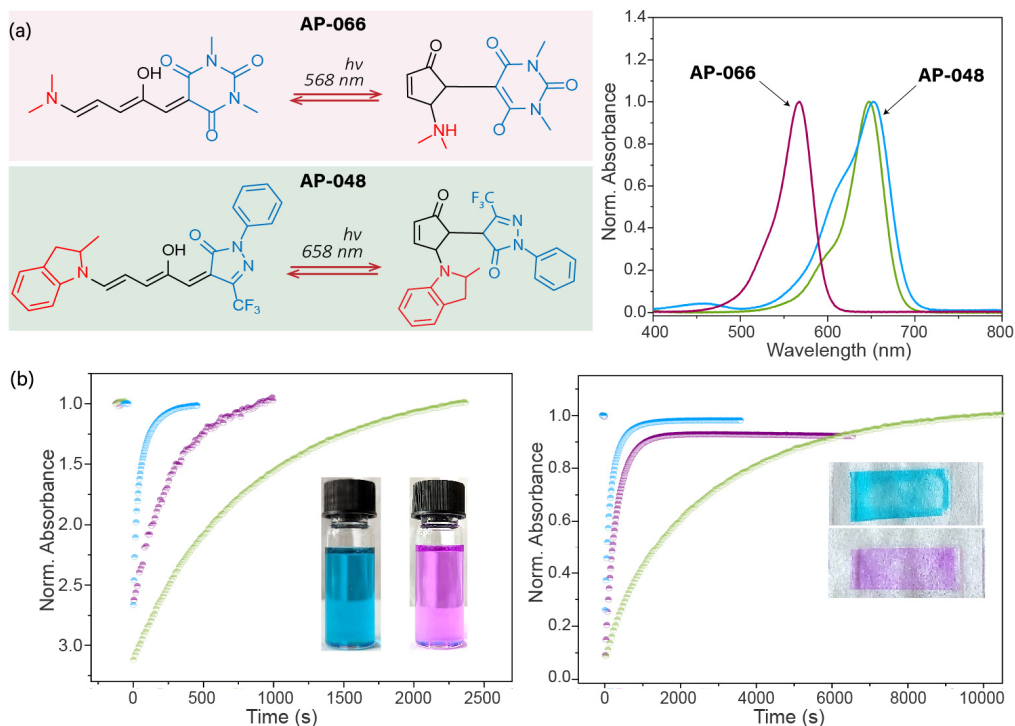


**Figure 2.13:** Photoswitching of DASA molecules. (a) Different molecular derivatives exhibit distinct absorption wavelengths (568 nm and 658 nm). (b) Switching kinetics of the dye in solution (toluene) and solid (PAMS).

A particularly interesting class of such molecules that are combined in devices described in the thesis are photochromic compounds, whose absorption spectrum changes reversibly under illumination. These molecules can switch between two structural configurations with different optical properties, allowing them to function as optically programmable absorbers or molecular switches, among which, DASAs<sup>20,131</sup>. DASA molecules exist in two stable configurations: a colored open form and a colorless closed form. When illuminated with light of corresponding wavelength (Figure 2.13a), photoexcitation in the open molecule redistributes electrons which alters the electronic distribution, weakens the atomic structure and triggers rearrangement that converts the molecule into its closed form and photobleaches it. When the illumination is removed, the closed form thermally relaxes back to the open configuration, restoring the original absorption spectrum. The process is therefore reversible and can be repeatedly cycled.

The spectral position of the absorption maximum and the recovery dynamics depend on the specific chemical structure of the DASA molecule. Different derivatives can therefore be designed to respond to different wavelengths of light and exhibit different relaxation times, resulting in exponential bleaching at varied speeds during illumination and exponential recovery after the light is removed. In addition, the surrounding environment, such as the host polymer or whether the molecules are in solid state or in solution, also affects the dynamics. For example, Figure 2.13b illustrates how the molecular dynamics differ depending on whether the molecules are embedded in a solid matrix or dissolved in a solution<sup>20,101,132</sup>. In solution,

the molecules can undergo structural rearrangements more easily, leading to faster switching, whereas in the solid state their motion is more restricted.

When combined with nanostructures, the natural separation of writing and reading mechanisms may allow the switching concept to be applied to neuromorphic device architectures that use optical signals to modulate synaptic weights, with neuron non-linear behavior transduced by semiconductor nanostructures. By incorporating multiple DASA derivatives with distinct absorption spectra into a single molecular layer, wavelength-selective control can be achieved within a single device. Furthermore, because different DASA molecules exhibit distinct recovery times, the temporal dynamics of the optical response can be tuned accordingly.



# Experimental techniques

While the development of new materials and nanostructures continues to widen the range of possible nanoscale systems, equally important are advances in experimental techniques that make it possible to observe and resolve their underlying dynamics. In this context, light is used both to excite electronic processes and to probe system response.

This chapter first briefly outlines some of the experimental techniques used to explore optically driven phenomena in hybrid nanostructures. A central technique is time-resolved photoemission electron microscopy (TR-PEEM) from FOCUS GmbH, which provides high spatial and temporal resolution under ultrafast excitation. Recent advances in ultrafast and nanoscale techniques enable access to processes on nanometer length scales and femtosecond timescales, which makes such approaches particularly powerful for studying nanoscale dynamics.

In addition, complementary techniques are employed, including optical, structural, and electrical characterization methods, providing further insight into the investigated systems. Finally, sample preparation strategies are introduced, with emphasis on PEEM-compatible samples and nanowire-molecule hybrid structures.

## 3.1 Optical and structural characterization methods

Before using PEEM to study ultrafast processes, the optical, electrical and structural characteristics of the system need to be characterized, as these experiments are essential to understand optical response and carrier dynamics. The methods are widely used in studies of nanostructures and molecular systems (such as dye molecules), where optical measurements are key to understanding absorption, switching, and relaxation dynamics.

### 3.1.1 Optical and electrical measurements

Optical and electrical measurements are used to characterize the interaction of light with the investigated systems and to link optical excitation to device response. Optical spectroscopy, including transmission, reflection, absorption, and scattering measurements, provides information on absorption, reflection, and scattering processes, which reveal electronic transitions and optical resonances. At the same time, electrical measurements allow direct probing of charge carrier generation and transport under illumination.

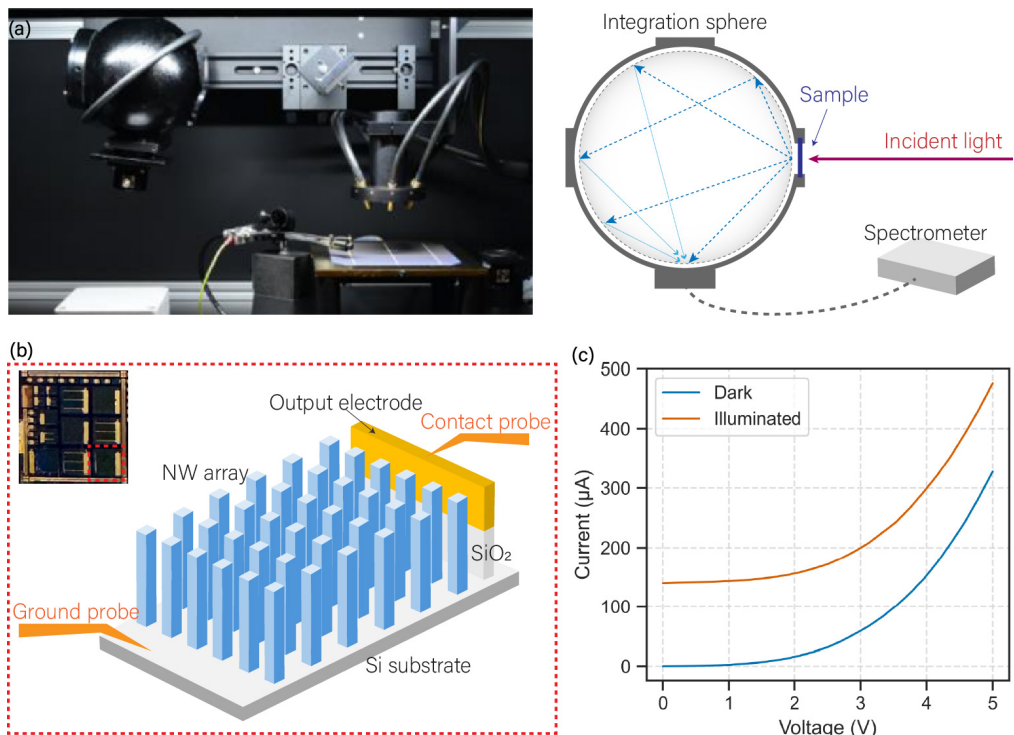


**Figure 3.1:** Optical and electrical characterization of samples. (a) Experimental setup and its schematic for optical measurements using an integrating sphere, where transmitted light in DASA-coated samples is collected by a spectrometer. (b) Image of the nanowire device and schematic of the contacted nanowire array used for electrical measurements. (c) Current-voltage characteristics of the nanowire array measured in the dark and under illumination, demonstrating the increase in current due to photocurrent generation.

A central and the simplest tool used for some optical measurements in this work is the integrating sphere (Figure 3.1a). It allows the collection of both directly transmitted and diffusely scattered light<sup>133</sup>. Its purpose is to measure the total optical transmission of a sample independently of the direction in which the transmitted light leaves the surface. The inner wall of the sphere is coated with a highly reflective material that repeatedly scatters the incoming light, and the detector measures the total transmitted optical power, including both directly transmitted and diffusely scattered light. This makes the integrating sphere particularly suitable for samples with scattering, rough surfaces, or non-uniform optical properties.

For studies involving DASA dyes, a sapphire substrate coated with a layer of the molecular dye is placed at the entrance port of the sphere, as shown in Figure 3.1a. The transmitted light is subsequently monitored as a function of wavelength and time. The entire optical response of the dye is obtained, including bleaching by illumination and recovery in darkness<sup>20</sup>. In addition, the switching behavior of the molecular system can be directly probed, providing information on the extent of bleaching and the relaxation processes during recovery.

To connect the optical response to device performance, the electrical characteristics of the

nanowire-based devices are measured under controlled illumination. An optical image of the device chip and a schematic of the contacted nanowire array are shown in Figure 3.1b. The nanowire array is contacted using probe needles, with one probe connected to the substrate and the other to the output electrode, allowing current-voltage measurements across the device.

A solar simulator is used as a programmable light source to provide stable and controllable illumination conditions with varied intensity and spectral composition. The electrical response of the nanowires to incident light can be measured, and for example, the diode current-voltage characteristics of the nanowire array can be defined (Figure 3.1c). Under illumination, a clear increase in current is observed, reflecting photocurrent generation in the nanowires.

To quantify the optical-to-electrical conversion efficiency further, external quantum efficiency (EQE) measurements<sup>55,56</sup> can be performed, which describe the number of collected charge carriers resulting from excitation and indicate how efficiently the system converts light into an electrical signal. With a similar approach to connecting the nanowire array with the probes as before, the photocurrent is recorded and compared to the incident optical power, and the EQE spectrum is extracted. This allows direct correlation between optical absorption features and electrical response and in the case of the hybrid nanowire-molecule systems, EQE measurements provide insight into how the molecular layer modifies the effective light reaching the nanowires and, consequently, the generated photocurrent.

### 3.1.2 Electron and scanning probe microscopy

Electron and scanning probe microscopy techniques are used to characterize the geometry, surface morphology, sample quality, and, depending on the specific technique, chemical composition and local physical properties in nanostructures. These measurements provide the structural information required to interpret the optical and photoemission experiments presented in this thesis.

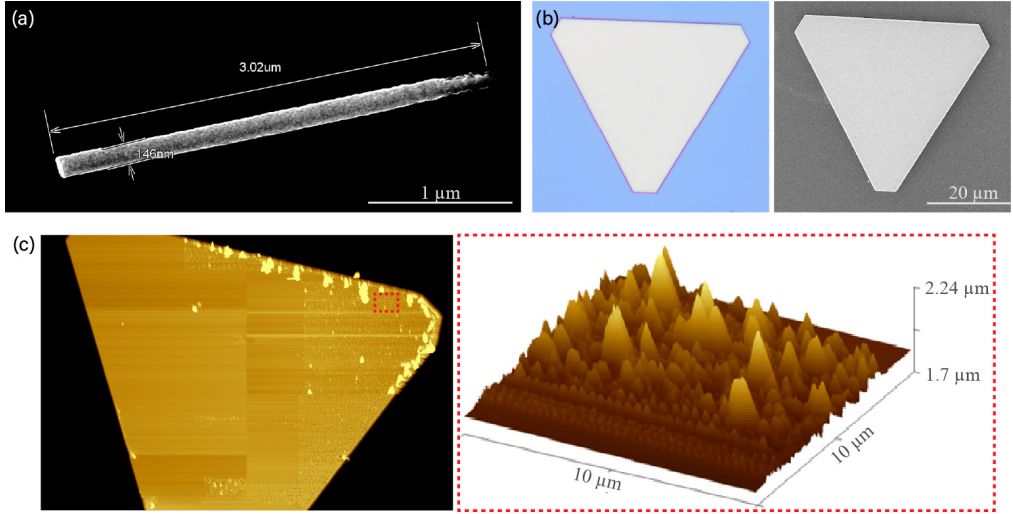


**Figure 3.2:** Electron and atomic force microscopy examples. (a) SEM on InP nanowire highlighting the nanowire size, (b) optical and SEM images of a gold flake, (c) AFM images of a gold flake with a zoomed-in area on defects.

Scanning electron microscopy (SEM) is the primary characterization tool used in this work prior to the main measurements. It is useful for imaging nanowire arrays and assessing their geometry, including diameter, length, and spatial distribution, as shown in Figure 3.2a-b. In the case of single nanowire samples, controlled placement and identification of individual structures is not trivial, and SEM plays an important role in locating and selecting suitable regions for further measurements.

Atomic force microscopy (AFM<sup>134</sup>) illustrated in Figure 3.2c is used as a complementary technique to probe surface morphology with high spatial resolution. In this work, AFM is particularly useful for detecting surface impurities and defects and assessing surface roughness, for example, in gold nanoflake samples. Since PEEM is highly surface sensitive, even small variations in surface structure can influence the measured signal, making AFM an important tool for sample evaluation.

Additional techniques, such as scanning tunneling microscopy (STM)<sup>135–137</sup>, Transmission electron microscopy (TEM)<sup>138–140</sup>, ellipsometry<sup>141–143</sup> and profilometry, can be used where relevant to determine atomic structure, thin film thickness and optical constants. All these parameters are important for understanding light absorption and propagation in the nanostructures and for correlating structural properties with optical and electrical response.

## 3.2 Photoemission electron microscopy

PEEM is a powerful surface-sensitive imaging technique for the visualization of electronic and optical properties of nanostructures. It does not rely on detecting light, but instead uses illumination of a material to induce the emission of electrons, which are then used to image

the surface and its dynamics.

The technique is based on the photoelectric effect: photons incident on the sample excite electrons, which are emitted from the surface and subsequently used to form an image. Because only electrons originating from the top few nanometers can escape into the vacuum due to inelastic scattering in the solid, PEEM is inherently sensitive to surface and near-surface phenomena<sup>144,145</sup>. This makes it suitable for studying nanostructures, thin films, and surface chemistry. PEEM is widely used to investigate a variety of phenomena, including optical near-field distributions in plasmonic nanostructures<sup>92,146–148</sup>, electronic structure and workfunction variations<sup>10,149,150</sup>, chemical composition<sup>108</sup>, and ultrafast carrier dynamics.

For example, in the scope of this thesis, PEEM has been used to image localized plasmonic fields in metallic nanostructures, such as silver nanowires and gold flakes, where confined plasmon modes and their propagation can be visualized. It has also been applied to more complex hybrid systems, such as InAs nanowires with Al rings, where the interplay between semiconductor and metallic components can be studied, as well as InP nanowires, as model semiconductors, where excitation with few-femtosecond UV pulses was used to access information about the energy distribution of the emitted electrons. In addition, by using X-ray excitation, PEEM allows access to chemically sensitive information, for example, to study the surface decomposition of layered structures (TiAlN) under thermal stress.

### 3.2.1 Working principle and electron optics

The working principle of the PEEM is based on the generation, acceleration, and imaging of photoemitted electrons. When the sample is illuminated with a suitable light source (e.g. UV or laser radiation), photoelectrons are emitted from the surface via the photoelectric effect. These electrons carry spatial information about where they originated on the sample.

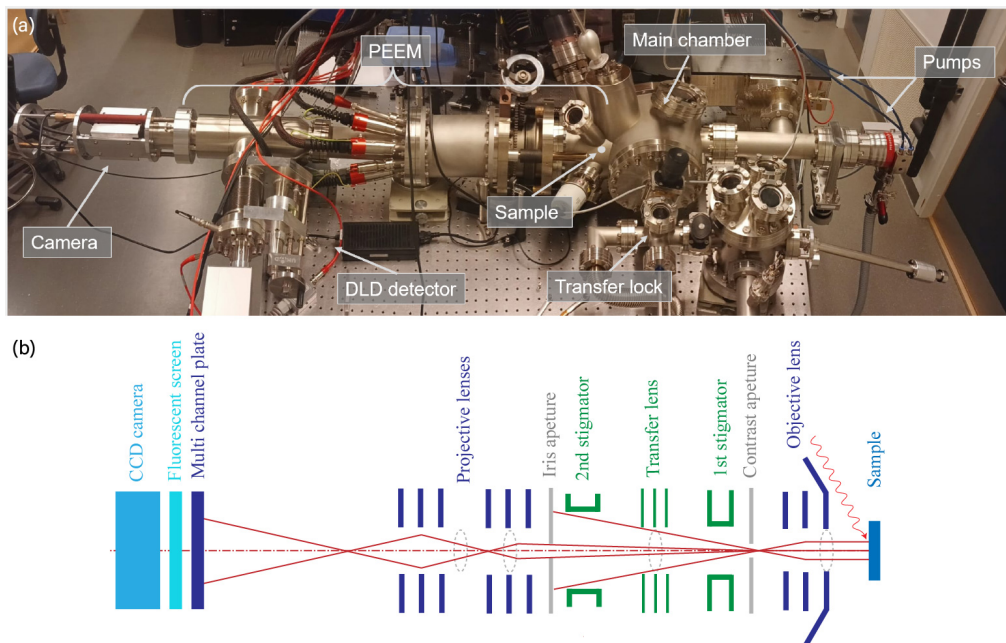


**Figure 3.3:** Photoemission electron microscope. (a) Photograph of the PEEM setup integrated with the vacuum system, showing the main chamber, sample position, transfer lock, pumps, and detection components. (b) Schematic of the PEEM electron optics, illustrating the electron trajectories from the sample through the objective lens, contrast aperture, transfer and projector lenses, to the detector (from Focus GmbH)<sup>151</sup>.

The PEEM, constructed and integrated on the laser table, is illustrated in Figure 3.3a. Several chambers and pumps are connected to create an ultrahigh vacuum (UHV) and to provide sample transfer and cleaning/annealing. A mercury lamp (Hg lamp with a wavelength up to 600nm) is incorporated to excite samples for reference pictures and focus adjustments, and a CCD camera at the end of the PEEM column images the collected electrons.

The magnification system in PEEM is based on electrostatic lenses, which control trajectories and focus the emitted electrons (Figure 3.3b)<sup>151,152</sup>. Unlike conventional optical lenses, which refract light, electrostatic lenses use electric fields to manipulate charged particles. This force in the electric field that electrons experience changes their velocity and direction, allowing them to be focused and guided along the PEEM column<sup>151</sup>.

The electrons are then guided through an electrostatic lens system consisting of an objective lens which operates in a strong extraction field close to the sample, where electrons are rapidly accelerated. This initial acceleration reduces the influence of their initial energy spread and improves spatial resolution. A strong electrostatic field is applied between the sample and the objective lens, which accelerates the emitted electrons to kinetic energies of typically 10–16 keV.

The subsequent lenses (transfer and projective lenses) further shape and magnify the electron distribution to form the final image. Behind the objective lens, a contrast aperture is inserted

to balance spatial resolution and signal intensity by blocking electrons with large angular spreads. Optical elements such as stigmators and deflectors are used to correct aberrations and align electron trajectories, thereby improving focus. Finally, the electron image is detected using a multichannel plate (MCP), where electrons are amplified, converted into visible light on a phosphor screen, and recorded with a CCD camera.

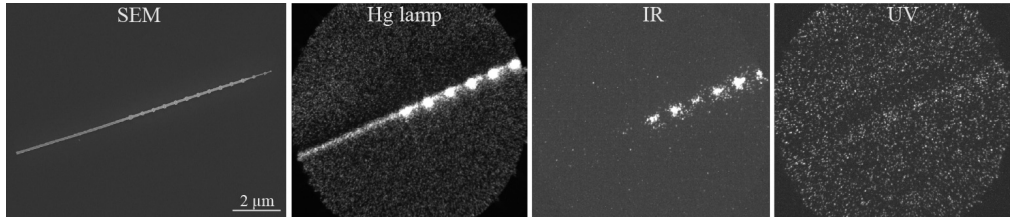


**Figure 3.4:** Images on InAs nanowires with Al rings recorded from left to right: with SEM, Hg lamp, IR laser source (830 nm), UV laser source (308 nm).

Depending on the excitation scheme and detection configuration, different parameters can be accessed. But in all cases, the photon energy must be sufficient to overcome the work function of the material in order to emit electrons from the surface. This can be achieved either by single-photon absorption or by multiphoton processes, as discussed in the previous chapter, depending on the excitation wavelength (Figure 3.4).

In the case of IR or visible light, the photon energy is mostly below the work function, and the photoemission process would be multiphoton. This regime is useful for studying optical near field and ultrafast dynamics, as the photoemission yield becomes sensitive to the local electromagnetic field intensity. In the case of UV excitation, the photon energy is typically high enough to induce single-photon photoemission, which can be suitable for probing workfunction variations, electronic structures, and absorption-related processes like interband excitation or structural variations. For synchrotron-based X-ray excitation, where the photon energy is much higher, deeper electronic states can be probed, which makes it possible to distinguish between different chemical elements.

### 3.2.2 Ultrafast laser systems

Since PEEM relies on optical excitation, the laser systems become an integral part of the experiment. As discussed in Chapter 2, ultrashort laser pulses can provide pulse durations of only a few tens of femtoseconds or even a few optical cycles. This allows the study of photoemission, nonlinear optical processes, and ultrafast dynamics. Although the laser architecture depends on the specific experiment, most systems share a common design consisting of pulse generation, amplification, wavelength conversion, and pulse delivery.

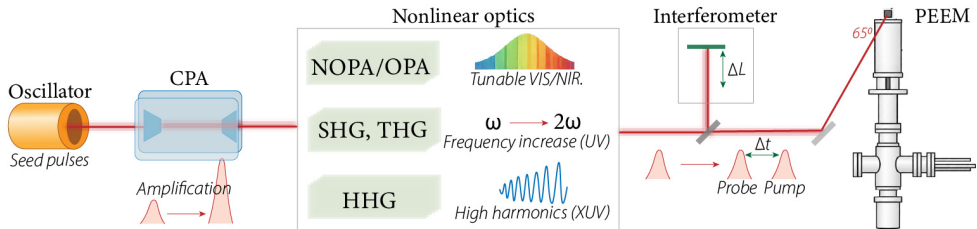


**Figure 3.5:** General architecture of an ultrafast laser system for PEEM experiments. Seed pulses from a mode-locked oscillator are amplified by a chirped-pulse amplifier (CPA), followed by wavelength conversion using nonlinear optical techniques (OPA/NOPA, SHG/THG, or HHG). An interferometer generates two time-delayed pulses for pump-probe measurements, which are directed onto the sample in the PEEM at an angle.

As briefly discussed in Section 2.3 and illustrated in 2.10b, ultrashort laser pulses are formed by the constructive interference of many phase-locked optical frequencies. In practice, as illustrated on Figure 3.5, these pulses are initially generated in a mode-locked laser oscillator, which produces a stable train of femtosecond pulses. Because the energy of these pulses is low and insufficient for many nonlinear optical experiments, they are typically amplified using a regenerative chirped-pulse amplifier (CPA). In this technique, the pulses are first temporally stretched to reduce their peak power, amplified, and then recompressed to recover femtosecond pulse durations while increasing the pulse energy by several orders of magnitude.

After amplification, the output can be modified depending on the desired wavelength with nonlinear optics. Optical parametric amplifiers (OPAs or NOPAs) generate tunable visible and infrared pulses with broad bandwidths and short pulse durations. In this thesis, this approach was used to generate SWIR pulses for multiphoton photoemission for silver nanowires, presented in Paper I. Frequency doubling or tripling (SHG and THG) produces light to wavelengths that are two or three times shorter than the original wavelength, creating ultraviolet radiation, as for studies on InAs nanowires in Paper II. For even shorter wavelengths, high-harmonic generation (HHG) focuses pulses in a gas, where nonlinear interactions convert intense femtosecond laser pulses into coherent extreme ultraviolet radiation, enabling attosecond photoemission experiments.

For time-resolved measurements, the laser beam is divided into two beams using an interferometer where a beam splitter divides the pulse into two beams. A mechanical delay stage in one arm of the interferometer controls the optical path length and therefore the arrival time of one pulse relative to the other one. Depending on the experiment, the pump and probe beams may have identical wavelengths or may be independently converted further with nonlinear optics. The generated laser pulses are subsequently used in PEEM excitation schemes, including the pump-probe described in the following section.

### 3.2.3 Time-resolved PEEM

The ultrafast laser pulses provide the basis for time-resolved PEEM measurements, which allow to follow electronic processes on femtosecond timescales. This is important because most

photoemission processes, such as electron movement and plasmon dynamics, occur on very short timescales due to the short lifetimes of the excited states. By combining nanometer spatial resolution with femtosecond temporal resolution, it enables direct observation of light-matter interaction and electron dynamics on their intrinsic length and time scales<sup>78,92,149</sup>.

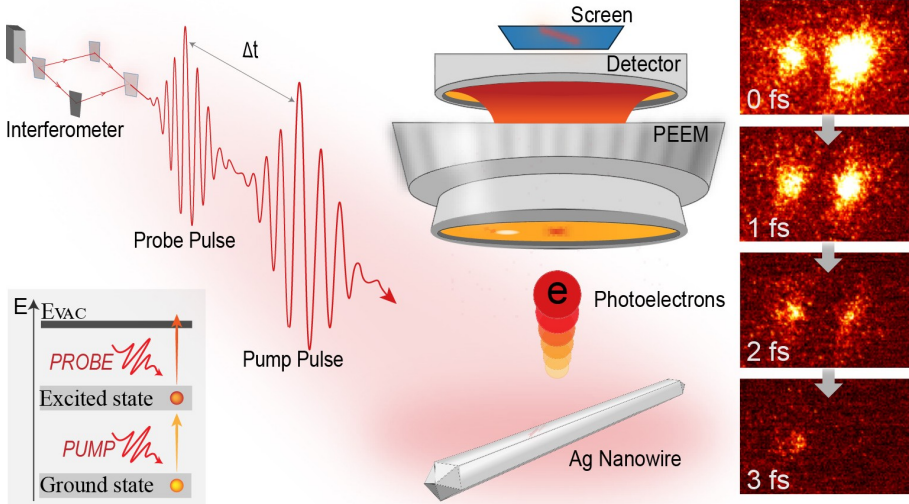


**Figure 3.6:** Pump-probe scheme for time-resolved PEEM measurements. An interferometer generates two time-delayed pulses, where the pump pulse excites the nanostructure and the probe pulse triggers photoemission. The emitted electrons are imaged by PEEM, producing time-dependent snapshots of the sample. The image sequence on the right shows the evolution of plasmonic near-fields in a silver nanowire at different delay times.

This technique is based on a pump-probe scheme, where a first laser pulse (pump) excites the system and initiates a dynamic process, such as plasmon excitation, carrier generation, or charge redistribution, but a second delayed pulse (probe) induces photoemission, providing a snapshot of the system as a function of this delay time (Figure 3.6). By scanning with this delay, the temporal evolution of the system can be reconstructed with femtosecond resolution. As demonstrated, for example, in silver nanowires, the spatial and temporal evolution of plasmonic near-fields can be mapped before those plasmons disappear.

The power of time-resolved measurements lies in its sensitivity to the localized near fields. The photoemission yield depends nonlinearly on the electric field, and in the multiphoton regime, the emitted electron intensity scales approximately as

$$EI_{\text{PEEM}} \propto |E(r, t)|^{2n}, \quad (6)$$

where  $n$  is the number of photons absorbed. This makes the technique highly sensitive to field enhancement and, as a result, regions with enhanced fields, such as plasmonic hotspots, appear significantly brighter in the PEEM image.

This nonlinear dependence makes the pump-probe technique suitable for direct imaging of

optical near-fields and tracking them over short timescales before they degrade, so imaging the near-field dynamics and not only probing where electrons are emitted.

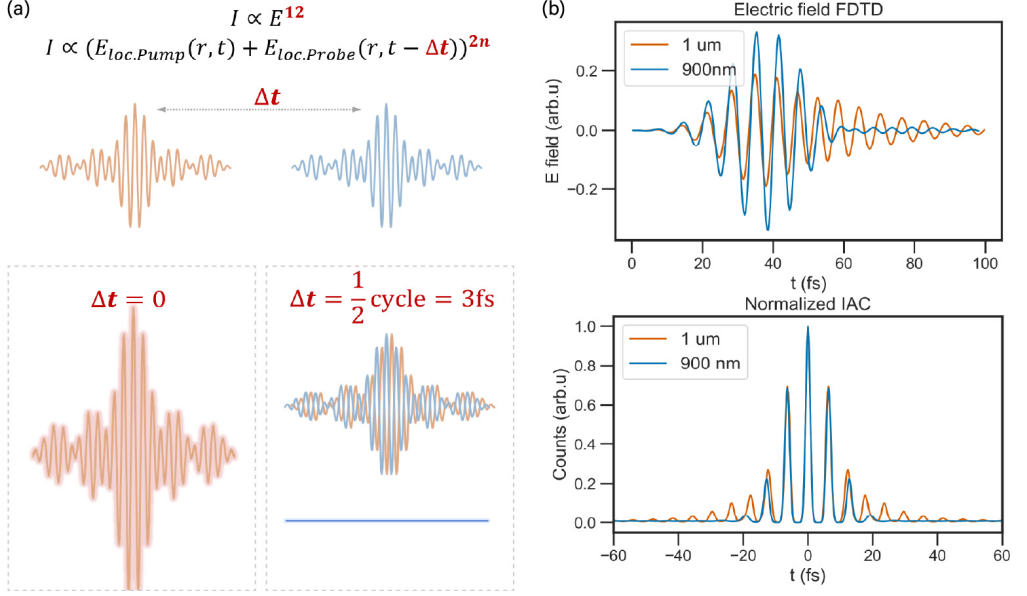


**Figure 3.7:** ITR-PEEM schematic and example results from simulated model nanowire. (a) Schematic of delay-dependent interference of two few-cycle pulses and resulting nonlinear photoemission signal. (b) An electric near-field temporal profiles at two ends of the 1  $\mu\text{m}$  and 900 nm nanowires (top). Corresponding nonlinear IAC traces, derived from the near-fields, representing the signal measured in ITR-PEEM experiments (bottom).

As an extension of standard pump-probe measurements, interferometric time-resolved PEEM can be employed. In this approach, two phase-locked pulses with a variable delay are used, and the photoemission signal depends on their coherent superposition. So, because of the locking, few-cycle pulses interfere with each other, and the delay can be scanned with even sub-femtosecond precision.

This resulting photoemission signal depends nonlinearly on this combined field because of the excitation, enabling interferometric autocorrelation measurements of the local response:

$$I(r, \Delta t) = \int |E_{\text{tot}}(r, t)|^{2n} dt = \int |E_{\text{pump}}(r, t) + E_{\text{probe}}(r, t + \Delta t)|^{2n} dt. \quad (7)$$

The measured dynamics and IAC traces originate from the interference of the two electric fields of incident pump and probe pulses, as also illustrated in the schematic in Figure 3.7a. When the delay  $\Delta t = 0$ , the electric fields of the two pulses overlap constructively, leading to a maximum in the total field amplitude and therefore a strong photoemission signal.

When the delay reaches half an optical cycle, the electric fields are out of phase and can partially cancel one another. As a result, the total field is reduced, which leads to a suppression of the

photoemission. This process repeats periodically with the optical cycles as the delay keeps changing and forms the basis of the interferometric autocorrelation measurement. Since the photoemission process is nonlinear, the resulting IAC traces (Figure 3.7b) show a pronounced peak at the constructive interference of the field and minima at the destructive interference.

Depending on the experiment, different excitation schemes can also be used in addition to two-pulse pump-probe excitation, involving three pulses. This allows additional control of the optical field dynamics and the photoemission process, enabling more flexible and detailed measurements of ultrafast phenomena.

### 3.2.4 Alternative PEEM modes

Modern PEEM systems can be extended with additional capabilities that provide access to complementary information.

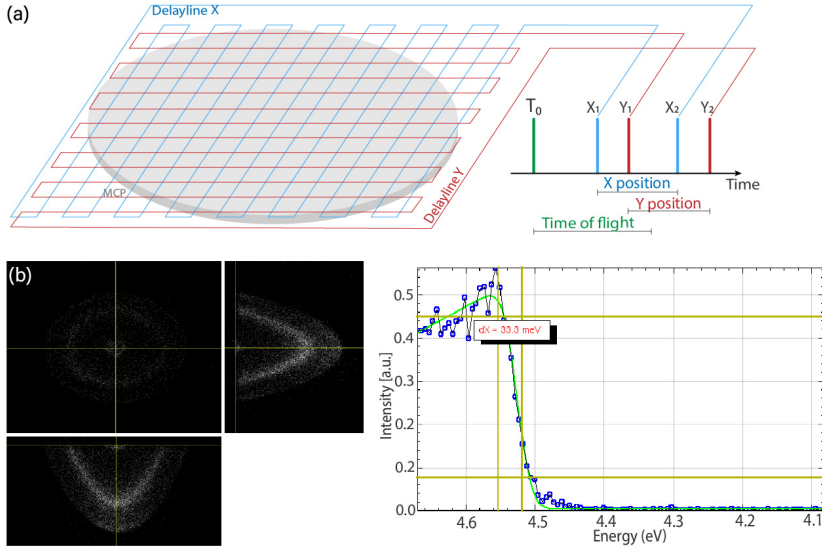


**Figure 3.8:** TOF-PEEM measurements and synchrotron-based PEEM setup. (a) Schematic illustration of the DLD working principle. (b) Momentum-space electron distributions and corresponding work function cut-off, obtained from ToF-PEEM measurements on Ag(111). (c) Experimental PEEM system at the MAX IV beamline used for synchrotron-based measurements.

One important extension installed was time-of-flight (ToF), in which the kinetic energy of electrons is determined from their flight time to the delay-line detector (DLD)<sup>153,154</sup>. In this approach, as illustrated in Figure 3.8a, electrons emitted from the sample travel through a drift tube before reaching a MCP where they are multiplied. The resulting electrons hit a position on the delay line detector, generating electrical pulses that propagate along the delay lines in opposite directions. From the differences in the arrival times, the  $x$  and  $y$  positions are reconstructed and the absolute arrival time indicates the electron time-of-flight from which the electron kinetic energy is determined using the calibrated instrument response.

Fast electrons correspond to photoelectrons with higher kinetic energy, typically originating from states with lower binding energy or regions with lower work function. In contrast, slower electrons often include contributions from scattering processes. So mainly fast electrons with low binding energy near the Fermi level are recorded. This results in energy-resolved imaging with spatial resolution, allowing access to the electronic structure and real-space, momentum-space, and energy information to be analyzed simultaneously.

The example results recorded on Ag (111) with ToF-PEEM are illustrated in Figure 3.8b. Momentum-space (k-space) images show the angular distribution of emitted electrons and the workfunction cut-off, demonstrating the capability of ToF-PEEM to access spatial, angular, and energy information within a single experiment. In this way, ToF-PEEM can also operate in a mode analogous to angle-resolved photoemission spectroscopy (ARPES)<sup>155,156</sup>. Having the UV excitation via single-photon processes, probing of the electronic structure near the Fermi level is possible. This is reflected in the k-space images obtained from ToF-PEEM measurements, where both angular and energy-resolved information can be accessed.

Another advanced upgrade is the MAX-PEEM beamline, which combines PEEM with synchrotron radiation with a wide photon energy range, from UV to soft X-rays. By selecting the photon energy, specific absorption edges of elements can be probed, allowing mapping of chemical composition and electronic states with high spatial resolution<sup>108</sup>. Here, we could measure how high temperatures affect the material structure, surface decompositions, oxidation processes, perform spectromicroscopy and x-ray absorption spectroscopy with spatial resolution<sup>157,158</sup>.

### 3.2.5 Sample preparation, measurement procedure and limitations

Sample preparation is a critical step in PEEM experiments due to the strong surface sensitivity of the technique. Any contamination on the sample or its holder can even cause damage to the optics. Samples and holders are normally cleaned before insertion into the vacuum or into a separate chamber coupled to PEEM, using sputtering and annealing. Heating facilitates the removal of adsorbates and improves surface quality, which is essential for obtaining reliable, reproducible measurements<sup>159,160</sup>. In addition, the sample geometry must be compatible. Flat, oriented parallel, and well-mounted surfaces are required to ensure homogeneous fields and avoid distortions in the electron trajectories.



**Figure 3.9:** PEEM sample holder and assembly. Photograph of a PEEM sample holder together with schematic illustrations of its assembly. The sample is mounted in the central aperture and secured using a clip and back plate, ensuring good electrical contact and proper alignment for PEEM measurements.

PEEM systems are equipped with specialized sample holders designed to ensure precise alignment. Figure 3.9 shows sample holders provided by FOCUS compatible with PEEM, which are made of molybdenum or stainless steel. The sample is pressed against the opening using a clip as illustrated on Figure 3.8, which also ensures good electrical contact and mechanical stability.

A typical PEEM measurement begins with sample preparation and insertion into the UHV chamber through the preparation chamber. Clean surfaces are essential, as PEEM is highly surface sensitive. Additional annealing can be done to clean and even the surface. After sample positioning in the stage and reaching proper UHV conditions, the electron optics are initialized and aligned.

One has to be careful during the first imaging. Before stable operation, the system components, such as the extractor, lenses, and imaging assembly, must be gradually “formed” by slowly increasing the applied voltages to stabilize the electron optics. The extractor voltage, which plays a crucial role in determining resolution, must also be increased carefully. Too high voltages can lead to field emission or arcing, which not only distorts the image but can damage the instrument.

Experimental parameters such as magnification, field of view, and aperture size are adjusted to optimize resolution and signal. After the best focus is reached and the reference picture is taken with an Hg lamp, the sample is illuminated with a laser, and the emitted electrons are imaged according to the lens configuration. In time-resolved experiments, the delay between pump and probe pulses is scanned, and image series are recorded as a function of time delay.

One of the main key limitations in PEEM experiments is the space-charge effect<sup>161,162</sup>. At high excitation intensities, a large number of electrons are emitted simultaneously, leading to electron repulsion and energy broadening. This results in spectral shift, reduced resolution, and image distortions. This effect scales strongly with the number of emitted electrons per pulse and therefore has a nonlinear dependence on incident excitation intensity. For this reason, the excitation intensity must be carefully controlled, and intense laser pulses should be attenuated, especially in ultrafast experiments.

Additionally, PEEM should not be operated at intermediate pressures, as this can lead to arching and unstable imaging. Also, mechanical stability is important, as vibrations or sample drift can reduce resolution and pose a problem for long acquisition and time-resolved scans.



# Main findings from PEEM studies

Based on the concepts introduced in the previous chapters, this chapter focuses on experimental results from PEEM measurements, linking the electronic processes discussed in Chapter 2 with some of the light-interaction processes from Chapter 3. Here, PEEM provides a direct way to connect the two by measuring electron emission driven by optical excitation.

In PEEM, as also illustrated in Figure 2.11, electrons are emitted when their energy exceeds the local surface barrier. The kinetic energy of emitted electrons can be calculated as

$$E_{\text{kin}} = nh\nu - \Phi, \quad (8)$$

where  $n$  is the number of absorbed photons,  $h\nu$  is the photon energy, and  $\Phi$  is the work function. In the case of SWIR excitation, where photon energies are low ( $\sim 0.5\text{--}0.8$  eV), this requires a multiphoton process because the work functions of common metals and semiconductors are in the range of 4–5 eV. As a result, several photons should be absorbed to give electrons enough energy to overcome the surface barrier, leading to a nonlinear process. In case of UV excitation, on the other hand, photon energies are typically in the range of  $\sim 4\text{--}6$  eV, which can exceed the work function. Photoemission can then occur via a single-photon process, where the absorption of a single photon is sufficient to overcome the surface barrier. In the presence of strong local electric fields, however, the effective surface barrier may be reduced, allowing photoemission to occur at somewhat lower photon energies.

## 4.1 TR-PEEM measurements on silver nanowires

As for nanostructures for this part of the work, silver nanowires are chosen as model systems. They provide a relatively simple and well-defined geometry, with homogeneous surfaces and reproducible plasmonic properties. In addition, silver exhibits low optical losses and supports strong plasmonic resonances, making it particularly suitable for studying field enhancement and ultrafast near-field dynamics.

### 4.1.1 Measurement setup

While plasmonic dynamics have been extensively studied at visible wavelengths, significantly less is known about plasmon excitation in the SWIR range. In the case of SWIR, longer wavelengths enable stronger plasmonic field enhancement and confinement at sharp nanoscale features and provide access to regimes where strong-field and highly nonlinear photoemission processes may occur. Understanding plasmonic behavior under SWIR excitation is therefore important both for fundamental studies of light-matter interaction and for future infrared nanophotonic technologies.

To access the ultrafast dynamics of localized fields in silver nanowires, TR-PEEM measurements are performed using few-cycle SWIR pulses. Two identical pulses are used with a variable time delay, allowing us to probe the evolution of the local field over time.

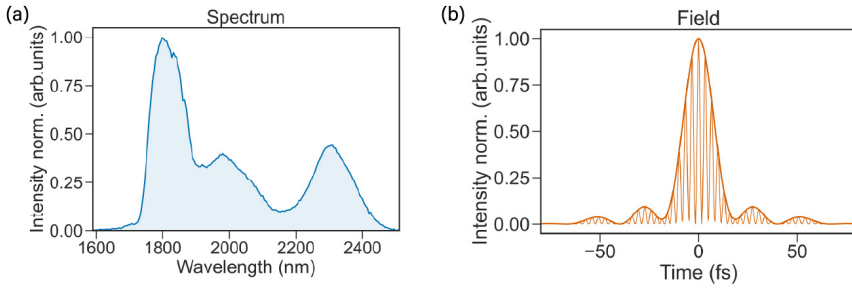


**Figure 4.1:** Parameters of laser pulses for SWIR studies. (a) Spectrum spans 1600–2500 nm and (b) around 17.5 fs in HWHM in time.

Experimentally, the pulses used in this part of the work are directed onto the sample inside the PEEM at a  $65^\circ$  angle relative to the surface normal. The emitted photoelectrons are then collected and imaged, providing spatially resolved information about the local field distribution. The excitation pulses are approximately 1500–2500 nm, as shown in Figure 4.1a<sup>111</sup>. This broad spectral bandwidth is required to support ultrashort pulse durations, as also discussed in Chapter 3. The corresponding temporal profile of the pulse, shown in Figure 4.1, has a FWHM of approximately 17 fs. This corresponds to about 2–3 optical cycles of the electric field, placing the experiment in the few-cycle regime.

### 4.1.2 Interpretation of the results

Following the description of the excitation scheme, the TR-PEEM measurements on silver nanowires can now be interpreted in the context of the concepts introduced in Chapters 2 and 3. In Figure 2.12, the excitation of localized plasmonic modes and the formation of enhanced near fields at metallic nanostructures were discussed, while Chapter 3 described how PEEM can image these local optical fields through photoemission.

A schematic representation of the expected dynamics is shown in Figure 3.7, where localized regions of enhanced electric field (“hotspots”) appear and oscillate with the optical excitation. The spatial distribution of these hotspots depends on the plasmon mode excited within the

nanowire. As illustrated in Figure 2.12, different excitation wavelengths can couple to different plasmon resonances, resulting in distinct field distributions and numbers of hotspots along the wire.

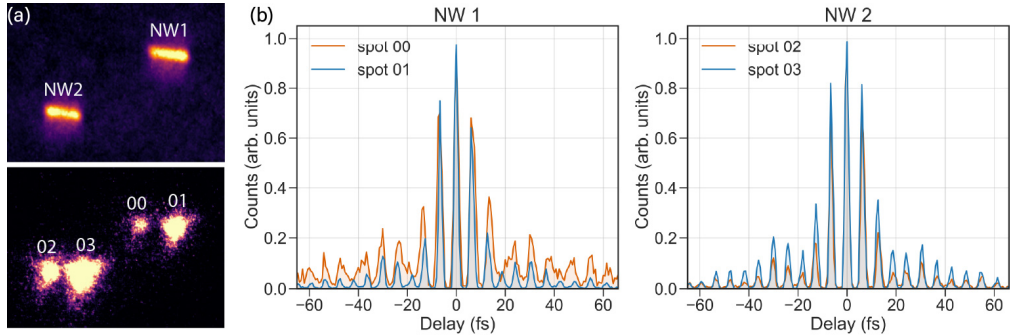


**Figure 4.2:** Spatial and temporal response of silver nanowires under SWIR excitation. (a) Comparison of Hg-lamp and SWIR PEEM images, where SWIR excitation produces strong photoemission localized at the nanowire ends. (b) IAC traces recorded from the hotspot regions, demonstrating few-cycle dynamics and hotspot-dependent temporal responses.

When nanowires are illuminated with SWIR pulses, photoemission is primarily localized at the ends of the nanowires (Figure 4.2a), forming bright regions, also sometimes termed as hotspots. These regions correspond to areas of enhanced local electric field.

For the  $1\text{ }\mu\text{m}$ -long nanowires, SWIR optical excitation drives longitudinal plasmon modes (LSPs), which lead to a concentration of the electric field at the nanowires' ends. Since the length of the nanowire is comparable to the excitation wavelength, those LSP modes can also be understood as standing wave-like oscillations. The central part of the nanowire contributes only weakly to the signal, as the local field enhancement is significantly smaller in this region compared to the nanowire's ends, where the surface has curvature (creating a lightning rod effect). Besides, due to the nonlinear nature of the photoemission process, only regions with the strongest local electric field contribute significantly to the measured signal, and weaker modes in the middle of the nanowire are suppressed in the PEEM images.

The IAC traces (Figure 4.2b) recorded from different positions on the nanowire, in particular at the hotspot locations, show oscillations that follow the optical cycle of the driving field (7–8 fs). However, an important observation is that the temporal response is not identical for all hotspots, even for the two ends of the same nanowire.

Small differences in IAC peak positions and shapes are observed in Figure 4.2b. Since the laser spot is in the range of  $200\text{ }\mu\text{m}$  in size and much larger than the nanowire dimensions, ensuring uniform illumination, the excitation field is identical at both nanowire ends. These differences in IAC traces must then originate from the local plasmonic response of the structure.

The orientation of the nanowires with respect to the incident light, particularly its polarization, also strongly influences the observed response, as we also briefly discussed in Subchapter 2.2.2. When a nanowire lies parallel to the polarization of the incident light, strong field enhancement occurs, leading to pronounced hotspots. But when the nanowire axis is oriented perpendicular to the polarization, excitation is suppressed, resulting in weaker or no emission.

All this together shows that the nanowire geometry strongly influences both the spatial and temporal characteristics of the near-field response. And to better understand those processes, FDTD simulations are performed.

### 4.1.3 Numerical simulations of near-field dynamics

The simulations using the Finite-Difference Time Domain (FDTD) method are carried out in the commercial software package Lumerical. With this approach, Maxwell's equations that describe the evolution of electric and magnetic fields in space and time are solved numerically, allowing the propagation, reflection, scattering, and interference of electromagnetic waves to be followed throughout the structure. Material properties, such as the wavelength-dependent refractive index and absorption, are incorporated directly into the calculations, enabling realistic modeling of complex nanostructures.

Electromagnetic fields are excited using a user-defined optical source with the desired wavelength, polarization, angle and temporal profile. During the simulation, field monitors record the spatial and temporal evolution of the electric and magnetic fields, from which quantities such as the local electric-field intensity, field enhancement, and cross-sectional field distributions can be extracted. This makes it suitable for studying plasmonic nanostructures, where both geometry and material properties influence the local field distribution.

Accurate FDTD simulations require careful selection of the computational parameters. The spatial mesh must be sufficiently fine to resolve nanoscale features and regions of strong field confinement. The simulation boundaries are terminated using perfectly matched layers (PMLs) to suppress artificial reflections. Accurate wavelength-dependent dielectric functions are required to correctly reproduce the optical response and local field enhancement of the simulated materials. During the post-processing, the local electric-field intensity, field enhancement, cross-sectional field distributions, and time-dependent field evolution are extracted, allowing direct comparison with the experimental observations.

Silver nanowires and SWIR excitation were modeled to match the experimental setup, and in addition, different lengths, tip shapes, and substrates were tested to provide a comparison. The simulation setup reproduces the main conclusions observed in the PEEM measurements, where we see strong field localization at the nanowire ends, just as observed in the PEEM images. But in addition to reproducing the experimental observations, the simulations provide further insight into the physical mechanisms governing the near-field dynamics.

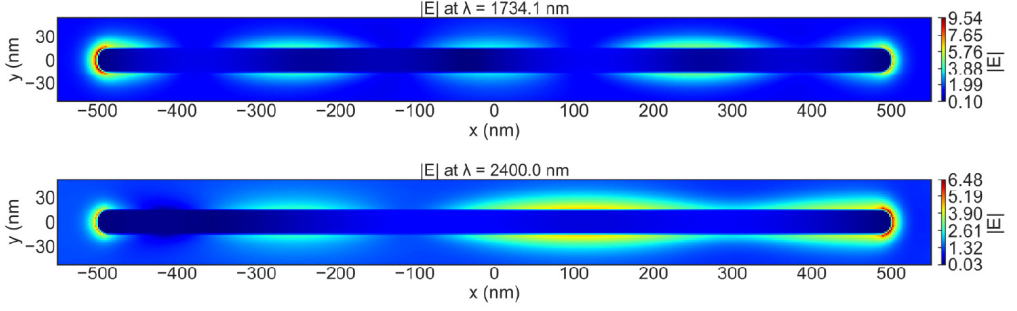


**Figure 4.3:** FDTD simulations of the near-field intensity distribution along a silver nanowire for two excitation wavelengths within the SWIR spectral range. The number and position of field maxima depend on the excited plasmon mode, illustrating the wavelength-dependent character of the nanowire response. The nanowire ends exhibit the strongest field enhancement for both excitation wavelengths.

This interpretation of the field dynamics is not always straightforward. First of all, due to broadband excitation, different spectral components excite plasmonic modes with varying propagation behavior along the nanowire, as illustrated in Figure 4.3.. For the simulated nanowire geometry, excitation in the 1600–1800 nm range produces a field distribution with five local maxima, whereas excitation in the 2200–2400 nm range results in four maxima. But either way, the ends of the nanowires exhibit the strongest field enhancement, and the temporal evolution over the full excitation bandwidth, as extracted from FDTD, mainly shows oscillations localized at the nanowire ends. The temporal evolution of the simulated fields and the shift of IAC peaks also match the experimentally measured IAC traces.

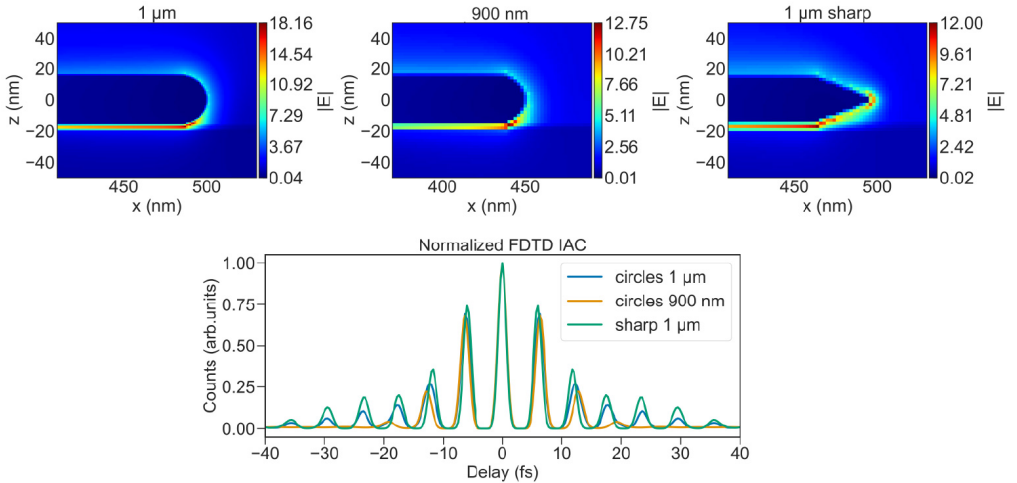


**Figure 4.4:** Influence of structural parameters of the nanowire on the plasmonic response. FDTD simulations of 1  $\mu\text{m}$  and 900 nm long nanowires with circular tips, and a 1  $\mu\text{m}$  long nanowire with sharp tips, demonstrate that nanowire length and tip morphology affect both the near-field enhancement and the temporal evolution of the plasmonic excitation, leading to shifts in the corresponding IAC traces.

To further understand the observed variations in the IAC traces, nanowires with slightly different geometries are compared. Comparing the simulated field distribution (Figure 4.4) and IAC traces of silver nanowires with different geometries, it is observed that even small changes in nanowire length lead to measurable differences in the temporal evolution. Longer nanowires show slower decay of the signal and larger temporal shifts, while shorter nanowires exhibit faster dynamics and more symmetric IAC traces.

The shape of the nanowire ends also plays an important role. Because of geometric coupling or lightning rod effect, sharper tips lead to stronger field localization, resulting in higher photoemission intensity and stronger near-fields. More rounded ends produce broader and weaker hotspots, with reduced emission intensity.

Besides, the length of the nanowires and the shape of the ends define whether part of the near-field distribution is “hidden” underneath the nanostructure, since the PEEM measurements primarily detect electrons emitted from the surface of the nanowire, which are visible to the electron optics. As a result, field enhancements located on the sidewalls or at the nanowire-substrate interface can be partially obscured and contribute less to the measured signal.

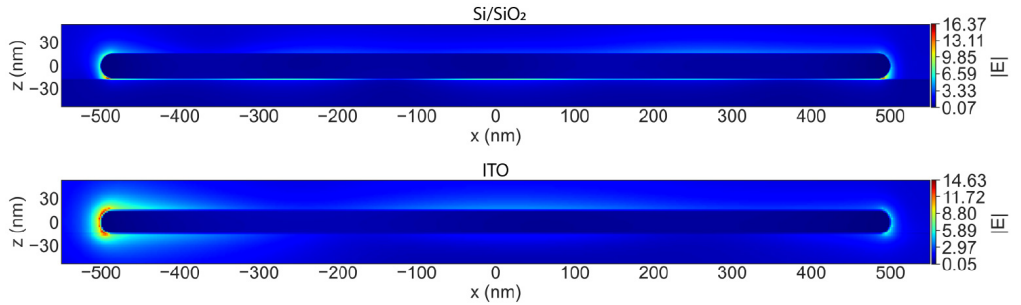


**Figure 4.5:** Side-view near-field distributions simulated by FDTD for silver nanowires on Si/SiO<sub>2</sub> and ITO substrates. The Si/SiO<sub>2</sub> substrate introduces additional substrate-coupled field components beneath the nanowire, modifying the local electromagnetic environment compared to the ITO-supported structure.

The substrate can also influence the near-field distribution as illustrated in Figure 4.5. The freestanding nanowire itself (or placed on an ITO substrate) supports the dominant longitudinal plasmon modes, but in the presence of the Si substrate, the electromagnetic environment is modified and these modes can also be affected. In particular, the substrate can introduce additional transverse modes under the nanowire that are not directly visible by PEEM, but which still affect the overall field distribution.

It should be noted that the simulated system is ideal and represents the ideal result. Even though we also see the IAC peaks shift that matches the experiment, especially at the first 20 fs delay time, the real nanowires can have small surface variations. For example, they have a surfactant coating to avoid aggregation, and their crystal structure may have slightly different from standardized dielectric constants. But overall, the simulations confirm that the near-field dynamics are strongly influenced by the nanowire geometry, including its length and tip shape, as well as by the surrounding environment. These simulations not only reproduce the experimentally observed behavior but also provide a deeper understanding of the spatial and temporal variations in the plasmonic response.

#### 4.1.4 Power dependence and photoemission type

The nonlinear nature of the photoemission process is further investigated by measuring the emission intensity as a function of excitation power. Experimentally, this is done by varying the incident laser power while recording the corresponding PEEM signal from a selected region of the nanowire, typically at the hotspots. The measured intensity is then plotted on a logarithmic scale to determine the order of the emission process.

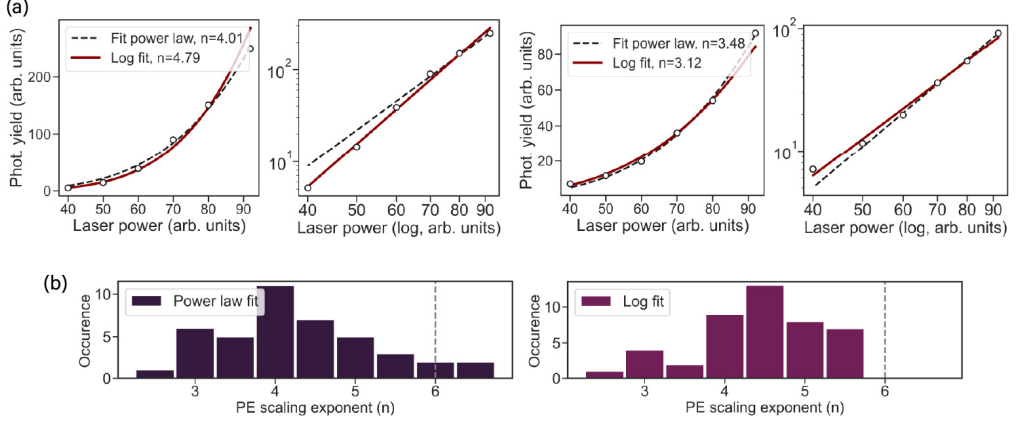


**Figure 4.6:** Photoemission yield as a function of incident excitation power. (a) Power-dependent photoemission intensity measured from representative nanowire hotspots 00 and 01 together with power-law and logarithmic fits. (b) Extracted power-law and logarithmic fit exponents for all investigated nanowires, showing consistent scaling behavior across the measured structures (the dashed grey line indicates the expected scaling).

The observed scaling follows a power-law behavior, with an effective exponent between 3 and 5 (Figure 4.6a). And those values are consistent with all the measured nanowires (Figure 4.6b). This is lower than what would be expected from a simple multiphoton emission based on the work function of silver. With  $\Phi \approx 4.3\text{--}4.7$  eV, typically 6–7 photons would be required for emission under SWIR excitation.

This lower photon count suggests that additional processes may contribute to the emission mechanism. Then we can refer back to Subchapter 2.3.2 where photoemission was described. First, due to high fields, local heating of the nanostructure may contribute to the emission process, as increased electron temperature can enhance the probability of emission through thermally assisted processes. However, under the ultrafast excitation conditions used in this work, this contribution is expected to be small.

Then, strong-field effects such as field-induced barrier lowering and optical tunneling can play a role under the experimental conditions. These may lower the effective emission threshold and reduce nonlinearity. This behavior can also be described using the Keldysh parameter ( $\gamma$ ), which characterizes the emission regime. For  $\gamma \gg 1$ , the process is dominated by multiphoton absorption, but for  $\gamma \sim 1$ , which was estimated for our case, tunneling contributions become more significant. Under the present experimental conditions, the enhanced local fields at the nanowire ends can reduce  $\gamma$ , bringing the system closer to the strong-field regime.

These power-dependence results show that the emission mechanism in silver nanowires cannot be fully described by the multiphoton regime alone, and that it may be governed by contributions from both multiphoton absorption and strong-field effects.

## 4.2 PEEM measurements on hybrid nanostructures

In addition to metallic nanowires, this thesis investigates hybrid metal-semiconductor nanostructures based on InAs nanowires patterned with epitaxial Al-rings, shown in Figure 3.4. Epitaxial Al/InAs nanostructures are of considerable interest for quantum technologies because of their interface that enables efficient carrier transport while simultaneously supporting plasmonic excitation in the metallic component. This provides a platform to investigate how metallic particles can tailor local optical excitation, modify carrier dynamics, and potentially facilitate hot-electron transfer in metal-semiconductor systems.

### 4.2.1 Measurement principle and excitation scheme

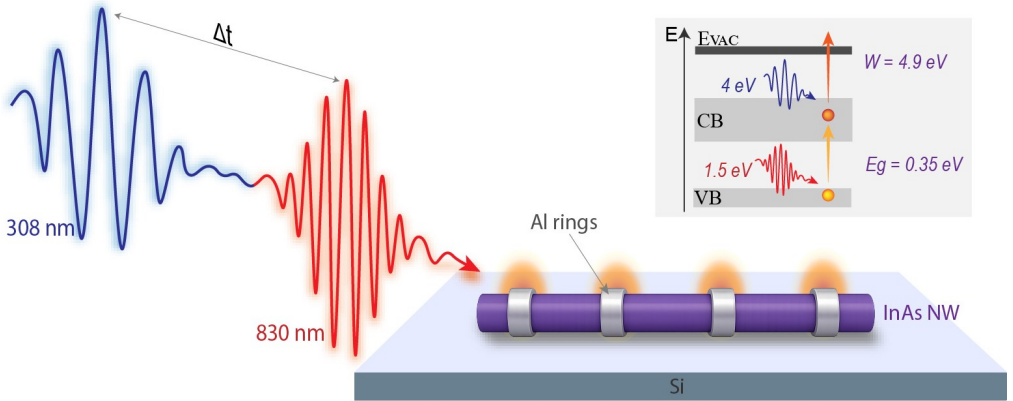


**Figure 4.7:** Schematic illustration of the IR-UV pump-probe excitation scheme used for TR-PEEM measurements of InAs nanowires with Al rings. An 830 nm (1.5 eV) IR pump pulse excites carriers across the InAs bandgap ( $\approx 0.35 \text{ eV}$ ), while a delayed 308 nm (4.0 eV) UV probe pulse photoemits electrons from the transiently excited states.

The measurements are performed using a time-resolved pump-probe PEEM scheme, as illustrated in Figure 4.7. An IR pump pulse with a wavelength of 830 nm (photon energy of approximately 1.5 eV and pulse duration of approximately 20 fs) is used to excite the nanowire, as this energy is higher than the InAs bandgap ( $\sim 0.35 \text{ eV}$ ). Then, a delayed UV probe pulse at 308 nm (photon energy of approximately 4.0 eV, lasting around 18 fs) is used to photoemit electrons from the excited regions. The high photon energy of the UV pulse provides sufficient energy to overcome the material work function and access the transient electronic population created by the IR excitation. By varying the time delay between the IR pump and UV probe pulses, the evolution of these short-lived excited electronic states can be tracked with femto-pico-second

temporal resolution. In contrast to the SWIR interferometric measurements in silver nanowires, which probe the coherent evolution of plasmonic near fields on the optical-cycle timescale, the pump-probe measurements on semiconductor-metal nanowires access transient carrier excitation and relaxation dynamics.

The investigated nanowires were approximately 10–11  $\mu\text{m}$  long and contained a segment patterned with epitaxial Al rings together with an uncovered InAs section on the same nanowire. This geometry enabled direct comparison between plasmonically modified and bare semiconductor regions within a single measurement.

## 4.2.2 Main results and electromagnetic simulations

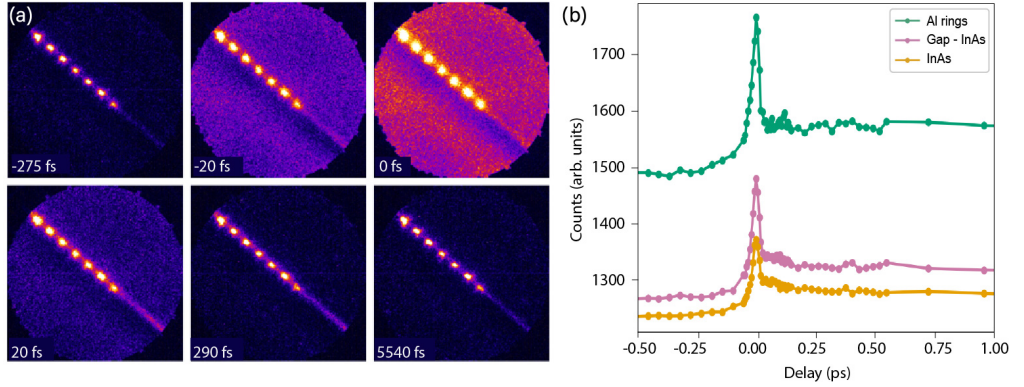


**Figure 4.8:** TR scans of an InAs nanowire patterned with Al rings under IR-UV pump-probe excitation. (a) Representative PEEM images acquired at selected pump-probe delays, illustrating the evolution of the photoemission signal. (b) Delay-dependent photoemission intensity extracted from the Al rings, the InAs regions between neighboring rings, and the uncovered InAs nanowire at 0 degrees incident light polarization.

Figure 4.8 shows representative time-resolved PEEM images of an InAs nanowire patterned with Al rings, together with the delay-dependent photoemission signal extracted from different regions of the nanowire. Before the temporal overlap of pulses, the photoemission signal remains nearly constant creating a photoemission background due to photoelectrons generated both by the UV and IR individually. At temporal overlap (time zero) a sharp increase in the signal is observed due to the simultaneous overlap of the IR and UV pulses. At longer positive delays, the remaining photoemission originates from hot electrons generated by the pump pulse that gradually relax on picosecond timescales.

The spatial distribution of this photoemission signal is strongly dependent on the local nanowire geometry. The highest intensity is observed at the Al rings, an enhanced signal is also detected in the InAs regions located between Al rings, while the uncovered InAs sections show the weakest response, which indicates that the influence of the metallic structures extends beyond the Al-covered regions and may modify the electromagnetic environment also along the neighboring semiconductor regions. This enhancement may originate from stronger local optical

excitation produced by the neighboring Al rings or from hot electrons transferred across the epitaxial Al/InAs interface.

The transient response also depends on the polarization of the incident IR excitation. The overall strongest photoemission signal is obtained for a polarization angle of  $30^\circ$ , while the response decreases for larger angles and is weakest for  $90^\circ$ . Besides, the bare InAs nanowire exhibited nearly constant decay times for all excitation polarizations, but the InAs located between neighboring Al rings showed pronounced polarization-dependent variations.

The transient dynamics observed can be described by two characteristic decay components: a fast component of approximately 10–20 fs which is observed for all regions and is dominated by the temporal overlap of the pulses with possible contribution from plasmon relaxation in Al, and a slower component ranging approximately 0.4–2 ps, which reflects the relaxation of hot carriers after excitation. The uncovered InAs nanowire exhibits a nearly constant slow decay of about 0.6 ps for all polarizations, but the Gap-InAs region shows pronounced polarization-dependent variations, with decay times extending to about 2 ps. These longer decay times can indicate a change in the dominant relaxation mechanism from electron–hole scattering in bare InAs toward additional electron–phonon scattering or hot-electron transfer processes around Al rings.

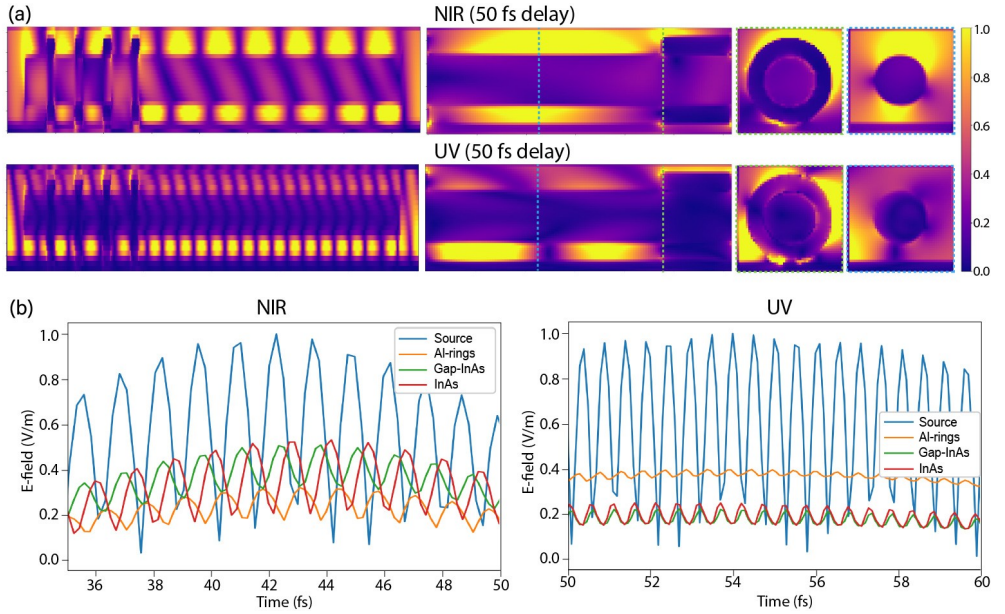


**Figure 4.9:** FDTD simulations of the electromagnetic field distribution in InAs nanowires with Al rings. (a) The spatial field distribution along the nanowire and cross-sectional field profiles for different regions of the structure under NIR and UV optical excitation. (b) The temporal evolution of the averaged electric field under NIR and UV excitation centered at the corresponding wavelengths, probed for Al rings, gap-InAs in between Al, and purely InAs regions.

To understand the spatial distribution of the field measured in the PEEM experiments better, FDTD electromagnetic simulations were performed. The calculated field distributions, shown

in Figure 4.9a, reveal that the Al rings modify the local electromagnetic environment of the InAs nanowire under both NIR pump and UV probe excitation. Distinct spatial field distributions are obtained for the two wavelengths, reflecting the different optical response of the hybrid metal–semiconductor structure and the wavelength-dependent coupling of the incident light to the nanowire geometry. The simulations reproduce the strong field localization at the Al rings while also demonstrating that the metallic patterning influences the optical field inside the neighboring semiconductor regions. Pronounced field localization is observed at the outer edges of the Al rings, forming asymmetric hotspots also due to the oblique angle of incidence of the excitation beam.

Further insight is obtained from the temporal evolution of the averaged electric fields in the different regions of the nanowire shown in Figure 4.9b. Under NIR excitation, the InAs region between Al rings exhibits a different temporal response compared with the corresponding InAs region without metallic rings, including a shift of the field oscillations and changes in the field amplitude. This indicates that the Al structures modify the propagation and local phase of the optical excitation within the semiconductor. In the case of UV excitation, stronger field enhancement is observed at the Al rings, whereas the temporal response of the two InAs regions remains comparatively similar, indicating that the probe field predominantly enhances photoemission from the metallic structures.

Importantly, the simulations show that although the Al rings substantially modify the local optical fields, the calculated field strength in the InAs region between neighboring rings is comparable to, and for some polarizations even weaker than that of the uncovered InAs nanowire. Consequently, the experimentally observed enhancement of the photoemission signal in the Gap-InAs region cannot be explained solely by local optical field enhancement. Instead, the PEEM contrast reflects the combined influence of pump-induced carrier excitation and probe-induced photoemission, with the results being consistent with an additional contribution from hot-electron transfer across the epitaxial Al/InAs interface, although further experiments are required for direct confirmation.



# Optical studies of dye-based systems

Based on the concepts introduced in Chapter 2, DASA molecules can act as optically tunable absorbers. In their colored open form, they absorb visible light strongly, while after illumination they switch into a more transparent closed form. In Chapter 3, it is introduced how optical transmission and electrical measurements can be used to follow these changes. Here, we will look at how this molecular switching can be combined with semiconductor nanowires to create hybrid optical–electrical devices.

The main idea is that the DASA layer controls how much light reaches the nanowires, while the nanowires convert the transmitted light into an electrical signal. In this way, the molecules provide optical memory and the semiconductor nanowires provide the electrical readout. This separation of optical control and electrical detection is useful because the DASA molecules do not need to be contacted electrically, while the nanowires can still report their state through photocurrent.

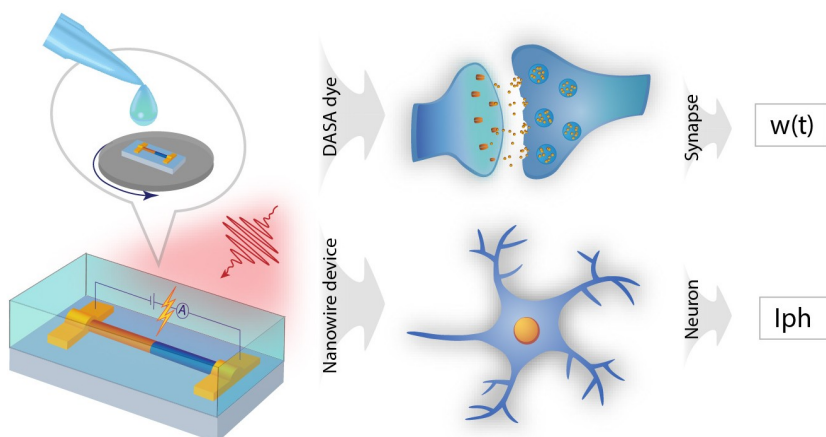


**Figure 5.1:** Biological inspiration and implementation of the hybrid DASA–nanowire artificial network. In the hybrid device, functions of synapses and neurons are reproduced by the DASA molecular layer, which stores the history of optical stimulation through reversible changes in optical transmission (artificial synapse), and the semiconductor nanowire photodetector, which converts the transmitted optical signal into an electrical response (artificial neuron)<sup>21</sup>.

In biological systems, synapses store and modify the strength of communication between neurons, whereas neurons integrate incoming signals and generate an output. In this hybrid architecture, these functions are implemented by DASA molecules, which act as artificial synapses by storing the history of previous optical stimuli, and nanowires, which function as artificial neurons by converting the transmitted optical signal into an electrical output, as illustrated in Figure 5.1. This concept was demonstrated using DASA embedded in a poly( $\alpha$ -methylstyrene) (PAMS) matrix integrated with III–V nanowire photodetectors, where the dye acts as the optically programmable element and the nanowire device acts as the light receiver.

Unlike many photochromic molecules that require UV light or exhibit no reversibility, DASAs can be switched using visible light and display tunable recovery dynamics spanning a broad range of timescales. In addition, they can be incorporated into polymer matrices, which can be deposited onto a wide variety of substrates and semiconductor devices. DASA-based films also exhibit stable and reversible photoswitching over repeated cycles. These properties make them particularly attractive for optoelectronic devices, adaptive systems, and neuromorphic applications where controllable memory and wavelength-selective responses are desired.

## 5.1 Bleaching and recovery dynamics of DASA dyes

The first step is to understand the optical response of the DASA dyes themselves. When the molecules are illuminated with light within their absorption wavelength, the colored open form is converted into the more transparent closed form. This process is observed as bleaching – the absorption decreases and the transmission through the dye layer increases. When the illumination is removed, the molecules relax back toward the colored open form. This recovery restores the color and absorption of the dye and decreases the transmission again.

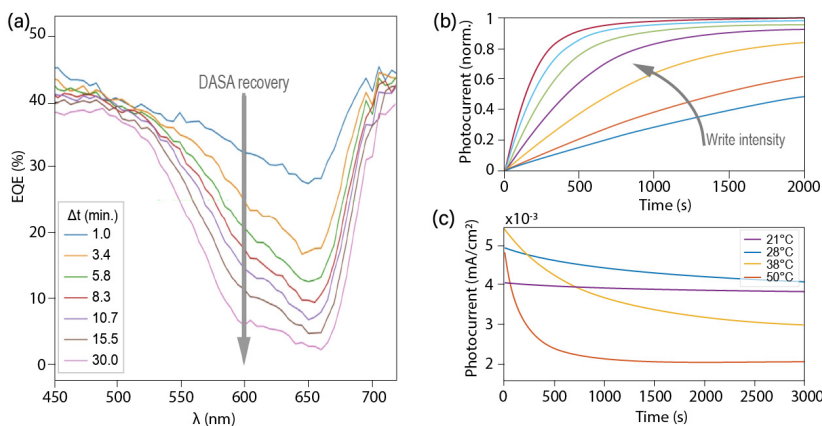


**Figure 5.2:** Optical switching dynamics of DASA dyes deposited on the substrate. Transmission recovery (a) following optical bleaching is shown together with the influence of excitation intensity (b) and temperature (c) on the switching behavior.

The transmission of the deposited dye may be measured as a function of wavelength and time, so that the spectral response and temporal response may be monitored together (Figure 5.2a).

While the recovery is often well described as a relaxation with an exponential time constant, the quantitative dynamics depend on the dye structure, polymer matrix used, film thickness and local conditions.

In addition to the previous main parameters, switching is also strongly dependent on excitation intensity, as illustrated in Figure 5.2b: the higher the intensity, the more molecules are switched to the closed state within a short period of time, and the faster the bleaching. At lower intensities, the bleaching process is slower and may remain incomplete, resulting in a reduced change in transmission. This change in transmission can be made dependent not just on the wavelength, but also on the intensity, thereby enabling control over the optical state of the system, and this feature is used later during optical switching with nanowire devices.

The supporting measurements also show that recovery depends strongly on temperature<sup>101</sup> (Figure 5.2c). At higher temperatures, the thermal back-conversion from the closed to the open form is accelerated, leading to faster recovery dynamics. In contrast, at lower temperatures, the recovery becomes slower and the bleached state can persist for a longer time. As a result, heating can be used as an effective reset mechanism for the molecular memory, allowing the system to be brought back to its initial state in a controlled way.

This means that the DASA layer is not only an absorber, but a dynamic optical element. Its state depends on previous illumination, as well as on the excitation intensity and temperature, and this state can persist for a certain time before recovering. This is the origin of the memory-like behavior used later in the nanowire devices.

## 5.2 Integration of DASA dyes with nanowire devices

To make use of the optical switching of DASA molecules in a device, the molecular layer is combined with semiconductor nanowire photodetectors. As discussed in Chapter 2, nanowires provide efficient light absorption and carrier transport, making them suitable for converting optical signals into an electrical response.

### 5.2.1 DASA dyes with nanowire arrays

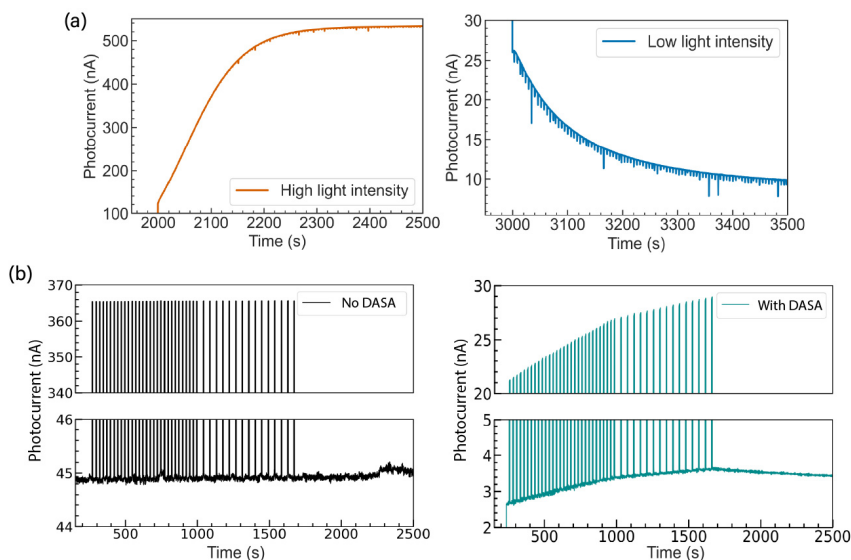


**Figure 5.3:** Optical writing, recovery, and pulse-train response of a DASA-functionalized nanowire device. (a) Photocurrent evolution during optical switching and recovery. High-intensity illumination bleaches the DASA layer, increasing transmission and causing a gradual rise in photocurrent, while subsequent low-intensity illumination allows thermal recovery of the molecules and a corresponding decrease in photocurrent. (b) Pulsed optical excitation of nanowire devices without and with DASA. In the absence of DASA, successive pulses generate an approximately constant photocurrent response, while with the DASA layer, the molecular state evolves between pulses, resulting in a history-dependent photocurrent response and gradual modulation of the pulse amplitudes.

In this hybrid architecture, the DASA layer, which is deposited on a nanowire array device, functions as an optically tunable filter to control the amount of light reaching the nanowires. The nanowires then convert the light into photocurrent, providing a direct electrical read-out of the molecular state.

In the measurement scheme, the intensity of the light is varied to distinguish readout and switching of DASA molecules (reading and writing, respectively). First, molecules will be exposed to weak light, which will have almost no effect. Therefore, a baseline signal is obtained as the photocurrent corresponds to the initial absorption of the dye in its open form. When the illumination intensity is increased, the DASA molecules are gradually bleached and switch to the transparent form. As the transmission through the dye layer increases, more light reaches the nanowires, leading to a gradual increase in the photocurrent. This behavior is illustrated in Figure 5.3a.

After switching, the intensity is reduced again to the low readout level. Under these conditions, the dye is no longer actively switched, and the system evolves through thermal recovery. As the molecules become colored again, the transmitted light decreases, and the photocurrent slowly returns to its original value, resulting in a gradual current change (Figure 5.3a).

The device therefore exhibits a history-dependent response: the electrical output depends not only on the instantaneous illumination but also on the previous optical excitation. This behavior provides the basis for optical memory and adaptive functionality in the hybrid nanowire-molecule system. As illustrated in Figure 5.3b, repeated optical pulses can progressively modify the transmission of the DASA layer, leading to changes in the photocurrent response of the nanowire device. The magnitude of the response therefore depends on the timing, intensity, and number of previous optical spikes. Such history-dependent behavior resembles synaptic plasticity in biological neural systems, where the response of a synapse depends on previous activity. In this way, the DASA layer acts as an optically controlled synaptic weight, while the nanowire acts as a neuron-like element by receiving the weighted optical signal, integrating and converting it into the electrical output<sup>6</sup>.

### 5.2.2 Single nanowire-DASA device

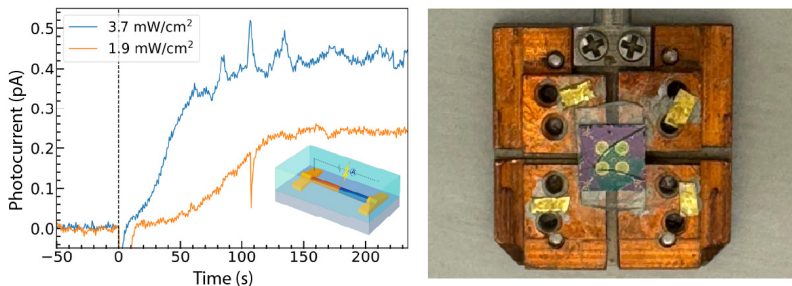


**Figure 5.4:** Optical switching in a single DASA-functionalized nanowire device. Left: photocurrent response under optical excitation at two different illumination intensities, showing a gradual increase in photocurrent due to bleaching of the DASA layer. Right: photograph of the measured single-nanowire device.

Experiments on single-nanowire devices are also performed (in addition to using nanowire arrays) in order to understand how the hybrid structure could be scaled (Figure 5.4). Although the photocurrent signal is weaker than that of nanowire arrays due to reduced active volume and absorption, the switching behavior is similar. In particular, the bleaching-induced increase in photocurrent is preserved at the level of an individual nanostructure.

The observation of the same switching mechanism in a single nanowire demonstrates that the functionality is not an ensemble effect but can be maintained at the level of an individual nanoscale device. This is an important step toward highly integrated optoelectronic architectures, where individual nanowires may serve as functional building blocks for optical memory, sensing, and neuromorphic information processing. A detailed investigation of these effects is beyond the scope of this work but represents an interesting direction for future studies.

### 5.2.3 Reproducibility and dynamic switching behavior

To evaluate the stability and dynamic properties of DASA and nanowires, we perform repeated switching using controlled light illumination sequences. In these experiments, the device is

periodically exposed to high-intensity illumination, which induces bleaching, followed by lower-intensity conditions that allow recovery, while the photocurrent of the nanowire device is continuously recorded.

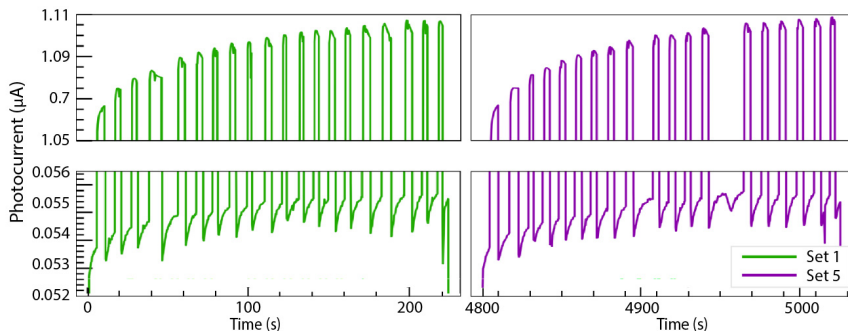


**Figure 5.5:** Repeated optical switching cycles of a DASA-functionalized nanowire device. The photocurrent response remains reproducible over multiple write–recovery cycles, demonstrating reversible and stable operation.

As shown in Figure 5.5, the response has been shown to be reproducible over numerous cycles of switching on and off, indicating that the switching effect is reversible and stable. These measurements have provided more information about the dynamics of the system. When the device is pulsed or illuminated, each time it is excited, the layer of molecules stays partially bleached. Upon further pulsing, if the time between pulses is shorter than the recovery time of the molecules, the bleaching will accumulate, and the photocurrent will gradually increase.

This leads to a memory-like behavior, in which the photocurrent depends not only on the excitation present at the time, but also on the history of excitation. For high repetition rates or for short delay times, the bleaching will accumulate. For longer times between pulses, the effect of thermal recovery becomes more significant. This behavior is similar to short-term plasticity, where the system response depends on the timing and sequence of the input signal. This temporal response can be tuned by varying the illumination intensity and illumination duration, in effect entering different dynamical regimes.

### 5.3 Extension to two dyes and wavelength-selective channels

Once a first DASA-nanowire system is successfully demonstrated using only one dye, the system can be extended by combining two DASA derivatives with distinct absorption spectra, in which case each dye best absorbs a different wavelength of light. This allows wavelength-selective control to be achieved using a single DASA-nanowire device.

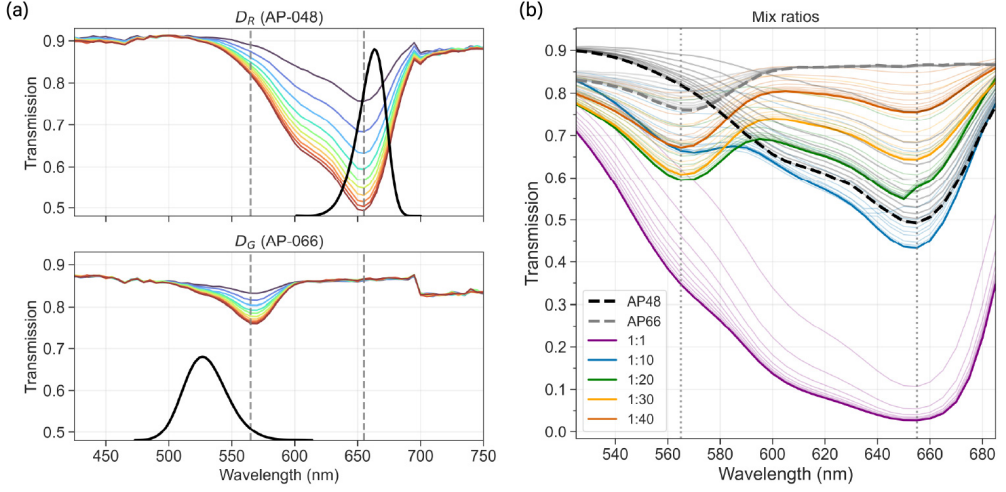


**Figure 5.6:** Spectral properties of a dual-DASA system. (a) Transmission spectra of the individual DASA derivatives,  $D_R$  and  $D_G$ , during optical recovery. Dashed lines point out the central wavelength of the dye, while black lines indicate the wavelength at which optical bleaching was done. (b) Transmission spectra of mixed dye layers with different concentration ratios.

Figure 5.6 shows the transmission spectra of recovery of two DASA derivatives functioning in 565 and 655 nm. Because the absorption bands of the two dyes are spectrally separated, illumination at one wavelength can predominantly bleach one dye while having a smaller or no effect on the other. As a result, different optical wavelengths can be used to independently modify the state of the molecular layer.

The transmission spectrum of the mixed dye system is the superposition of the two individual dye absorption bands, and therefore depends on their relative concentrations. By adjusting the mixing ratio, the strength and spectral position of the transmission features can be tailored (Figure 5.6b). This introduces an additional degree of freedom compared with single-dye systems and allows multiple optical channels to coexist within the same device.

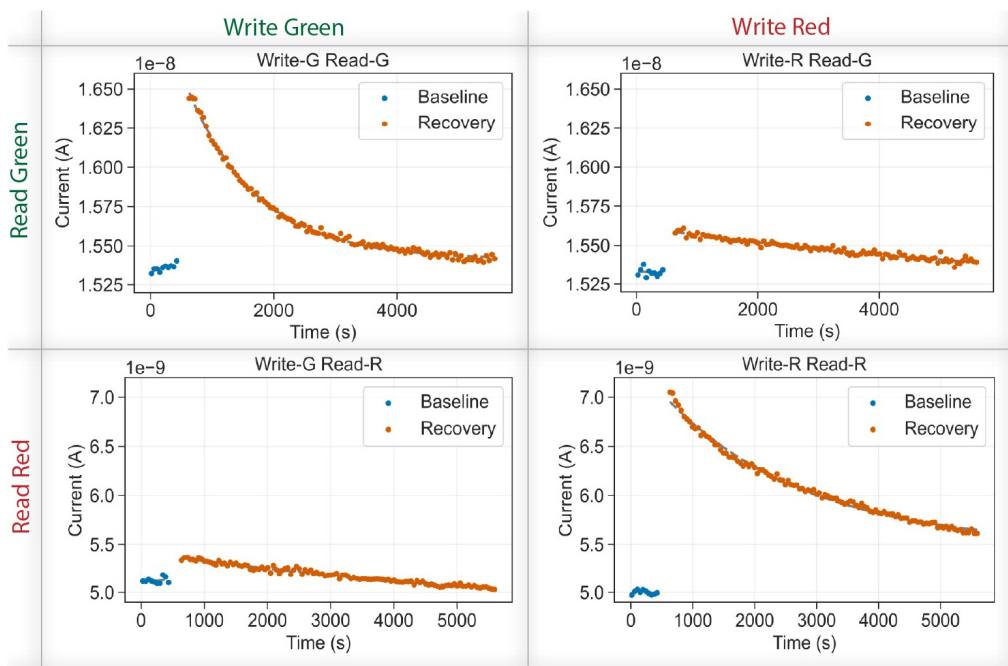


**Figure 5.7:** Selective optical writing and readout in a dual-DASA nanowire system via wavelength-selective addressing and partially independent operation of multiple optical memory channels within a single molecular layer. Writing and reading within the same spectral channel produce the strongest response, whereas cross-channel measurements show significantly reduced changes in photocurrent.

Similar pulsed illumination schemes that were used for single-dye systems can be extended to the multi-dye case by using different excitation wavelengths. Here, two low-intensity pulses at two wavelengths (R – 655 nm and G – 565 nm) are applied to the dye mixture on the nanowire array, producing a small current but not enough to bleach the dye. Then, a high-intensity “write” pulse of either red or green is turned on to bleach the corresponding dye. After full bleaching, the same low-intensity two pulses are “reading” the dye response at the two corresponding wavelengths. In this way, one dye can be selectively written while the other remains unchanged, or both can be written at the same time, enabling parallel or sequential control of multiple optical channels.

Figure 5.7 presents the photocurrent response measured for different combinations of write and read wavelengths. When the writing and readout wavelengths correspond to the same dye channel, for example, addressing “Green AP048” (655 nm) dye, the largest changes in photocurrent are observed for reading the corresponding “Green AP048” dye. When one dye is “written”, but the other is “read”, the response is significantly weaker, and the remaining response observed in the cross-channel measurements originates from partial spectral overlap between the absorption bands of the dyes.

This selective mixing of different molecular dyes permits the spectral and temporal characteristics of the system to be tuned. Such multi-channel molecular systems may therefore provide a platform for wavelength-multiplexed optical memories, adaptive photonic devices, and neur-

omorphous architectures with independently addressable synaptic weights.

The polarization of the incident light provides another degree of freedom for controlling DASAs. Because photoisomerization is most efficient when the electric field of the incident light is aligned with the molecular transition dipole moment, the bleaching efficiency depends on the polarization of the writing beam. In polymer films, the transition dipoles may exhibit a preferred orientation defined by the molecular arrangement within the polymer matrix. Consequently, the underlying nanowire photocurrent can be modulated not only by wavelength but also by the polarization state of the incident light. Even though the molecular orientation in the polymer is not perfectly uniform, resulting in only partial polarization selectivity, polarization still provides an additional degree of freedom that could be combined with wavelength-selective addressing to increase the number of independently accessible optical states in future molecular photonic memory and neuromorphic devices.



# Concluding remarks and outlook

This thesis explores a variety of nanoscale systems and how light can be used to probe, control, and exploit electronic processes within them. The work spans a broad range of length and time scales, from femtosecond plasmon dynamics in semiconductor and metallic nanostructures to memory effects persisting for hours or days in molecules. A common theme throughout this thesis is the rich and complex interaction between light and matter. In metallic nanostructures, ultrafast optical excitation is used to uncover the fundamental mechanisms governing electronic and plasmonic dynamics, whereas in molecular systems, light is employed to actively control material properties and improve device functionality.

## *Understanding fundamental insights*

Papers I and II address one of the main challenges in nanophotonics of understanding how optical excitation is converted into electronic motion on nanometer length scales and femtosecond timescales.

Paper I presents the first time-resolved PEEM study of plasmon-driven electron dynamics in metallic nanowires excited by few-cycle SWIR pulses, extending ultrafast photoemission measurements into a previously unexplored spectral regime. Paper II applies a similar approach to hybrid semiconductor–metal nanostructures, revealing how metallic patterning modifies ultrafast carrier excitation in semiconductor nanowires. Together, these studies provide new insight into the ultrafast light–matter interactions that underpin nanoscale optoelectronic and plasmonic devices.

This part of the thesis focused on ultrafast dynamics in metallic and semiconductor nanostructures using time-resolved PEEM. The experiments on silver nanowires, presented in Paper I, demonstrated that TR-PEEM can directly visualize plasmonic near fields excited by few-cycle SWIR pulses with nanometer spatial and femtosecond temporal resolution. Plasmonic fields are highly sensitive to nanowire structural details, and fields can change both in spatial distribution and time evolution due to small nanometer variations in nanowire length, tips, and the surrounding. This observation highlights the importance of structural control in the design of nanoscale plasmonic devices.

Furthermore, the power-law scaling of the photoemission yield indicates that strong local fields can push the system towards non-perturbative photoemission regimes via processes that cannot be simply described as multiphoton absorption. Instead, strong local fields may drive the system toward non-perturbative photoemission regimes. Future studies using energy-resolved

TR-PEEM could help distinguish between these mechanisms by measuring not only where and when electrons are emitted, but also their kinetic energy distribution. One possible implementation is time-of-flight PEEM, where the arrival time of emitted electrons at the detector is used to reconstruct their kinetic energy. Combined with pump-probe excitation, this would make it possible to follow how the energy distribution of photoelectrons evolves in femtosecond time and to separate multiphoton, thermally assisted, and strong-field emission contributions. Understanding and exploiting this regime opens a window into the fundamental limits of light-matter interaction at the nanoscale and can help to develop efficient emitters, electron sources, and nonlinear optical devices.

The extension of the work to semiconductor-metal nanostructures in Paper II provided new insight into how IR-UV pump-probe excitation drives carrier dynamics across material interfaces. This study shows how near-field coupling and distribution in InAs are affected by metal within picoseconds and with varied polarization of incident light. This interaction between plasmons and semiconductor carriers creates opportunities to tailor local optical responses and charge transport processes. To extend it even further in the future, energy resolution combined with bias-dependent measurements could separate the contributions of optical field enhancement, carrier excitation, and interface-mediated transport.

Throughout this work, electromagnetic simulations played a crucial supporting role. In silver nanowires, FDTD simulations confirmed that while the strongest photoemission signal originates from localized fields at the nanowire ends, additional substrate-coupled modes remain largely hidden. These reveal the substrate effect and local-geometry dependence and show that small nanoscale geometrical changes affect the near-field distributions. For the metal-semiconductor nanowires, FDTD simulations provided a close-up view of how the near fields in InAs are altered in the presence of Al. Overall, FDTD simulations also established a direct link between nanostructure geometry and photoemission, which could guide the design of future plasmonic and hybrid optoelectronic nanostructures.

### *Functional light-responsive systems*

The second part of the thesis explored hybrid systems based on semiconductor nanowires functionalized with DASA molecules. The work demonstrated that molecular switching can be directly integrated with nanoscale optoelectronics, enabling optical control of electrical response. The resulting structures exhibit reversible switching, tunable recovery dynamics, and history-dependent responses that provide a form of optical memory, which originates from the molecular layer itself rather than from conventional electronic charge-storage mechanisms. Besides, this functionality is preserved down to individual nanowire devices, demonstrating that the concept is minimized down to the nanoscale.

Extending this concept to multiple DASA derivatives that work within two separated absorption spectra revealed that wavelength-selective control can also be achieved within a single device architecture. This introduces the possibility of independently addressable optical channels and more complex forms of memory and information processing, which are relevant for neuromorphic systems.

For DASA-functionalized nanowire devices, the main future challenges are stability and the need for controlled integration. More refined deposition methods, homogeneous coating and film thickness, patterned coating, temperature-dependent recovery, optimized polymer matrices, and long-term cycling studies are needed to understand device-to-device variability and to make DASA bleaching and recovery dynamics more universal. An additional challenge comes

from the spectral matching between the nanostructures and the molecular switches. The wavelengths that most efficiently drive DASA switching do not necessarily align with those that produce the strongest response in the nanowires, making it difficult to maximize coupling between the two components. There is also a challenge in quantifying optical changes when dyes are integrated with nanostructured devices using purely optical tools. In dual-DASA systems, spectral overlap between dyes is both a limitation and an opportunity. It can lead to cross-talk between wavelength channels, making independent readout more difficult, but it can also enable analogue and mixed memory states useful for neuromorphic processing. Future work should therefore focus on controlling the degree of overlap and developing models that predict their temporal and spectral response.

### *Future perspectives*

The results presented in this thesis open several promising directions for future research, both in the context of fundamental light–matter interactions and the development of functional nanoscale devices.

Recent developments in energy-resolved PEEM offer new opportunities to investigate not only where and when photoemission occurs, but also the energy distribution of emitted electrons. Such measurements could provide direct insight into hot-electron generation, energy relaxation pathways, and carrier transport in plasmonic and semiconductor nanostructures<sup>163–165</sup>. Combined with ultrafast excitation sources<sup>166</sup>, this may enable studies of nonequilibrium electronic processes with spatial, temporal, and energy resolution, and future work could investigate ultrafast charge transfer<sup>164,167</sup> and interface-mediated processes<sup>168</sup> in complex heterostructures.

For plasmonic nanostructures, the observed strong field localization under SWIR excitation suggests the possibility of entering regimes where strong-field and field-driven photoemission become dominant<sup>169,170</sup>. Such studies are relevant not only for fundamental plasmonics but also for emerging concepts such as lightwave electronics, ultrafast electron sources, and optical control of electronic motion on sub-cycle timescales<sup>171</sup>. These advances point toward a future where electronic processes can be steered with the precision of an optical waveform, a subtle shift from conventional electronics.

The hybrid molecular–nanowire platform developed in this thesis offers many opportunities for future device concepts. The integration of molecular functionality with nanoscale optoelectronics points toward possibilities of information processing performed not only by electronic circuits but also through the optical switching of molecules themselves. Inspired by biological signal processing principles, improved future hybrid photonic and molecular systems may perform computation, memory storage, and signal processing directly through their material response. Besides, recent demonstrations of optical neuromorphic computing<sup>172</sup>, ultrafast nano-optoelectronic nodes<sup>173</sup>, and wavelength-adaptive polymer-based optical synaptic transistors<sup>174</sup> also illustrate the rapid progress in this field. Technological approaches as such could contribute to the development of adaptive sensors, photonic neural networks, and low-power neuromorphic hardware that operate closer to the efficiency and functionality of natural systems.

Besides, future implementations could integrate multiple, more than two, photoswitchable molecules with different absorption spectra and recovery times. Such systems may support multiple independently addressable optical memory channels, wavelength-selective processing, and analogue synaptic weights. By exploiting both spectral and temporal selectivity, the functionality of molecular optoelectronic devices could be significantly expanded. Beyond

neuromorphic computing, such systems could find applications in adaptive optical filters, programmable photonic circuits, and smart sensors.

The overall results presented in this thesis demonstrate that advances in both experimental techniques and material systems are required to understand and exploit nanoscale electronic processes. By combining ultrafast microscopy, nanostructured materials, and molecular functionality, it becomes possible not only to observe electronic dynamics on their natural scales, but also to harness them for future technologies. The journey from fundamental understanding to functional devices is long, but the path forward is rich with many possibilities.

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# Scientific publications

## I Ultrafast near-field dynamics in silver nanowires driven by few-cycle short-wave infrared pulses

**Nelia Zaiats\***, Lukas Wittenbecher, Ivan Sytceвич, Anne-Lise Viotti, Chen Guo, Chandni Babu, Xiaolu Zhuo, Luis M. Liz-Marzán, Eduardo J. C. Dias, F. Javier García de Abajo, Jan Vogelsang, Anne L’Huillier, Cord Arnold, Anders Mikkelsen  
ACS Photonics 2026, 13(8), 2248–2259

*I was involved in the sample preparation, including nanowire deposition on substrates and mounting of samples. I participated in the experimental measurements, where the samples were evaluated using PEEM in the laser laboratory. I conducted the final data analysis and performed all FDTD simulations to support the experimental results. I was main responsible for writing the paper.*

## II Ultrafast dynamics of InAs nanowires with epitaxial Al-rings studied by TR-PEEM

Mahesh Saini, Saurabh Bokar, Thomas K. Jensen, **Nelia Zaiats**, Joachim E. Ses-toft, Jesper Nygård, Donatas Zigmantas, Anders Mikkelsen  
Manuscript in writing

*I assisted in the PEEM setup prior to experiments, participated in the measurements, and contributed to discussions regarding measurement procedures. I provided input during the data analysis, interpretation of the results, and manuscript writing.*

## III Integrating molecular photoswitch memory with nanoscale optoelectronics for neuromorphic computing

David Alcer\*, **Nelia Zaiats\***, Thomas K. Jensen\*, Abbey M. Philip, Evripidis Gkani-as, Nils Ceberg, Abhijit Das, Vidar Flodgren, Stanley Heinze, Magnus T. Borgström, Barbara Webb, Bo W. Laursen, Anders Mikkelsen

\*These authors contributed equally to the work  
Communications Materials 6, 11 (2025)

*I performed the initial testing of the dye integration, carried out measurements of the dye dynamics, and participated in measurements on the nanowire devices. I took part in the discussions regarding the experiments, interpretation of the results, and development of the project. I contributed to the data analysis and manuscript writing.*

## IV All-optical molecular synapses with wavelength-selectivity on III-V nanowire devices

**Nelia Zaiats\***, Xinyu He\*, Thomas K. Jensen, Abbey M. Philip, Bo W. Laursen, Anders Mikkelsen

\*These authors contributed equally to the work  
Manuscript in writing

*I performed the initial testing, including the deposition and integration of two dyes. I carried out a significant part of the experiments on the dual-dye dynamics and dye integrated with nanowire devices. I performed a substantial part of the data analysis and contributed to the planning and evaluation of the experiments. I took part in discussions of the results and am responsible for writing the main text of the manuscript.*





