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# **Progress Report**

## **2001 - 2002**

**Editor: Marie Holmdahl-Svensson**

**Lund Reports on Atomic Physics**  
**LRAP-306**

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# Introduction

The Division of Atomic Physics, Lund Institute of Technology (LTH), is active in basic physics teaching in engineering disciplines and specialized teaching in optics, atomic physics, spectroscopy, laser physics and applications. Research activities at the Division are mainly carried out in the fields of basic and applied spectroscopy, largely based on the use of lasers. The Division is also one of nine divisions comprising the Physics Department at Lund University. Since 1980, biennial progress reports have been issued within the series *Lund Reports on Atomic Physics (LRAP)*. Our latest report, covering 1999-2000 was *LRAP-270*, preceded by the reports *LRAP-20*, *LRAP-43*, *LRAP-85*, *LRAP-90*, *LRAP-119*, *LRAP-144*, *LRAP-172*, *LRAP-228*, and *LRAP-244*. The present report describes the activities of our division during the calendar years 2001 and 2002.

Research at the Division of Atomic Physics takes place in a multidisciplinary atmosphere, in which informal collaboration with external scientists and industry forms an important part. The Division is part of the Lund Laser Centre (LLC) which, based on a long informal existence, was officially established at Lund University in March 1995, directly under the Rectorate of Lund University. Other members are the Division of Combustion Physics (Prof. Marcus Aldén), the Division of Laboratory Astrophysics at the Astronomy Department (Prof. Se. Johansson) and the Division of Chemical Physics at the Chemical Centre (Prof. Villy Sundström). The Lund University Medical Laser Centre is also part of the LLC, while two other umbrella organisations, the Combustion Centre and The Centre for Environmental Measurement Techniques are associated members. The Board of the LLC includes members from the Technical, Science and Medical Faculties of Lund University. The chairman of the board, Prof. Bengt E.Y. Svensson, is appointed by the vice-chancellor of Lund University as is the director of the LLC, Sune Svanberg.

The European Commission provides funding for researchers in European research groups visiting the LLC within the Access to Large-Scale Infrastructures Scheme. The Division has benefited considerably from this programme, which has resulted in many joint projects (more than 70 since the start). The LLC is part of a cluster of Large-Scale Infrastructures, which now also includes LENS - University of Florence, LOA - Ecole Polytechnique, Palaiseau, the Max-Born Institute, Berlin, the Amsterdam Laser Centre, CUSBO at Politecnico di Milano, ULF-FORTH, Heraklion, and finally, SLIC at Saclay. Interaction with our sister facilities has also strengthened our European links.

At the High-Power Laser Facility, which is operated by the Division of Atomic Physics, an active research programme is being pursued, co-ordinated by Prof. Claes-Göran Wahlström. The facility was inaugurated at the end of 1992 and the equipment, spearheaded by a multi-terawatt, chirped-pulse amplification titanium-

sapphire system, is successively being upgraded. The facility is the main experimental resource for our basic atomic physics research programme, but is also used for applications. High harmonics have been studied extensively. The efficiency response with regard to atomic properties and phase-matching is now well understood, and the radiation is being utilised both for spectroscopy and interferometry. Schemes for attosecond pulse formation are being studied. An important aspect is to develop novel schemes for the measurement of ultra-short pulses. The experimental programme is complemented by theoretical studies on the description of the phenomena studied. The research on harmonics is headed by Prof. Anne L'Huillier, who is also co-ordinating a European research network on attosecond research.

Another aspect of the high-power laser/matter interaction programme is the generation of broadband X-rays by focusing terawatt radiation pulses on rotating solid targets. The properties of the radiation are being studied with regard to spectral content and temporal evolution. Radiological applications have been investigated, including gated X-ray imaging for the suppression of scattered radiation. Recently, the kHz high-power system of the Division has been used for X-ray pulse generation. The temporal behaviour of soft X-ray pulses has been studied with time-resolved X-ray diffraction studies in mind. Dr Jörgen Larsson is heading this programme, which also includes the use of fast synchrotron radiation pulses, for example, at ESRF, Grenoble. A very close link has been established with the MAX-lab facility in Lund, where extensive laser equipment is being integrated.

A recent development at the High-Power Laser Facility is a programme initiated by Prof. Claes-Göran Wahlström, to study relativistic channelling and the acceleration of electrons and protons to multi-MeV energies. Feasibility studies have been performed and research in the area of laser-induced nuclear processes is being prepared.

Extensive research activities concerning time-resolved laser spectroscopy in the VUV and XUV spectral regions have also been pursued. In a ns-pulse, fully tuneable set-up, four-wave mixing or Raman shifting is used in the generation process, while a ps system utilises low-harmonic generation. Rydberg sequences in free atoms have been investigated, as well as resonance lines in atoms and ions, observed by the Hubble Space Telescope. A laser-induced plasma has been used in many experiments to produce atoms and ions, also in metastable states. Rare-earth atoms, refractory atoms and iron are mostly being investigated. XUV spectroscopy has included studies of astrophysically interesting molecular lifetimes and excited-state photo-ionisation experiments on the fundamental atom helium.

Photon echoes are being used for the investigation of relaxation processes in rare-earth-ion-doped crystals at liquid-helium temperatures and are being tested as a means of optical storage and processing. Different all-optical operations are being implemented using photon-echo techniques. This programme, headed by Prof.

Stefan Kröll, also includes basic quantum optics studies of single-photon self-interference, and recently this fundamental effect has been demonstrated.

Extensive applied molecular spectroscopic studies are being pursued at the Division of Atomic Physics, including atmospheric remote sensing using differential absorption lidar monitoring of atmospheric pollutants, and fluorescence lidar studies of vegetation and historical monuments. Industrial effluents are being studied, in particular atomic mercury from a chlor-alkali plant, within the European EMECAP project. Another activity in the atmospheric work is focusing on geophysical gas emissions from mining, geothermal and volcanic activities. Recently, techniques for IR differential absorption lidar have been further developed for hydrocarbon monitoring. Optical parametric oscillator technology is being employed and extensive control and steering systems have been constructed. Diode laser spectroscopy for applied gas monitoring is being pursued with the frequency modulation technique. The available wavelength region has been strongly augmented by the use of blue GaN lasers and by using sum-frequency mixing techniques. A new method for measurements of gases dispersed in strongly scattering media (GASMAS; GAs in Scattering Media Absorption Spectroscopy) was recently introduced at the Division, allowing concentration, pressure and diffusion studies in previously inaccessible media. Measurements are being extended to the biomedical field. Passive IR gas correlation imaging of gas leaks has been further developed with spectacular visualisation of, for instance, hydrocarbons. A spin-off company from the division, GasOptics AB, has been established, and has recently obtained funding from the oil and gas industry. A further aspect of the lidar techniques is remote fluorescence spectroscopy and imaging. Following spectacular mapping of the facades of Lund Cathedral, and the Parma Cathedral and Baptistry, the techniques are now also being employed for characterising surface degradation of large insulators in the power grid.

The research activities within the Lund University Medical Laser Centre have further developed during the past two years. An important part of the research deals with malignant tumour detection and treatment. A core group consisting of 10 or more physicists and physicians is located together at the Department of Physics, ensuring close and daily interaction. The activity is headed by Prof. Stefan Andersson-Engels (physics) and Dr Katarina Svanberg (medicine). Members of this group also participate in a large number of projects at other departments and clinics. Particularly active clinical departments in this collaboration are Oncology, Dermatology, ENT, Surgery, Urology, Radiology and Pathology at the Lund University Hospital. Fluorescence diagnostics is widely used, while Raman and near-IR spectroscopy are being developed as alternative diagnostic techniques, in particular for cardiovascular diseases. Photodynamic treatment is firmly established in Lund with the treatment of hundreds of tumours. The use of the haem precursor ALA, applied topically to the lesion or administered orally, has meant a breakthrough in clinical applications. Interstitial and interactive tumour treatment is presently a focus of the group activities. A substantial grant from the Swedish Foundation for Strategic Research is supporting this activity. In order to detect



deeper lesions we are developing techniques for tissue transillumination. The long-term goal of this research is to achieve an optical mammographic method for screening without the use of ionising radiation. Promising results have been obtained with techniques varying from terawatt laser-induced, white-light illumination to diode-laser, time-resolved spectroscopy.

Fluorescence and scattering spectroscopy are being employed for the characterisation of paper and pulp in an industrially oriented programme headed by Adjunct Professor Lennart Malmqvist. Optical and laser techniques are also being utilised in another industrial project, in which the insulators for grid applications are being studied.

The division is pursuing a programme aimed at strengthening research in developing countries. These activities are supported by the International Programme in the Physical Sciences (IPPS), Uppsala, SIDA/SAREC and the Abdus Salam Centre for Theoretical Physics (ICTP), Trieste. A one-month workshop was arranged in Lund in April 2001, where groups from 6 developing countries could integrate equipment based on diode laser spectroscopy, and take this equipment home with them. The programme has been followed up by visits to Dakar, Harare and Quito for research purposes. Researchers from different locations are also spending time pursuing research in Lund.

In our report series, *Lund Reports on Atomic Physics (LRAP)*, material which is not published in international journals is presented. The reports include Master's dissertations, doctoral theses and special investigations. So far, more than 300 papers have appeared in this series.

The main teaching activities in physics, regarding students at the Lund Institute of Technology, have been connected to the Atomic Physics Division for many years. Undergraduate teaching at the Physics Department is now being reorganised, with the long-term goal of achieving better integration between the physics teaching at the engineering and the science faculties. As a step in this direction, physics teaching at LTH was recently organized as a separate unit. We wish our colleagues dedicated to basic course teaching continued success in their important endeavours, and look forward to continued good collaboration. A number of elective courses continue to be the main responsibility of the Atomic Physics Division: *Atomic and Molecular Spectroscopy, Advanced Atomic Physics, Optical Techniques, Laser Physics, Non-linear Optics, Tissue Optics, and Multi-spectral Imaging*.

At the end of the reporting period the research staff and graduate students at the Division of Atomic Physics totalled 42, the other 14 teaching staff now forming a separate unit. It is through the dedicated work of all the research, teaching and support personnel that the accomplishments reported here have been made possible.

We are very grateful for the support of a large number of funding agencies, in particular the European Commission, the Swedish Research Council (VR), the Swedish Foundation for Strategic Research, VINNOVA, the Swedish Medical Research Council (MFR), the Knut and Alice Wallenberg Foundation (KAW) and the Crafoord Foundation. Grants for research collaboration with industry were also received from AstraZeneca, and Science and Technology International.

Special thanks are due to Marie Holmdahl-Svensson for collecting and editing the material for this report, and to all division members for their contributions to the contents.

Sune Svanberg  
Head of the Atomic Physics Division

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### **Ph.D. Theses**

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Lena Roos	01-09-28	Optimisation and application of intense high-order harmonic pulses LRAP-276
Petter Weibring	02-08-30	Environmental monitoring by multi-spectral lidar techniques LRAP-284
Thomas Johansson	02-09-04	Applications of laser spectroscopy to analytical chemistry, environmental monitoring and medicine LRAP-286
Carl Magnus Nilsson	02-09-27	New possibilities using modern optics in paper production
Anders Sjögren	02-11-22	Laser matter interactions at extreme irradiance: X-ray generation and relativistic channelling LRAP-288
Johan Norin	02-12-06	Characterisation and applications of ultrashort extreme ultraviolet pulses LRAP-289
Johannes Swartling	02-12-13	Biomedical and atmospheric applications of optical spectroscopy in scattering media LRAP-290

**DIVISION OF ATOMIC PHYSICS  
LUND INSTITUTE OF TECHNOLOGY**

Head: S. Svanberg

Deputy head: C.-G. Wahlström

<b>Basic Atomic Physics and X-ray Science</b>	<b>Quantum Electronics, Quantum Optics and Applied Optics</b>	<b>Applied Molecular Spectroscopy</b>	<b>Medical Applications</b>
Z. Dai	S. Kröll	M. Bengtsson	Ch. Abrahamsson
C. Delfin	L. Rippe	H. Busk	S. Andersson-Engels
M. Gaarde	K. Mohan	H. Edner	L. Gustafsson
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J. Larsson		G. Somesfalean	P. Snoer Jensen
A. L'Huillier		P. Weibring	M. Soto Thompson
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# A. Basic Atomic Physics and X-Ray Science

Our research programme in basic atomic physics is gradually changing character. From being mainly devoted to the structure and the radiative properties of free atoms, the establishment of the Lund High-Power Laser Facility, in 1992, caused our efforts to be redirected towards the interaction of high-intensity radiation with matter. The generation of high-order harmonic radiation in gases and hard X-rays from laser-solid interactions quickly became two new important areas of research. At the same time, our laser spectroscopic research programme became more focused on challenging investigations in the vacuum ultraviolet and extreme ultraviolet (VUV and XUV) spectral regions.

In this chapter we briefly present our different research activities, and give examples of results obtained during the past two-year period. We begin in Section A1 with time-resolved studies of short-lived atomic and ionic states of astrophysical interest. Atoms in strong laser fields are discussed in the following two sections. Experimental research on high-order harmonics, and their applications to, e.g., XUV spectroscopy and attosecond pulse generation, is presented in Section A2, followed by our theoretical work in this field in Section A3. Generation, characterisation and applications of hard X-rays from laser-produced plasmas are discussed in the next two sections; in Section A4 with respect to imaging applications in, for example, diagnostic radiology, and then in Section A5 with emphasis on time-resolved studies in material science and biochemistry. This latter activity is closely linked to the MAX-lab in Lund, as part of our activities involve the establishment of a dedicated beam-line there with an integrated femtosecond laser system. Finally, in Section A6, we describe how our fundamental studies of laser-matter interactions at ultra-high intensities are being pursued at increasingly higher peak intensities. With peak intensities now exceeding  $10^{19}$  W/cm<sup>2</sup>, relativistic plasma dynamics and laser-plasma particle acceleration are being explored.

Our research programme has clearly evolved from being characterized as *basic atomic physics* to a programme better characterized as *extreme optical physics with applications in X-ray science, spectroscopy and plasma physics*.

Most of our experimental research is carried out at the Lund High-Power Laser Facility, which is part of the Atomic Physics Division. The main system is the *10 Hz, femtosecond terawatt laser*. It is based on chirped-pulse amplification in titanium-doped sapphire, and provides 35 fs pulses of multi-terawatt power. It operates with two separate laser beams; one with a peak power of about 2 TW, propagating in air, and the other exceeding 30 TW, with pulse compression and subsequent beam propagation in vacuum. The *kilohertz laser system* is also a titanium-sapphire-based system but operates at a 1 kHz repetition rate with a peak

power of 0.1 TW. Laser pulses of 30 fs duration are spectrally broadened by self-phase modulation in a hollow capillary, and subsequently compressed to about 10 fs duration, for ultrafast investigations. Our *XUV laser system* is based on a mode-locked picosecond Nd:YAG laser, pumping a short-pulse dye laser, followed by a solid-state power amplifier. In combination with high-order harmonic generation in gas jets, this system provides tunable, short-pulse radiation in the XUV spectral range. Finally, the *VUV system* is a narrow-bandwidth, tunable, system with a pulse duration in the nanosecond range. It is designed for time-resolved laser spectroscopy in the UV and VUV spectral ranges.

During the two-year period of this progress report, 2 MSc projects [A1, A2] and four PhD theses [A3-A6] have been completed and successfully defended.

## **A1. Time-resolved laser spectroscopic lifetime measurements of atomic and ionic excited states**

*Zhenwen Dai, Zhankui Jiang, Zhongshan Li, Vladimir Lokhnygin, Huailiang Xu, Zhiguo Zhang, Hans Lundberg, Anders Persson, Claes-Göran Wahlström, Sune Svanberg, Kiril Blagoev\*, Per Jönsson\*, Sveneric Johansson\*, Hans Karlsson\*, Ulf Litzén\*, Glenn Wahlgren\*, Hampus Nilsson\*, Veselin Penchev\*, H. Pihlemark\*, Carl-Magnus Sikström\*, Emile Biémont\*, Henri-Pierre Garnir\*, Xavier Tordoir\*, Pascal Quinet\*, Michael Schultz-Johanning\* and R. Schnabel\*.*

*\* Visiting scientists*

Time-resolved laser spectroscopy is a powerful method for the determination of excited-state lifetimes in atoms and ions. Experimental data are important in verifying theoretical calculations and for normalising transition probabilities required in astrophysics and plasma physics. With the wealth of new data provided by the Hubble Space Telescope, also in the vacuum ultraviolet spectral region, there is a great need to supply the corresponding atomic data. Our preferred experimental technique is to produce free atoms and ions in a laser-produced plasma, expanding in a vacuum system. By refining the techniques it has become possible to study up to triply ionized atoms.

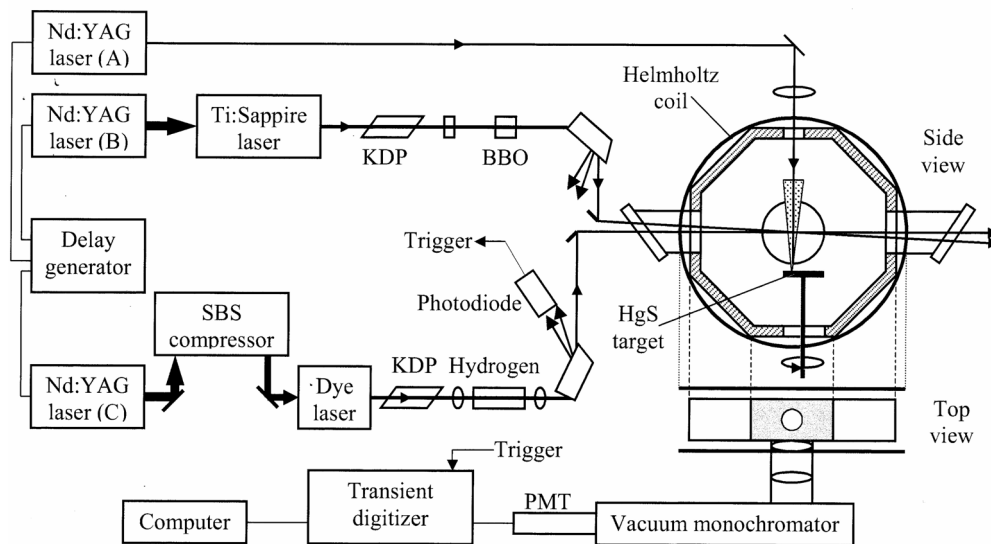
Even refractory elements (MoII, PdII, WIII) constitute no problem when using the laser ablation technique [A7-A9]. Experimental lifetime data and theoretical calculations were compared for BI, AuII, HgI and PbI [A10-A13]. For Pb [A13] also a large number of Landé factors were determined by Zeeman quantum beat techniques, and the data were analysed using multichannel quantum defect theory [A14-A15].

Extensive work has been performed on the rare-earth ions to meet the need for reliable data in astrophysics. Experimental lifetime determinations have been complemented by theoretical calculations using a relativistic Hartree-Fock method. The following rare-earth spectra have been studied: CeII, CeIV, PrIII, NdIII, SmIII, GdI, GdII, GdIII, TbIII, DyIII, HoIII, ErI, ErIII, TmI, TmII, TmIII, YbII, YbIII, and LuI [A16-A36]. The need to include core polarization effects was frequently very manifest. Transition probabilities for tens of thousands of lines have been

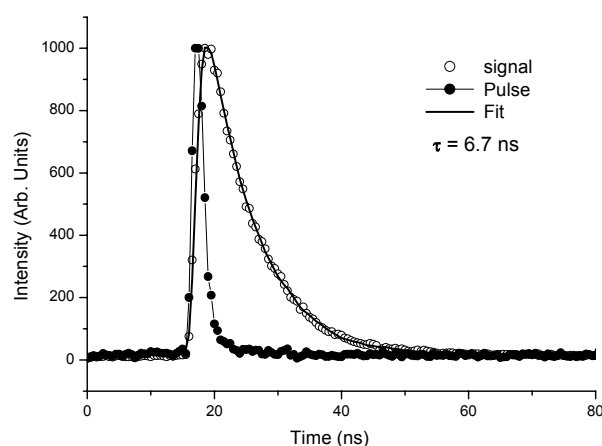
determined and the data are stored in the DREAM database at Université Mons-Hainaut: <http://www.umh.ac.be/~astro/dream.shtml>.

The iron spectral lines are very important in astrophysics. Within the FERRUM project we have performed extensive lifetime evaluations for the singly ionized atom [A37-A39]. We have also participated in experiments on lifetime measurements on metastable ions at the CRYRING facility in Stockholm, regarding  $\text{Fe}^+$  [A40] and  $\text{Ca}^+$  [A41], in the latter case employing blue diode lasers for the excitation of the stored ions.

Spectral lines in the heavy elements Th and U have been suggested for cosmochronological evaluations. Studies on ionic species of these elements were also successful, and extensive data have been analysed and published [A42-A45]. The research activities within this programme have also been reported at several conferences [A46-A49].



**Fig. A1.** Experimental set-up for measurements of lifetimes in atoms and ions generated in a laser-produced plasma. In the example chosen, Hg and S atoms and ions are formed from HgS.



**Fig. A2.** *Experimental recording of the fluorescence signal from the  $4f^{12}(^3H_6)6 p_{3/2}$  ( $29640.863 \text{ cm}^{-1}$ ) level in ionized erbium. The recorded data points are shown as open circles, while the filled circles show the laser pulse. The lifetime deduced by deconvolution fitting is 6.7 ns.*

## A2. High-order harmonic generation

High-order harmonic generation in gases, using short-pulse high-intensity lasers, is one of our main areas of research (for reviews, see [A50-A52]). We focus our research partly on the optimisation and characterisation of high-order harmonics [A53-A61], partly on applications of this novel source of coherent radiation in the extreme ultraviolet range to different areas in physics [A62-A71]. During the two years covered in this report, we have mainly focused our effort on developing a cross-correlation technique to characterize the high-order harmonics in time and frequency [A54-A56], using the kHz laser system of the Lund High-Power Laser Facility. We also present results from projects performed in collaboration with European colleagues, concerning:

- (i) the temporal confinement of high-order harmonics by manipulating the laser ellipticity in time [A57],
- (ii) studies of the effect of the relative polarization in the "sideband" intensities, (The sidebands are photoelectron peaks resulting from the ionization of an atom by absorption of a harmonic together with absorption or emission of a laser photon) [A58]
- (iii) first-order autocorrelation measurements of high-order harmonics, and
- (iv) lifetime determinations in nitrogen using harmonics of picosecond duration [A66-A67].

Finally, two optical set-ups adapted for the extreme ultraviolet region have been proposed. One is a SPIDER design (spectral interferometry for direct electric field reconstruction) using ponderomotive shearing [A60] and the second is a grating

monochromator allowing selection of one harmonic without stretching it in time [A61].

One of the goals of our efforts to characterize the high-order harmonics in time and frequency is to better understand the conditions in which harmonics are emitted from the generating medium as a train of attosecond pulses. To see evidence for such a train of attosecond pulses is our next ambition. A European network, "*Generation and characterisation of attosecond pulses in strong laser-atom interactions: A step towards attophysics*", was formed in 2000 by seven European laser laboratories, coordinated by us, to encourage work and collaboration in this area. Our work has been presented also at conferences and workshops [A76-A82].

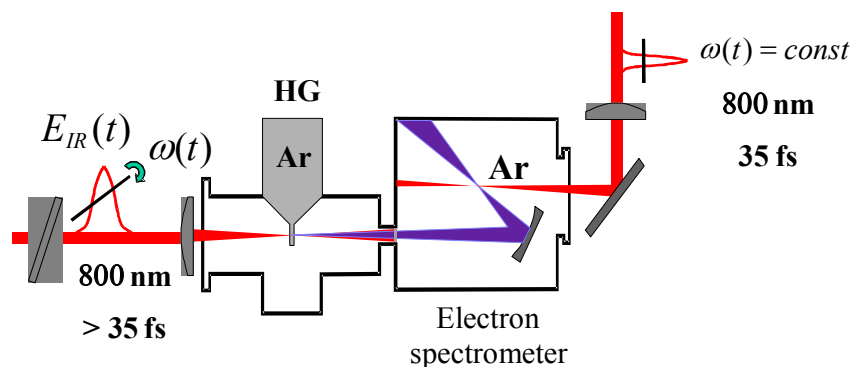
### Time-frequency characterisation of high-order harmonics

*Johan Mauritsson, Rodrigo López-Martens, Johan Norin, Per Johnsson, Allan Johansson, Anne L'Huillier, Mette Gaarde, Anders Persson, Claes-Göran Wahlström, Ursula Keller\*, Wouter Kornelis\*, Jens Biegert\* and Kenneth Schafer\**

*\*Visiting Scientist*

Our method is based on energy-resolved cross correlation between harmonics and a fraction of the 800 nm infrared laser pulse. When the two pulses overlap in time, sidebands appear in the photoelectron spectrum. By delaying the two pulses with respect to each other, the duration of the harmonic pulse can be inferred from the time-dependent amplitude of the sidebands. The location of the sideband in the photoelectron spectrum also depends on the delay between the pulses, and can be used to measure the time-dependent frequency of the harmonic.

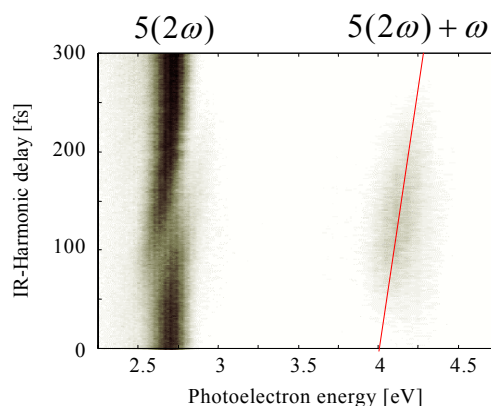
The harmonic radiation is generated by focusing the fundamental pulses from a 3 mJ, 1 kHz, 800 nm laser in a small cell containing argon gas. The weakly diverging harmonic beam is passed through a 200 nm thick Al filter, to eliminate the residual infrared light, and focused by a gold-coated mirror into the sensitive region of a magnetic bottle spectrometer (MBES) filled with argon. Before the laser is sent in to generate the harmonics, a fraction of similar energy, constituting the probe, is separated from the total laser output and re-routed through an independent delay stage and made to coincide in space and time with the harmonic beam inside the MBES (See Figure A3).



**Fig. A3.** Experimental set-up for the time-frequency characterisation of the harmonics

Our first experimental results [A54] were obtained by using harmonics generated by 50 fs 400 nm radiation and probed with 50 fs infrared radiation. The advantage of this configuration is that the sidebands from two consecutive odd harmonics do not interfere, allowing us to characterize one harmonic at a time. A typical result obtained on the fifth harmonic is shown in Figure A4. The trace on the left is the main peak, corresponding to ionization of Xe gas at the fifth harmonic of the frequency-doubled light; the trace on the right is a side-band peak, obtained by absorption of the fifth harmonic plus an infrared photon. As shown by the tilted line passing through the sideband signal, the energy of the photoelectron increases linearly as the time delay increases. In this case, this provides evidence of a linear positive chirp of the fifth harmonic. The right trace clearly shows that the main peak, corresponding to absorption at the fifth harmonic frequency, is also strongly influenced by the presence of the probe beam. The photoelectron signal is slightly broadened, and depleted in an asymmetric way, reflecting the shape of the side-band trace. Three cases in which different amounts of chirp were imposed on the fundamental beam by passing it through glass plates were investigated. We could impose positive as well as negative chirp on the fifth harmonic and measure it. These results led us to improve our experiment in order to be able to measure the chirp of higher order harmonics, with better characterisation of the pump and probe beams [A55-A56]. We present in Figure A5, results obtained with 40 fs infrared pulses used to generate harmonics 13-25 in argon and 40 fs probe pulses. In contrast to the results obtained with the frequency-doubled radiation, the observed sidebands now include contributions from the harmonics directly below and above, since they can be created through absorption of one harmonic plus or minus one laser photon. If the chirp induced were very different for two consecutive harmonics, the sidebands would exhibit a complex variation in energy and time delay. The present results indicate that the frequency variation is rather small and similar for the different harmonics.

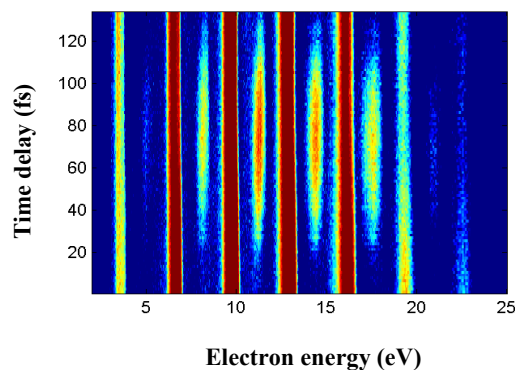
A limiting factor in these experiments is the long duration of the probe pulse, as well as the small (7 degrees) angle between the pump and probe beams. We have recently improved our set-up by setting pump and probe beams in an almost collinear



**Fig. A4.** Electron signal as a function of kinetic energy and time delay.

As shown by the tilted line passing through the sideband signal, the energy of the photoelectron increases linearly as the time delay increases. In this case, this provides evidence of a linear positive chirp of the fifth harmonic. The right trace clearly shows that the main peak, corresponding to absorption at the fifth harmonic frequency, is also strongly influenced by the presence of the probe beam. The photoelectron signal is slightly broadened, and depleted in an asymmetric way, reflecting the shape of the side-band trace. Three cases in which different amounts of chirp were imposed on the fundamental beam by passing it through glass plates were investigated. We could impose positive as well as negative chirp on the fifth harmonic and measure it.

These results led us to improve our experiment in order to be able to measure the chirp of higher order harmonics, with better characterisation of the pump and probe beams [A55-A56]. We present in Figure A5, results obtained with 40 fs infrared pulses used to generate harmonics 13-25 in argon and 40 fs probe pulses. In contrast to the results obtained with the frequency-doubled radiation, the observed sidebands now include contributions from the harmonics directly below and above, since they can be created through absorption of one harmonic plus or minus one laser photon. If the chirp induced were very different for two consecutive harmonics, the sidebands would exhibit a complex variation in energy and time delay. The present results indicate that the frequency variation is rather small and similar for the different harmonics.



**Fig. A5.** Electron signal as a function of time delay and energy

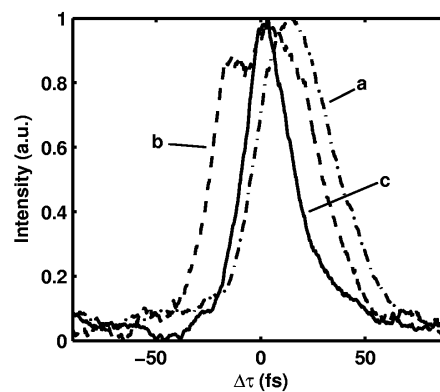
geometry as well as by using shorter probe pulses. In addition, the infrared pulses including the probe pulses have been characterized on-line by using SPIDERS. These sub-10-fs duration pulses are obtained by using self-phase modulation in a hollow fibre followed by compression with chirped mirrors. The cross correlation width of the sidebands is now much shorter than in the previous measurements, leading to pulse durations of the harmonics of the order of 20 fs. The harmonic chirps can now be determined with much better resolution.

### Time-gating of high-order harmonic pulses

*Rodrigo López-Martens, Johan Mauritsson, Allan Johansson, Per Johnsson, Anne L'Huillier, Olivier Tcherbakoff\*, Eric Mével\*, Armelle Zair\*, Jonathan Plumridge\* and Eric Constant\**

*\*Visiting scientists*

By modulating the polarization of 35 fs laser pulses from the kHz laser system in time, we were able to temporally confine the emission of harmonics 13 to 21 generated in argon [A57]. The technique used to modulate the ellipticity of the laser consists of transmitting the 35 fs driving pulses through two quarter-wave quartz plates, one multiple order, the other zero order, with their neutral axes oriented at 45 degrees to each other. The polarization of the created pulse sweeps from circular through linear back to circular. Using this arrangement, we could create a 7 fs gate where the laser ellipticity is less than 0.13, corresponding to a decrease in efficiency of a factor of two. The duration of the harmonics generated in argon was measured by cross-correlating the harmonic pulses with an ultrashort (10 fs) probe pulse (see above). Figure A6 presents the results obtained for sideband 18 in the photoelectron spectrum, which acts as a simultaneous probe for harmonics 15 and 17. Here, the confinement of the harmonics is shown in the case of the large and narrow gate configurations, as well as in the case when no time gate was implemented. The confinement of the harmonic emission due to the ellipticity modulation is clearly visible in the narrow-gate case. The durations of the harmonic pulses were estimated to be 61 fs (large gate), 41 fs (no gate) and 22 fs (narrow gate), with an uncertainty of about 10 fs. Moreover, we could continuously tune the duration of the individual harmonics by changing the orientation of one of the plates.



**Fig. A6.** Temporal confinement of harmonics 15 and 17 by ellipticity gating. (a) no gate (b) large gate (c) narrow gate.

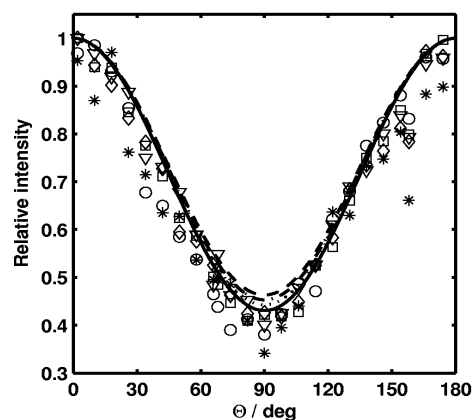


## Polarization effects in two-photon, non-resonant ionization of argon with extreme ultraviolet and infrared femtosecond pulses

Rodrigo López-Martens, Johan Mauritsson, Allan Johansson, Anne L'Huillier, Valérie Vénier\*, Richard Taïeb\*, Alfred Maquet\*, Michael Meyer\* and Patrick O'Keeffe\*

\*Visiting scientists and external collaborators

The two-photon ionization of the Ar 3p shell has been studied using high-order harmonics generated in argon with 40 fs, 815 nm laser pulses in combination with infrared photons from the same laser [A58]. The intensity variation of the sidebands has been studied as a function of the relative orientation of the electric field vector of both excitation sources. The sideband intensity was maximal when the polarization vectors are parallel and decreased by a factor of about 60%, almost independently of the harmonic order, when they are perpendicular (see Figure A7). The results agree well with theoretical calculations, which underline the importance of the relative phases of the harmonics as well as the phases and the angular momentum of the outgoing electron for the complete understanding of the observed polarization effect. We have also varied the degree of ellipticity of the infrared probe pulse, but did not find any significant variation of the sideband intensity.



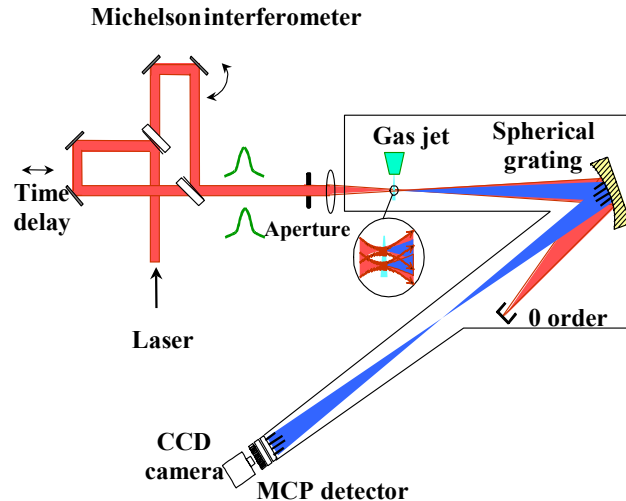
**Fig. A7.** Variation of the sideband intensity as a function of the relative polarization of the harmonic and the infrared laser pulse.

## First-order autocorrelation of high-order harmonics

Johan Norin, Anders Persson, Anne L'Huillier, Pascal Salières\*, Hamed Merdji\*, and Milutin Kovacev\*

\*Visiting scientists

Two spatially-separated sources of high-harmonic radiation created by the same laser pulse using, e.g., a Michelson interferometer with a slightly misaligned arm, are phase locked and interfere when superposed in the far field. An interferometer based on this principle has been built and used in many experiments, e.g. in the measurement of the temporal coherence, as well as for XUV interferometry of thin films and dense plasmas. It has recently been used to perform first-order field autocorrelation of high-order harmonics, by selecting a given region in the far field with a narrow aperture and by studying how the signal varies as a function of the time delay between the two pulses (Figure A7). High-order autocorrelation traces of the infrared laser pulse could also be recorded by superposing the two foci.



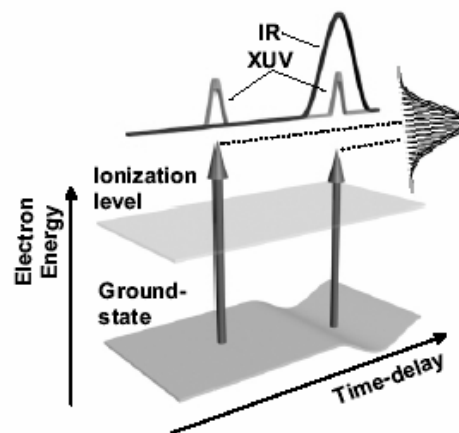
*Fig. A7. Principle of the XUV interferometer*

## Optical set-ups in the extreme ultraviolet range

*Johan Mauritsson, Ken Schafer, Karoly Osvay, Claes-Göran Wahlström, Anne L'Huillier, Johan Norin, and Rodrigo López-Martens*

A new method for the complete characterisation of ultrashort XUV pulses in time and frequency has been proposed [A60]. It is an adaptation of the SPIDER technique to this energy range. Two electron wavepackets are coherently produced by photoionizing atoms with two time-delayed replicas of the XUV pulse. For one of the pulses, photoionization occurs in the presence of a strong infrared pulse which ponderomotively shifts the binding energy, thereby providing the spectral shear needed to reconstruct the spectral phase of the XUV pulse. (See Figure A8.)

For many applications it is necessary to select radiation of only one particular harmonic order without affecting the ultrashort pulse duration. A three-grating monochromator meeting this demand has been designed and modelled by ray tracing as well as wave optical simulations [A61]. The only remaining temporal lengthening of the XUV pulse is due to pulse front distortion by the gratings and is predicted to be around 1 fs. The design has been successfully tested in the near-infrared region. Finally, the monochromator is also capable of eliminating any linear chirp in the harmonic pulses, therefore compressing them to shorter duration.



*Fig. A8. Principle of the XUV SPIDER*

## Pump-probe lifetime measurements in molecules

Allan Johansson, Anne L'Huillier, Claes-Göran Wahlström, Patrice Cacciani\*, Wim Ubachs\*, Rudiger Lang\* and Arjan Sprengers\*

\*Visiting scientists

The short pulse duration of the harmonic source makes it an interesting source for time-resolved studies of atoms and molecules in the VUV and XUV range. In addition, it can easily be synchronised with another colour, in the infrared, visible or ultraviolet range. Many two-colour pump-probe experiments have been performed during the past decade, in collaboration with European or Swedish scientists [A66-A71]. Recently, lifetime measurements have been performed in CO and N<sub>2</sub> using the 8th harmonic of the picosecond laser (produced by frequency-mixing of the fundamental and second harmonic generated in a KDP crystal) [A66-A68]. Some of the N<sub>2</sub> results, of astrophysical interest, are shown in the table below. Special attention was paid to the  $b^1\Pi_u$ ,  $v = 1$  state in <sup>14</sup>N<sub>2</sub> for which an exceptionally long lifetime was found. This result motivated new theoretical calculations, which are currently in progress.

State	$v$	Rotational lines	$\tau$ (ps)	$\lambda$ (nm)
$b^1\Pi_u$	1		2610±100	98.57
	6		380±40	94.91-94.93
	7	$R(0-2)$	550±40	94.236
		$R(5), Q(2)$	500±40	94.246
		$R(8), Q(5)$	320±40	92.262
		$R(11), Q(9), P(7)$	250±40	94.296
	8		95±70	93.51-93.52
9		≤50-110	92.88-92.90	
$c_3^1\Pi_u$	10		1320±150	91.89-92.30
	1	$R(0-13), Q(1-6), P(2-3)$	170±30	93.86-93.88
		$R(15), Q(10), P(6)$	97±70	93.90
$o_3^1\Pi_u$	1		≤50-110	92.88-92.90
	2		1150±150	91.26

**Table A1.** Lifetimes of the singlet ungerade states studied in <sup>14</sup>N<sub>2</sub>

### A3. Theory of strong-field dynamics

Lena Roos, Mette B. Gaarde and Kenneth J. Schafer\*

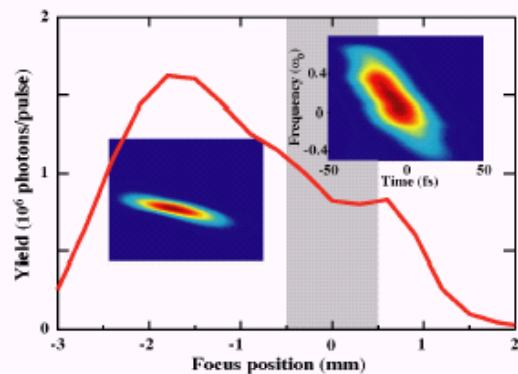
\*Visiting scientist

It has recently been experimentally demonstrated that high-order harmonics can be used to generate light pulses of sub-femtosecond duration. These attosecond pulses can be produced either individually or in a periodic train of pulses, via spectral manipulation of the high-harmonic radiation generated in the interaction between atoms and a strong laser field. In both cases it is important to characterize and control the temporal and spectral coherence properties of the harmonic radiation.

Our theoretical description of atoms interacting with strong laser fields is in two parts. We treat both the interaction between a single atom and the intense field, by direct numerical integration of the time-dependent Schrödinger equation, and the macroscopic effects of propagation and phase matching in the non-linear medium. The macroscopic harmonic field is calculated by numerically solving the Maxwell wave equation, using the atomic dipole moment as a source of the non-linear part of the polarization field. Combining these allows us to realistically model harmonic generation experiments.

### Characterization of the time-dependent phase of high-order harmonics

We have studied the temporal coherence properties of high-order harmonics through the theoretical characterization of their time-frequency behaviour [A72, A73]. The time-dependent frequency of the harmonics strongly depends on the macroscopic focusing and phase matching conditions, as shown in Figure A9. The figure shows the yield of the 27th harmonic generated in argon as a function of the relative position of the laser focus and the non-linear medium. The insets show the time-dependent frequency of the 27th harmonic for two different focusing configurations. In collaboration with the experimental group, we have also studied how a chirp on the driving laser pulse affects the time-dependent harmonic phase.



**Fig. A9.** *The yield of the 27th harmonic in argon is calculated as a function of position of the laser focus relative to the non-linear medium. The insets show the time-dependent frequency of the 27th harmonic for two different focusing configurations.*

To study the control of the harmonic output we have developed a tool to shape the (calculated) harmonic field to fit a specific application. We use an evolutionary algorithm to (for instance) minimize the spectral bandwidth of the harmonic profile, by changing the macroscopic phase matching conditions, such as the pressure or the size of the laser focus [A74].

### Phase locking of high-order harmonics

Only high order harmonics that are phase locked to each other can produce an attosecond pulse train. We have studied how phase locking is influenced by the interplay between the microscopic single atom dipole phase and the macroscopic phase matching conditions. We find that both the temporal and the spatial structures of the harmonics are important in achieving phase locking [A75]. Since the harmonics do not present a point source, true phase locking is only achieved when the different harmonics are phase locked at all points in their radial profiles.

This means that it is not possible to force harmonics to phase lock through purely temporal manipulation of their phases.

#### A4. Generation of hard X-rays from laser-produced plasmas

*Anders Sjögren, Michael Harbst, Claes Olsson\*, Sune Svanberg and Claes-Göran Wahlström*

*\*Department of Diagnostic Radiology, Lund University Hospital*

Since the establishment of the Lund High-Power Laser Facility in 1992, a programme on laser-produced hard X-rays and their applications has been continuously pursued. The meaning of *hard* X-rays differs between different communities, but here we mean photon energies above a few keV. In this section we describe some of our recent work on laser-based X-rays for medical imaging. Time-resolved aspects of laser-produced X-rays are discussed in the next section.

The X-rays are produced by focusing high-power femtosecond laser pulses tightly with an off-axis parabolic mirror onto solid targets of high-Z metals. With peak laser intensities on the target of up to  $10^{18}$  W/cm<sup>2</sup>, hot, dense plasmas are formed, emitting intense bursts of hard X-rays. The bremsstrahlung spectrum can extend up to the MeV region. The applications of such X-rays are numerous, but the most apparent is medical diagnosis: i.e. imaging of parts of the body and diagnosing diseases. One of the underlying aims of our X-ray work is to investigate the possibility of using laser-produced X-rays for medical imaging. The long-standing collaboration between scientists at the Lund High-Power Laser Facility and the Lund University Hospital makes comparative investigations of laser-based X-rays and radiation from the common X-ray tube feasible.

Most of our previous studies have utilised the 10 Hz terawatt laser. With that system, the feasibility of many interesting aspects has been explored. However, these studies showed that for laser-produced X-rays to become a realistic source for medical imaging, the average flux must be increased, but *not* the peak photon energies: almost all applications of X-rays require a high average flux of photons with energies below 100 keV. Instead of increasing the laser peak power, or



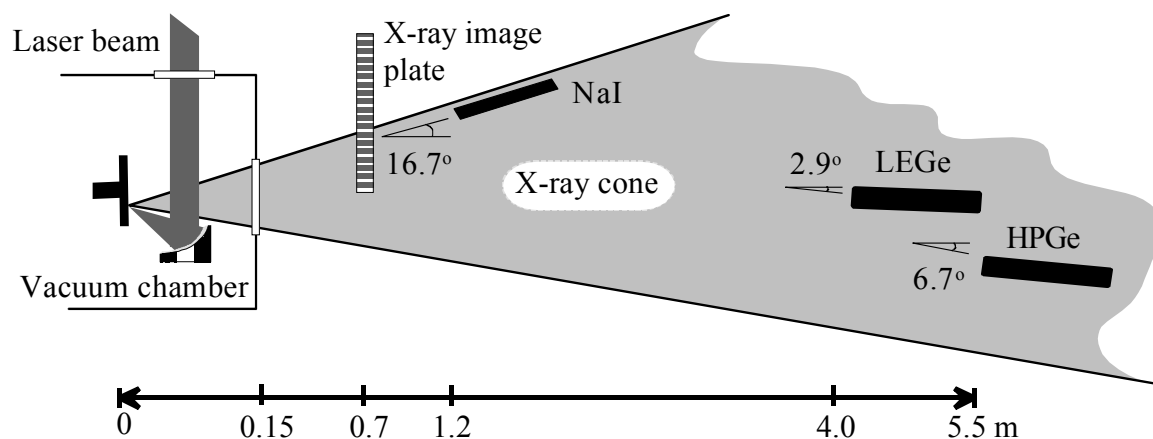
**Fig. A10.** A one-minute exposure of a sacrificed Wistar-Furth rat. The grey-scale extends from 2  $\mu$ Gy (white) to 13  $\mu$ Gy (black) over 256 levels.

intensity, the repetition rate should be increased. At present, the average emitted X-ray power from an X-ray tube is 10-1000 times higher than from a laser-produced plasma. In addition, the laser system must become more compact and user friendly. The aim of our most recent work, as described in A. Sjögren's thesis [A5], was therefore to investigate the use of a more compact laser system with less energy per pulse, but with a 100 times higher repetition rate (1 kHz).

When using the kHz laser we found that medical applications in which the properties of laser-produced X-rays can be beneficial are not far away. The laser-based X-ray source is not only an interesting alternative to the X-ray tube, it also exhibits important complementary features. For example, the short pulse duration might be used in combination with gated imaging to lower the absorbed dose to the patient, thus lowering the risk of radiation-induced diseases.

When evaluating the usefulness of the laser-produced X-rays for medical applications, a computed radiography (CR) system at the Lund University Hospital was used. The hospital utilises image plates, which are used to directly transfer image data to digital form in a computer system. Since the image plate response is proportional to the exposure over a wide range (at least 0.01 mR to 10 mR), the digitised data can be calibrated to the exposure or the absorbed dose provided that the X-ray spectrum is constant. This makes it possible to estimate the X-ray dose absorbed by the image plate when recording medical images, such as the one in Figure A10 [A5, A83, A85]

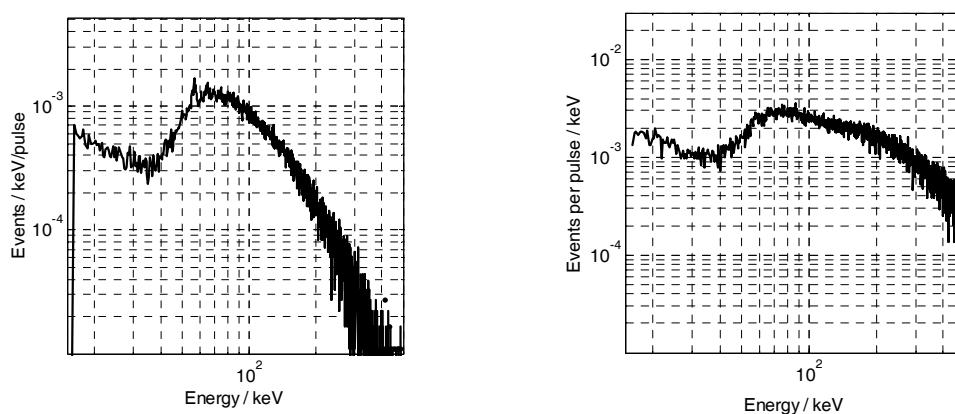
One of the main difficulties associated with energy-resolved measurements of laser-produced X-rays is pile-up. Most detectors are designed to record only one photon at a time, with a finite dead time between consecutive pulses. However,



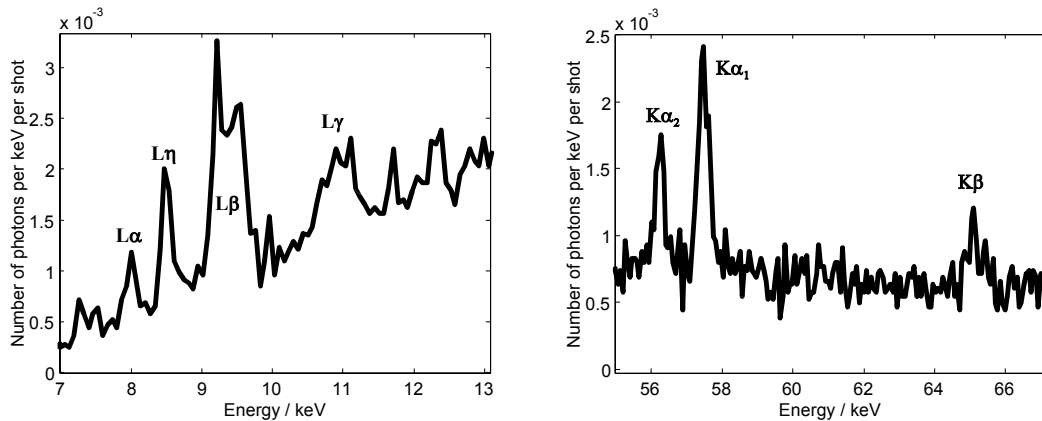
**Fig. A11.** The irradiated tantalum target with the laser-produced plasma is seen on the left. The X-rays are emitted more or less isotropically, but only a cone of X-rays will leave the vacuum chamber through its thin plastic window, as indicated. The lead bricks that shield the image plate and the Ge detectors have been omitted for clarity. The image plate, and its shield, block the path to the NaI detector but are only present when recording radiographs.

laser-produced X-rays are emitted in extremely intense, short bursts. This leads to a significant risk of many photons being detected simultaneously, but being interpreted as a single photon with an energy equal to the sum of the energies of all the photons detected. This problem requires special measures to be taken. The use of a crystal monochromator, in an alternative discussed in [A84]. We have previously also explored X-ray spectroscopy using a CCD camera, with each pixel acting as an individual detector. This allows a very large number of photons to be recorded per pulse, without pile-up. However, most CCD cameras have a limited spectral response ( $<20$  keV). In our most recent work the kHz laser was used. Its higher repetition rate, compared to the 10 Hz laser used in our previous studies, allowed germanium (Ge) detectors [A5, A83] to be used. In the set-up schematically drawn in Figure A11, two germanium detectors were used to sample different parts of the X-ray spectrum; a low-energy (LEGe) detector and a standard-electrode coaxial (HPGe) detector. The detectors are connected to individual multichannel analysers (MCAs). A NaI scintillator-coupled photomultiplier tube is used to assess the integrated X-ray spectrum on a shot-to-shot basis.

Figure A12 shows typical spectra recorded with the HPGe detector, with and without the effect of pile-up. Measuring the spectrally integrated X-ray yield with the NaI detector allowed us to control the effect of pile-up by rejecting X-ray bursts with too high (or low) flux, thereby accepting only pulses corresponding to a sufficiently low detection probability per shot, in spite of large shot-to-shot fluctuations. More detailed discussions on the measurements and pile-up can be found in Refs. [A5, A83]. Information is also available there on plasma



**Fig. A12.** Two X-ray spectra as they appear in the MCAs. The left spectrum was recorded over 540,000 laser pulses from the 1 kHz laser. The laser energy on target was 0.7 mJ per pulse and the pulse duration 25 fs. The right spectrum illustrates the effect of pile-up when the detection probability per laser shot is too high. In this case, the detection probability is higher than 50% per laser shot.



**Fig. A13.** Magnified views of a LGe spectrum. Left image: Characteristic L radiation from tantalum. It is possible that the peak structures between 10.5 keV and 13 keV are caused partly by L radiation from the lead aperture in front of the detector. Right image: Characteristic K radiation from tantalum.

temperatures as deduced from the spectral measurements. The spectra from the LGe detector, which has a better energy resolution, clearly indicate the existence of several characteristic emission lines. In Figure A13, characteristic lines of tantalum are visible.

## A5. Ultrafast X-ray science

In our research, we are applying ultrafast time-resolved X-ray probes as tools in solid-state physics and chemistry. This type of work will lead to the understanding of how light induced processes change the structure of matter. The dynamic properties of solids is due to atomic motion and the relevant timescale is that of a vibrational period ( $\sim 100 \text{ fs} = 10^{-13} \text{ s}$ ). This is the timescale on which molecular dynamics, chemical reactions and phase transitions in solids occur. In contrast to existing, fs laser probes, X-rays have a wavelength approximately equal to the distance between atoms, and hence enable atomic movement to be visualised directly. Thus, the natural technique for studying evolving atomic structures is through X-ray diffraction. Since their discovery, X-rays have been the dominant tool for determining atomic structures. We believe that the availability of time-resolved structural probes with sub-ps time resolution which employ X-ray diffraction, would enable groundbreaking work by allowing the investigation of rapidly changing structures. However, the lack of both ultrashort-pulse X-ray sources and ultrafast X-ray detectors has, until very recently, prevented such studies. During recent years, the development of pulsed X-ray sources has been rapid in the two main categories: laser-based table top sources; and sources based on particle acceleration (e.g. synchrotrons). Currently work is pursued in collaboration with research groups worldwide in order to utilise existing X-ray sources coupled to fs laser facilities. Our work involves both instrument development and basic science. It has been reported on in journals [A86-A91] and at conferences and workshops [A93-A95].



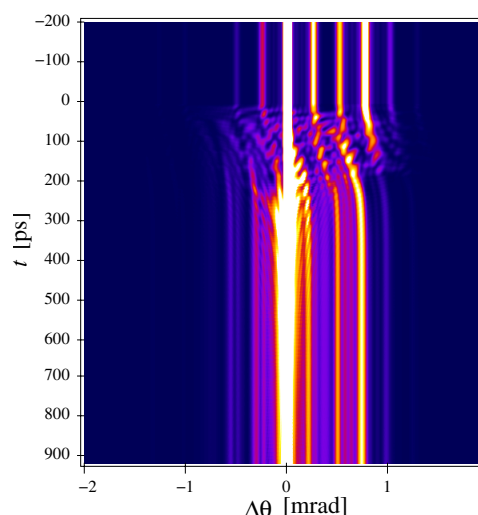
## Time-resolved X-ray diffraction studies of phonons in epitaxial semiconductor heterostructures

*Peter Sondhauss\*, Ola Synnergren, Michael Harbst, Adrian Allen\*, Kees Scheidt\*, Graham Naylor\*, Michael Wulff\*, Justin Wark\* and Jörgen Larsson*  
*\*Visiting scientists*

The fact that X-rays can resolve atomic positions makes Time Resolved X-ray Diffraction (TRXD) a powerful tool for the study of lattice dynamics, in particular for the study of non-thermal phonons. In the experiment presented below, coherent phonons in a superlattice, i.e. a stack of alternating epitaxial layers of different materials, were studied. The superperiodicity of the superlattice leads to the enhancement of certain phonon modes and allows generation of higher frequency phonons compared with a bulk crystal. This is due to the effect of “phonon branch folding”. This means that due to the composition of the superlattice, the phonon dispersion curves looks like those of the bulk crystal phonons, but the branches have been folded into a mini Brillouin zone. The size of the mini Brillouin zone is given by the superlattice period.

Coherent acoustic phonons were generated in a GaSb/InAs superlattice by ultrafast heating with a femtosecond pulse from a Ti:sapphire laser. The immediate thermal expansion launches an acoustic wave into the superlattice equivalent to a set of coherent phonons. Intense 16.45 keV X-rays from an undulator at the ESRF synchrotron were diffracted from this superlattice and recorded with an ultrafast streak camera. A prerequisite for this type of measurement is the synchronisation of the laser to the synchrotron clock, on the one hand, and the streak camera to the laser on the other. The first was achieved by tuning the cavity length of the laser oscillator, and the second by triggering the streak camera with the laser itself via a photoconductive switch.

The acoustic superlattice phonons can be modelled to a good approximation with an isotropic continuum model where phonon dispersion is neglected. In this model the acoustic wave equation has to be solved considering the boundary conditions at each layer interface. Figure A14 shows the simulated evolution of the (002) reflection of a 5-layer GaSb/InAs superlattice with 147 nm periodicity, irradiated by a laser pulse with 8 mJ/cm<sup>2</sup> at t=0. For t<0 the image shows the unperturbed reflection curve of the superlattice. At the centre the intense, fairly narrow GaSb



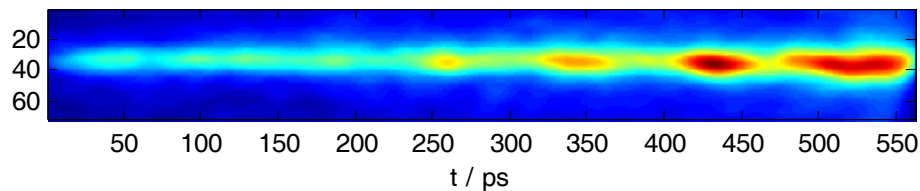
**Fig. A14.** The diffraction pattern, as a function of time, from a superlattice is complex and uniquely identifies the periodicity and composition.

substrate peaks can be seen. On the left and on the right are weaker peaks originating from the periodic superstructure. The onset of phonon oscillations at  $t=0$  is quite obvious. Certain modes are enhanced and manifest themselves as higher oscillation amplitudes at certain diffraction angles.

### Coherent control of acoustic phonons

*Ola Synnergren, Peter Sondhauss\*, Michael Harbst, Adrian Allen\*, Kees Scheidt\*, Graham Naylor\*, Michael Wulff\*, Justin Wark\* and Jörgen Larsson*  
\*Visiting scientists

After heating the surface of a semiconductor, an acoustic wave is launched into the crystal. The acoustic wave consists of the superposition of coherent acoustic phonons with a broad frequency bandwidth. By using a train of laser pulses to heat



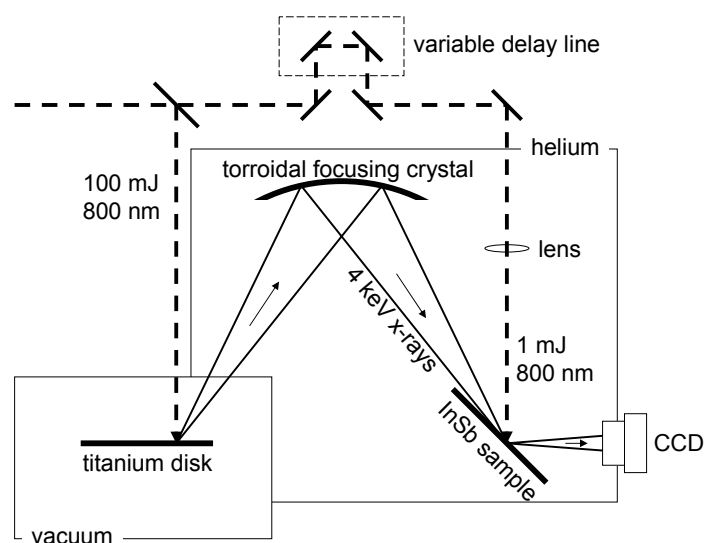
**Fig. A15.** *Experimental data showing how a selected phonon increases in amplitude after each pulse from the laser.*

the surface instead of a single pulse, certain phonon frequencies can be enhanced and others can be suppressed. This results in sidebands in the X-ray diffraction rocking curve. We investigated this feature during the summer of 2001 at ESRF in Grenoble, France. However, the two days of beam time were not enough to collect an adequate amount of data. Following the experiment, we simulated the experiments using a computer code developed by Sondhauss and Wark. The experiment has been redesigned and will be continued at beamline D611 at MAX-lab in Lund.

### Coherent phonons studied with laser-produced X-rays

*Peter Sondhauss\* Tue Hansen, Michael Harbst, Jörgen Larsson, Ola Synnergren, Ben Lings\*, Katarina Rosolonkova\* and Justin Wark\**  
\*Visiting scientists

We investigated the potential for monochromated laser-based K-alpha sources for the study of laser-irradiated crystals. A pump-probe scheme was used. The probe was a Ti K-alpha X-ray source, focussed with a toroidally bent crystal onto an InSb 111 crystal, which was pumped with  $\sim 10 \text{ mJ/cm}^2$  sub-picosecond light pulses, generating a coherent strain pulse in the crystal.



**Fig. A16.** Experimental set-up for coherent phonon studies with laser-produced X-rays.

We have observed a dramatic increase in spectral resolution compared with that which has been achieved in previous experiments using laser K-alpha sources [Rose-Petruck et al., Nature 398310 (1999)]. In these previous experiments it was only possible to see the effects of surface expansion, but due to the reduced source bandwidth size we can clearly see evidence of the coherent strain propagating into the bulk of the crystal.



**Fig. A17.** Laser-induced coherent strain wave propagation into an InSb crystal.

## Development of a dedicated time-resolved beamline at MAX II

*Tue Hansen, Ola Synnergren, Michael Harbst and Jörgen Larsson*

Beamline D611 is a bending magnet beamline dedicated to time-resolved studies. MAX II is a pulsed source operating at 500 MHz. Using a streak camera the duration of the pulses was measured and found to be approximately 150 ps. The beamline has a double-crystal monochromator. Until a toroidal focusing mirror has been installed, one of the crystals in the monochromator is cylindrically bent to

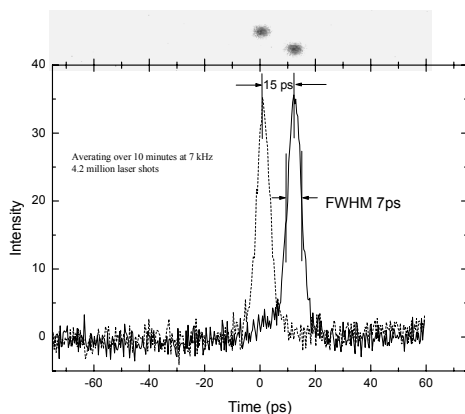
provide horizontal focusing. A laser providing pulses with a duration down to 30 fs has been synchronised to the ring, and a streak camera yielding sub-ps time resolution will be installed. The temporal resolution will not depend on the relative jitter between the laser and the synchrotron (10 ps) but rather the jitter between the streak camera and the laser. The laser operates at a maximum of 10 kHz, which sets the data accumulation repetition rate.

## Development of a low-jitter streak camera for beamline D611

*Tue Hansen, Kees Scheidt\* and Jörgen Larsson*

*\*Visiting scientist*

Ultrafast time-resolved X-ray diffraction at synchrotron facilities can be performed using a streak camera synchronised to the synchrotron pulses. During the past decade X-ray streak cameras with sub-picosecond time-resolution have been the subject of intense development. With the very high time-resolution it is usually necessary to accumulate many time-resolved signals at an adapted frequency to obtain a reasonable signal-to-noise ratio. The total time resolution of the detector is then a convolution of the intrinsic camera resolution and the trigger jitter. To reduce this trigger jitter a number of groups have implemented a photoconductive switch (PC switch) to trigger the sweep voltage on the deflection plates in the streak camera. We describe here initial tests with a laser-pumped GaAs PC switch used to trigger the sweep plates of a commercial Kentech X-ray streak camera undertaken at beamline D611. The laser is operated at 7 kHz frequency with a pulse length of  $<50$  fs at a central wavelength of 780 nm, and with an average output power of 5 W. Part of the main beam is split off and directed onto a GaAs PC switch used to switch a high-voltage pulse to the deflection plates. The laser



**Fig. A18.** The temporal resolution and jitter of the streak camera was measured using short-pulse UV-radiation. This image is the raw data and the lineouts are included to show the actual temporal resolution.

fluence on the PC switch is about  $57 \mu\text{J}/\text{cm}^2$ ; at this level the switch is saturated. The voltage ramp of the sweep can be up to 6 V/ps with a photoconductive switch which, together with the deflection sensitivity of the streak tube, provides a fundamental limit of the time resolution of the camera. In the work described here a UV laser pulse was generated by third-harmonic generation of the fundamental laser pulse. A gold cathode in the streak camera was used to convert the impinging third-harmonic laser pulse to electrons. A  $50 \mu\text{m}$  slit at the input of the streak camera defines the laser pulse size on the cathode. The electrons are subsequently accelerated in a

4.5 kV/3 mm DC electric field and focused onto a phosphor-coated screen at the

back of the streak camera. According to the manual the static deflection sensitivity of the tube is 17.6 V/mm at the output screen. An intensified CCD camera in close contact with the output coupling fibre plate is used to record the images on a computer. Figure A18 shows the recorded image of 4.2 million shots at 7 kHz. A 1 cm thick fused silica window is inserted half-way into the laser beam, giving a time delay between the part of the pulse that travels through the glass and the half that does not of about 15 ps.

### **Jitter measurement of two independent short-pulse sources**

*Vladimir Tenishev, Anders Persson and Jörgen Larsson*

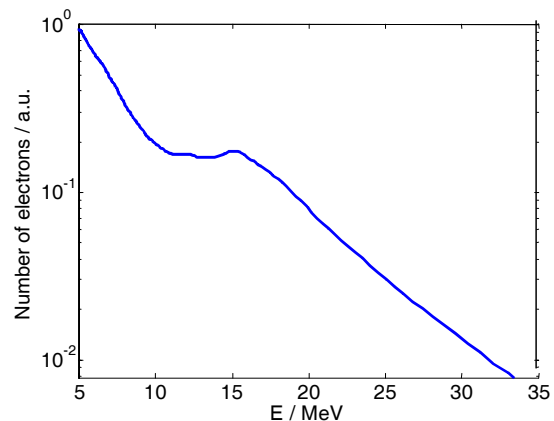
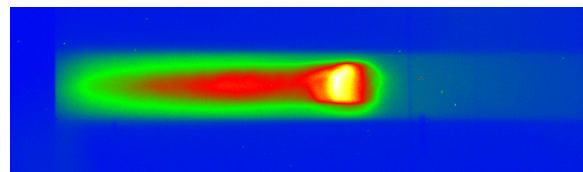
A method of measuring the jitter between a short pulse laser and a weak beam has been developed and tested in the laboratory at Max-Lab/LLC. This will be used as a clock signal between a SASE (Self-Amplified Spontaneous Emission) Free-electron laser and a femtosecond pump laser. Our laser energy temporal distribution measurement technique requires splitting the laser pulses to be measured into two replicas, which can be variably delayed with respect to one another. We measure intensity autocorrelation (ACC) or crosscorrelation by crossing the undelayed and delayed pulses in a second harmonic generation (SHG) crystal and detecting the SHG energy as a function of delay and the number of photons in one arm of the autocorrelator. In the final set-up spontaneous radiation from the electron bunch will be used, and the spatial position of the SHG signal will be linearly dependent on the relative delay of the laser and the electron bunch. Two types of detectors are being used: a diode array capable of 10 Hz operation and a position-sensitive detector with 100 ns responsetime. Particular care has been taken to ensure that the technique can be used with a photon flux as low as  $10^6$  photons in a 10% bandwidth, which we believe will be achievable at HASYLAB in Hamburg, where this device will be used.

## **A6. Relativistic laser-plasma interactions and particle acceleration**

The multi-terawatt arm of the 10 Hz terawatt laser has been used to study the interaction between intense laser light and target plasmas (produced by the leading edge of each laser pulse) at relativistic intensities, i.e. exceeding  $10^{18}$  W/cm<sup>2</sup>. The extreme longitudinal electric fields supported by plasmas can accelerate electrons to tens of MeV over short distances. This makes compact accelerators possible, and various particle acceleration mechanisms have been investigated.

At intensities greater than  $10^{18}$  W/cm<sup>2</sup>, free electrons oscillating in the laser field reach relativistic velocities, and the magnetic component of the laser field becomes very important. The increased relativistic mass of the oscillating electrons gives rise to a self-focusing effect, extending the laser focus into a channel of several Rayleigh lengths. A condition for the formation of channels is that the laser power exceeds a critical level,  $P > P_c$ . Experiments have been carried out to investigate how the channel length depends on laser parameters, such as pulse length and energy [A3, A5, A96, A97, A99]. In these experiments, the laser was focused on the edge of a high-pressure helium gas jet with an off-axis  $f/3$  parabolic mirror. It was found that the channel length decreased with shorter pulses at constant pulse energy, i.e. longer channels at lower power. An explanation of this, which has been tested with Particle-In-Cell (PIC) simulations, is pulse front erosion. Energy is drained from the front of the pulse at a rate proportional to the square root of the intensity until the power falls below  $P_c$  and channelling stops. Longer (less intense) pulses with the same total pulse energy, thus lose energy at a lower rate, allowing them to produce longer channels. Varying the sign of the chirp for the temporally stretched pulses did not affect the channel length, indicating that chirp-dependent loss sources, such as Raman forward scattering, were not very substantial.

Electrons can be accelerated to high kinetic energies in the plasma wake produced by the laser as it propagates through the medium. The energy distribution and the total number of these electrons have been characterized experimentally. Using a magnetic spectrometer, electron energies above 30 MeV have been recorded, with the total charge in each electron bunch found to be several nC [A5,A97]. In general, the energy spectrum is Maxwellian, but under certain conditions (to be investigated further) enhancement of the number of electrons at a given energy is observed. This is illustrated in Figure A19. This is a very interesting feature as it could confirm theoretical predictions (simulations) [Appl. Phys **B74**, 355, (2002)] suggesting that a monoenergetic electron beam can be produced by a very short, high-energy laser pulse.



**Fig. A19.** Energy distribution of laser-accelerated electrons. The top figure shows a false-colour representation of the fluorescence intensity on a scintillating screen behind a bending magnet. The lower figure shows the number of electrons per unit energy interval in the top figure. A non-Maxwellian distribution is clearly visible.

Focusing an ultra-high intensity laser onto a very thin film of solid material can produce an energetic beam of protons at the back of the target. The laser pulse ionizes the target, and accelerates the released electrons very abruptly in the forward direction. This results in a very high but transient electrostatic field between the electron bunch and the remaining positively charged plasma. This field, which can be of the order of TV/m, accelerates protons, or other positively charged ions present, in the forward direction. We have performed initial experiments, and observed beams of protons with energies up to 3 MeV. Not only very high intensity is required for this mechanism to be efficient, but the temporal pulse contrast of the laser is also a critical issue. Efforts are therefore currently being devoted to further improvements of this particular aspect of our multi-TW laser system.

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## B. Quantum Electronics, Quantum Optics and Solid State Spectroscopy

In this chapter we report on our work on observing single-photon interference using non-overlapping wave packets (Section B1), quantum optics and quantum computing in rare-earth-ion-doped inorganic crystals (Section B2) and diode laser development and the use of coherent transient techniques to explore all-optical techniques for data storage and data processing (Section B3).

### B1. Delayed interference for single photons

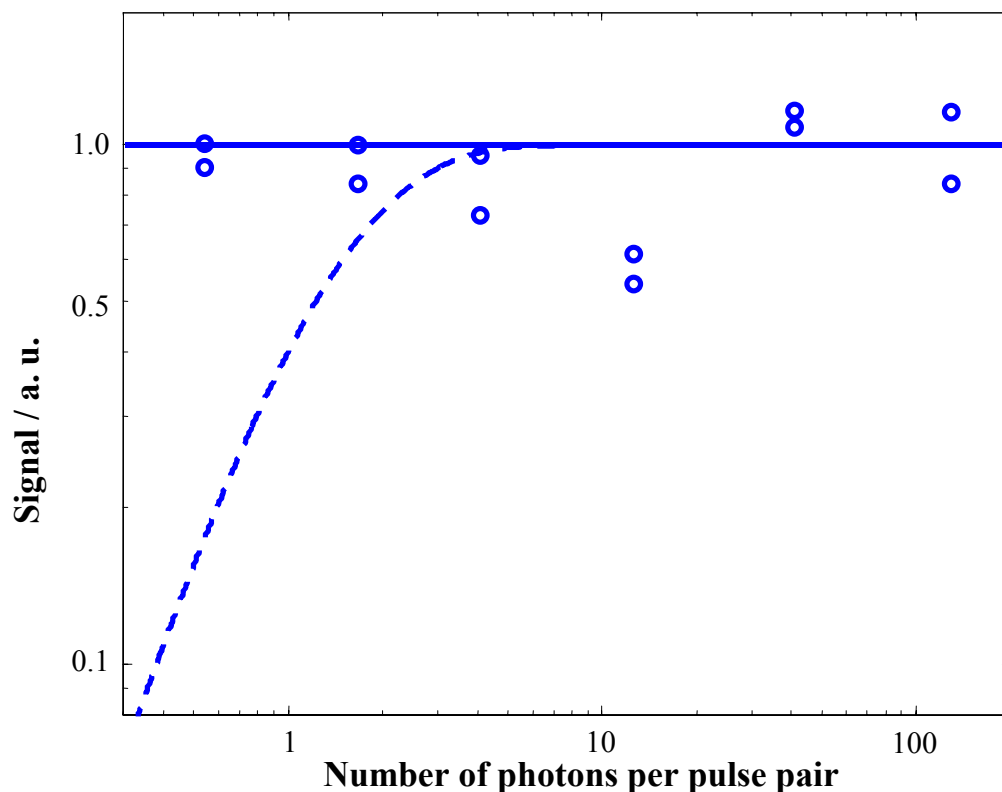
*Stefan Kröll, Serguei Moiseev\*, Mattias Nilsson, Nicklas Ohlsson and Robert Saers*  
*\*visiting scientist*

During the past two years we have been working on the realisation of an interference experiment for single photons that was originally suggested by Kessel and Moiseev (Kessel & Moiseev, JETP Lett. **58**, 80 (1993)). In the original proposal for the experiment, a wave packet consisting of a single photon was to be split into two by a beamsplitter. The two wave packets would then travel along two different paths to an absorbing material, as in a Young type double-slit interference experiment. If the two paths have different lengths so that the two wave packets did not reach the absorbing material at the same time, no interference pattern would normally be formed in the material. However, if the absorbers in the material are chosen so as to have coherence times for their optical transitions that are longer than the time separation between the wave packets, the interaction with the first wave packet will be stored in the wave functions of the absorbers, and these wave functions can interfere with the second wave packet to form an interference pattern in the material. This interference can later be detected by sending a strong read-out pulse into the material. If an interference pattern is present in the material, the read-out pulse will give rise to coherent emission of a light pulse from the absorbers in the material. This pulse will be sent out at a time after the read-out pulse that equals the time separation between the single-photon wave packets. In order for an interference pattern with sufficient contrast to allow for detection to be formed in the material, a large number of single-photon events must be accumulated in the material before read-out.

In the experiment, two wave packets separated in time give rise to an interference pattern in the frequency domain. This interference pattern is stored as a frequency-dependent modulation of the absorbers in the absorbing material. The interference can be described as a photon-echo process, which is a non-linear optical process. It is interesting to see whether this process can be performed with only a single photon acting as two of the optical fields involved in the process.



In our experimental realisation of the proposed idea, a few modifications were made compared with the original proposal. Instead of using a beamsplitter to create the two wave packets, two pulses were created by gating a CW laser. In order for an interference pattern to be formed as a result of the accumulation of many pulse pairs, the difference in phase of the light in the two pulses within each pair must be the same for all pulse pairs. To fulfil this, the time separation between the pulses in the pulse pairs must be shorter than the coherence time of the laser. The light in the pulses was then attenuated with the aim of reaching a level where only a total of one photon was present in the two pulses. The use of attenuated light from a laser instead of true single photon states means that the photon statistics in the pulses must be taken into account.



**Fig. B1.** *The experimentally obtained signal strengths for pulse pairs containing different average numbers of photons are shown (circles). The number of pulse pairs has been adjusted to keep the total excitation energy constant. The data have been modified to take the relaxation of the hyperfine levels into account. The solid trace shows the expected scaling of the signal if the delayed single photon interference effect exists, and the dashed trace shows the expected behaviour if at least two photons are required in each pulse pair.*

Different inorganic crystals doped with rare-earth ions have been tested as absorbing materials, and the best results were obtained for praseodymium ions doped into yttrium silicate ( $\text{Pr}:\text{Y}_2\text{SiO}_5$ ). The number of photons in each pulse pair has gradually been decreased [B1]. In order for the accumulation of several pulse pairs to work, the frequency of the laser light must be stable during the whole accumulation sequence. The long-term stability of the dye laser was improved by locking the frequency of the laser light to a hyperfine transition in molecular iodine [B2]. As a result of this, the energy in the pulse pairs could be further decreased [B3].

After having improved the set-up with better detection and better discrimination of stray light from the laser, the average number of photons per pulse pair could finally be reduced to 0.54 [B4]. In this final experiment,  $7 \cdot 10^9$  pulse pairs were accumulated during almost one hour before read-out. Each pulse had a duration of 44 ns and the time separation between the pulses in the pulse pairs was 175 ns. The accumulation time that can be used in the experiment is limited by the time that the interference pattern can be stored in the material, which for  $\text{Pr}:\text{Y}_2\text{SiO}_5$  is the relaxation time between the hyperfine levels of the ground state. To obtain the long accumulation time used in the experiment, this lifetime was increased by placing the material in a magnetic field of approximately 10 mT. The experimentally obtained signal strength for pulse pairs containing different numbers of photons can be seen in Figure B1. The data have been compensated for the relaxation time between the hyperfine levels and is compared with theoretical models where the delayed single-photon self-interference effect has been assumed to exist and not to exist.

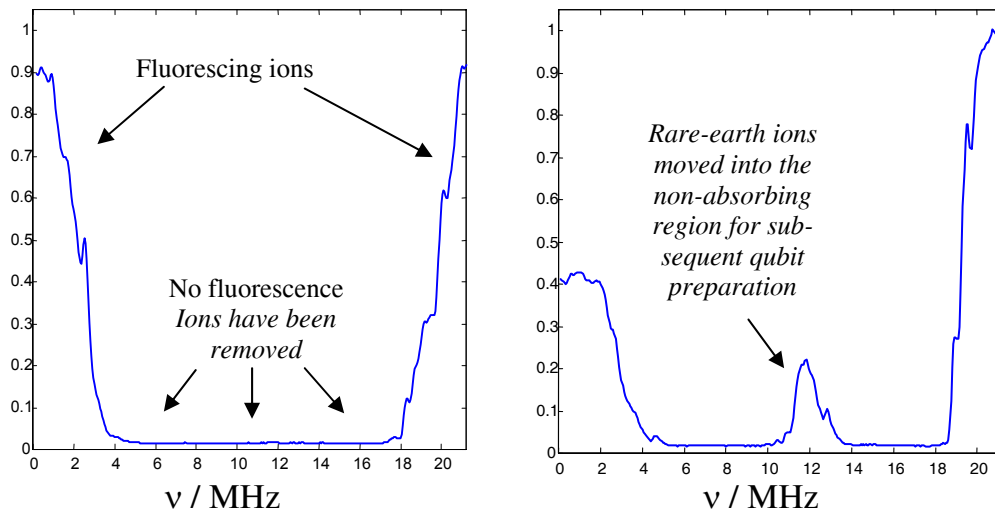
## **B2. Quantum computing and quantum optics in rare-earth-ion-doped inorganic crystals**

*Tomas Christiansson, Marito Forsberg-Olsson, Stefan Kröll, R. Krishna Mohan, Serguei Moiseev\*, Mattias Nilsson, Nicklas Ohlsson, Markus Persson, Lars Rippe, Ingela Roos and Fredrik Vestin*

*\*visiting scientist*

Rare-earth-ion-doped inorganic crystals have properties that make them suitable for experiments on quantum optical phenomena and quantum information processing, in particular, the long phase memory of the absorbers at cryogenic temperatures, and many new ideas for such experiments have emerged [B5]. We have initiated a research programme, including international collaboration, aimed at the experimental realisation of quantum computing in these crystals.

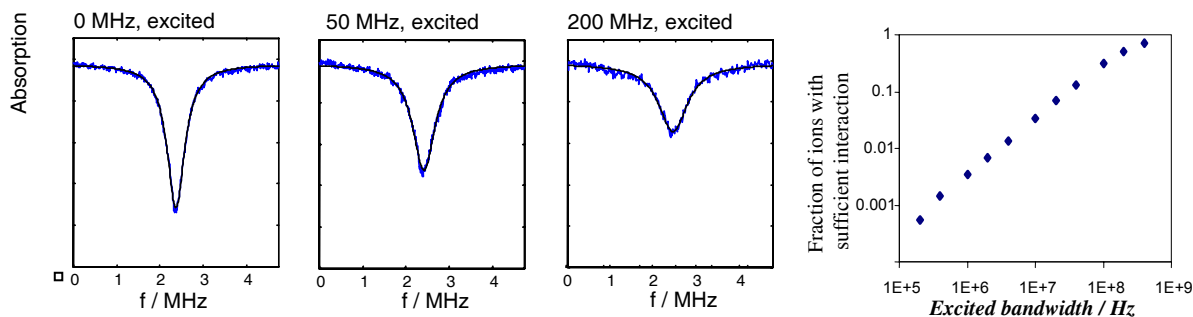
We have presented a scheme for generating qubits and demonstrating quantum logic gates in rare-earth-ion-doped crystals [B6]. The qubits were chosen to be two of the ground-state hyperfine levels of the dopant ions. These hyperfine levels can



**Fig. B2.** Preparation of an ensemble of ions that is addressable by frequency. Left, a spectral interval is emptied of absorbing ions, through optical pumping. Right, a group of ions, absorbing from a single qubit state, is moved into the region.

have lifetimes of hours, and decoherence times of ms or longer. Different qubits consist of ensembles of ions absorbing light in different spectral regions, within an inhomogeneously broadened absorption line. Controlled logic between the qubits can be accomplished using the change permanent dipole moment that occurs in an ion when it makes an optical transition between the ground and excited state. We believe that a system with several interacting qubits can be set up using existing techniques and technology.

Our work is progressing towards the experimental realisation of this quantum computing scheme [B7-B10]. The initial experiments have shown that it is possible to move the ions between the different hyperfine levels in a controlled fashion (see Figure B2) [B11] and we have demonstrated the physical mechanisms for qubit interaction [B12, B13]. Figure B3 shows the broadening of a spectral hole when ions absorbing at other frequencies are excited.



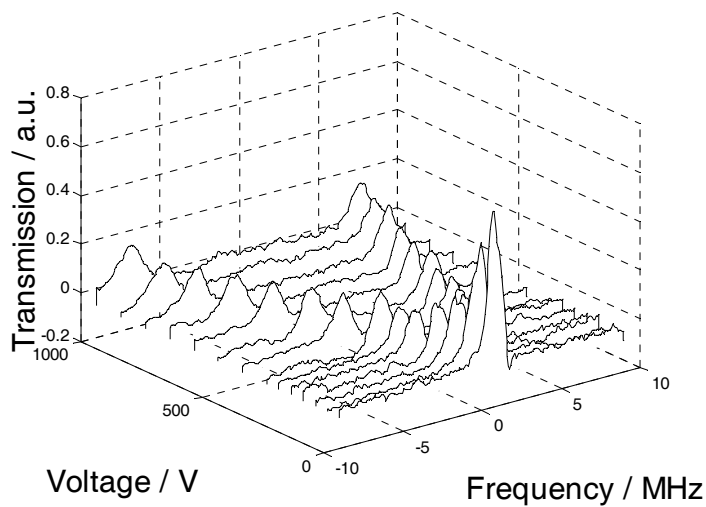
**Fig. B3.** Broadening of a spectral hole. The absorption frequencies of the ions shift when ions within a 50 or 200 MHz interval elsewhere in the absorption profile are excited.

**Fig. B4.** The simulations show the fraction of ions shifted enough to enable controlled logic.

The broadening occurs because of the dipole-dipole interaction which also is the mechanism that creates the entanglement in the quantum computing scheme.

The experiments have been complemented with simulations as shown in Figure B4 showing that a sufficiently large fraction of the ions interacts sufficiently strongly to be used for the quantum gate operations. Theoretical investigations have also been made of how the quantum systems can be controlled using optical pulses [B14].

The existence of a permanent electric dipole moment is essential to the quantum computing scheme since this is the basis of the mechanism for interaction between qubits. This has been explored by applying an external electric field to a Eu:YAlO<sub>3</sub> crystal. Figure B5 [B15] shows a spectral hole that splits in an electric field because the permanent dipole moment can have two different directions.



**Fig. B5.** A spectral hole split into two components when an external electric field is applied and increased. The hole was probed using transmission measurements.

Another important material property is the coherence of the hyperfine levels (spin states) that are used as qubit states. Coherent superpositions of the spin states have been created and detected using various types of Raman heterodyne detection [B16]. From these the dephasing time of the states have been calculated. The spin states have also been examined with respect to transition strengths and selection rules for optical transitions between different spin states in the ground and excited states [B17]. Among other things, this may be relevant for the implementation of quantum optical experiments that have previously been performed using ensembles of atoms in gases and in rare-earth-ion-doped crystals.

Closely related to the work described above and the work presented in Section B1 is a scheme we have presented in which the state of a single-photon wave packet can be completely reconstructed after being absorbed by a gas with a Doppler-broadened transition [B18].

### **B3. Time-domain optical data storage and processing**

*Stefan Kröll, Mattias Kuldkepp, R. Krishna Mohan, Mattias Nilsson, Nicklas Ohlsson and Lars Rippe*

The time-domain optical data storage and processing project is concerned with the physics and the concepts of photon-echo-based techniques for optical storage and processing of information. Data storage densities above Gbits/cm<sup>2</sup> (Opt. Lett. **20**, 1658 (1995)), and data rates above THz (Opt. Lett. **20**, 749 (1995)) have been achieved using photon-echo-based techniques, and storage densities >Tbits/cm<sup>2</sup> have been predicted. The projected and demonstrated performance makes photon-echo techniques extremely interesting for future optical storage and processing concepts. However, many problems must be addressed and solved in order to make time-domain optical storage and processing competitive with existing technology. We have recently written a review paper describing and discussing the photon-echo data storage and processing experiments that have been performed worldwide during recent years [B19].

In time-domain or photon-echo storage many bits of information can be stored and addressed within a single diffraction-limited point (area) because atoms within this small area absorb at different frequencies. In some rare-earth-ion-doped materials, more than 10<sup>7</sup> different spectral intervals can be addressed within any such single spatial point. This is the basis for the high storage densities that have been achieved and predicted in these materials. In the time-domain or photon-echo approach to data storage and processing in these materials it is the frequency Fourier spectra of temporal wave forms (data streams) that are stored. This makes it possible to perform a variety of all-optical operations on temporal data, for example, temporal pattern recognition, header/address decoding, optical time domain encryption, bit rate conversion, serial-to-parallel and parallel-to-serial conversion.

#### **Development of frequency-agile external-cavity diode lasers for optical processing and space-based microwave radiation detectors**

As information is stored and sometimes also addressed in the spectral domain, suitable frequency-agile lasers are very important in implementing many of the ideas in photon-echo storage and processing. Rapidly chirping the laser over large frequency intervals can increase the data storage rate. There are several other interesting aspects regarding the use of chirped pulses to generate photon echoes. We have used this type of laser for all-optical conversion of data bit rates. It is also possible to perform a spectral analysis of the stored input signal. Individual Fourier frequency components can be read with a chirped input pulse as it is the frequency Fourier spectrum of a temporal input sequence that is stored. In a project supported by the European Space Agency (ESA) we have co-operated with Jean-Louis

LeGouët's group at Laboratoire Aimé Cotton, France. The objective of this project was to construct a broadband analyser of microwave signals based on spectral hole-burning/photon-echo materials. This analyser is intended to be carried by satellites on scientific missions. Our task in this project was to construct a first prototype of an external-cavity diode laser which features rapid frequency scanning and good frequency stability, that can be used for spectral selection of the different microwave signals. This device that has been constructed within this project has a 50 GHz single-mode continuous tuning range without mode hops, it can be tuned at rates up to 2 GHz/ $\mu$ s, and it has a frequency stability of 1 MHz over a 1 ms timescale [B20, B21].

Using a previously designed frequency-agile external-cavity diode laser source we demonstrated multiple bit storage at a single spatial location and pulse conversion. We believe these experiments can be significantly improved using the new diode laser. In addition, both the bit-rate conversion and the multi-bit storage would benefit from higher laser powers, and a laser amplifier based on a broad-area laser diode that can be operated together with the new external-cavity diode laser has been constructed [B22].

The data storage work we have performed was carried out in a Tm-doped YAG crystal and, in fact, most of the recent work in photon-echo data storage has been carried out in this material. While this material has many favourable properties the storage time of the material is only about 10 ms. However, we have shown that by placing the Tm YAG crystal in a magnetic field it is possible to increase the storage time by at least two orders of magnitude [B23]. This can have significant effects on many of the applications that have been discussed for this crystal.

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## C. Applied Molecular Spectroscopy

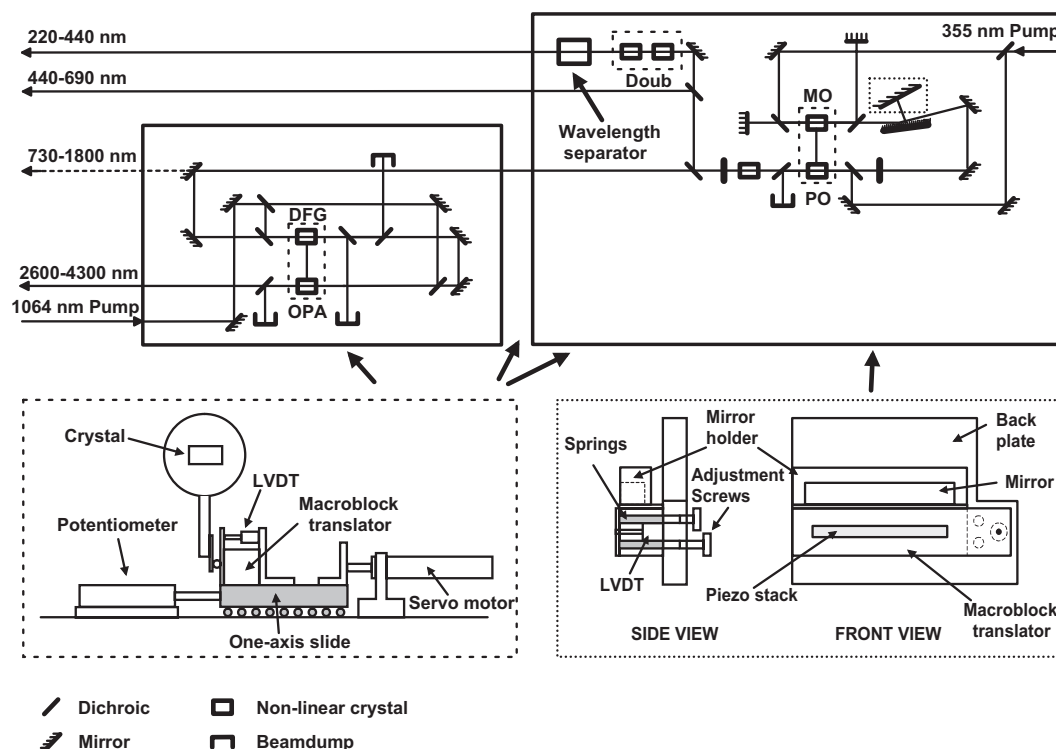
Research projects in the field of applied molecular spectroscopy are directed towards the development and application of optical techniques, both laser and non-laser, to detect mainly gases and particles in various environmental studies. Several of the techniques are used for remote sensing over long distances. The method most used in studies of air pollutants is the differential absorption lidar (DIAL) technique, where a mobile DIAL system has been employed in several field campaigns. The lidar system has also been used in the remote monitoring of environmental effects on historical stone buildings and monuments using laser-induced fluorescence (LIF) techniques, and special multicolour imaging methods have been developed for this purpose. The method of imaging gas flows using an IR camera and gas correlation techniques has been further pursued. The aim here is to develop a method that can produce images of a certain gas at a certain location using only the thermal background radiation. Finally, ultrasensitive gas measurement methods are being developed in a project utilising diode laser spectroscopy and frequency modulation (FM) techniques. During the period, one PhD thesis has been completed within the group [C1], and results are also included in two other theses [C2,C3].

### C1. Lidar measurements of atmospheric gases

*Christoffer Abrahamsson, Hans Edner, Mikael Sjöholm, Sune Svanberg, Johannes Swartling and Petter Weibring*

An all-solid-state, fast-tuning lidar transmitter for range- and temporally resolved atmospheric gas concentration measurements has been developed and thoroughly tested [C4,C5]. The instrument is based on a commercial OPO (Optical Parametric Oscillator) laser system which has been redesigned with piezoelectric transducers mounted on the wavelength-tuning mirror and on the crystal-angle-tuning element in the OPO, as shown in Fig. C1. Piezoelectric transducers similarly control a frequency-mixing stage and doubling stage, which have been incorporated to extend system capabilities to the mid-IR and UV regions. The construction allows the system to be tuned to any wavelength, in any order, in the range of the piezoelectric transducers on a shot-to-shot basis. This extends the measurement capabilities far beyond the two-wavelength DIAL method and enables simultaneous measurements of several gases. The system performance in terms of wavelength, linewidth and power stability is monitored in real time by an etalon-based wavemeter and gas cells. Tests showed that the system was able to produce radiation in the 220-4300 nm wavelength region, with an average linewidth better than  $0.2 \text{ cm}^{-1}$  and a shot-to-shot tunability up to  $160 \text{ cm}^{-1}$  within 20 ms. The utility of real-time linewidth and wavelength measurements is demonstrated by the ability to identify occasional poor-quality laser shots and to discard these measurements. Also absorption cell measurements of methane and mercury demonstrate the ability

to obtain stable wavelength and linewidth during rapid scans in the mid-IR and UV regions. Earlier measurements of  $\alpha$ -pinene gas, a biogenic volatile organic compound, have now been published [C6].

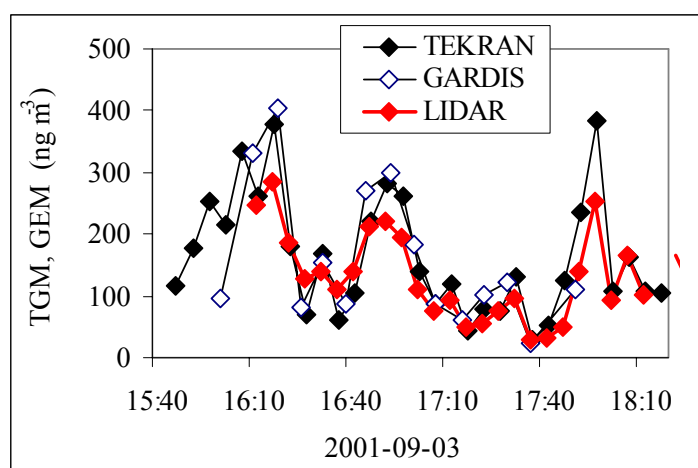


**Fig. C1.** Optical arrangement for the modified OPO and the mixing unit. The upper right part of the figure shows the layout of the OPO. It consists of a Littman cavity (MO), creating narrow-band photons, which are seeded into an unstable cavity (PO), generating high output power. The output radiation can either be sent out directly or frequency doubled in an integrated doubling stage. The upper left part of the figure shows the mixing unit consisting of a Difference Frequency Generation (DFG) stage, followed by an Optical Parametric Amplifier (OPA) stage. To enable fast tuning the Littman cavity mirror in the OPO is modified according to the lower right part of the figure, while all crystal holders are modified according to the lower left part of the figure.

Most DIAL measurements are made on single compounds by switching the transmitted beam between two wavelengths. The use of more than two wavelengths is a mathematical necessity for simultaneous measurements of multiple species, or for resolving interference effects between a compound of interest and a background gas, such as water vapour or carbon dioxide. Especially in the mid-IR region there are many important hydrocarbon compounds which have fundamental rotational-vibrational transitions that require a multiwavelength approach. Multivariate statistical techniques have been explored for the analysis of the composition of hydrocarbon mixtures from their combined absorption spectrum [C7,C8]. Generic Algorithm (GA) and Partial Least Squares (PLS) techniques have been successfully

implemented and tested. The tests were carried out with lidar signals from a remote, open-ended flow cell emitting light hydrocarbons such as methane, ethane and propane. When larger hydrocarbons are studied, the spectral overlap will be more severe and it can be difficult to determine the individual concentrations of the different hydrocarbons, without increasing the number of wavelengths substantially. Recording of whole spectral structures over hundreds of nanometers is not possible due to prolonged measurement time and possible shorter time constants in the pollutant emission and atmospheric aerosol distributions. Instead, establishing the concentration of different groups of hydrocarbons, using fewer wavelengths, could be a useful approach.

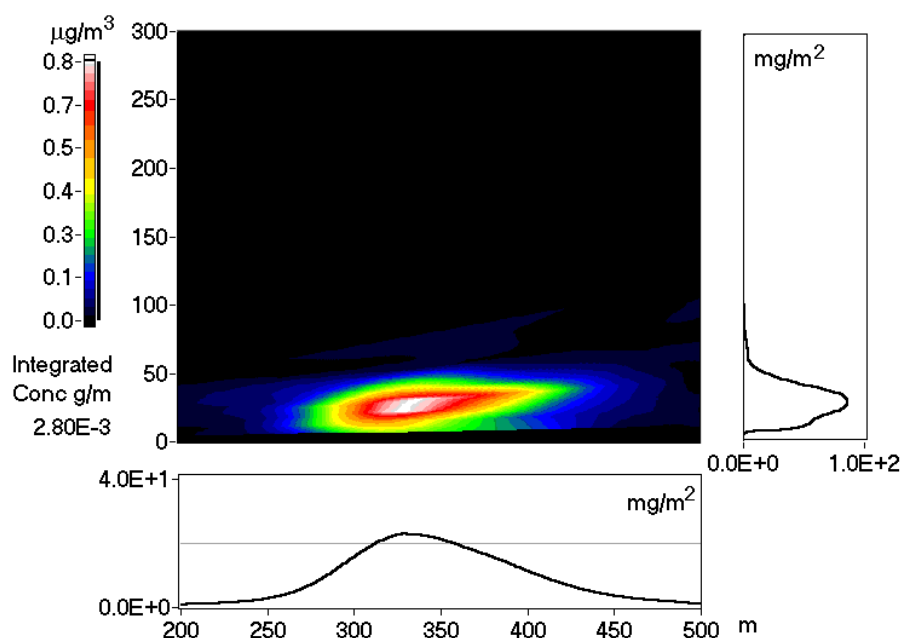
The mobile lidar system is being used in a large cross-disciplinary project, financed by the European Commission, concerning European mercury emission from chlor-alkali plants (EMECAP). The aim of the project is to provide decision makers with an improved tool for evaluating the risk to the human health and the environment around mercury cell chlor-alkali (MCCA) plants. During the period of the project, April 1, 2001 to March 31, 2004, environmental, as well as epidemiological, studies around the pollutants are being performed. The environmental issues concern measurements of mercury in fish and vegetables, as well as point monitoring of atmospheric mercury concentrations around the MCCA plants. The lidar system is used to measure the flux of elemental mercury from the plants. The results obtained with the system will serve both as input to an improved mathematical dispersion model for mercury, and a large database to which smart software for correlating different parameters of interest will be applied by other partners within the project. During the first field campaign in August-September 2001 at the EKA Chemicals chlor-alkali plant in Bohus, Sweden, an inter-comparison between the lidar system and two point-monitoring systems was performed, see Fig. C2 [C9].



**Fig. C2.** Mercury time series obtained using three different measurement techniques.

The point monitors were based on gold amalgamation of the mercury followed by thermal desorption and atomic fluorescence spectrophotometry (TEKTRAN) and

atomic absorption spectrophotometry (GARDIS) and were placed as close to the lidar light beam as possible. Keeping in mind that the sampling volumes of the instruments were slightly different, the agreement between the concentrations measured by the different instruments is good. Apart from the first field campaign in 2001, winter campaigns in Bohus and Rosignano Solvay, Italy, have been performed during 2002 [C10,C11]. Each campaign lasted about two weeks and provided long time series of mercury flux data that will serve as input in other parts of the EMECAP project. The mercury flux is obtained by scanning the lidar light beam in 7-15 directions in vertical planes downwind from the main cell house at the plant, resulting in vertical 2-D concentration maps (see Fig. C3). The concentration ( $\mu\text{g}/\text{m}^3$ ) in the 2-D map is then integrated to give an integrated concentration ( $\text{g}/\text{m}$ ) which, multiplied by the wind component perpendicular to the 2-D map, gives the mercury flux ( $\text{g}/\text{h}$ ). During the summer of 2003 field campaigns will be performed in Sweden, Italy and Poland.



**Fig. C3.** A vertical concentration distribution acquired with the lidar system at the chlor-alkali plant in Rosignano Solvay on February 2, 2002.

The emission of sulphur dioxide from the Italian volcanoes Mount Etna and Stromboli has been studied through ship-borne underpasses of the volcanic plumes [C12-C14]. Active DIAL measurements were compared with passive remote sensing techniques. All these techniques utilise the spectroscopic signature of the gas in the wavelength region around 300 nm. The differential optical absorption spectroscopy (DOAS) technique provided spectrally resolved absorption spectra. In one configuration the absorptive imprint in the sky light, recorded with a vertically looking telescope was studied, while a different DOAS implementation utilised the sun disc as the light source in slant-angle, long-path absorption measurements. Parallel measurements with the customary correlation spectroscopy (COSPEC)

method were also performed. It was found that there is general agreement between the DIAL and the sun-tracking DOAS results, while the COSPEC and the vertically looking DOAS system employing the diffuse sky radiation also showed internal agreement. However, the two groups of results exhibited a discrepancy of a factor of about 0.9 to 1.6, with the instruments employing diffuse sky light usually overestimating the total overhead SO<sub>2</sub> burden. This is in agreement with earlier observations. Path length simulations of the down-welling radiation through the plume at various heights and azimuth angles of the sun were performed. A scattering model was developed using Monte Carlo simulations of traced photon histories, and solutions to the transport equation for the atmospheric scattering conditions pertaining to the measurement situation provide the necessary correction factors to allow straightforward and convenient passive measurements for improved volcanic SO<sub>2</sub> assessment. The modelling can be further improved by simulations of the exact plume shape in three dimensions yielding even better agreement, but the expected error reduction is of the same magnitude as the present measurement error and the result cannot be used by passive-instrument users due to the fact that it is difficult to access the 2-D geometry surface without an active system. Clearly, the correction factors will depend on the scattering conditions, which leads to a residual uncertainty.

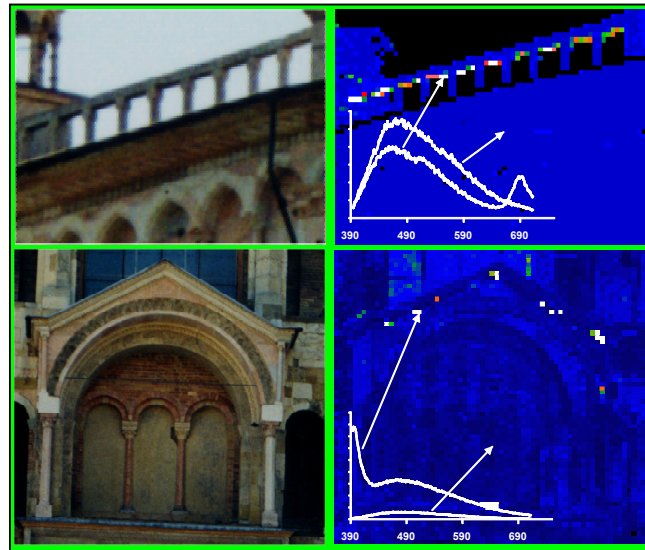
## C2. Fluorescence lidar applications

*Hans Edner, Thomas Johansson, Sune Svanberg and Petter Weibring*

The fluorescence lidar technique offers a powerful means for the monitoring of the surface of the sea, of terrestrial vegetation and of the weathered facades of historical buildings and monuments. While the distributed scattering along the laser beam is analysed at selected wavelengths in atmospheric lidar studies, the spectral content of the fluorescence echo from topographic targets is utilised in the fluorescence lidar approach. The mobile lidar system can be adapted for laser-induced fluorescence measurements on solid and liquid targets. For excitation, the frequency-tripled output at 355 nm of one Nd:YAG laser is particularly suitable. High pulse energies of up to hundreds of mJ can be obtained, but normally the energy is limited to tens of mJ. With the availability of a wide variety of excitation wavelengths from the OPO transmitter, additional discriminating power in fluorescence analysis, of e.g., different types of vegetation or different types of surface materials in historical buildings, is obtained.

Fluorescence lidar measurements have been performed on the cathedrals of Lund and Parma [C15-C19]. Here the 355 nm laser beam is directed towards the building wall from a distance of 50-100 m, and the induced fluorescence is detected by the receiving telescope. The fluorescence from the illuminated spot is then imaged onto the entrance of an optical fibre assembly and conducted to the entrance slit of a spectrometer. An image intensifier in front of the CCD detector is gated to match the arrival time of the signal. The beam is swept row by row till a full matrix covering a certain area at the target is covered. As an example an 8 x 8 m<sup>2</sup> area of the facade of the cathedral in Parma, Italy, is shown in the lower part of Fig. C4.

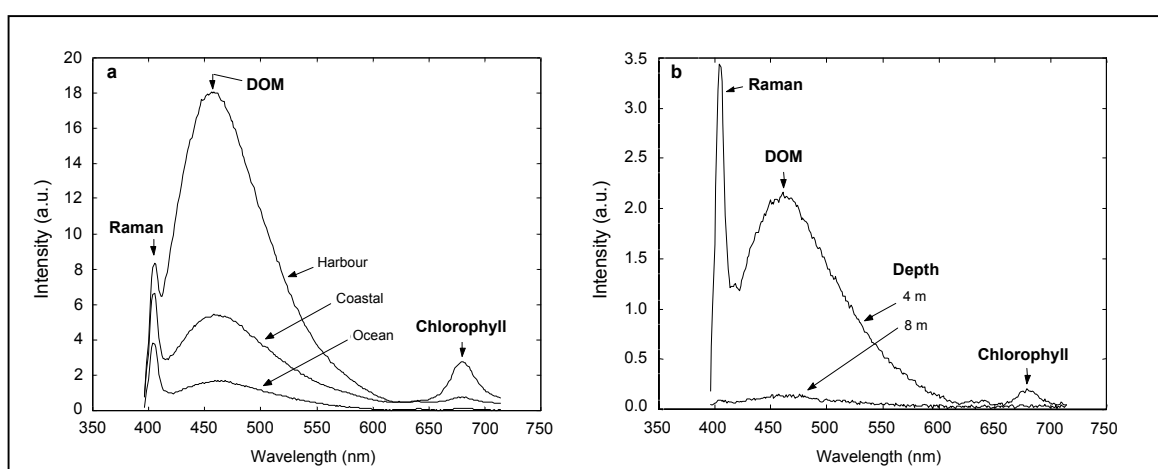
Part of the stone area had been treated with a restoration solution binding the surface and reducing decay. A processed image where the fluorescence intensity ratio  $I(400\text{ nm})/I(445\text{ nm})$  is displayed clearly demarcates the treated areas. The origin of the demarcation is evident from the inserted spectra shown for treated areas and normal stone. Also included in Fig. C4 is a different measurement of the upper part of the Baptistery next to the cathedral. Here, instead the  $I(685\text{ nm})/I(645\text{ nm})$  ratio is displayed, sensitive to chlorophyll and demarcating areas of biodeterogen invasion, as can easily be understood from the inserted spectra for invaded and non-invaded stone.



**Fig. C4.** Photographs and fluorescence images from two portions of the Parma cathedral, showing areas that had been subject to surface protection treatment (below) and algae coverage (above). Individual spectra are shown as inserts to indicate the spectral basis of the discrimination.

The most common application of laser systems in marine monitoring is bathymetry, where the elastic echo from the sea floor is detected. Blue-green radiation penetrates water best and a depth range down to tens of metres can be achieved. If the elastic scattering is suppressed, the laser-induced fluorescence can instead be detected. Major chromophores in water are dissolved organic matter (DOM), giving rise to a broad fluorescence light distribution in the blue-green region, and chlorophyll from microscopic algae (plankton) featuring a main peak around 685 nm. In addition, a strong sharp signal due to the O-H stretch Raman scattering in the water molecules occurs with a characteristic shift of  $3400\text{ cm}^{-1}$ , corresponding to a spectral position of 404 nm for 355 nm excitation. By normalising the chlorophyll and the DOM signal to the freestanding water Raman signal, effects due to observational geometry, light penetration properties etc. can be eliminated. Examples of water fluorescence data are shown in Fig. C5. The data were recorded during a ship-borne measurement campaign around Sicily, when atmospheric as well as marine experiments were performed [C20]. 355 nm pulses were transmitted horizontally from the dome over the compartment roof to hit a 45-degree mirror

placed outside the railing of the ship. The fluorescence was collected in a retrograde beam path and was dispersed and recorded in a gated and intensified OMA system. The strong elastic scattering from the water surface and column was suppressed with a coloured glass filter. In part a) of the figure the signal evolution as the ship enters the Italian harbour of Civitavecchia is shown with increasing DOM concentrations. Depth-resolved curves are shown in part b). A 40 ns gate in the image intensifier could be activated at different delays, resulting in fluorescence curves pertaining to water slices about 4 m thick. Data from two depths are shown. Gating the fluorescence signal from the bottom vegetation would be of particular interest, but due to the constraints on shallow-water passage of the research vessel, we were not able to obtain a signal from the bottom. With the system mounted on a smaller vessel or barge, *in situ* monitoring of, e.g. *Posidonia Oceanica*, which is an important environmental bioindicator, would be possible.



**Fig. C5.** Fluorescence data obtained for Mediterranean water during a measurement campaign on board the Italian research vessel *Urania*. The data in part a) of the figure show increasing levels of DOM as the ship enters the port of Civitavecchia. b) Two depth-resolved fluorescence curves showing the signal integrated over a 4 metre column centred at depths of about 4 and 8 metres depth.



### C3. Gas imaging using gas correlation spectroscopy

*Tomas Christiansson, Hans Edner, Jonas Sandsten and Sune Svanberg*

Real-time gas imaging is of great interest in many contexts. Inspection of leaks from chemical installations, petrochemical plants, tank farms or pipelines is important from economical, environmental and safety viewpoints. Easily deployable surveillance techniques for assessing sites of accidents involving gas tankers or trains are desirable to ensure public safety. We have further developed a method for remote visualisation of gas flows based on infrared absorption or emission and gas-correlation techniques using a sensitive IR camera combined with optical filters and gas cells. The method relies on simultaneous multispectral imaging and computer processing of the data. A suitable IR wavelength window where the gases of interest absorb is chosen using a bandpass filter, but the high-resolution "holistic" filtering is performed by the optically thick gas cell. New algorithms have been developed which enable calibrated gas concentration images to be produced typically at video rates.

Applications demonstrated so far are concerned with ammonia, ethylene and methane monitoring. New studies have been performed to measure releases of sulphur hexafluoride, SF<sub>6</sub> [C21]. This is a gas that is extensively used for providing electrical insulation in high-voltage installations. Furthermore, it is frequently used as a tracer gas to follow the movements of gas clouds, and mass spectrometry of air samples is then often employed. SF<sub>6</sub> has a very sharp and prominent absorption structure around 10.6 μm. Experiments on SF<sub>6</sub> monitoring were performed on open flows in the laboratory, as shown in Fig. C6. The gas image, coded in shades of red, is overlaid on a visible image from a CCD camera. The heavy gas can clearly be seen expanding from the hose and subsequently "falling". The potential for monitoring of sulphur hexafluoride leaks using the IR gas correlation technique appears to be considerable.



**Fig C6.** Gas correlation imaging of a sulphur hexafluoride flow.

## C4. Diode laser spectroscopy

*Benjamin Anderson\*, Mikael Sjöholm, Gabriel Somesfalean, Sune Svanberg and Zhiguo Zhang*

*\*visiting scientist*

Diode laser spectroscopy can provide low-cost, real-world implementation of techniques developed using more advanced laboratory equipment. Diode lasers are suitable for high-resolution laser spectroscopy applications since they can usually run in a single longitudinal mode and are easily tuned by current or temperature control. By using different types of semiconductor compound combinations, a large span of wavelengths can be covered and further extended by frequency mixing techniques. The lasers can also be efficiently modulated electrically with rf sources.

For a long time diode laser action was limited to the infrared, near-infrared and red spectral regions. Recently, our group has investigated the spectroscopic use of the new violet and blue continuous-wave (CW) diode lasers from the Nichia Corporation [C22]. One use of violet diode lasers is as excitation sources for laser-induced fluorescence. In a basic research experiment, laser-induced fluorescence with a violet diode laser was used for measurements in an ion storage ring of the lifetime (of the order of 1 s) of a metastable state in  $\text{Ca}^+$  [C23]. The laser was operated in a Littrow-type external-cavity set-up, which ensured single longitudinal mode operation, and generated a wavelength corresponding to an electronic transition of this ion.

Laser absorption spectroscopy is a powerful method for quantitative measurements with high sensitivity, especially when using frequency modulation (FM) techniques. Common to a variety of FM techniques is the shifting of the detection band to higher frequencies to avoid laser source ( $1/f$ ) noise. The absorption signal is detected either at the same frequency as the applied modulation frequency, an overtone or an intermediate frequency. Our group has specialised in a certain approach called two-tone frequency modulation spectroscopy (TTFMS), which provides high dynamic range, favourable detection bandwidth and shot-noise-limited detection, i.e. ultrasensitive absorption measurements. This high sensitivity is suitable for work on small volumes of absorbing gases and for measuring weak molecular transitions in the near-infrared region.

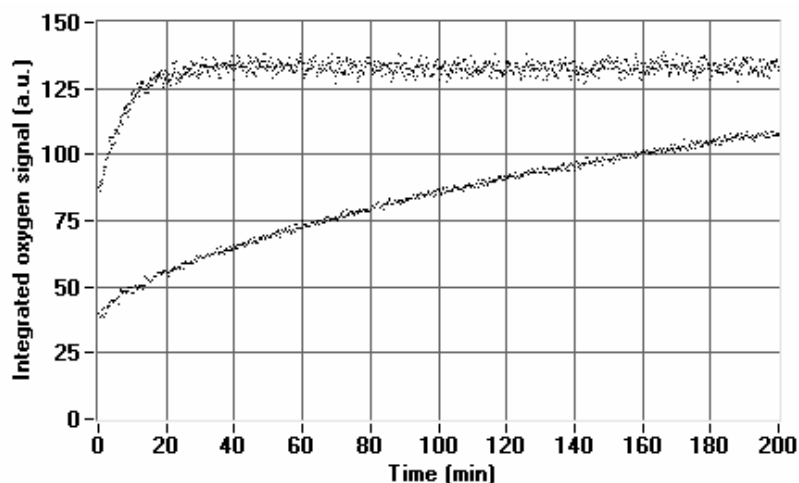
We have demonstrated the advantages of TTFMS in several applications, e.g. in combination with long-path detection for trace gas monitoring at atmospheric pressure [C24]. In this way, traffic-generated  $\text{NO}_2$  could be remotely detected, even at the readily available wavelength of 635 nm, which is situated far from any strongly absorbing lines of this gas. A real-time laser absorption spectrometer was accomplished by repetitively applying a rectangular current pulse to the diode laser DC drive current, allowing detection of isolated  $\text{NO}_2$  absorption lines. A detection limit of  $10 \text{ }\mu\text{g/m}^3$  for  $\text{NO}_2$  at atmospheric pressure with a 160 m absorption path was

demonstrated. Continuous monitoring of NO<sub>2</sub> over a road intersection at peak traffic load was performed.

Recently, an international project involving our group and researchers at the Harbin Institute of Technology and Jilin University in China was started within the field of molecular laser spectroscopy. The objective of the project is to pursue a research and development programme using tuneable diode laser spectroscopic techniques to monitor important atmospheric pollutants. Efficient pollution monitoring systems will, for example, help major Chinese cities to meet environmental challenges. A specific project is the simultaneous monitoring of the important pollutant gases nitrogen dioxide and sulphur dioxide and, in addition, water vapour, which is of major climatological importance. This will be done by sum-frequency mixing of a blue diode laser (spectrally overlapping NO<sub>2</sub>) with a near-IR laser (spectrally overlapping H<sub>2</sub>O) to produce UV radiation (spectrally overlapping SO<sub>2</sub>). During initial experiments we were able to demonstrate sum-frequency generation of UV light around 300 nm in a BBO crystal and detection of well-resolved spectral structures of SO<sub>2</sub> in both low- and atmospheric-pressure samples [C25].

Many other trace species also have strong electronic transitions in the blue and UV spectral regions. Our blue lasers, combined with doubling and mixing techniques, show great promise. Here the availability of periodically poled nonlinear crystals is of particular interest. We plan to explore FM diode laser spectroscopy in combination with sum- and difference-frequency mixing. Full advantage of the modulation techniques will be achieved only when very efficient generation can be obtained so that shot noise will not be a primary limiting factor.

A completely new aspect of gas spectroscopy, called Gas in Scattering Media Absorption Spectroscopy (GASMAS) has been proposed and demonstrated by us for the unique analysis of gas enclosures in turbid solids and liquids [C26-C32]. Many substances, frequently of organic origin, are porous and contain free gas distributed through the material. Many examples can be given: living tissue, paper, powder, sintered materials, catalysts, foams and liquids with dissolved gas are but some. The normal method of gas absorption spectroscopy fails for porous media, since the radiation is greatly scattered in the material containing the gas. Thus, there are no well-defined path lengths, as required by the Beer-Lambert law, but light emerges diffusely. The free gas molecules dispersed in porous materials have unique, sharp absorptive signatures, typically ten thousand times narrower than the spectral features of the bulk material. The small absorption imprint in the emerging, multiply scattered, diffuse light can be detected sensitively by the use of frequency modulation techniques, easily achievable using diode laser spectroscopy. In the case of strong scattering, which is the most interesting aspect of the proposed gas-detection technique, a long effective path length is achieved, giving rise to a strong gas signature. An absorption sensitivity of  $2.5 \times 10^{-4}$ , corresponding to 1.25 mm of air column, was demonstrated in measurements on dispersed molecular oxygen. The spectroscopic measurements were performed at around 760 nm on lines belonging



**Fig. C7.** Re-invasion of oxygen into a 10 mm thick slab of Norway spruce (fast curve) and a 10 mm thick slab of balsa (slow curve).

to the oxygen A band. Two measurement geometries are particularly useful in this new field for performing basic studies: transillumination and backscattering monitoring. In both cases optical fibres can be used to inject the light and collect scattered radiation. The absorption and scattering properties of the bulk material have been deduced independently by spatially resolved and time-resolved measurements, providing a direct and independent assessment of the photon history. The mean path length of the impinging photons through polystyrene foam, used as a generic test material, was estimated, which together with the magnitude of the gas absorption was used to determine the concentration of the dispersed oxygen, which in this case was 20.4%.

The GASMAS technique provides new opportunities for non-destructive studies of gas in highly scattering, natural and man-made porous materials. In recent experiments on wood, anisotropy related to the fibre structure was studied [C33-C35]. This kind of measurement may be of fundamental interest in the understanding of light transport in inhomogeneous porous materials. The new possibility to observe free gas in scattering media not only allows static gas assessment but also the study of dynamic processes, i.e. how gas is exchanged with the environment. Studies of gas transport dynamics in different wood samples were also performed [C33-C35]. Recordings for two types of wood are shown in Figure C7. In our experiments, slabs of evacuated polystyrene foam and of different kinds of wood were exposed to the surrounding air and the successive penetration of the atmospheric oxygen was studied. One practical application is the assessment of the resistance to gas penetration of various protective covers, such as paint layers on wood [C36]. The new technique can be applied to any gas having narrow absorption features that are reachable with tunable diode laser radiation. Both penetration of atmospheric pollutants into porous stone materials, e.g. those used in historical buildings, and their transport in porous catalysts, e.g. zeolites, are particularly interesting areas of application.

## **C5. Optical diagnostics of charged surfaces and dielectric materials**

*Magnus Bengtsson, Andreas Dernfalk\*, Hans Edner, Rasmus Grönlund, Stefan Kröll, Anders Larsson, Albin Roslund, Mikael Sjöholm, Sune Svanberg, Petter Weibring*

*\* Division of High Voltage Engineering, CTH*

In this section the development of techniques for remote non-intrusive optical diagnostics of materials and events of relevance in electrical insulation systems is described.

### **Mid-gap laser-triggered electrical breakdown in dielectric liquids**

Pre-breakdown and breakdown phenomena in liquid dielectrics have been studied extensively during recent decades. Our understanding of these phenomena has been greatly improved by the advent of new diagnostic tools and techniques. One such new technique is the combination of laser triggering and the laser shadow method, as introduced by researchers at the Division of Atomic Physics [C37]. In laser triggering, a laser pulse is focussed in the electrode gap where a plasma is formed in the focal region. The plasma develops into a high-pressure gas-filled cavity that expands and eventually triggers the disruptive discharge. In the laser shadow method (shadowing), the probe volume is illuminated with collimated laser light. An event in the probe volume, with a refractive index different from that of the surroundings, diverts the collimated light and a shadow appears in the line of sight. The expansion of the cavity and pre-breakdown discharge phenomena in mid-gap laser-triggered dielectric breakdown in transformer oil were imaged using the laser shadow method. The physical mechanisms and the timing sequence of this dielectric breakdown were revealed from the analysis of these measurements. Detailed calculations of the electric field inside and outside the expanding cavity were performed [C38, C39].

### **Measurement of surface charge**

All electrical systems rely on the dielectric strength of their electrical insulation. The electrically weakest point of insulator systems is normally the surface of the insulators. One property of the surface that determines the probability of a surface discharge propagating is the ability of the material to bind electric charge at the surface. Extensive effort has been devoted to developing a non-invasive technique for remote sensing of the charge density on dielectric materials. In particular, we have tried to use non-linear optical processes for this purpose. When a high-power laser beam is reflected or scattered at an interface, it may be subjected to a non-linear process in which two photons of one frequency are annihilated and one photon of twice the frequency is emitted. This process is called second-harmonic generation (SHG). SHG is greatly enhanced in the presence of spatial asymmetry. Surfaces and surface charge are examples of such spatial asymmetries. In the work

carried out at the Division, SHG signals have been detected from metal surfaces. However, the signal due to surface charges appears too small to be detected under realistic operating conditions in the field. The work has therefore instead been directed towards remote detection of other type of surface properties.

### **Remote and non-intrusive diagnostics of high-voltage insulation materials using laser-induced fluorescence spectroscopy**

A new method for remote and non-intrusive diagnostics of high-voltage insulation and insulation materials is being developed in collaboration with the Division of High-Voltage Engineering at the Chalmers University of Technology. The method is based on laser-induced fluorescence (LIF) spectroscopy of the surfaces of polymeric insulator materials. High-voltage insulators made of polymeric materials are successively being used to replace traditional insulators made of porcelain or glass. The new insulators have several advantages, such as having a lower weight, being shatterproof and being easier to work with. However, the main advantage is that insulators made of polymer material have significantly better insulation properties. Their better performance is due to their surface properties; a polymeric surface is highly water-repellent (i.e. hydrophobic) whereas water forms a film on a porcelain surface (i.e. hydrophilic). Severe environmental conditions can, however, cause permanent or temporary loss of hydrophobicity. High-voltage insulators of polymeric materials with good hydrophobicity are therefore often pessimistically designed for a hydrophilic state. This means that the benefits of using a polymeric material instead of porcelain are not utilized to their full potential. Today, there is still no standardized or widely used technique for inspecting polymeric insulators in the field. However, several techniques, applicable to live insulators, are now being used to assess the state of non-ceramic insulators in service, and to organize and plan maintenance work. These techniques include visual inspection, image intensification (using a night-vision camera), infrared thermography, electric field distribution measurements and directional wireless acoustic emission. One disadvantage of these techniques is that only large defects can be detected (punctures and cracks) and not deterioration or changes in the hydrophobic properties of housing materials. High-resolution methods, such as electron spectroscopy for chemical analysis (ESCA) and time of flight – secondary ion mass spectroscopy (ToF-SIMS), give detailed information about the surface, but are intrusive methods. LIF surface spectroscopy is a method that is both non-intrusive and can give detailed information about the properties of the surface under study. Initial measurements have shown a difference between outdoor insulators that have been exposed to different weather conditions [C40-C43]. In further work algae have been remotely detected at distances of approximately 60 metres. Work is presently in progress to more fully assess the potential of this technique [C44, C45].

Further work concerning electrical breakdown in air has been carried out in connection with the High Performance Outdoor Electrical Insulation project

(ELIS), which is supported by the Swedish Foundation for Strategic Research (SSF). This includes simulation of streamer discharges and measurements of streamer current and streamer charge [C46-C49].

## **C6. Optical diagnostics of paper and print**

A joint research group consisting of scientists from the Divisions of Atomic Physics and Nuclear Physics at LTH and Computer and Electrical Engineering at the School of Information Science at Halmstad University, has been formed. Together with four newsprint-producing paper mills within the Stora Enso and the Holmen groups, it is engaged in the areas of paper physics, printing and print quality evaluation. The research involves the use of optical methods, nuclear techniques, signal analysis, colour classification in the development of fast, non-destructive sensor systems [C50].

### **New possibilities using modern optics in paper production**

*Carl Magnus Nilsson and Lennart Malmqvist*

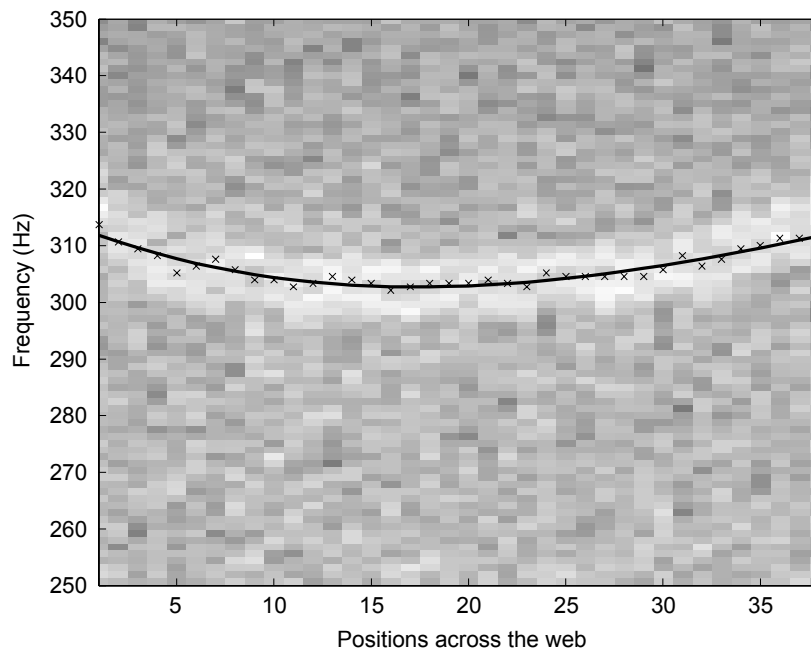
Recent developments in sensors and fast, high-capacity computers open up new opportunities for the development of high-capacity monitoring systems for the paper industry. Optical methods can offer the possibility of fast and high-spatial-resolution measurements without any contact with the paper. However, this will require an increase in measurement capacity and the development of signal processing, as well as a deeper understanding for the interpretation of the results.

One important quality parameter of paper is its homogeneity, i.e. having as small variations as possible over the paper sheet in terms of different paper properties. Therefore, the on-line monitoring of the distribution of different substances in the paper has become important. In newsprint the main constituent is wood fibres, containing cellulose, hemicellulose and lignin. Measuring the distribution of these constituents in the paper could, for example, be used as an indication of the homogeneity of the final paper product. The print quality, however, is governed by how well the smallest print entity, the individual halftone dot, is reproduced on the paper. Therefore, the properties of the newsprint at the precise position on the paper where the dot is to be printed govern the print quality. Consequently, optical sensors with high spatial resolution are required to measure such variations in paper.

A robust optical sensor with high spatial resolution has been developed and tested for on-line measurements in paper production. The sensor is designed to measure the fluorescence response from lignin in paper. A fast system for monitoring and recording the signal from the new sensor has also been designed and built. With this system a whole tambour, about 60 km of web length, can be monitored along a line with a physical distance between adjacent samples of 25  $\mu\text{m}$ , at a web speed of

30 m/s. With this system the performance of the paper machine can be studied on different length scales, from sub-millimetre to the scale of many kilometres of paper. The sensor system has been evaluated and tested on-line at newsprint mills. From these measurements it was concluded that the system can be adapted to the industrial environment and give valuable results reflecting the variations in the process.

The high geometrical resolution of the fluorescence sensor has opened up the possibility of measuring small variations in the paper structure. An example that demonstrates this is the possibility to measure imprints in the paper made by the forming fabric. This property has also been used to develop a fast and robust method for measuring the paper shrinkage [C51, C52]. In Figure C8 a shrinkage profile is shown as an another example. The measurement was performed in an off-line test rig where a cross-directional shrinkage profile was recorded.



**Fig. C8.** A shrinkage profile from newsprint paper measured with the red fluorescence sensor.

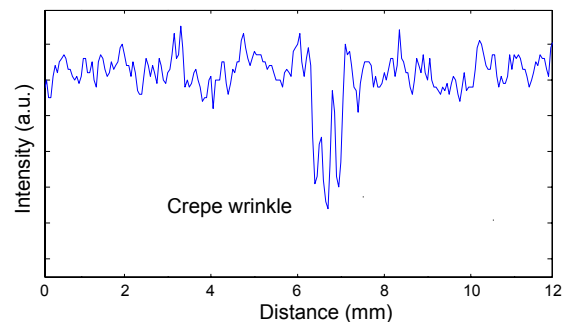
The identification of crepe wrinkles in the paper has long been a challenge for newsprint mills. A system that can map the position of crepe wrinkles has also been developed. In Figure C9 photograph is shown of the crepe wrinkle detection device mounted on a rewinder. This illustrates the opportunities provided by the fast high-resolution measurements combined with an algorithm designed to look for this quality-degrading phenomenon in newsprint.

To be able to design special optical sensors and algorithms for the identification of specific quality-related properties in the paper web, a multipurpose instrument for two-dimensional paper analysis has been developed. The instrument is an adaptive



test bench in which different properties and parameters can be measured simultaneously, using different sensors arranged with high geometrical precision. Such a test bench is a key tool in the study of paper properties and for the development of new optical sensors [C53].

Our work has contributed to the creation of a platform for the development of future sensors and data acquisition systems for on-line quality monitoring in paper production. The experiments conducted at the paper mills have demonstrated that the use of high-resolution optical sensors and high-capacity data acquisition systems can provide new information to improve our understanding of the dynamics of the manufacturing process.



**Fig. C9.** The crepe-wrinkle detector mounted in a rewinder at a paper mill (left). Typical response due to a large crepe wrinkle (right).

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## **D. Laser Applications in Medicine and Biology**

All research conducted at the Division concerning laser spectroscopy applied to biomedicine is conducted within the framework of the Lund University Medical Laser Centre. The "medical group" at the Division can be considered the core group of the Medical Laser Centre, with a highly interdisciplinary approach to developing and evaluating novel optical tools for both diagnostic and therapeutic purposes. The aim of the Centre is to support multidisciplinary projects using lasers in medical research, to give courses and seminars in related topics, to act as a base for grant applications and to act as a partner in multicentre research studies. The activities within the Centre cover a broad field of interdisciplinary research, involving clinicians from various clinical specialities, physicists and chemists. In our presentation of the personnel involved in the different projects, only those affiliated to the Division of Atomic Physics are listed. A large number of collaborators, within and outside the Medical Laser Centre, also contribute to our work.

The research at the Division of Atomic Physics involves mainly tissue characterization using laser spectroscopic techniques and laser treatment of malignant tumours. Research in tissue diagnostics is mainly directed towards early detection and identification of premalignant and malignant lesions. Other areas of research include developing techniques for concentration measurements in aqueous solutions and in highly scattering material, such as pharmaceutical preparations. We are currently participating in three European projects within the field: Optical Mammography (OPTIMAMM), Medical Photonics (MedPhot) and Virtual European Laser Institutes (VELI). Below, the different projects are briefly presented with citations to the original publications. A selection of presentations of our activities that have been given at various conferences can be found in Refs [1-9]. A large number of Masters students have also conducted projects within the group within the various projects [10-16].

### **D1. Photodynamic therapy**

Photodynamic therapy (PDT) is a treatment modality for various diseases under development and evaluation. The procedure is based on a photochemical reaction, in which a photosensitizer absorbs light and reacts with tissue oxygen. The sensitizer is applied to the patient systemically or topically and accumulates selectively in lesion cells. Illumination with light at the wavelength of the absorption peak of the photosensitizer results in the formation of free radicals in the

tissue (e.g.  $^{10}\text{O}_2$ ) which lead to immediate tissue necrosis. Secondary effects include vascular damage, induction of apoptosis and inflammatory reactions.

The limitations of the treatment procedure are the selectivity of the photosensitizer, the penetration depth of the photosensitizer after topical application, tissue oxygen concentrations and light transport in tissue. These limit the treatable tumours to superficial lesions of limited thickness. To understand and improve the treatment procedure we have conducted various measurements in connection with PDT of skin malignancies. Also, by inserting optical fibres into the lesion, interstitial PDT, it is possible to improve the light penetration for the treatment of thicker lesions.

### **Superficial PDT**

*Stefan Andersson-Engels, Niels Bendsøe, Lotta Gustafsson, Claes af Klinteberg, Sara Pålsson, Marcelo Soto Thompson, Maria Stenberg, Katarina Svanberg, Sune Svanberg and Ingrid Wang*

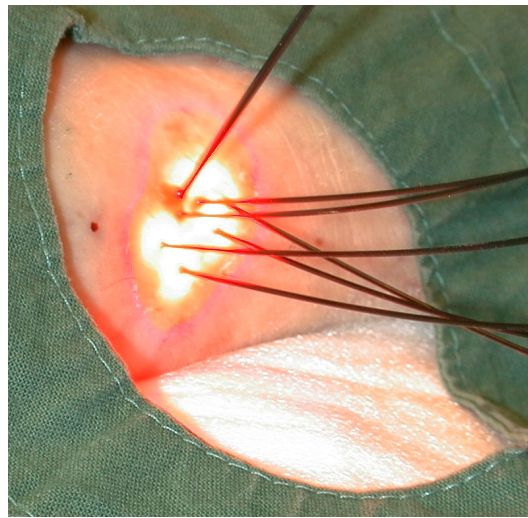
The results of a clinical trial have been published, in which PDT was compared with cryosurgery in the treatment of non-melanoma malignant lesions[17]. The clinical results showed that PDT is comparable to conventional treatment methods in terms of response. PDT causes less scar formation and more normal tissue is saved using PDT compared with cryosurgery. Several studies have also been conducted to investigate how the local blood perfusion in the lesion altered as a response to the treatment [18,19], showing an increased blood perfusion after the treatment. This is of importance in understanding the mechanisms of PDT in more detail. The tissue fluorescence and temperature were studied in other studies [20-22], with the aim of optimizing the treatment parameters.

### **Interstitial PDT**

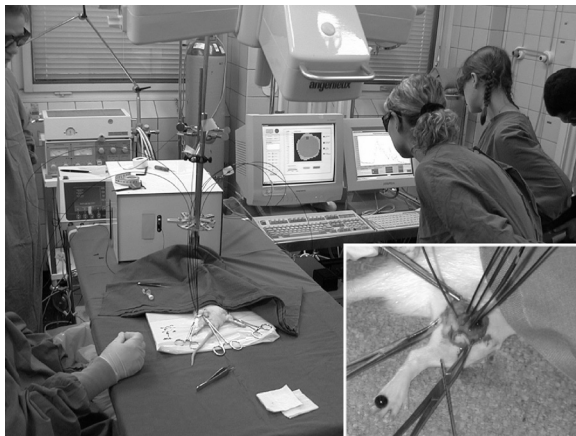
*Stefan Andersson-Engels, Niels Bendsøe, Lotta Gustafsson, Thomas Johansson, Claes af Klinteberg, Sara Pålsson, Eva Samsoe, Marcelo Soto Thompson, Maria Stenberg, Unne Stenram, Katarina Svanberg and Sune Svanberg*

To develop PDT beyond the treatment of thin superficial tumours, for the efficient treatment of deeply located and/or thick tumours, a system based on interstitial illumination using multiple fibres has been developed [23-25]. A patent application describes how such treatment could be performed [26]. Examples of conditions that could benefit from such a treatment modality are malignant brain tumours and tumours in the oral cavity. In interstitial PDT (I-PDT), multiple fibres should be used for light delivery in order to allow the treatment of tumours larger than a few millimetres in diameter. Our system consists of a laser light source, a beam-splitting system dividing the light into three or six output fibres and a specially developed computer software for the treatment dosimetry. The concept is then to use these fibres not only to deliver the treatment light, but also to measure parameters of interest for the outcome of the treatment. The fluence rate of the light

emitted by each fibre is measured at the positions of the other fibre tips. From these results, the light dose at all positions can be calculated. Changes in optical properties as well as concentration of the photosensitizer by photobleaching during treatment could thus be monitored and compensated for by adjusting the dose. Tumours have been treated both in experimental studies and in patients with thick Basal Cell Carcinomas (BCCs). An image of a BCC under treatment is shown in Fig. D1, while a photograph of experimental treatment is shown in Fig. D2. In order to obtain relevant parameters for the mathematical dosimetry model, a study of the threshold dose for tumour destruction was conducted [27]. A photograph of the excised tissue following treatment is given in Fig. D3. The diameter of the necrotic lesion was measured from these samples.



**Fig. D1.** Treatment of a human basal cell carcinoma.



**Fig. D2.** Photograph from experimental treatment of a tumour with interstitial PDT.



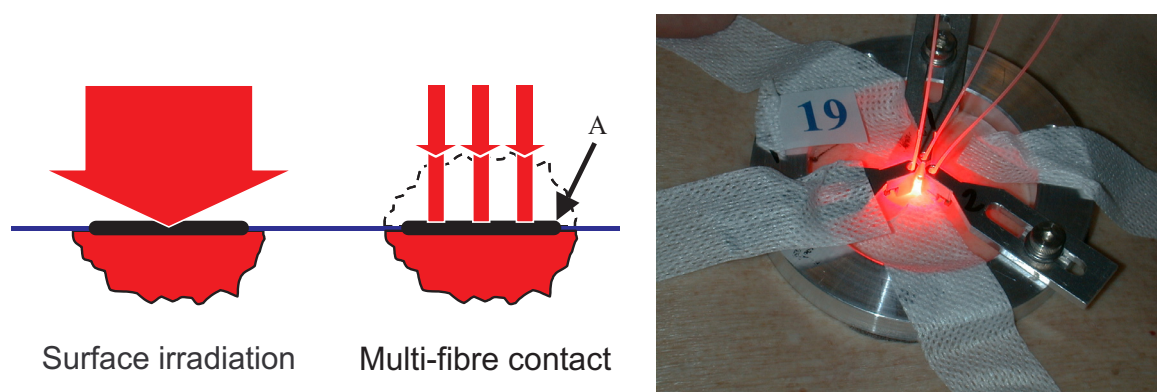
**Fig. D3.** Slices of a tumour resected three days after IPDT are shown. The slightly lighter areas in the slices indicate tumour necrosis, while the dark areas are due to bleeding.



## Multi-fibre contact PDT as a step towards interstitial-PDT

*Stefan Andersson-Engels, Niels Bendsøe, Thomas Johansson, Sara Pålsson, Marcelo Soto Thompson, Unne Stenram, Katarina Svanberg and Sune Svanberg*

A clinical trial was performed at the Latvian Oncology Centre, Riga, Latvia [28]. PDT was performed on 29 nodular basal cell carcinomas, of which 17 lesions were treated with conventional superficial PDT. The remaining 12 lesions were treated with multi-fibre contact PDT (MFC-PDT) in which the light was delivered with clear-cut fibres placed in direct contact with the tumour area instead of inserting them into the tumour (see Figure D4).



**Fig. D4.** The conceptual difference between superficial, surface illumination PDT and MFC-PDT is shown to the left. As can be seen, with an imaginary upper part of a tumour considered, as indicated by the dashed line in the figure, MCF-PDT could serve as a coarse model for interstitial PDT. An example of MFC-PDT with three fibres positioned in the fibre holder is shown (right).

As indicated in Fig. D5, the cosmetic and clinical results of the trial were very good with comparable or better results for the patients treated with MFC-PDT compared with those treated with superficial PDT. On a technical level the trial resulted in valuable experience for the continuing work on improving I-PDT.



**Fig. D5.** Photographs before, 6 days after and 7 month after MFC-PDT of a  $15*15*3 \text{ mm}^3$  nodular basal cell carcinoma.

## **Development of diode lasers for I-PDT**

*Stefan Andersson-Engels, Eva Samsøe, Katarina Svanberg and Sune Svanberg*

One limitation of the compact light sources available for PDT today is that the coupling efficiency to thin optical fibres is poor. We have thus devoted considerable effort to the development of diode lasers with greatly improved coupling efficiency. This work is being conducted in close collaboration with Risø National Laboratory outside Roskilde in Denmark. The results are very promising. The beam quality out of the laser is very satisfying, with M2-values of 2.1. The results have been published in a number of papers [29-35], and a patent application has been submitted to commercially protect the intellectual property [36].

## **D2. Optical spectroscopy for biomedical applications**

Taking into account the different spectral properties of healthy tissue, pre-malignant and malignant tissue, several studies have been performed in various clinical areas. Much work has also been devoted to the development of methodologies and instrumentation to measure the fundamental optical properties of tissue. Further development of existing treatment modalities and equipment, and improvements in the methods of data analysis are of major importance, and have resulted in two theses [37,38].

### **Fluorescence spectroscopy**

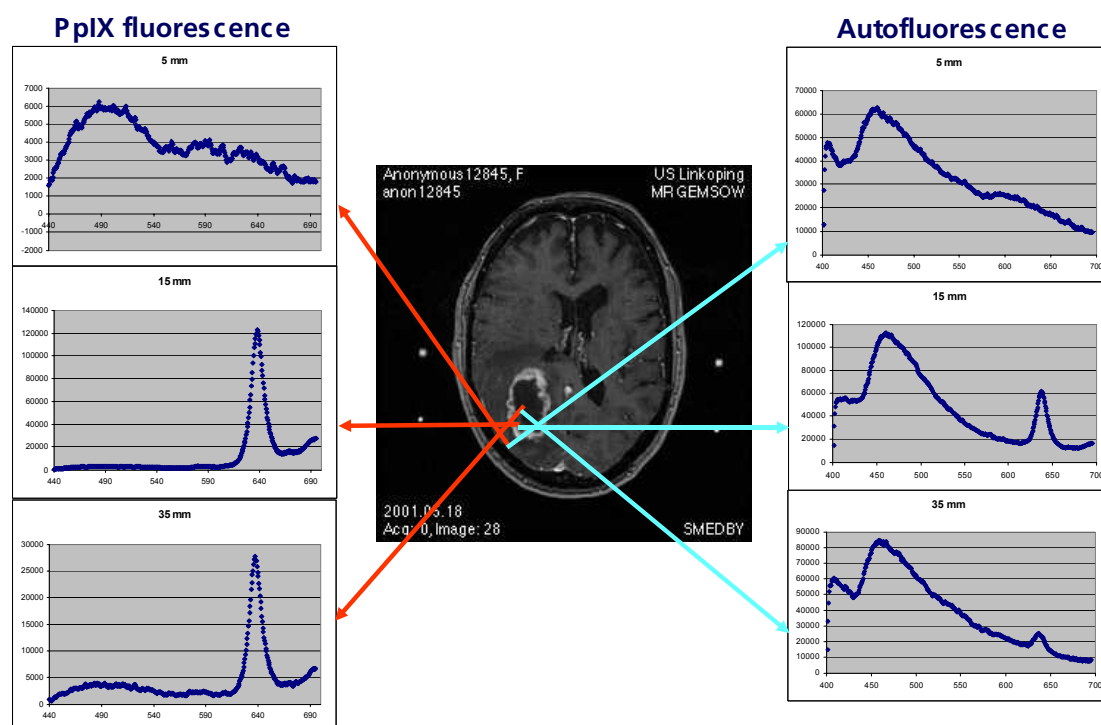
*Janis Alnis, Stefan Andersson-Engels, Charlotta Eker, Johanna Ekström, Thomas Johansson, Claes af Klinteberg, Sara Pålsson, Jonathan Roth, Gabriel Somesfalean, Marcelo Soto Thompson, Maria Stenberg, Katarina Svanberg, Sune Svanberg*

Two new fluorosensors have been developed within the group [13,39-41]. The first is a relatively advanced system using several excitation wavelengths and with the capacity to measure time-resolved fluorescence spectra. The second is a compact fluorosensor, consisting of a blue diode laser, fibre optics and a small, integrated spectrometer. The box is only 22 x 13 x 8 cm<sup>3</sup> and it is thus easy to transport together with a laptop computer.

A clinical trial has been conducted to investigate the possibility of using fluorescence spectroscopy for the differentiation of pre-cancerous lesions versus healthy tissue in the cervical region. The study was performed in Vilnius in collaboration with the Women's hospital in Vilnius and the company Science and Technology International (STI). Over one hundred (111) women were included in the study, in which autofluorescence spectra were measured using a point-monitoring fluorosensor with excitation at 337 nm. Suspicious areas were biopsed and the biopsies were processed and analysed according to a very detailed protocol

of sectioning and histopathological evaluation [42]. This protocol allows precise orientation and localisation of the biopsies and it includes evaluation of all tissue parameters that could potentially correlate with fluorescence spectra. Reflectance and fluorescence images were recorded by the imaging instrument provided by STI [43,44]. The material has not yet been fully evaluated but preliminary results indicate differences between spectra from normal tissue and pre-malignant changes. However, spectra from inflamed tissue resemble spectra from pre-malignant lesions and definite conclusions can only be drawn after a more comprehensive statistical evaluation.

An ongoing study of  $\delta$ -aminolevulinic acid (ALA) induced protoporphyrin IX (PpIX) fluorescence in brain tissue has continued together with brain surgeons in Linköping, Sweden [45]. Our aim is to evaluate the possibility of determining the boundaries of infiltrating brain tumours during stereotactic biopsy using fibre optics utilizing both changes in autofluorescence and PpIX fluorescence spectra. Although only a rather limited number of patients are included in the evaluation up till now, the statistics indicate that this method may be useful in the development of dissection techniques. A magnetic resonance image from one of the studied patients is shown in Fig. D6, together with fluorescence emission spectra from indicated points.



**Fig. D6.** Magnetic resonance image of an astrocytoma captured just prior to a stereotactic biopsy procedure. Fluorescence was recorded during the incision of the biopsy needle. The fluorescence spectra shown to the left are excited at 405 nm, while those to the right are excited at 337 nm.

Another study has been carried out to measure the kinetics of two different types of ALA [20]. In addition to normal ALA, methyl-esterified ALA (ALA-ME) was also

used, the latter being more lipophilic than ALA, possibly resulting in a greater penetration depth. A total of 30 BCCs on 11 patients were included in the LIF part of the study, the results of which indicated a higher and more selective build-up of PpIX in the tumours treated with ALA-ME than those treated with ALA. To improve the statistics, a study is in progress including 50 lesions each with ALA and ALA-ME.

Furthermore, fluorescence imaging studies have been performed on skin lesions [22,46]. The aim of this study was to investigate the demarcation of skin tumours after the topical application of ALA, by detecting the PpIX fluorescence in the spectral domain. A hyperspectral fluorescence imaging system was used to identify BCCs after topical application of ALA. Another system was based on time-gated fluorescence imaging, relying on differences in the fluorescence lifetime. Both systems rely on the comparison between the exogenous and the endogenous fluorescence. Clear demarcation of skin malignancies was seen *in vivo*, noninvasively, with both fluorescence imaging systems. The two complementary approaches applied in this study show promise for skin tumour detection and delineation based on specific fluorescence features.

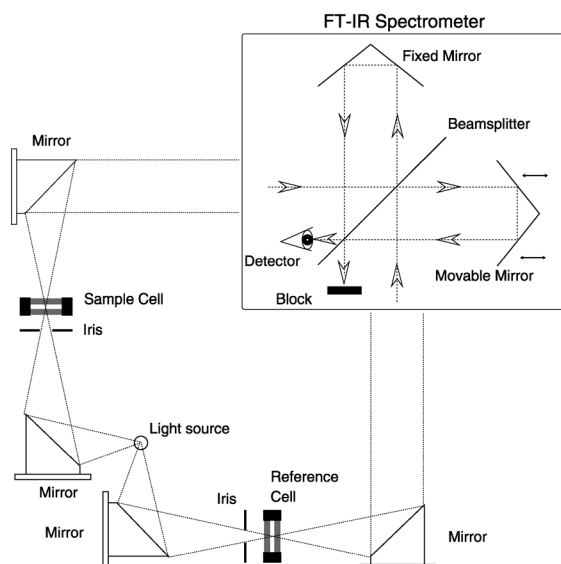
In several other studies, fluorescence has been recorded from small suspected lesions in order to evaluate the potential of laser-induced fluorescence (LIF) as a tool to assist the examining doctor in tumour diagnosis [47-50]. Two of those studies were aimed at estimating the extent and severity of lesions on the vocal fold suspected of being malignant [48,49].

Measurements have been performed on biopsies collected from hearts of transplant patients following the transplantation, with the aim to develop a technique to optically assess whether the heart is about to be rejected by the new host [50]. The results showed that it is possible to distinguish fibrotic myocardium from normal tissue. However, there seems to be no apparent difference between rejected and normal tissue, but the number of measurements is very small.

## **Vibrational spectroscopy**

*Stefan Andersson-Engels and Peter Snoer Jensen*

During the period covered by this report we have had close collaboration with Risø National Laboratory in Denmark, in the development of an optical subtraction method during Fourier transform IR spectroscopy to improve the sensitivity of the technique. The developed instrument is shown in Fig. D7. The aim of the work was to develop a technique with which to measure concentrations of biomolecules in aqueous solutions. One application of interest is dialysis. Improved control of the dialysis process may be possible with simultaneous on-line measurements of several of the important molecules of importance in this process. This work has resulted in a number of publications and a PhD thesis [51-55].



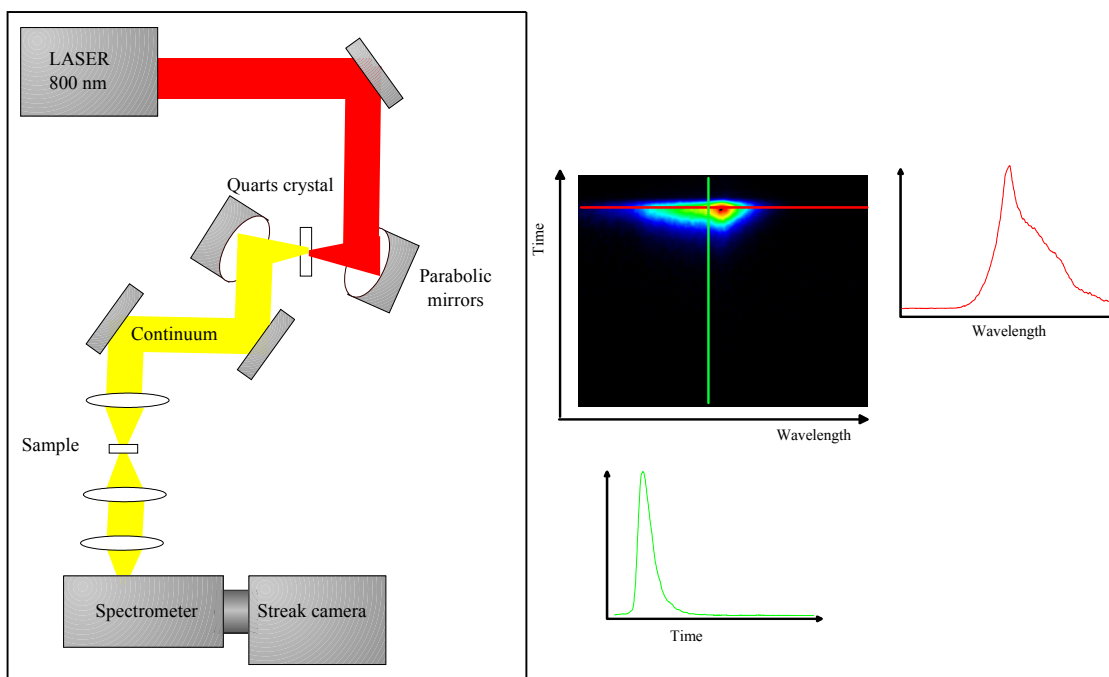
*Fig D7. Dual beam Fourier transform spectrometer setup for difference signal measurements.*

## Spectroscopy on Solid Pharmaceuticals

*Christoffer Abrahamsson, Stefan Andersson-Engels, Sune Svanberg*

Near-infrared (NIR) spectroscopy has become a valuable technique for measurements on pharmaceutical preparations and tablets, which can replace some of the expensive and time-consuming wet-chemistry methods currently used in the pharmaceutical manufacturing process. The technique does, however, suffer from limitations due to the fact that changes in the physical properties of a solid sample greatly changes the optical properties of the sample and therefore also affect the recorded spectra. Several computational methods, e.g. multiple scattering correction (MSC), have been proposed to compensate for the variations in the physical properties between different tablets, but despite all computational efforts it is still difficult to obtain robust quantitative results from NIR spectroscopic measurements on highly scattering samples.

Time-resolved NIR-visible spectroscopy is utilized to study the light propagation through solid pharmaceuticals, see Fig. D8. The time-resolved approach, combined with a newly developed Monte Carlo model makes it possible to fully characterize the optical properties of the measured tablets, and thus separate the scattering properties from the true light absorption of the sample. Since scattering in tablets is mainly affected by the physical parameters, e.g. compression force and granulate quality, while the absorption is mainly affected by the chemical content of the tablet, this new technique not only has the ability to measure the concentration of different constituents in the tablet, but also to provide more information about tablet quality than ordinary continuous wave spectroscopy [3,56].



**Fig. D8.** Set-up for time-resolved measurements on intact tablets, and an example of data from a measurement.

Apart from time-resolved measurements other efforts have been devoted to improving spectroscopic methods for measurements on intact tablets. Variable selection is a method that reduces the number of data points in the spectra while improving the predictive ability and robustness of the model. A study was made using transmission NIR spectra, in which four different methods of variable selection in the partial least squares (PLS) regression method were studied and compared with calibration performed with manually selected wavelengths. The methods used were genetic algorithm (GA), iterative PLS (IPLS), uninformative variable elimination by PLS (UVE-PLS) and interactive variable selection for PLS (IVS-PLS). All methods improved the predictive abilities of the model compared with the model in which the wavelengths were selected manually. For the data set used in this work IVS-PLS and GA provided the best results, with a reduction in prediction error of 20% [57].

## Optical properties of tissue

*Stefan Andersson-Engels, Charlotta Eker, Claes af Klinteberg, Aruna Pakrasa, Tuan Pham, Johannes Swartling, Sune Svanberg and Jan Sørensen Dam*

Knowledge of the optical properties of tissue forms the fundamental basis of most activities in the field of biomedical optics. For therapeutic applications, such as PDT and laser-induced thermotherapy, it is important to achieve optimal light doses by combining optical properties and light propagation models to predict the effects of the treatment. For diagnostic techniques, such as LIF and tissue transillumination, the diagnostic information depends on the optical response of the

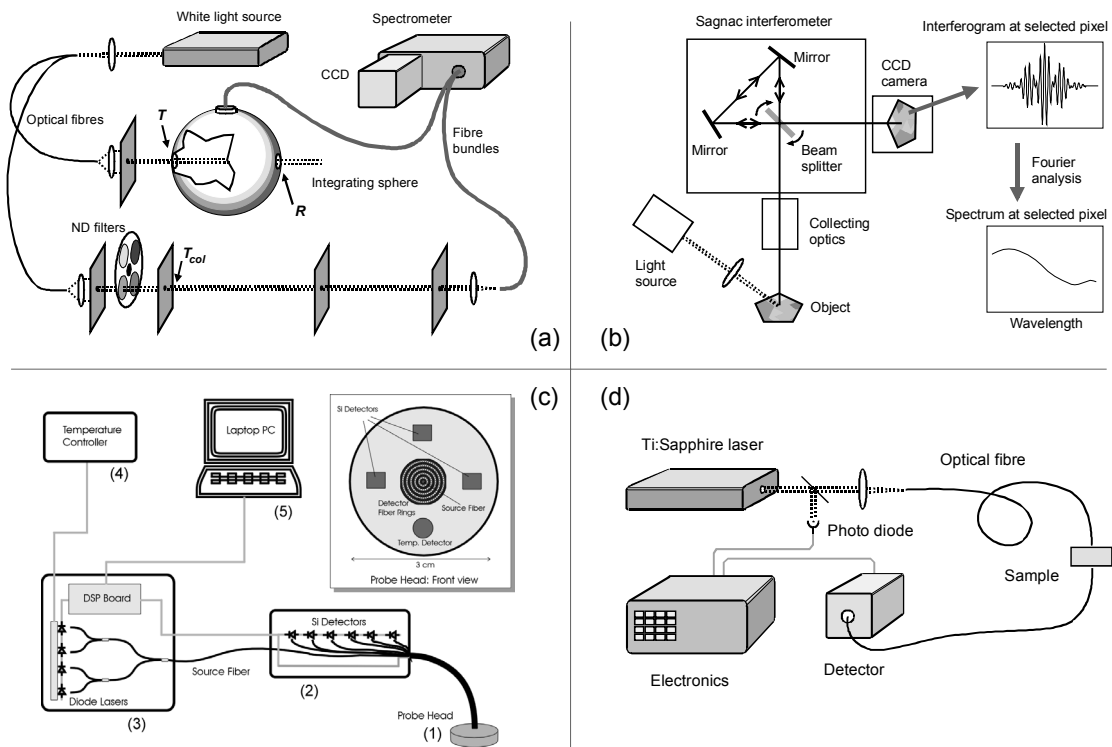
tissue, which again can only be predicted by the optical properties and suitable light propagation models. In December 2002, Johannes Swartling defended his doctoral thesis "Biomedical and atmospheric applications of optical spectroscopy in scattering media" [37]. His work was closely related to the measurements of the optical properties of tissue.

At the Division, a number of methods of measuring the optical properties of tissue have been developed and investigated. Several permanent systems have been built and are available to support day-to-day measurements. The properties most often used to describe the optical characteristics of tissue are the absorption coefficient,  $\mu_a$ , and the scattering coefficient,  $\mu_s$ . Sometimes, the scattering phase function, which describes the amount of forward scattering by the tissue, is of importance, and is usually expressed as the mean cosine of the scattering angle, denoted  $g$ . These properties vary with wavelength, so spectral resolution adds one more degree of freedom.

The measurements of optical properties in tissue presents an inverse problem, in the sense that the response of an illuminated tissue sample is measured using an experimental set-up, and is then indirectly related to the optical properties by means of a light propagation model. The problem is nonlinear, which means that sophisticated numerical algorithms are necessary.

Several systems are available at the Division for determining the optical properties of tissue [58-60]. For accurate measurements of the properties of excised tissue samples, the integrating sphere method is ideal (see Figure D9a). The integrating sphere method allows the measurement of the total transmitted as well as total reflected light intensity from thin samples. As a third measurement the collimated transmittance can be measured in a separate set-up to suppress the scattered light.

Another type of method involves spatially resolved diffuse reflectance measurements of the surface of the tissue. The advantage of this method is that it can be applied *in vivo*, e.g. directly on the skin of the patient. The surface is illuminated by a spot, and the reflected intensity is measured at different distances from the spot. Two such systems are available at the Division. The first is an imaging Fourier transform spectrometer, which can record multispectral images of the tissue surface [61] (see Figure D9b). The second is a specially designed fibre probe, developed by Jan Sørensen Dam at Bang & Olufsen Medicom A/S, where diode lasers are used as light sources and optical fibre bundles collect the reflected light [62] (see Figure D9c).



**Fig. D9.** Four systems for measurements of the optical properties of tissue are depicted. (a) The optically integrating sphere system. (b) The imaging Fourier transform spectrometer. (c) The Bang & Olufsen fibre probe system. (d) The time-resolved measurement system.

A third means of extracting optical properties is based on time-resolved measurements of the dispersion of very short laser pulses propagating through the tissue (see Figure D9d). This method requires slightly more advanced and expensive technology than the others, but has several important advantages. For example, it is suitable for investigating deep into the tissue, and it is possible to perform tomographic reconstruction of the optical properties inside the body. At the Division, both pulsed diode lasers and a mode-locked Ti:sapphire laser are available as light sources. A time-correlated, single-photon-counting detection instrument is used to obtain the time-resolved data.

White-light, ultra-short, high-power pulses can be employed to obtain spectral information from time-resolved measurements, see figure D8. Either the 10 Hz terawatt laser, or the new kHz terawatt laser, at the Division can be focused in a suitable material, such as water, to give femtosecond pulses with a broad spectrum due to nonlinear effects. By using a streak camera to detect the light, it is possible to obtain virtually the same information as with the time-resolved system, but at many wavelengths simultaneously.

To extract the optical properties from the measurements, advanced numerical methods have been developed. For the integrating sphere and the diffuse reflectance systems, Monte Carlo simulation is usually employed as the model for light propagation. It is also possible to directly calibrate the system against a set of



phantoms with known optical properties. This was done for the fibre probe system. Irrespective of which method is used, the nonlinear nature of the problem calls for special care. A powerful method that relies on fitting the results from the light propagation model (or calibration measurements) to multidimensional polynomials has been developed. The optical properties can be determined by comparing the measured data with the polynomials using a generalized Newton-Raphson algorithm.

The time-resolved measurements can be modelled successfully using diffusion theory. Since the distance between the light source and detection point is relatively large, and the scattering is high, the propagating light resembles a diffusing "gas" of photons.

Applications of these types of measurements have mainly been focussed around diagnostic measurements of breast cancer - optical mammography [63,64] and to measure concentration of photosensitizers during PDT [65].

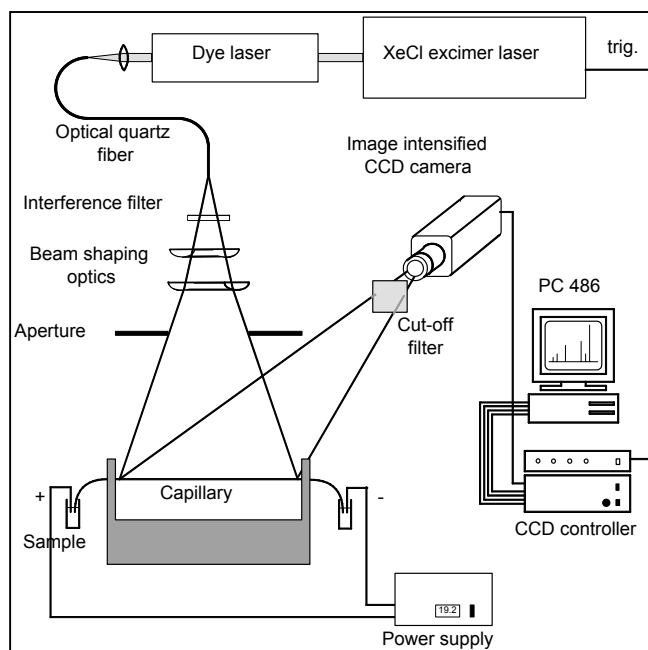
Besides investigating methods for measurements of the optical properties of tissue, one project at the Division has been aimed at measuring the optical properties of flowing blood, and another to construct an eye model with controllable lens scattering [66]. Knowledge of the scattering and absorption of blood is important in diagnostic applications, when monitoring parameters such as haemoglobin concentration, oxygenation, and various other constituents of blood. The optical properties were measured in the integrating sphere set-up, and yielded new information on how the scattering from red blood cells changes due to varying flow conditions [67].

Research has also been conducted to measure gas content inside turbid media using the new technique developed at the Division called GASMAS - Gas in scattering media absorption spectroscopy [68-70]. This work has been conducted together in a collaboration between the environmental and medical groups at the Division, and is described in Chapter C of this report.

### **D3. Analytical chemistry**

*Thomas Johansson*

The different detection techniques used in analytical chemistry constitute an interesting area of research. UV absorption and LIF are the two main techniques used for detection in liquid samples. Collaboration between the Divisions of Atomic Physics and Technical Analytical Chemistry at the Lund Institute of Technology was initiated a few years ago, and several projects are now in progress.



**Fig. D10.** Set-up used for fluorescence imaging of a capillary

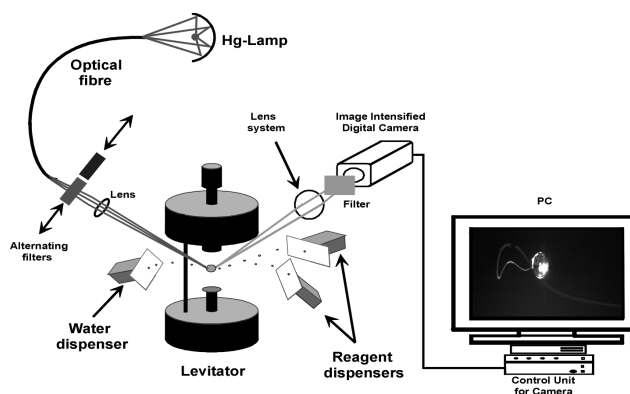
### Analysis in the area of Analytical Chemistry

A common approach in analytical chemistry is to use some sort of separation technique. In one technique, called capillary electrophoresis, the analytes of the sample are separated in a thin capillary under the action of an electrical field. Detection is usually performed at the end of the capillary and light absorption is one detection technique frequently used. Another way to detect the analytes is to use fluorescence detection, but in this case the analyte must be self-fluorescing. If the analyte has no self-fluorescence one can use a technique in which the analyte is bound to a fluorescent particle [71]. The detection system used for fluorescence imaging for this purpose is shown in Figure D10.

Levitation is a technique in which the sample volume floats in the air, i.e. in a wall-free test-tube, with an air interface instead of a boundary with other materials. In this way, no walls can interact with the species in the sample. Also, adsorption of the substances onto the sample container is avoided. For detection purposes, this is an advantage because the background signal is lower, since there are no walls for the species to adsorb onto. Another advantage is the low consumption of sample and reagent, although other micro-techniques also exist. When such small volumes are used, the surface-to-volume ratio increases and strong surface tensions will influence the sample. This wall-free technique is favourable compared with other micro-container techniques, because of the low adsorption at the air-solution interface, as mentioned above.

### Monitoring of a levitated drop

The complete system used in the experiments is shown in Figure D11. The central part is the levitator and for sample delivery a droplet dispenser is used. This dispenser delivers 30-100 pl droplets at a rate of up to 9000 per second. Reagent

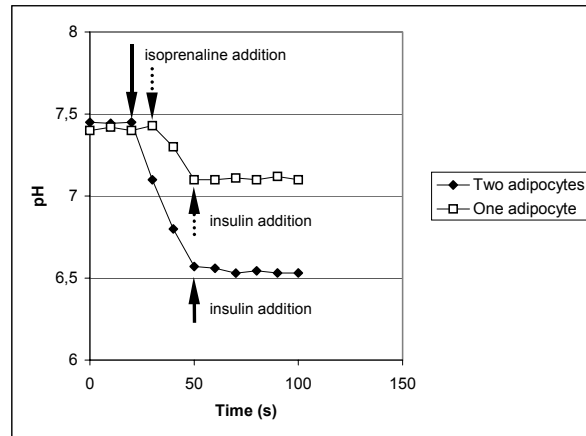


*Fig. D11. Instrumental set-up for fluorescence imaging of a levitated drop.*

solutions are also added to the drop with this technique. Due to the wall-free sample containment, the volume of the drop decreases with time due to evaporation. To compensate for such losses and keep the concentration of the reagents constant, a dispenser is delivering droplets of the solvent to the drop.

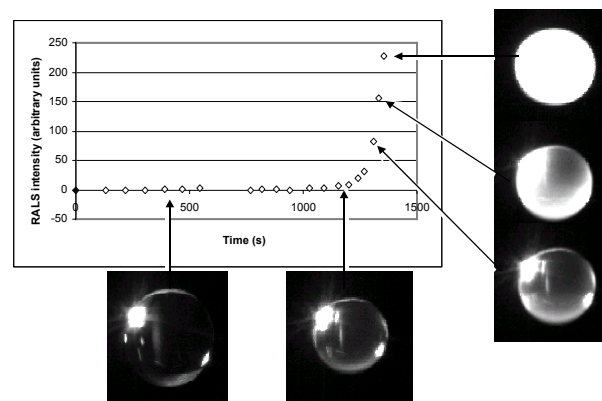
Different techniques can be used for the determination of the properties of the levitated drop. To determine the pH of the drop, LIF was used. Here, the change in pH is monitored by the use of 8-hydroxypyrene-1,3,6-trisulphonic acid (HPTS), which is a pH-dependent fluorophore. HPTS has two major absorption wavelengths, 405 and 450 nm, and it fluoresces at 511 nm independently of the excitation wavelength. The excitation light used with the wavelength 435 nm is absorbed in a lower amount compared with light of 450 nm. The cross-section for absorption at the different wavelengths depends on the deprotonation of the HPTS molecule. By acquiring images of the emitted fluorescence from the drop at the two excitation wavelengths and calculating the intensity ratio, a relative pH is determined. To calibrate the system, different pH solutions were used.

In one experiment the aim was to detect the increase and decrease of the pH due to adipocyte (fat cell) lipolysis (fat mobilisation), where every free fatty acid (FFA) can release a proton. FFA:s are produced when adipocytes are stimulated with isoprenaline, which is a synthetic beta-adrenergic agonist. By adding insulin, the reaction is inhibited. Figure D12 shows the change in pH in an experiment and when insulin is added, the reaction is inhibited.



**Fig. D12** Changes in pH during an experiment with fat cells

In another experiment, protein crystallisation was studied [72]. When molecules, e.g. salts or proteins, in a solution are slowly brought into supersaturation, small aggregates of the molecules are formed. This is the beginning of a precipitate or a crystal. The theoretical and practical knowledge of the process is not yet fully described for the formation of macromolecules. Here, the formation of protein nuclei was detected by imaging of scattered white light. The experimental set-up is similar to the one for fluorescence measurements. A technique named right angle light scattering (RALS) was used for these experiments. Here, a light beam is directed towards the levitated drop and the camera (at 90 degrees angle), without any filter, collects the images of the scattered light. By monitoring the mean intensity of the scattered light from an area in the image, excluding specular reflections from the light source, a change in intensity is detected when the concentration of the aggregates increases as shown in Figure D13. However, calibration of the concentration of aggregates was not made because the aim was to indicate the onset of precipitation. This was recorded when the scattered light intensity was above the limit of detection, indicating the formation of aggregates.



**Fig. D13.** Image series of a levitated drop during a precipitation experiment of a protein

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# E. Teaching

The personnel from the Atomic Physics Division contribute to the teaching of basic courses in physics at the Lund Institute of Technology. The Division is responsible for a number of elective courses which are listed below.

## E1. Specialized courses

The specialized course *Laser Physics* deals with the physical principles of lasers, the most common types of lasers and their applications. In laboratory practicals the students learn how to study the fundamental properties of different lasers and to use the laser as a tool in optical measurements.

The specialized course *Atomic and Molecular Spectroscopy* provides knowledge about modern atomic and molecular spectroscopy with special emphasis on technical applications. Together with the laser physics course, this course forms the natural introduction to graduate studies at the Division of Atomic Physics.

The course in *Multispectral Imaging* deals with the extraction of physical and chemical information from images. The course covers imaging using radiation ranging from X-rays to microwaves, and applications from astronomy to microscopy. Four advanced laboratory exercises are included in the course.

A specialized course in *Advanced Optics* has been established at the Division. This course emphasizes Fourier optics, interferometry, fibre optics, holography and phase-conjugation techniques. There is a course in *Medical Optics*, covering light transport in strongly scattering media (such as living tissue) and laser-based therapeutic methods.

The course in *Non-linear Optics* is given in alternate years. It is essentially a theoretical course, which provide the background to non-linear interactions between light and matter.

The course *Advanced Atomic Physics* takes the students from the atomic physics foundations relying on quantum mechanics to the research front, laser cooling and trapping, atoms in strong laser fields, antihydrogen etc. It is partly a theoretical course aiming at providing to the students the basic time-dependent and time-independent theoretical methods for diverse atomic physics calculations and partly an experimental course with laboratory exercises in modern atomic physics.

## E2. Graduate teaching

Several of the specialized courses in Section E1 are given jointly for graduate and undergraduate students. These include Advanced Optics, Atomic and Molecular Spectroscopy, Laser Physics, Multi-Spectral Imaging, Medical Optics, Non-linear Optics and Advanced Atomic Physics.

## E3. Master's Projects

Several undergraduate students are performing their Master's projects within the Atomic Physics Division. Below, those who completed their projects during the present period are listed, together with the title of their dissertations.

<b>Tomas Christiansson</b>	<i>A first step towards quantum computing in rare-earth-ion-doped crystals LRAP-266</i>
<b>Jenny Pettersson Mikael Sebesta</b>	<i>Evaluation of microscopic FRET measurements to analyse proteine interactions in human cells LRAP-268</i>
<b>Matilda Träff</b>	<i>Improvement of the temporal contrast of a femtosecond laser pulse with a non-linear Sagnac interferometer LRAP-269</i>
<b>Mikael Sjöholm</b>	<i>Development of a laser spectroscopic technique for gas in scattering media LRAP-271</i>
<b>Mattias Kuldkepp</b>	<i>Construction and design of a high-power double-pass laser-diode amplifier LRAP-273</i>
<b>Anna Fragemann</b>	<i>Design and construction of a laser display and a new electro-optic modular LRAP-274</i>
<b>Albin Roslund</b>	<i>Fluorescens från isolatorytor LRAP-275</i>
<b>Daniel Karlsson Henrik Nilsson</b>	<i>A study of two-photon excitation in turbid media - Possibilities in photodynamic therapy LRAP-277</i>
<b>Kristina Nilsson Mikael Otendal</b>	<i>A first step towards detection of biofilm using fluorometry LRAP-278</i>

<b>Markus Persson</b>	<i>Stark shifts and ion-ion interaction in Europium doped YAlO<sub>3</sub> -On the road to quantum computing LRAP-279</i>
<b>Jenny Svensson Anna Ralsgård</b>	<i>Biomedical fibre optic thermometer based on Cr<sup>3+</sup> -doped crystal fluorescence - A feasibility study  LRAP-280</i>
<b>Johanna Ekström</b>	<i>Development of software for a diode-based fluorosensor LRAP-281</i>
<b>Robert Saers</b>	<i>Stabilising a ring dye laser to iodine transitions for quantum computing and photon echoes LRAP-282</i>
<b>Martin Rupinski Henry Lehtiniemi</b>	<i>An innovative method of measuring the optical properties of tissue LRAP-283</i>
<b>Per Johnsson</b>	<i>Adaptive laser pulse shaping using an acousto-optic programmable dispersive filter and genetic algorithm LRAP-291</i>



