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Table of Contents

Introduction

The Division of Atomic Physics, Lund Institute of Technology (LTH), is responsible for basic physics teaching in all engineering disciplines and for specialised teaching in Optics, Atomic Physics, Spectroscopy, Laser Physics and Non-linear Optics. Research activities of 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 seven divisions comprising the Physics Department, Lund University. Since 1980, biennial progress reports have been issued within the series Lund Reports on Atomic Physics (LRAP). Our latest report, covering 1991-92 was LRAP-144, preceded by the reports LRAP-20. LRAP-43, LRAP-85, LRAP-90 and LRAP-119. The present report describes the activities of our division during the calendar years 1993 and 1994. The research programme consists of a number of basic and applied projects some of which are pursued jointly leading to mutual benefits.

Research at the Division of Atomic Physics takes place in a multi-disciplinary atmosphere, in which informal collaborations with external scientists and industry form an important part. This network is sometimes referred to as the *Lund Laser Centre*, in which the new Division of Combustion Physics (Prof. Marcus Aldén) forms an important part. During this two-year period the Centre has been strengthened by the formation of a new research group in Chemical Dynamics at the Chemical Centre. Villy Sundström, a renowned researcher in the field of ultra-fast spectroscopy with photosynthetic studies as his speciality, was appointed professor and head of this group. We wish him and his colleagues all success in their research activities. Three umbrella organisations, the Combustion Centre, The Environmental Measurement Techniques Centre and the Medical Laser Research Centre, continue to play important roles in fostering collaboration within laser applications in energy, environmental and medical research, respectively. An Institutional Fellowship grant within the Human Capital and Mobility programme of the EC has been received for work at the Lund Laser Centre.

Research activities at the nationally available High-Power Laser Facility, which is operated by the Atomic Physics Division, have been very intense during the first two years of operation, since its inauguration at the end of 1992. All laser systems have been efficiently utilised by local and national researchers and their international collaborative partners. In particular, the Division is participating in three European networks (Human Capital and Mobility) with intense activities at the Facility in Lund. Presently, one European postdoc. is active in Lund within each network (a French, a British and a German). Many foreign guests have spent research periods at the facility in Lund. High harmonics have been studied extensively and the first application experiments have been performed, where the lifetime of the 2p state of helium was measured following XUV excitation at 58.4 nm. A narrow-band fully tuneable laser system in conjunction with low harmonics generation has been used in these spectroscopic measurements. Within our X-ray laser research programme, which has been pursued together with Max-Planck Institut für Quantenoptik, amplification has been observed in the XUV region following optical field-ionisation (OFI) pumping in nitrogen gas. Many more gaseous elements have been studied and experiments with solid targets are also being pursued. Broadband X-rays are produced by focusing on rotating solid targets. The properties of the radiation are being studied and radiological applications are being investigated together with medical experts. Our terawatt laser system has operated very satisfactorily. The beam quality is being upgraded by

using a saturable absorber, frequency doubling and spatial filtering. Upgrading of the laser to higher brightness is also in progress.

Extensive research activities concerning time-resolved laser spectroscopy in the VUV region have also been pursued, using four-wave mixing or Raman shifting in the generation process. Rydberg sequences in free atoms have been investigated, and resonance lines in atoms and ions, observed by the Hubble Space Telescope, have been studied. Atomic beams or laser-produced plasmas have been used in the experiments.

High-resolution (CW) laser spectroscopy is being utilised in several projects. High-contrast transmission spectroscopy has revealed sub-natural linewidths that are now being interpreted theoretically. Collisional laser spectroscopy is a new research field in Lund, where several aspects of alkali-dimer/atom collisions are being investigated in a collaboration with the Riga University, Latvia. Our diode-laser spectroscopic programme has now entered a very productive phase where many studies involving frequency-modulation techniques have been completed. In particular, accurate studies of lineshapes have been made experimentally and theoretically, allowing accurate gas concentration data to be presented.

Theoretical atomic physics at the Division supports our experimental programme and is centred around multi-configuration Hartree-Fock methods for the calculation of radiative properties, hyperfine structures and isotopic shifts. Relativistic methods have now also been introduced into the programme which is being pursued in close collaboration with Vanderbilt University, USA, Université de Bruxelles and the Quantum Chemistry Group, Chemical Centre, Lund.

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.

The programme concerning novel methods for high-resolution microscopy is also progressing well with further developments of a debris-free, laser-produced plasma X-ray source. Trapped particles used for second-harmonic generation have been demonstrated to provide an interesting light source for near-field microscopy, and first images have been produced. Both methods share the common goal of developing small, reasonably priced microscopes suitable for use in research.

Optical remote sensing at the Division of Atomic Physics consists of differential absorption lidar monitoring of atmospheric pollutants and fluorescence lidar studies of vegetation. Apart from monitoring of industrial effluents, the atmospheric work is focused on geophysical gas emissions from mining, geothermal and volcanic activities. Thus, the total atomic mercury emission at the largest mercury mine in the world at Almaden in Spain, was determined. Last summer, emissions from the Italian volcanoes Etna, Stromboli and Vulcano were again studied on board the Italian research vessel "Urania" making passes under the volcanic plumes. Our collaborative partners were CNR-IROE (Firenze) and Istituto Nazionale di Vulcanologia (Catania). The lidar group is now in the process of updating the mobile laboratory, developing new software and exchanging the dye laser transmitter for a solid-state source. Fluorescence lidar work is being pursued within the

EUREKA project LASFLEUR. Spectral (point) monitoring and multi-colour imaging of vegetation are being performed. Important experience was gained in a field test at Avignon, France. A new concept for fluorescence imaging using "push-broom" techniques (successive line imaging), very suitable for airborne operation, has been demonstrated.

The research activities within the Lund University Laser Centre have further developed during the last two years. A main part of the research deals with malignant tumour detection and treatment. A core group consisting of 8 physicists, 4 physicians and sometimes also biochemists is now located together at the Physics Department ensuring a close and daily interaction daily and close interaction. Members from 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 and Pathology at the Lund University Hospital. A project concerning optical brain diagnostics is being pursued together with the Brain Surgery Clinic at Linköping University Hospital. Several joint measurement campaigns have also been pursued at foreign clinics where Lund researchers introduced, tumour diagnostic systems, for example at the Medical Laser Centres in London and Lübeck, at the ENT Department in Porto, Portugal and at the Urology Clinic of the University of Leuven, Belgium.

Point spectral monitoring and multi-colour fluorescence imaging of malignant tumours in different clinical specialitites are being performed, allowing small and early tumours to be detected. Photodynamic treatment has now been firmly established in Lund with treatment of hundreds of tumours. The use of the haem precursor ALA, applied topically on the lesion or administered orally, has meant a breakthrough in the clinical application. A clinical study for basal cell carcinomas is now being performed with support from a major health-care company. Apart form assessing the therapeutical results, fluorescence and Doppler perfusion imaging are used to obtain insight into the processes involved. Photodynamic treatment has also been used for lung, ENT, oesophageal and gynaecology patients. Our multi-colour fluorescence imaging system has been tested in different clinical specialities and continuing research is partially supported by the Canadian company Xillix under a special contract with Lund University. In parallel with the clinical work, studies of new sensitisers are performed on animals. Work on the cell level involving two-photon and confocal fluorescence microscopy is also being pursued to increase our understanding of photodynamic imaging and treatment. A further new aspect of our medical work is laser-induced hyperthermia, which is being studied through both theoretical modelling and animal tumour treatment.

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 ionizing radiation. Promising results have been obtained with techniques varying from terawatt laser-induced white-light illumination to diode-laser time-resolved spectroscopy. The work, which also includes phase-modulation spectroscopy, is being carried out in a collaboration with the Department of Diagnostic Radiology in Lund.

Emission spectroscopy has proved to be a powerful technique for industrial monitoring of pyrometallurgical processes involving copper and steel. Fluorescence and scattering spectroscopy are employed for characterisation of paper and pulp. Steady-state as well as time-resolved techniques are being used. Optical and laser techniques are also utilised in an-

other industrial project, in which the insulating properties of oils are being studied. Laser-induced breakdown in the insulating fluid is used to trigger discharges in high-voltage devices. The project, which is supported by ABB, is aimed at an increased understanding of the origins of electric breakdown. Research at the Atomic Physics Division has now generated five spin-off companies: OPSIS AB, SEMTECH AB, SPECTRAPHOS AB, INSPECTRA ANALYS AB and LIGHTEN AB (the last two during the period reported here).

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 171 papers have appeared in this series. At the end of the period covered by this Progress Report the staff of the Division of Atomic Physics totalled about 67. This number includes 23 full-time postgraduate students and 4 long-term post-doctoral visitors. Four part-time students and a record number of Master's students (18) have been pursuing their projects at our division during the last two-year period. It is through the dedicated work of all the research, teaching and support staff that the accomplishments reported hare have been made possible.

We are very grateful for the support of a large number of funding agencies, in particular the Swedish Natural Sciences Research Council (NFR), the Swedish Research Council for the Engineering Sciences (TFR), the Swedish Board for Technical and Industrial Development (NUTEK), the Swedish Space Board (RS), the Swedish Cancer Society (RmC), the Swedish Medical Research Council (MFR), the Council for Planning and Coordination of Research (FRN), the Knut and Alice Wallenberg Foundation (KAW) and the Crafoord Foundation.

Special thanks are due to Dr Claes-Göran Wahlström, who has invested a great deal of time, patience and skill in serving as the editor of this progress report.

Sune Svanberg
Head of the Division of Atomic Physics

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Göran Werner

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Dr Uldis Berzinsh (University of Latvia, Riga)
Dr Bertrand Carré (CEA, Saclay)
Mirianos Chachisvilis, M.Sc. (Umeå University)
Dr Romualdas Danielius (Vilnius University)
Prof. Peter Erman (KTH, Stockholm)
Dr Ernst Fill (Max Planck, Garching)
Dr Leszek Frasinski (Reading, U.K.)
Matthias Grätz, Dipl. Phys. (University Kiel)
Dr Michel Godefroid (University of Brussels)
Dr Paul Hatherly (Reading, U.K.)
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Dr Roland Smith (Imperial College, London)
Christian Strömholm, M.Sc. (KTH, Stockholm)
John Tisch, M.Sc. (Imperial College, London)
Dr George Tsakiris (Max Planck, Garching)

Ph.D. Theses

Jonas Johansson	1993-09-24	Fluorescence Spectroscopy for Medical and Environmental Diagnostics, LRAP-148
Pär Ragnarson	1994-04-22	Optical Techniques for Measurements of Atmospheric Trace Gases, LRAP-152
Eva Wallinder	1994-04-29	Application of Lidar Techniques in Environmental Studies, LRAP-154
Jörgen Larsson	1994-06-03	Laser Spectroscopy on Atoms and Ions using Short-Wavelength Radiation, LRAP-156

Licenciate Degrees

Lars Malmqvist	1994-06-01	Nonintrusive Probes for Scanned Near-field Optical Microscopy, LRAP-160

DIVISION OF ATOMIC PHYSICS
LUND INSTITUTE OF TECHNOLOGY
Head: S. Svanberg
Deputy head: W. Persson

Research Programme
(December 1994)

Basic Atomic Physics	**Applied Optics and Quantum Electronics**	**Optical Remote Sensing**	**Medical Applications**	**Industrial Applications**
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(J. Carlsson)	M. Berglund	P. Andersson	R. Berg	P. Bårmann
M. Borg Gaarde	U. Elman	H. Axelsson	J. Johansson	J. Carlsson
P. Jönsson	U. Gustafsson	H. Edner	Ch. Lindquist	(S. Kröll)
A. L'Huillier	H. Hertz	B. Galle	Liu Lexin	L. Malmqvist
H. Lundberg	P. Kauranen	S. Wallin	A. Nilsson	W. Persson
Luo Caiyan	S. Kröll		I. Rokahr	A. Sunesson
I. Mercer	Luo Baozhu		Ch. Sturesson	W. Wendt
E. Mevel	L. Rymell		K. Svanberg	
A. Persson	L. Malmqvist		I. Wang	
(W. Persson)				
T. Starczewski				
J. Steingruber				
C. Tillman				
C.G. Wahlström				
R. Zerne				

Names in parenthesis are also given under their main activity heading

47 Research Personnel (19 Ph.D., 28 Grad. Student)
20 External Positions
Total Personnel, Div. of Atomic Physics: 67

I Basic Atomic Physics

Research activities in basic atomic physics during the last two years have been strongly influenced by the establishment of the *Lund High-Power Laser Facility* [1]. This facility, which was brought into operation at the end of the previous two-year period, has opened doors to new fields of research. This is true in particular for basic atomic physics, but also applies to different areas of, e.g., photochemistry, medical diagnostics and industrial applications.

The High-Power Laser Facility consists of three major laser systems, all operating at 10 Hz repetition rate. The first is a narrow-bandwidth, tunable system with a pulse duration in the nanosecond range. This is designed to be used, in particular, for pulsed laser spectroscopy in the UV and VUV spectral ranges. The second system, being a mode-locked, Q-switched Nd:YAG laser, gives pulses in the range 30 to 300 picoseconds. It can be used either directly or after frequency upconversion in non-linear crystals. Most frequently, however, it is used to pump a short-pulse dye laser. The third, and most exciting of the three systems, is the terawatt laser. This system is based on chirped-pulse amplification in titanium-doped sapphire and provides 150 fs pulses of terawatt power. An upgrade of this system to still higher peak power is in progress. Continuous tunability over a large spectral range can be obtained with a specially designed optical parametric amplifier (OPA), pumped by the terawatt laser [2]. The laser radiation from each system can be sent through special beam ports connecting the different laser rooms and the target area rooms. The lasers can hence be combined in various ways to allow complex experiments to be performed. The layout of the facility is shown in Fig. 1.

The Facility is nationally available and open for Swedish scientists and their international collaborators. However, it is part of the Division of Atomic Physics, and is operated entirely by the staff of this division. Sometimes, experiments initiated by external groups have also involved the participation of local scientists. Examples of this are the accurate determination of the ionisation potential of the CO molecule using the VUV laser system [3], chemical dynamics studied by means of white-light continuum generation and femtosecond absorption spectrometry [4] and the investigation of molecular fragmentation after multi-electron ionisation, using the terawatt laser [5].

Most of the work using the terawatt laser and part of the work with the picosecond laser has been devoted to the study of high-order harmonic generation in gases and plasmas, to X-ray laser related investigations and to the generation and applications of hard X-rays from laser-produced plasmas. This research is partly performed in international collaborations through our participation in three different European networks. It will be described in the following sections, together with a brief presentation of some experiments on emission spectroscopy of highly ionised species (Sect. A1-A4).

Our long tradition in time-resolved laser spectroscopy in the visible and UV spectral regions has been much extended during the last two years through work with the UV/ VUV laser system. A number of investigations of atomic and molecular excited states has been performed. In order to measure very short radiative lifetimes, and to perform spectroscopy in the XUV spectral range, new techniques using the picosecond laser

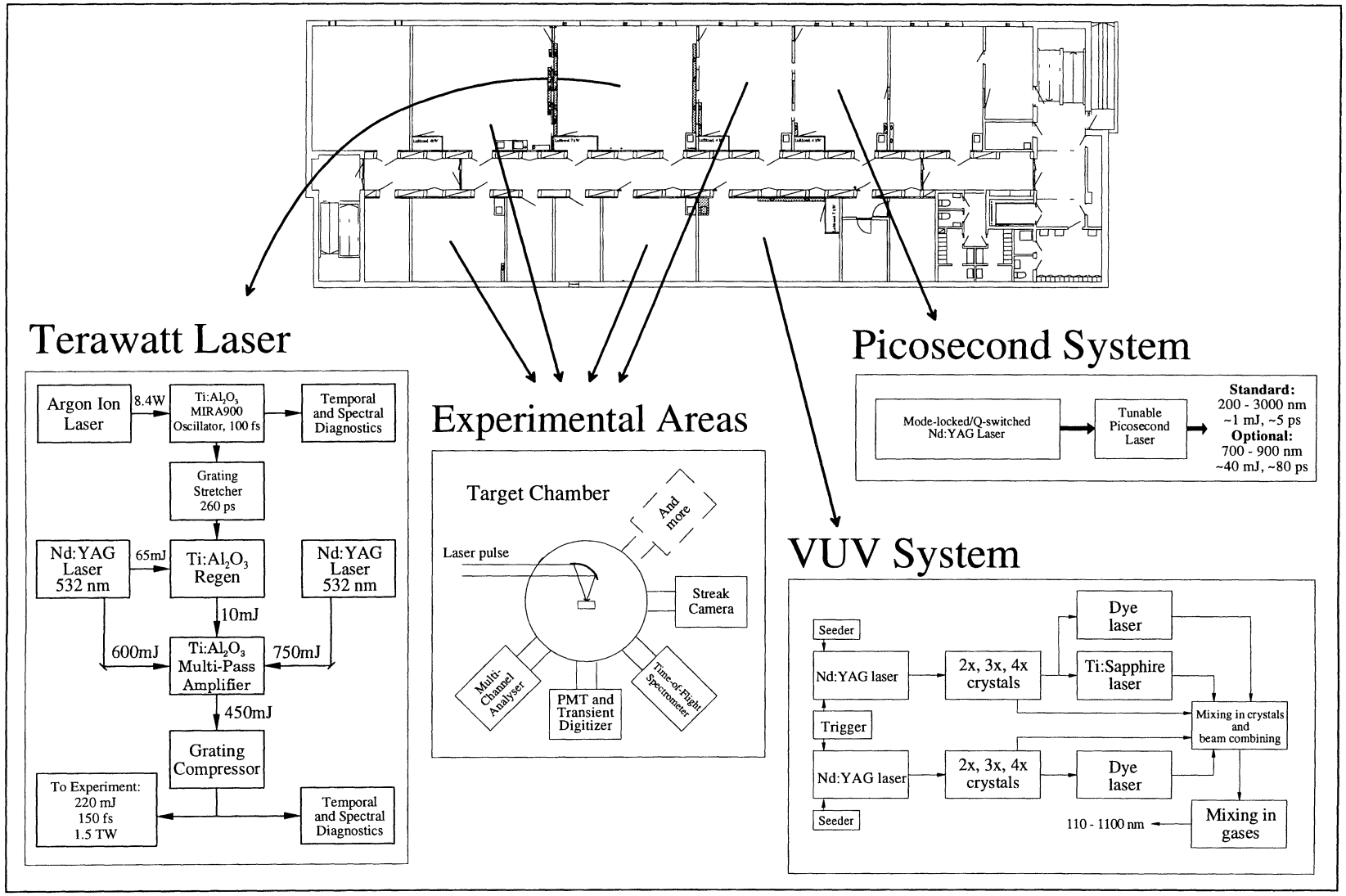

Fig. 1. *Layout of the nationally available high-power laser facility.*

system have been developed. In this way, the temporal and spectral ranges for spectroscopic investigations have been still further extended. This is all described in Section B. In Section C, we present our recent activities using continuous lasers in the visible region. They represent the continued development of high-contrast transmission spectroscopy, and the introduction of a new field of research into the group: the study of collision processes using high-resolution laser spectroscopy.

The activities in theoretical atomic physics, outlined in Section D, have continued along the same path as previously. Oscillator strengths, radiative lifetimes, hyperfine splittings and isotope shifts have been calculated with great accuracy from numerical atomic wavefunctions. The wavefunctions were obtained through large-scale calculations using the multiconfiguration Hartree-Fock approach.

During the past two-year period there has been a very international atmosphere in the basic atomic physics group. Senior scientists, post-doctoral researchers and PhD students from more than twelve different countries have visited the group and participated in experiments or in theoretical investigations. Much of the work in the group has been presented at international conferences on atomic physics, astrophysics, spectroscopy, strong-field interactions and quantum electronics [54]. The work is also described in a number of recent articles of review character [55,60]. During the period, two MSc projects have also been completed [61,62] and one of the students in the group has defended his PhD thesis [63].

A Atomic physics with high-power laser radiation

When laser pulses from a terawatt laser, such as the one in Lund, are focused, intensities of the order of 10^{18} W/cm^2 or higher can be achieved. Already at about 10^{14} W/cm^2 the electric field amplitude at such a focus is comparable to the field experienced by the outer electrons in heavy atoms. Clearly, conventional perturbation theory cannot properly describe the atomic response to laser pulses with peak intensities in this range. Indeed, recent experiments with high-power lasers have led to the discovery of several unexpected and interesting effects. The ultra-short pulse duration (~100 fs) of these pulses also introduces new types of effects related to non-linear optics, photo-ionisation and plasma dynamics.

A1 High-order harmonic generation

Stig Borgström, Mette Gaarde, Per Jönsson, Jörgen Larsson, Eric Mevel, Anders Persson, Sven-Göran Pettersson, Tomas Starczewski, Sune Svanberg and Claes-Göran Wahlström, Carlo Altucci, Bertrand Carré*, Anne L'Huillier*, Joanna Muffett*, Roland Smith* and John Tisch**

**Visiting scientists*

High-order harmonic generation, using high-power, short-pulse lasers, has been found to be a promising way of generating coherent radiation in the extreme ultraviolet (XUV) and soft X-ray spectral regions. This radiation has unique properties of short pulse length, high peak power and high spectral brightness. With a tunable laser, the generated radiation is

also tunable. Besides its basic research interest (as a probe of the behaviour of an atom strongly perturbed by an electromagnetic field), the harmonic generation process is a novel source of coherent radiation in this short-wavelength range. In the present section we will describe some of our work aimed at understanding the fundamental physics involved in the generation process and on the characterisation of the radiation generated. In Section B3 the application of high-order harmonic radiation in atomic spectroscopy will be discussed.

Study of the cut-off law

The formation and the extent of the characteristic plateau in the harmonic spectra have been investigated by performing systematic studies of the intensity dependence of individual harmonics [A1]. Numerical calculations of the single-atom response, valid in the regime of tunnelling ionisation, have shown that the cut-off energy, i.e. the photon energy of the highest harmonic in the plateau, is well approximated by the simple formula I_p+3U_p. I_p is the ionisation energy and U_p the ponderomotive energy, proportional to the intensity. (The limit is hence given by the maximum value of the intensity seen by the non-linear medium; the saturation intensity for ionisation.) In our experiments, we found that the cut-off energy in the macroscopic field also increased linearly with laser intensity. The constant of proportionality, however, is reduced due to intensity-dependent phase-matching effects [A2]. Harmonic generation in He and He^+ have also been investigated through numerical simulations [A3]. This was done for the KrF wavelength and at intensities beyond the corresponding saturation intensity.

Characterisation of the radiation

In order to make use of high harmonic radiation in different applications, its main features, such as spectral, temporal and spatial profiles, should be well characterised. First, ionisation of the non-linear medium leads to time-dependent changes in the refractive index. This causes a spectral broadening and a blue shift of the laser radiation propagating through the medium. We have investigated how this affects the harmonics generated in the ionising medium. The spectral profile of the 15th harmonic in xenon is illustrated in Fig. A1 for three different intensities. The spectral broadening and the shifts of the harmonics were found to be proportional to the broadening and the shift of the fundamental laser radiation [A1].

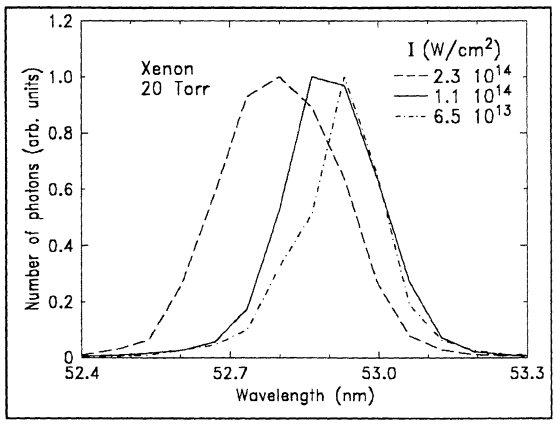

Fig. A1. *Normalised spectral profiles of the 15th harmonic in xenon at 25 mbar, recorded at three different peak intensities.*

Secondly, we performed temporal measurements of the harmonic radiation generated in an ionising gas [A4]. We studied, in particular, the occurrence in time of the fifth harmonic radiation relative to the generating laser pulse. In order to obtain an absolute time reference, we simultaneously generated the harmonics in unsaturated, non-linear crystals and in the ionising gas, recording both pulses with the same streak camera. We found that at low intensities, the harmonic radiation pulses coincided in time with the laser pulses, but as the laser intensity increased above a certain threshold intensity, an intensity-

dependent shift in time between the laser pulse and the harmonic pulse was observed. This behaviour was reproduced in numerical simulations.

Finally, the harmonic spatial profile and its dependency on various parameters, such as focusing, ionisation and intensity-dependent phase variations of the atomic dipoles, was studied theoretically [A5]. It was found that the spatial and spectral coherence depend strongly on the focusing geometry. These properties can be controlled and optimised by moving the laser focus position relative to the nonlinear medium [A6].

Optimisation of the efficiency with respect to the gas pressure

In order to determine the optimum gas pressure to be used when harmonic radiation is generated, we have studied in detail how the time- and space-integrated harmonic signal depends on the gas density [A7]. We found that at low pressures (a few mbars), the harmonic signal increased approximately quadratically with the pressure, as expected for a coherent process. At pressures of a few tens of mbars the yield saturated, and even decreased with further increase in pressure. The saturation pressure, and the pressure for maximum yield, were found to be order dependent for the high-order harmonics in the cut-off regime.

High-order harmonics from alkali-metal ions

To generate harmonic radiation with a very short wavelength (high cut-off energy), one should choose atoms or ions with high ionisation potential as the non-linear medium. The singly ionised alkali-metal atoms look like rare gas atoms, but with much higher ionisation potentials. We have studied high-order harmonic generation in laser-produced Na^+ and K^+ ions [A8]. However, the highest harmonic orders observed (~27) were not as high as expected from estimates based on the saturation intensities for these ions and on the focused intensities in the absence of the medium. The limited extents of the plateaus are explained, at least partly, as being the consequence of ionisation-induced defocusing of the high-power laser beam, reducing the peak intensity obtained inside the medium. The spatial far-field distribution of the harmonic radiation was also found to exhibit complex structures which varied with the focusing conditions.

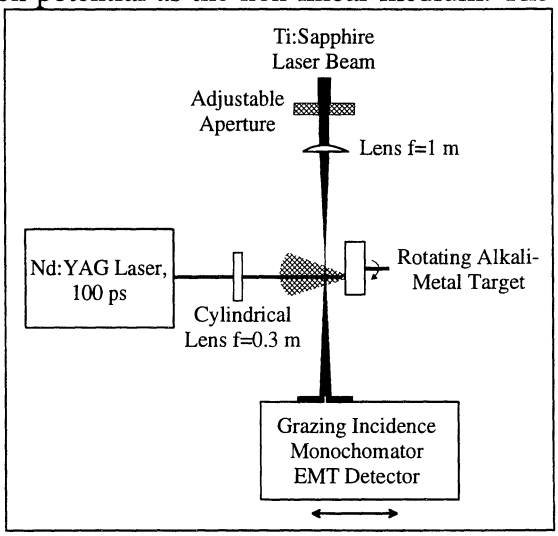

Fig. A2. *Experimental setup to study harmonic generation in laser-produced alkali-metal ions.*

A2 X-ray-laser-related investigations

Stig Borgström, Ernst Fill, Anders Persson, Sven-Göran Pettersson, Tomas Starczewski, Jürgen Steingruber*, Sune Svanberg and Claes-Göran Wahlström*

**Visiting scientist*

The research programme in Lund related to X-ray lasers started about two years ago, at the time of the inauguration of the new terawatt laser. Shortly afterwards, we were also accepted as one of ten members of the European Network for X-ray lasers. The Lund terawatt laser, with its ultrashort pulse duration, high peak power and high repetition rate, may be an interesting pump source for a "table-top X-ray laser". The interest within the scientific community in this kind of pump source is great, because it is much cheaper, more widespread and has a much higher repetition rate than the few existing, enormous X-ray laser establishments in the world. However, the successful realisation of a table-top X-ray laser requires considerable efforts, since a large number of requirements must be simultaneously fulfilled.

Our activities have been divided into studies with solid targets and with gas targets.
In the case of solid targets, we have explored the possibility of focusing the terawatt laser into a plasma, preformed by means of an additional laser. However, we found that the present laser wavelength (800 nm) is too long for sufficient penetration into the plasma. The experiments will be repeated, utilising a newly acquired frequency-doubling crystal, and interesting results are expected. An alternative approach, with solid targets, is to focus the terawatt laser along a line on the target. Experiments are underway together with Dr Ulf Litzén at the Division of Atomic Spectroscopy here in Lund. Preliminary spectroscopic results from a point focus revealed production of very highly ionised species (see section A4).

In our studies with gas targets, we have a fruitful cooperation with the Max-Planck-Institut für Quantenoptik in Garching, Germany. Using optical-field ionisation (OFI) we have investigated a number of gases [A9,A10]. We have found evidence for gain in four-times-ionised nitrogen on, e.g., the transition 3d-5f (Fig. A3). Also, using our new soft-X-ray streak camera we have observed a peculiar time behaviour of the emission from He^+ [52]. These experiments will continue with improved methods and with other gases.

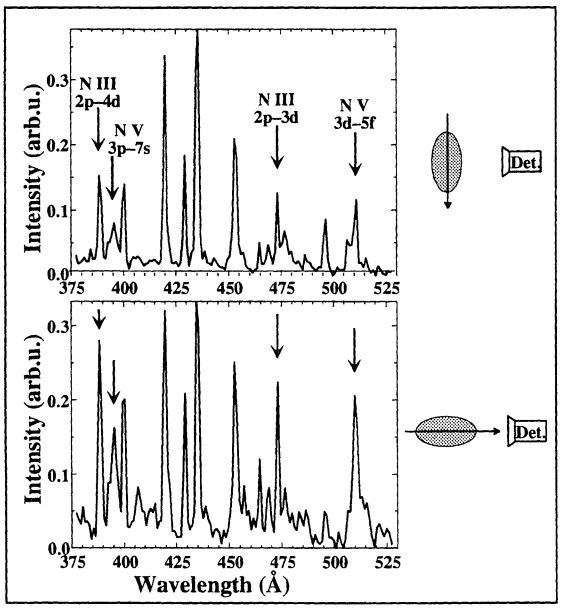

Fig. A3. *Spectra obtained through optical-field-ionisation of nitrogen. The spectra were recorded in the direction of the laser beam and in the perpendicular direction, respectively.*

At the 4th International Colloquium on X-Ray Lasers, held in the USA, we were given the honour of organising the next meeting. The 5th meeting in this series will thus be held in Lund in 1996.

A3 Hard X-rays from a laser-produced plasma - characterisation and applications

Matthias Grätz, Ian Mercer, Anders Persson, Sune Svanberg, Carl Tillman, Claes-Göran Wahlström, Christer Andersson, Anna Göransson* and Christofer Lindheimer**

**MSc students*

Focusing ultra-short pulses from the terawatt laser onto a high-Z solid target (Fig. A4) produces a micro-plasma which radiates X-rays with energies up to the MeV region. The laser intensity in the focal spot is believed to exceed 10^{18} W/cm^2. We have explored various techniques for the characterisation of this X-ray source, each with advantages and drawbacks [A11,A12]. Spectral measurements using an energy-dispersive Ge detector allow high-resolution spectra to be recorded in a single-photon counting mode, although the spectrometer is hampered by *pile-up*, i. e. more than one photon at the time reaches the detector. Operating an X-ray-sensitive CCD array in single-photon counting mode, means that each pixel in the array behaves almost as an independent detector, thus shortening the acquisition time and enabling single-shot measurement to be made. Passive filters and image plates offer an alternative method with high reliability with respect to *pile-up* but with lower resolution. Spectral recordings indicate a relatively larger ratio of characteristic line emission to *Bremsstrahlung* compared with conventional X-ray tubes. Imaging tests with a star test pattern, as well as objects with sharp edges, indicate an extension of the X-ray source of less than 60 μm, but it is believed to be one order of magnitude smaller. The emission time of X-rays has been measured by other groups to be of the order of a few ps.

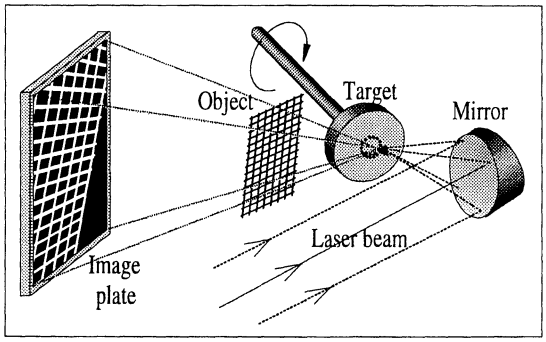

Fig. A4. *A laser beam is focused by an off-axis parabolic mirror onto a rotating solid target. The plasma created is used as an X-ray source for radiographic imaging.*

Some of the unique features of this laser source have been exploited for medical imaging. Since living tissue consists mainly of low-Z elements, contrast enhancement agents are frequently used for improvement of the image quality. For penetration of the human body, relatively hard X-rays are necessary, with X-ray energies in the range 20-100 keV. Most of our experiments have been performed with tantalum as the target material. Tantalum has its characteristic K-line emission in the range 55 to 67 keV and is therefore very suitable as an X-ray source.

Strong characteristic-line emission in combination with contrast enhancement agents allows for differential imaging with the possibility of producing high-contrast images with lower radiation exposure for the patients. Differential-absorption imaging is based on subtraction or division, pixel by pixel, of two images. One image should be exposed using characteristic radiation with an energy below the absorption edge energy, and the second image with characteristic radiation energy above the same edge. In order to use the absorption in the most efficient way, the characteristic line emission should be chosen to be as close to the absorption edge as possible. Preliminary tests have been performed with promising results.

The small dimensions of the X-ray source mean that it can be used for magnification imaging. Fig. A5a) shows a copper grid at high magnification, up to a factor of 80. This image was exposed to X-rays produced by one single laser pulse, showing that the high X-ray flux enables exposure times down to a few ps. One example of biological imaging is presented in Fig. A5b), showing a 4-day-old Syrian hamster with fine details resolved. Parts of this work have been performed in collaboration with the Department of Diagnostic Radiology at the Lund University Hospital.

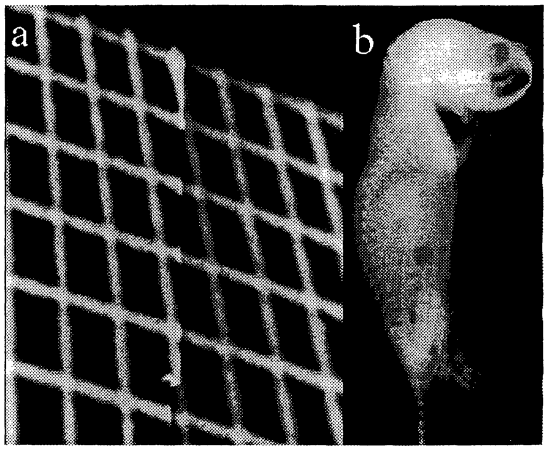

Fig. A5. *a) Copper mesh magnified by a factor of 80; single laser pulse. b) Syrian hamster, 4 days old; 3000 laser pulses.*

A4 Emission spectroscopy of highly ionised species

Stig Borgström, Ulf Litzén, Willy Persson and Claes-Göran Wahlström*
**Atomic Spectroscopy Division*

The plasma formed by focusing the terawatt laser beam onto a solid taget can be used as a source for emission spectroscopy. Due to the ultrashort pulse duration, interesting effects are expected. A project aimed at investigating the line radiation from this type of high-temperature plasma has been initiated as a collaboration with the Division of Atomic Spectroscopy. In a preliminary experiment the radiation from the terawatt laser was focused onto flat solid targets, and the emission from the plasma close to the target surface was recorded in the XUV region. Different focusing conditions using lenses and an off-axis parabolic mirror were tested. The XUV spectra were recorded by means of a 2.2 m grazing incidence Schwob-Fraenkel spectrometer with a 600 l/mm grating and a multi-channel MCP/CCD detector system. The full range of the spectrometer is 5 - 340 Å and the reciprocal linear dispersion at 100 Å is approximately 1 Å/mm.

Recordings were made with carbon, aluminium, vanadium and molybdenum targets. A preliminary examination of the spectra shows lines from highly charged states, such as Ni-like Mo XV, Ne-like V XIV, Li-like Al XI and H-like C VI. In future experiments, the charge-state distributions and the relative line intensities as a function of the focussing conditions will be sytematically investigated. Comparisons will be made with spectra obtained using pulses of much longer duration from the 1 GW, 4 J laser at the Division of Atomic Spectroscopy.

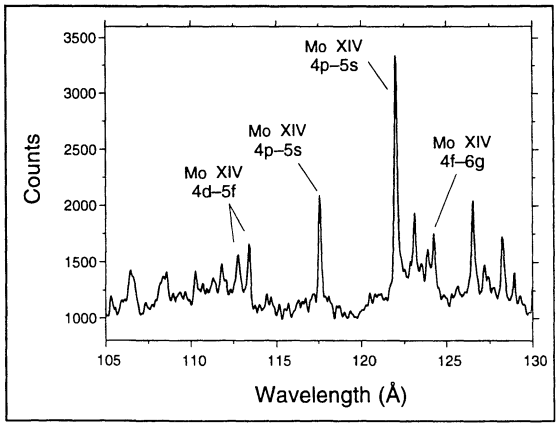

Fig. A6. *Part of a recording of the molybdenum spectrum, showing some 4-5 and 4-6 transitions in Cu-like Mo XIV.*

B Laser-spectroscopic investigations of atomic and ionic excited states in the short-wavelength region

Over many years, time-resolved laser spectroscopy has proved to be an accurate method for the determination of excited-state radiative lifetimes and the determination of energy splitting through quantum beats.

Radiative lifetimes, together with branching ratio measurements, can be used to deduce oscillator strengths. The oscillator strengths of spectral lines are used in astronomy to compute atomic and ionic abundances in the sun as well as in stars, comets and other celestial objects. Experimental lifetime values can also be used to test different theoretical models of atoms and ions. Currently, there is great interest in atomic/ionic lines in the VUV spectral range, since the spectrometer on board the Hubble Space Telescope (HST) can spectrally resolve observations of UV/VUV light. The demand for lifetime data for states requiring excitation by short-wavelength radiation has brought new challenges to the field of time-resolved laser spectroscopy. Apart from the fact that laser radiation in the UV/VUV must be generated, the duration of the laser pulse should be shorter than the excited-state lifetime of the studied atom/ion. Strong lines observed in stellar spectra are often lines connecting a short-lived ionic excited state to the ground state. The lifetimes of these excited states are often about 1-3 ns, too short to be measured with standard pulsed lasers (pulse duration of 10 ns), and other laser systems or other techniques have thus to be used [B1].

The Landé g_J factors, reflecting the coupling of the angular momenta, can also be determined using time-resolved laser spectroscopy. Degenerated atomic levels will split into several sublevels (Zeeman effect) in a magnetic field, where the g_J factor determines the magnitude of the splitting. If two of the sublevels are populated coherently by a short laser pulse, the time-resolved fluorescence intensity will decay exponentially with a superimposed modulation (quantum beat). The modulation frequency depends on the energy splitting and consequently on the g_J factor. The g_J factors can be used for monitoring perturbations and providing data for detailed comparisons with theoretical calculations.

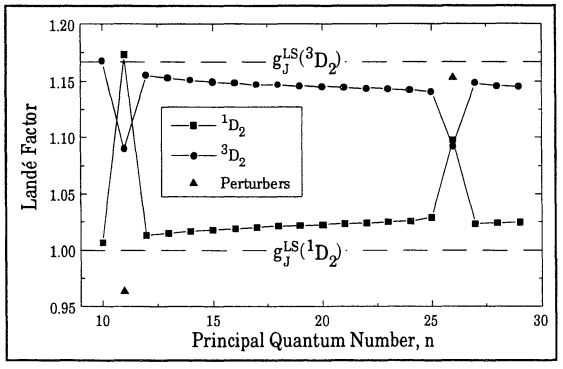

Fig. B1. *Experimental g_J values for the 6snd $^{1,3}D_2$ sequences in ytterbium. Strong perturbation effects for n=11 and n=26 are observed.*

The Landé g_J factors for the strongly perturbed 6snd $^{1,3}D_2$ Rydberg sequences in ytterbium, including both singlets and triplets as well as states from the perturbing configurations, are shown in Fig. B1. This figure shows our measurements of the g_J factors of more than 40 different states using Zeeman quantum beats [B2]. A strong perturbation effect is evident for the n=11 and n=26 states. The results provide a new set of data for checking the theories used in energy and lifetime analysis.

B1 Determination of radiative lifetimes in Rydberg sequences of different elements by time-resolved VUV laser spectroscopy

Uldis Berzinsh, Mette Borg Gaarde, Zhankui Jiang*, Jörgen Larsson, Caiyan Luo, Anders Persson, Sune Svanberg, Claes-Göran Wahlström and Raoul Zerne*

**Visiting scientist*

The long tradition in Lund of experimental determinations of atomic radiative lifetimes using time-resolved laser spectroscopy has been continued during the past two-year period [B3-B13]. Experiments requiring excitation wavelengths in the VUV have been performed and the radiative lifetimes of excited states in atomic copper, magnesium, gold and zinc have been reported [B4-B7]. The measurements on copper and gold will be discussed briefly below.

Short-wavelength laser radiation can be generated in different ways. Efficient frequency mixing in crystals can be achieved for wavelengths down to about 190 nm. To reach shorter wavelengths, stimulated Raman shifting is a feasible method. However, the efficiency of the Raman generation will decrease with wavelength since higher order anti-stokes components have to be used.

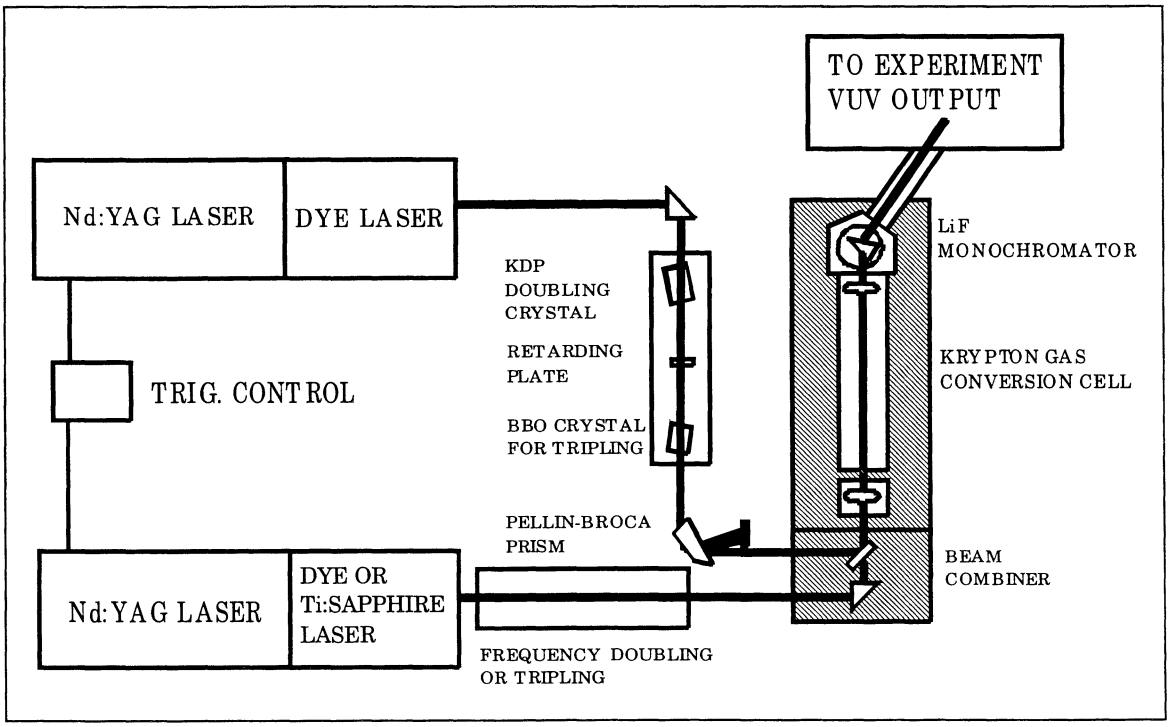

Fig. B2. *Setup for generation of VUV radiation using resonant sum-difference frequency four-wave mixing in krypton.*

One method of generating tunable VUV radiation is through sum-difference frequency four-wave mixing in rare gases or metal vapours with the scheme $\omega_{VUV}=2\omega_R-\omega_T$. By tuning the frequency ω_R to a two-photon resonance, a substantial enhancement in conversion efficiency can be achieved.

The transition primarily used in our VUV system is 4p-5p[1/2,0] in krypton with the wavelength λ_R=212.55 nm for the two-photon resonance. To produce tunable VUV radiation the UV radiation ω_R is mixed with tunable visible or near-UV radiation, ω_T. With this scheme, we can generate VUV radiation in the range 120-200 nm. The VUV radiation produced has a pulse energy of several μJ. A more detailed description of the VUV laser system can be found in [B5,1].

Investigating the $3d^{10}np\ ^2P$ sequence of neutral copper is a challenging task for both experimentalists and theoreticians. A strong configuration interaction with the doubly excited $3d^94s4p$ states gives rise to irregular fine structures, anomalously short lifetimes and highly irregular hyperfine structures. In one of our recent experiments, accurate lifetime data for the (n=6-9) $np\ ^2P_{1/2,3/2}$ states have been obtained [B5]. The excitation wavelengths for the measured states were produced through resonant sum-difference four-wave mixing, except for the $6p\ ^2P_{1/2,3/2}$ states, where stimulated Raman shifting was used.

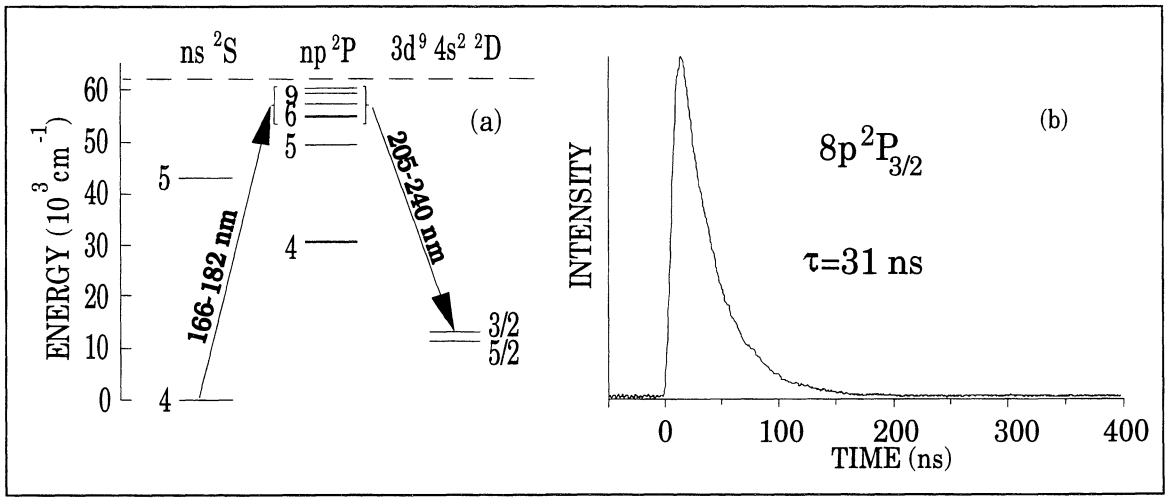

Fig. B3. *a) Partial energy-level diagram for the copper atom with relevant transitions indicated. b) Time-resolved fluorescence from the $3d^{10}8p\ ^2P_{3/2}$ state in copper.*

Gold is another interesting element to study since it is among the few heavy elements that have a simple electronic structure. However, the $5d^{10}np\ ^2P$ Rydberg sequence is strongly perturbed by the $5d^96s6p$ states. We have measured radiative lifetimes in the $np\ ^2P$ sequence for n=6-9, and some of these lifetimes were found to be very short compared with the expected behaviour of the lifetime of a Rydberg state. The lifetimes of four of the perturbing states were also measured.

B2 Ionic-state lifetime measurements for the analysis of space-telescope-observed stellar spectra

Jörgen Larsson, Hans Lundberg and Raoul Zerne

Spectral lines in Pt II, Pd II and other heavier ions have recently been observed in spectra from the chemically peculiar star χ-Lupi, by the Goddard High-Resolution Spectrograph (GHRS) on board the HST. We have made accurate determinations of the radiative

lifetimes for several of the relevant excited states in both Pt II and Pd II. The branching ratios will be determined later at the Division of Atomic Spectroscopy in Lund in order to obtain the oscillator strengths.

A special short-pulse laser was used since the lifetimes of the excited states are too short to be measured with a standard pulsed laser. The short pulses were obtained by pumping a distributed feedback dye laser (DFDL) with the output of an 80 ps mode-locked Nd:YAG laser operated at a 10 Hz repetition rate. Lasers with distributed feedback have been shown to produce pulses for which the product of the pulse length and the bandwidth are near the Fourier limit. The wavelength of such a laser can be changed by varying the angle between the pump beams or by changing the refractive index of the dye solvent. In this experiment we used wavelengths in the range of 720-770 nm [B9].

Using the radiation from the DFDL, together with various frequency-mixing schemes in crystals, we were able to obtain radiation with wavelengths as short as 194 nm. In order to reach the high-lying states to be studied, excitation was performed starting from metastable states. Free palladium and platinum ions were obtained from a laser-produced plasma created with a Q-switched Nd:YAG laser. A high population was obtained for metastable states higher than 20 000 cm^{-1} above the ground state. In order to minimise the effects of collisions in the plasma and other systematic effects which might influence our measurements, we investigated the effect of varying the delay between the plasma-generating and the spectroscopic laser pulse. As a result, we obtained radiative lifetimes of three states in Pt II and four states in Pd II, all shorter than 4 ns [B10,B11].

B3 XUV spectroscopy

Jörgen Larsson, Anne L'Huillier, Eric Mevel, Sune Svanberg, Claes-Göran Wahlström and Raoul Zerne*

**Visiting scientist*

In the extreme ultraviolet (XUV) range (typically below 100 nm), time-resolved laser-induced fluorescence techniques [B6] are limited by poor collection efficiency and temporal resolution. Nevertheless, accurate measurements of the radiative lifetimes of high-lying selectively-excited atomic or ionic states can be achieved by pump/probe time-resolved methods with the pump in the XUV. A synchrotron radiation light source, continuously tunable in the XUV, has recently been used as the pump. However, its synchronisation with pulsed lasers still remains a tedious problem and temporal resolution is limited to a few tens of picoseconds.

We have demonstrated that high-order harmonics emitted when an intense laser is focused into a rare-gas jet provided a table-top, tunable, short-pulse XUV source, ideally suited for pump/probe types of studies [B12]. The spectral bandwidth and the temporal duration of the XUV source should be compatible with the width of the upper level (for efficient pumping) and an accurate measurement of the lifetime respectively. A new laser system based on a distributed feedback dye laser oscillator delivering near-Fourier-transform-limited 80 ps pulses, tunable between 715 and 900 nm has been developed. These pulses were amplified in two dye cells and in a titanium-sapphire crystal up to an energy of 50 mJ. The laser bandwidth was 0.08 nm and the repetition rate was 10 Hz. Harmonics were

generated in a pulsed jet of krypton and separated by a normal-incidence spherical grating. The odd harmonics of the fundamental and those of the second harmonic generated in a doubling crystal, together provided a continuously-tunable, narrow-band XUV light source from 200 nm to 35 nm (21st harmonic) [B9]. We tuned the laser wavelength around 760 nm and selected the 13th harmonic (58 nm, 21 eV) to resonantly excite the $1s2p\,^1P$ state of He. The excited atoms were subsequently ionised by the third harmonic (355 nm, 3.5 eV) of a fraction of the 80 ps Nd-YAG laser pulses whose second harmonic was used to pump the dye laser (see Fig. B4).

The two beams crossed at 45 degrees inside a time-of-flight spectrometer. Both the 13th harmonic and the 355 nm light were approximately focused at the crossing point. A variable delay line placed in the beam path of the third harmonic allowed us to adjust the relative time between the two light pulses. The temporal resolution is limited by the duration of the probe pulse (~ 80 ps) (X-ray streak camera measurements showed that the 13th harmonic pulse duration was of the order of 30 ps). The generated ions were separated in mass in the time-of-flight tube and detected by a microchannel plate (MCP). We measured a radiative lifetime of 0.57 ± 0.03 ns, which is in good agreement with previous experimental data obtained by much more indirect methods (self absorption or electron scattering). The level of accuracy is approximately the same. Furthermore, the method was found to be very sensitive and the measurement could be performed at helium pressures as low as $2\ 10^{-7}$ mbar.

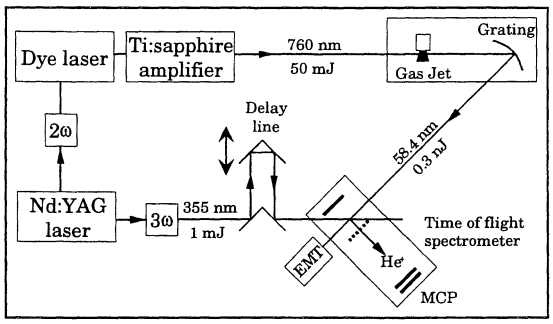

Fig. B4. *Setup for the pump-probe experiment in the XUV.*

The determination of the well-known helium 2p radiative lifetime has demonstrated the feasibility of pump/probe experiments in the XUV region using high-order harmonics. It has encouraged us to continue new investigations of fast processes for even higher-lying excited states. Shorter wavelength (50-10 nm) and (at the expense, however, of the harmonics bandwidth) better temporal resolution (~ 100 fs or less) can be attained by using a pump laser with a shorter pulse duration.

C Laser spectroscopy in the visible

C1 High-contrast transmission spectroscopy

Luo Caiyan, Stefan Kröll, Lennart Sturesson and Sune Svanberg

Sub-natural linewidths and coherent transient effects related to lasing without inversion, in high-density sodium gas, are being investigated in this project.

Saturated absorption spectroscopy linewidths as low as half the natural linewidth have been obtained in high-density sodium gas for the $3s\ ^2S_{1/2} \rightarrow 3p\ ^2P_{1/2}$ transition [C1,C2]. The line narrowing is a direct consequence of the light absorption and propagation properties in highly absorbing media. The sample transmission is proportional to

exp[-α(ν)L], where L is the absorption length and α(ν) is the frequency-dependent absorption coefficient. Assume a pump beam creates a Lorentzian hole in the absorption coefficient α(ν) with a linewidth equal to the natural linewidth. A probe beam can then sense this decreased absorption and experiences increased transmission. If the pump beam is used to carry out an optical pumping process it may, in principle, be possible to obtain zero absorption at the line centre and, for example, one percent transmission off the line centre. The separation between the frequencies of 50% transmission for such a case will clearly be less than the natural linewidth. In this way, subnatural (subhomogeneous) linewidths can readily be obtained. The experimental results have been modelled by rate equation calculations with reasonable agreement [C3]. The calculations specifically treat the optical pumping process. A model with two dimensions in space, one along the beams and one perpendicular to the beams, is used. This model explicitly takes into account the fact that if an atom has been optically pumped to an inactive state in the outer part of the beams, it cannot absorb any more photons as it traverses the beam. This means, for example, that the outer parts of the beams will experience a stronger absorption than the inner parts. This effect will also differ depending on the type of transition (upper state cross-over, lower state cross-over or normal transition) yielding different spatial profiles for different lines (these spatial effects do not require self-focusing).

The experimental setup for observing coherent transient effects is depicted in Fig. C1. AOM denotes acousto-optic modulators. With these, both pump and probe beams can be turned on and off at arbitrary times. Two experimental curves showing the probe beam transmission versus time when the pump and probe beams are turned on and off are shown in Fig. C2. The durations of the observed transients are all much longer than the upper state lifetime of 16 ns. Therefore, we assume that they must arise from ground state effects. Also, according to our understanding, optical pumping changing only the diagonal elements of the density matrix would not be sufficient to explain the observed peaks and dips. It is therefore also necessary to take coherence effects into account. Each state involved is split into 2*F+1 near degenerate Zeeman levels. A crossover resonance, such as the top trace in Fig. C2, thus in total consists of transitions between eleven different levels. In an attempt to simplify the problem, we have tried to model the system using only four levels. We have attempted to model the process by solving the Liouville equation for this four-level system, including the atom interaction with the pump and probe beams, radiative relaxation and the transit time for the atoms through the laser beams [C4]. However, although transient effects similar to those observed in Fig. C2 are obtained, the qualitative agreement is still unsatisfactory.

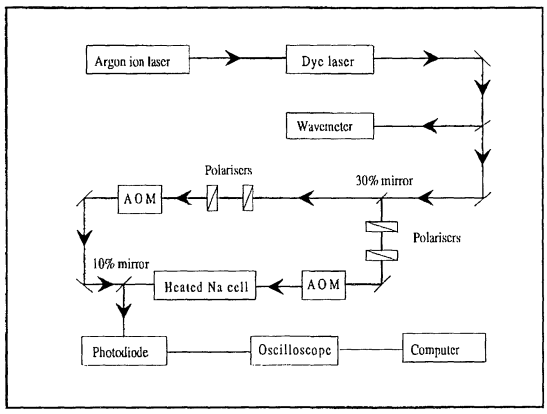

Fig. C1. *Experimental setup for investigating the temporal behaviour of the transmission of a probe beam through an optically pumped, optically thick sample. AOM = Acousto-Optic Modulator*

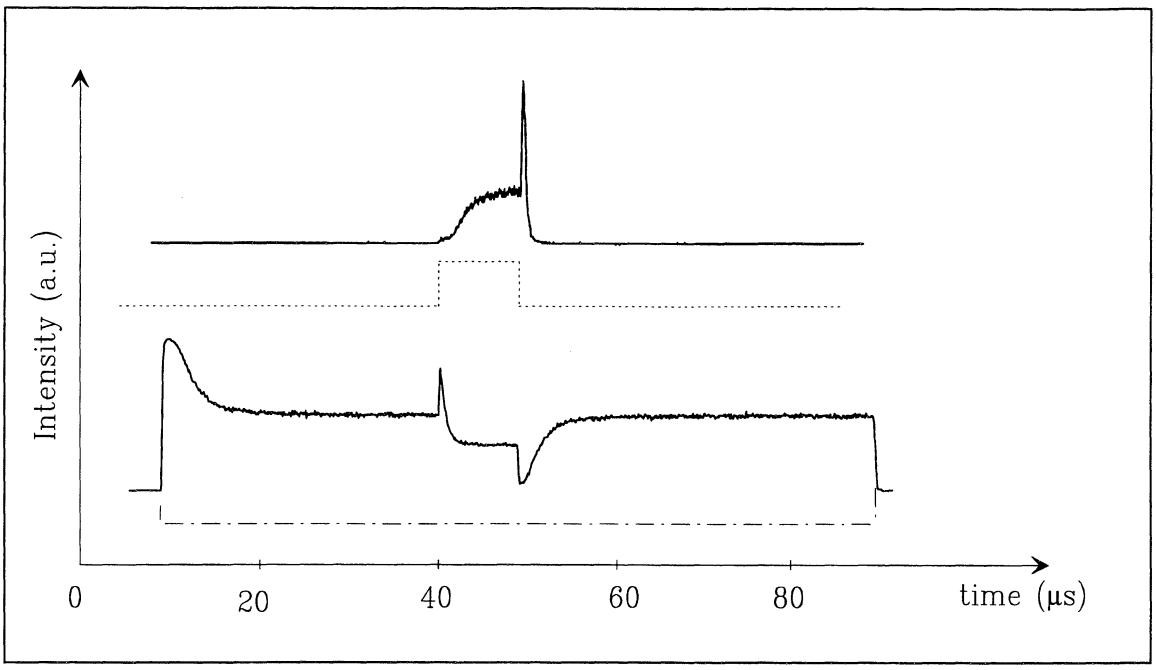

Fig. C2. *The probe beam transmission through optically pumped sodium vapour for two different cases is shown. The dashed lines indicate the duration of the pump pulse and the dashed-dotted line in the lower trace shows the duration of the probe pulse. The upper trace shows a crossover transition where the pump and probe beams have linear and parallel polarisation. In the lower trace, the probe beam is linearly polarised and the pump beam is elliptically polarised. An additional polariser is also inserted in the probe beam path in front of the detector. Thus not only optical pumping but also alignment effects are observed in this trace. As the probe beam is turned on, we believe that Faraday rotation caused by the uncompensated magnetic field of the earth rotates the probe beam polarisation. The probe beam field itself will, however, align the sodium atoms in a different way, causing the transmitted intensity to change with a time constant determined by the time it takes the atoms to traverse the laser beam. When the pump beam is turned on, it will also change the sodium atom alignment. This, in part, but not fully, explains the trace when the pump beam is turned on. Note that the photodiode cannot see the pump beam light. It can only see the effect of the pump light through the change in probe beam transmission.*

C2 Collisional processes studied with laser spectroscopy

Luo Caiyan, Aigars Ekers, Janis Klavinsh* and Sune Svanberg*

**Visiting Scientist*

CW laser spectroscopy has been used to study energy transfer in collisional processes between excited alkali dimers and alkali atoms. One or two dye lasers, pumped by argon-ion lasers were used in these studies, which were performed on high-temperature cells partly made of alkali-resistant glass. In one experiment, the quasi-resonant energy transfer in collisions between sodium dimers in the $Na_2(A^1\Sigma^+_u)$ state and ground-state potassium atoms [K(S)] was studied by observing laser-induced fluorescence [C5]. A strong increase in the cross section, by as much as an order of magnitude, was observed for the rovibronic

levels of the dimer, which are in close resonance with the 4S-4P transition of the potassium atom. Comparisons with first order perturbation theory calculations were performed.

The ionisation resulting from collisions between excited alkali dimers and atoms was studied in a further experiment observing fluorescence as well as the ionisation current [C6]. Sodium dimers were excited to a $Na_2(A^1\Sigma^+_u)$ state by one dye laser while a second one prepared sodium atoms in the 3p state. In the collisions, Na_3^+ ions were formed with an order of magnitude larger cross section than Na_2^+ formation from two excited sodium atoms. A further collisional study involving rubidium atoms has been performed and is being prepared for publication [C7].

D Theoretical atomic physics

The theoretical work at the Division is done in close co-operation with several other theory groups in Europe and in the United States. Recently, a joint project, SAMSCF, was started with the goal to provide very accurate atomic data of importance for theoretical modelling of astrophysical and other types of plasmas.

D1 Program development and large-scale calculations

Jörgen Carlsson, Per Jönsson and Lennart Sturesson

Much time is spent on the development of computer codes adapted to run on supercomputers as well as on the parallel computers that are now becoming available. As a part of the SAMSCF project, a program has been developed [D1] which calculates isotope shifts from multiconfiguration Hartree-Fock and configuration interaction wave functions. Using this program, isotope shifts have been calculated with high accuracy for a number of transitions in light atoms [D2-D4]. A program to calculate hyperfine structure splittings has also been developed [D5]. Using this, together with a new program [D6] for generating large configuration expansions in a systematic way, it has been shown that it is indeed possible to calculate hyperfine structures extremely accurately, in many cases with an error of less than 1% [D7-D11]. Fully relativistic hyperfine structure calculations have been performed for Sc^+, Y^+ and Li [D12,D13]. For the ground state of lithium the calculations are almost exact, and the remaining errors very small. For highly charged ions the hyperfine structure is sensitive to QED effects and to the distribution of the magnetisation within the nucleus. By comparing experimental values of the hyperfine structure with calculated ones it should thus be possible to probe the above mentioned effects. Although recent progress in computational techniques has made it possible to calculate many properties with very high accuracy, the calculation of transition probabilities has remained difficult. An example of this difficulty is the discrepancy between theoretical and experimental values of the transition probability for the resonance transitions for both lithium and sodium [D14], and it has been much debated why theoretical calculations are unable to predict oscillator strengths with an error less than 1%. Recently, however, a new computational method has been implemented and tested [D15], and it was shown that it is indeed possible to calculate transition probabilities very accurately. Using a program based on this new computational method, the transition probability for the resonance transition in sodium has been calculated and for the first time experimental and theoretical values are in

perfect agreement, that is, the error in the calculated value is estimated to be less than 0.2% [D16]. A similar low error was estimated for the transition probability of the resonance transition in BII [D17]. The lifetimes in the Rydberg series of the boron and nitrogen atoms have also been calculated [D18,D19].

D2 Astrophysical applications

Per Jönsson

Most of our detailed knowledge about the stars and the interstellar medium is based on the analysis of stellar spectra. Since the launch of the Hubble Space telescope (HST) the Goddard High-Resolution Spectrograph (GHRS) has produced stellar spectra of very high resolution. At high resolution, essentially all atomic lines are asymmetric because of isotope shifts and hyperfine structures, and to correctly interpret the spectra it is necessary to include these effects as well as the transition probability in the theoretical modelling of the line profiles. For many lines there are no laboratory values and therefore the modelling has to rely on calculated values. One such example is the determination of the abundances of the ^{10}B and ^{11}B isotopes as well as the isotopic ratio in different astrophysical objects . In this case the observed boron resonance line is only partly resolved, and in order to extract the abundances and isotopic ratios accurate values of the isotope shift, hyperfine structure and transition probability must be supplied [D2,D17]. The abundances of ^{10}B and ^{11}B are related to certain parameters used in the big bang models, and by determining these abundances it should be possible to distinguish between the different models that have been proposed. Another problem of current interest is the determination of electron densities in the stellar plasmas in connection with the present non-local thermodynamic equilibrium models. It has been recognised that metastable states are valuable probes of the electron density since these states live long enough to be affected by the electron collisions. The lifetime of metastable states can be appreciably shortened by hyperfine quenching and to investigate this a program that calculates the quenching effect has been developed and tested [D20].

To stimulate the contact between atomic theory and different applications, such as astrophysics, and to bring together scientists from these different fields, a number of international symposia have been and will be arranged together with the Division of Atomic Spectroscopy:

Trends in Atomic Structure Calculation and Spectroscopy, Lund March 3-4, 1993
Iron Group Atoms in the Laboratory and Space, Lund June 28-28, 1994
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II Applied Optics and Quantum Electronics

In this chapter, three projects based on lasers and non-linear optical techniques are presented. In the first project new methods for high-resolution microscopy on living biological matter are being developed, the second project concerns solid state spectroscopy and the investigation of new concepts for optical storage and optical signal processing, and the third utilises FM modulation of diode lasers for ultrasensitive absorption measurements.

A High-resolution microscopy

In this project we are developing novel methods for high-resolution microscopy suitable for living biological matter. For such non-intrusive studies, scientists are still basically limited to the classical optical microscope, the resolution of which is limited by diffraction to a few 100's of nanometres. Since many structures of considerable interest are smaller, we are investigating two potential approaches to obtain a higher-resolution microscope suitable for imaging living matter: table-top soft X-ray microscopy and non-intrusive scanned near-field optical microscopy. Both methods share the common goal of developing compact and reasonably priced microscopes suitable for research as well as practical applications.

A1 Soft X-ray microscopy

*Lars Rymell, Magnus Berglund, Hans M. Hertz and Johan Anderberg**

**MSc student*

The basic idea of soft X-ray microscopy is to use the large natural difference in absorption between proteins (i.e. carbon) and water (i.e. oxygen) in the wavelength range 2.3-4.4 nm (0.3-0.5 keV). In this wavelength range, at least an order of magnitude higher resolution than the optical microscope can be acheived. Furthermore, the samples may be studied without staining, sectioning or fixation in an aqueous environment and at atmospheric pressure. A major difficulty is that most X-ray microscopes for biological studies utilise synchrotron sources, which often results in a limited accessibility for the scientist. Below, our progress in the construction of a table-top soft X-ray microscope is described.

The laser-produced plasma (LPP) is a table-top source that has the potential to produce sufficiently high soft X-ray brightness to compete with synchrotron radiation. However, LPP is generally used with solid targets and thus produces debris, which contaminates and may damage sensitive X-ray optical components positioned close to the LPP. We use small (approximately 10 μm) liquid droplets as the target. The drops are produced using vibrating glass capillary nozzles. Since the target fully evaporates and ionises upon plasma formation, the debris problem is practically eliminated. Thus, the X-ray source can be

positioned close to the optics or sample thereby, increasing the soft X-ray flux. The work has been described in several papers and conference contributions [A1-A9]

A schematic experimental arrangement of the low-debris laser-plasma droplet X-ray source is shown in Fig. A1. In the first experiments, ~10 μm ethanol droplets were produced by a 1 MHz vibrating capillary ink jet printing nozzle [A1]. A frequency-doubled, 70 mJ/pulse, 120 ps, 10 Hz mode-locked Nd:YAG laser is focused on the droplets with a focal spot diameter of approximately 12 μm. Spectral measurements using a grazing-incidence monochromator show text-book C V, C VI, O VII and O VIII spectra in the λ=1.5-4.0 nm region. Flux measurements using an X-ray diode result in approximately 10^{12} photons/(ster·line·pulse) in the C V and C VI lines at λ=3.3-4.0 nm. Using a novel knife-edge method [A9], the diameter of the soft X-ray source has been determined to be less than 25 μm. The resulting brightness (photons/(ster·line·pulse·μm^2)) compares very well with alternative sources. Emission at longer wavelengths (λ= 10-20 nm) is also significant [A2].

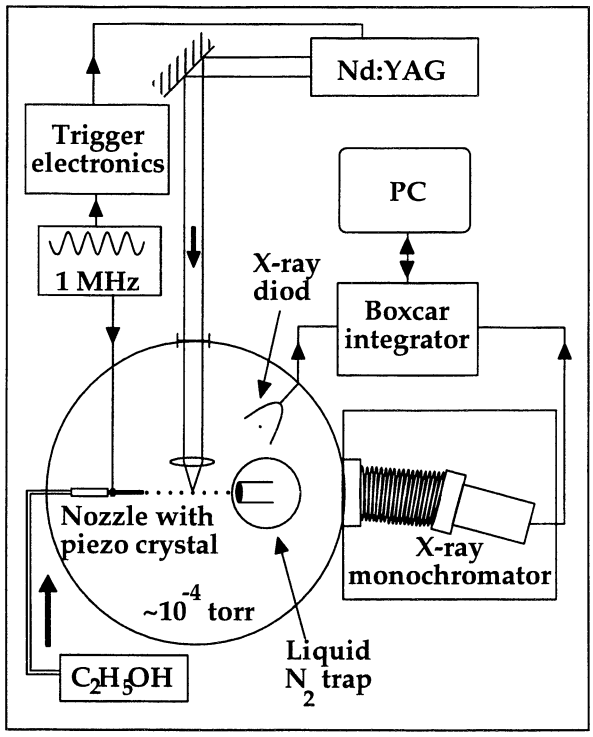

Fig. A1. Experimental arrangement for debris-free, high-brightness, soft X-ray laser-plasma droplet source.

However, the most important feature of the soft X-ray source is that it is nearly debris-free. Debris production is reduced by nearly 3 orders of magnitude compared with the best available conventional target (thin-film mylar tape). This is probably due to the fact that the whole droplet target is evaporated and ionised and no target material is available in the low-intensity Gaussian tails of the focused laser beam. The assumption is supported by the very high degree of ionisation that we have observed, both at X-ray and VUV wavelengths [A2]. In order to further reduce the effect of debris, we have invented a localised gas jet debris shield which reduces debris deposition another 1-2 orders of magnitude [A3]. Thus the debris reduction compared with conventional targets is 4 orders of magnitude, making this soft X-ray source useful for X-ray microscopy and lithography.

Due to their lower attenuation in water, the N VII and N VI lines, at λ= 2.5 and 2.8 nm, are better suited for microscopy than the carbon lines at 3.3-4.0 nm. Furthermore, single-line emission is preferable in order to avoid problems due to the chromatic aberration in zone-plate optics used for X-ray microscopy. For this purpose, we have utilised the versatility of our drop-formation technique to produce target droplets of ammonium hydroxide, resulting in N VII and N VI line emission [A4]. Combining this source with a thin-film Ti filter results in a practically single-line source at λ= 2.9 nm, as shown in Fig. A2. The source emits approximately $1 \cdot 10^{12}$ photons/(ster·line·pulse).

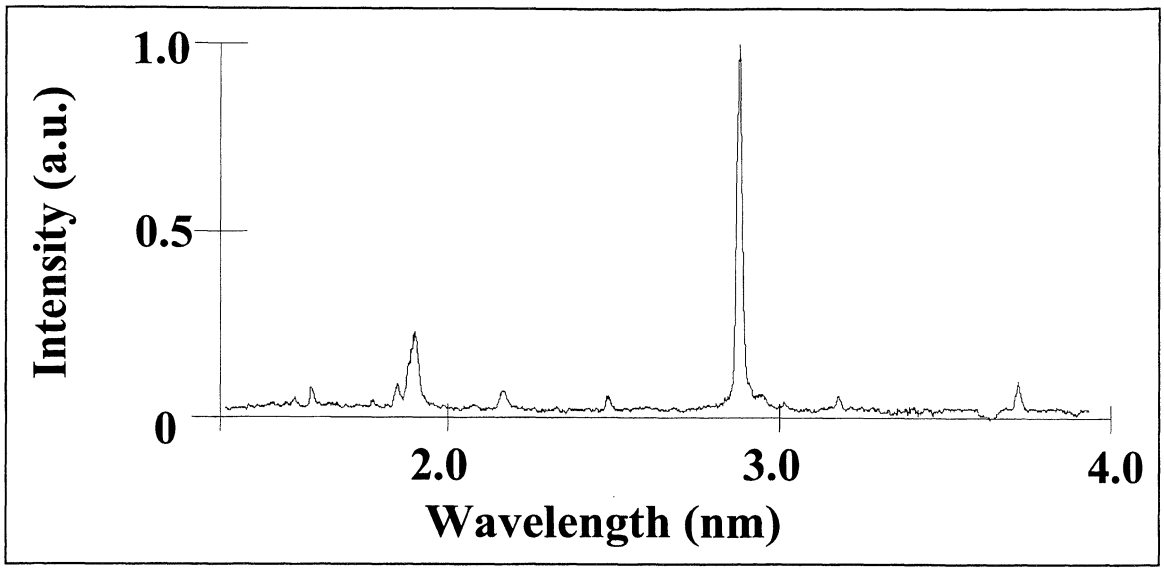

Fig. A2. *Filtered single-line N VI spectrum from droplet source, suitable for X-ray microscopy*

Our X-ray microscope will be based on this nitrogen source. We will commence the construction of the microscope early 1995. We are also investigating other applications of the laser-plasma droplet X-ray source, such as lithography.

A2 Trapped-particle optical microscopy

Lars Malmqvist and Hans M. Hertz

The non-intrusive character of classical optical microscopy is of great importance in the study of, e.g., living biological systems. Unfortunately, far-field diffraction limits the resolution of such microscopes. Near-field optical methods have demonstrated very high resolution by scanning a microscopic light source in close proximity to the studied object. However, the mechanical positioning of the probe restricts its use to mechanically accessible and smooth surfaces, limiting the method's applicability for studies of, e.g., living biological material with intervening membranes. Our work is focused on the development of a non-intrusive, near-field optical microscope. The work has been presented in several papers, conference contributions and a licientiate thesis [A10-A20].

We are investigating the use of optically trapped microscopic non-linear crystals as microscopic light sources for non-intrusive, scanned probe optical microscopy - Trapped Particle Optical Microscopy (TPOM) [A11, A12]. Fig. A3 shows the experimental arrangement. A sub-100 nm lithium niobate or KTP crystal is optically trapped by a strongly focused λ=1.06 μm Nd:YAG laser. Due to the non-linear properties of the crystal, green light (λ=532 nm) is generated when the particle is trapped close to the focus. This visible light source may be non-intrusively positioned in close proximity to the studied object. By accurate piezoelectric scanning, a sub-diffraction limited image may be recorded.

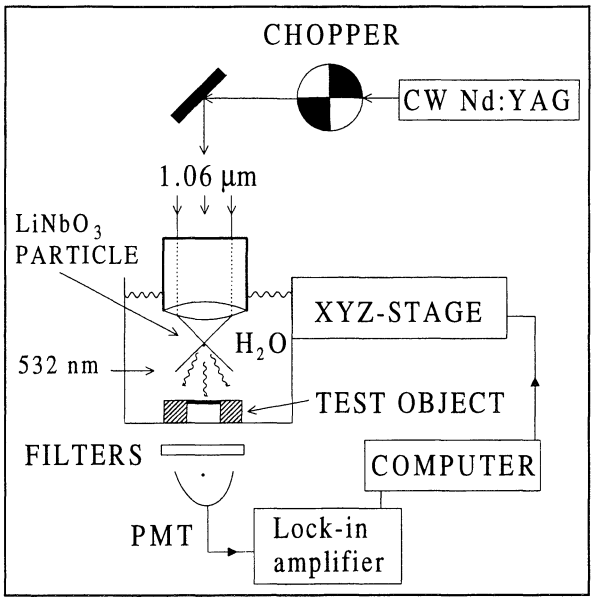

Fig. A3. Experimental arrangement for two-colour trapped particle optical microscopy.

In contrast to our initial experiments [A10], in which the Rayleigh scattered light from a 60 nm SiO_2 particle was used, the separation of the trapping and detection wavelengths employing a non-linear crystal results in higher resolution since scattered light from the strong trapping beam is spectrally eliminated. The best resolution obtained so far is <0.5 μm with an etched Si cantilever as the test object. Here van der Waals surface forces limit the minimum distance between the object and the particle due to the large refractive index of silicon. With the lower refractive index of biological objects, we expect to obtain subdiffraction resolution. Theoretical calculations of the resolution indicate that 80 nm should be possible with the current particles. In these calculations the effective size of the particle is determined by convolving the particle's physical size with its displacement due to Brownian motion in the trap. Further resolution improvement may be obtained by using other types of traps [A12, A13].

For studies of biological samples fluorescence offers a well-developed contrast technique. In order to perform sub-diffraction-limited fluorescence imaging with our trapped particle probe approximately a nW of emitted green power is needed. Since the average power of the trapping beam is limited to approximately 100 mW, due to the risk of biological damage, the emitted frequency-doubled light can only be increased by pulsing the trapping beam and thus increasing the peak power [A14]. High-repetition-rate, pulsed lasers are necessary in order to avoid the escape of the particle from the trap between the pulses due to Brownian motion. We have investigated the emitted power using a 100 ns, 25 kHz, Q-switched Nd:YAG laser and a 100 fs, 76 MHz Ti: Sapphire laser. The frequency-doubled power ranged from 0.1 nW to a few nanowatts with stably trapped particles.

Finally, we have developed a method for controlling the distance between the trapped particle and the sample with a few nm accuracy [A19]. Thus, the building blocks for a non-intrusive, near-field optical microscope have been established and we are currently constructing a dedicated instrument suitable for non-intrusive sub-diffraction limited imaging of biological samples.

B Photon echoes in rare-earth-ion-doped crystals

Ulf Elman, Baozhu Luo, R Kaarli, Stefan Kröll and Mikael Pålsson***

**Visiting Scientist, **MSc student*

Our recent work has focused on the ability of photon echoes to serve as all-optical recorders of temporal information. Theoretically, we have calculated that storage densities of 100 times the diffraction limit with terabit read and write speeds can, in principle, be obtained. Experimentally, all-optical processing and storage concepts have been tested, involving logical operations, erasure of stored data, programming in the frequency domain for time-domain readout, enhancing the data storage efficiency by using a combination of laser and radio-frequency fields and testing of new materials.

Our theoretical calculations [B1] of the obtainable storage density of photon echo storage are based on the Dicke model of superradiance. Since these calculations provide us with the number of photons emitted in the coherent photon echo process we can, in principle, obtain sufficient information to estimate the signal-to-noise limit of the ultimate performance of photon echo storage under the conditions stated. In short, we obtain a storage density two orders of magnitude better than the diffraction limit and THz bit rates. This work has attracted considerable attention [B2-B5], although theoretical calculations by other groups based on novel experimental schemes for the storage process have now shown that this performance can be further improved.

It seems sensible to concentrate the work to areas in which the properties of the photon echo storage process are truly unique. We have therefore focused on the ability of photon echoes to "remember" temporal information. There are many applications of this property, one is to use photon echoes to carry out all optical processing on temporal information. By simultaneously working in the time and frequency domains, we have demonstrated word-by-word logical processing (Fig. B1) [B2,B4,B6]. This work basically demonstrated one of, undoubtedly, many concepts for using photon echoes for all-optical processing on temporal data sequences.

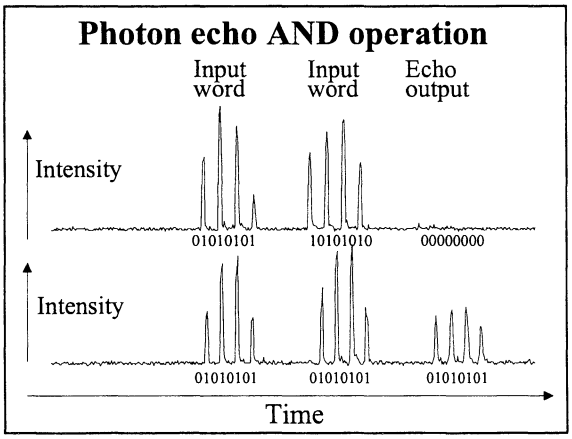

Fig. B1. *Experimental curves demon-strating a photon echo, bit-by-bit* AND *operation on 8-bit words. Two pairs of input words with their corresponding output sequences are shown. The laser frequency is swept while the input words are written. A high output bit will result only if the two corresponding bits in the input words both have the same frequency.*

The approach to processing mentioned above is sensitive to the amplitude of the electromagnetic excitation pulses but not to their phase. As a result of this, only a rather limited class of operations can, in reality, be performed. Full utilisation of the properties of photon echo processing also requires using the feature of photon echoes to record the relative phase of the excitation pulses. One process, perhaps even the simplest one, utilising this property is to erase a stored data bit by rewriting it with the relative phase

between write and data pulse shifted by 180 degrees (N.N. Akhmediev, Opt. Lett. **15**, 1035 (1990)). Our experimental findings (see e.g. Fig. B2) [B7-B10] when performing such erasure operations have caused us to consider the sensitivity of such processes to the phase and frequency variations of the excitation source that is used. Since the efficiency of the data erasure process is limited by the stability of our light source, we have realised that we have a powerful diagnostic tool regarding the phase and frequency fluctuations of our light source. On the other hand, these fluctuations prevent us from fully evaluating the possibility of erasing stored data. Our current efforts are being directed towards using our data to characterise the laser source. A future aim is to use a more stable source to instead characterise the coherent erasure process itself.

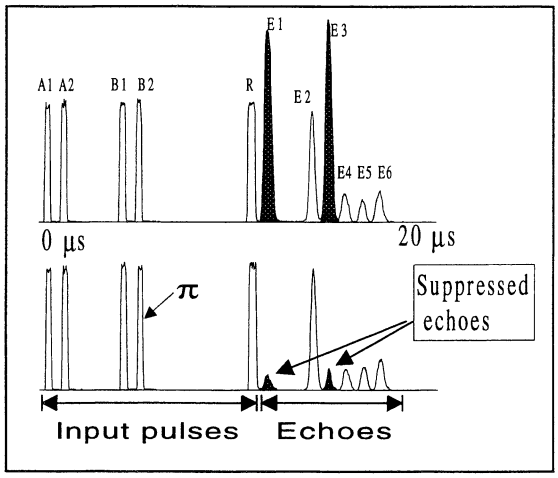

Fig. B2. *Experimental curves showing partial coherent photon echo erasure: intensity vs time. R is the read-out pulse, A1, A2, B1 and B2 are input pulses, Ex (x=1-6) are echo pulses. In the lower trace, a 180 degree phase shift was applied to pulse B2 leading to partial suppression of the echoes E1 and E3.*

In the writing process, a frequency-dependent upper-state population grating is created. In the theoretical calculations [B1] it was shown that one of the most critical parameters for obtaining a high storage density was the efficiency in transferring this transient upper-state grating into a permanent, frequency-dependent ground-state grating. For the rare-earth-ion-doped crystals, this ground-state grating is a frequency-dependent population of the ground state hyperfine levels. The hyperfine population can be affected by a radio-frequency (rf) field tuned to the hyperfine splitting frequency. A diploma work was therefore performed in order to construct rf coils and electronics which could induce such hyperfine transitions [B11]. The setup was, however, not able to increase the signal of the stored photon echo data. We presently do not fully understand this lack of success.

In photon echo storage, temporal information is stored as a frequency grating. Such a grating can also be created also by simply tuning the light source to some selected frequencies and exciting atoms absorbing at these frequencies to the upper state. This grating will emit a certain temporal sequence if excited by, e.g. a short pulse. In other words, by exciting certain frequency components and stimulating them to emit radiation coherently we can, in principle, generate an arbitrary temporal output sequence. In a collaboration with Rein Kaarli at the Estonian Academy of Sciences, Tartu, we have begun to investigate this so-called spectral programming approach (Fig. B3) [B12]. One of our motives here was also to be able to perform phase-sensitive storage and simultaneously eliminate the problem of temporal phase instability of our laser source. This should be possible to achieve because the phase information in spectral programming can be stored using a conventional spatial interference pattern created by splitting the laser output beam into two parts and overlapping them at the storage point. These experiments have given us improved insight into some aspects of the relation between instantaneous frequency and absorption linewidth. Specifically, we have understood that when changing the relative

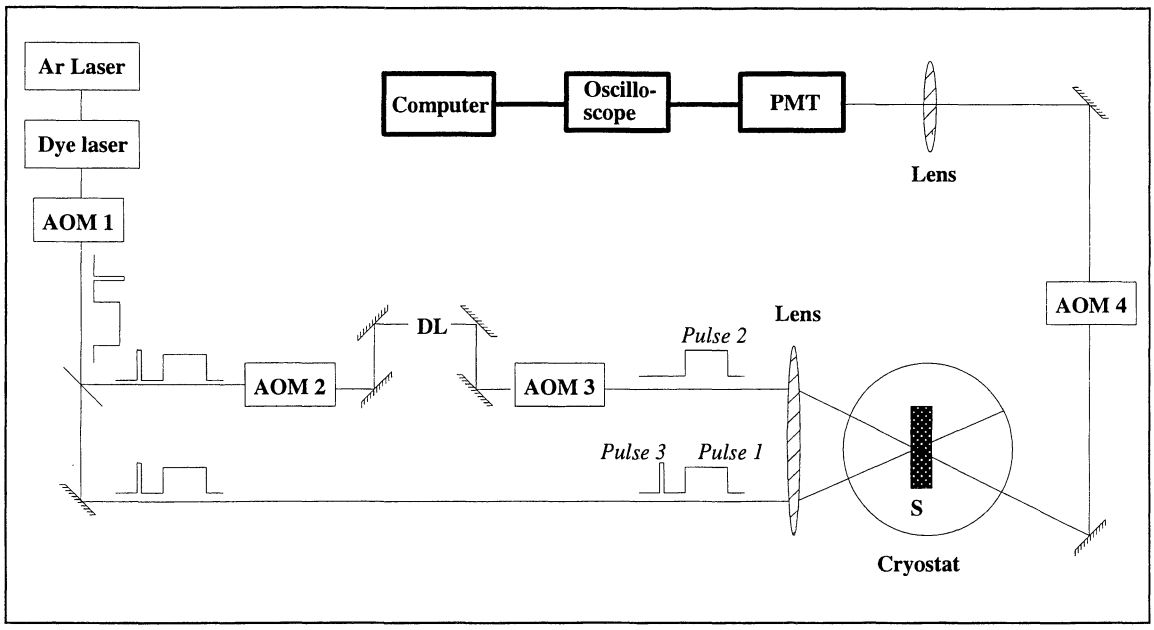

Fig. B3. *Experimental setup typical for photon echo experiments. The present setup is specifically for the spectral programming approach. AOM = Opto-Acoustic Modulator. The AOMs are used for creating pulse sequences from the cw laser beam and for shifting the excitation frequency of the exciting light pulses. DL = Delay Line. In combination with frequency shifts induced by AOM2 and AOM3 the fixed delay line can be used to induce controllable phase changes between the two legs in the setup. The pulses are numbered consecutively in the order they arrive at the liquid-helium-cooled crystal inside the cryostat. PMT = PhotoMultiplier Tube.*

phase between two interfering beams, the ratio between the relative instantaneous frequency, here defined as the derivative of the relative phase of the excitation pulses with respect to time, and the homogeneous linewidth of the absorbing ions, determines whether information is written using a time or a frequency domain approach. If the relative instantaneous frequency is larger than the absorption linewidth, photon echo storage is performed, if it does not exceed the linewidth, spectral programming is performed. We note that in the first case the phase stability of the light source is still important in order to fully control the phase information. Other current topics include the testing and development of new storage materials [B13].

C Diode laser spectroscopy

The value of laser absorption spectroscopy for trace species detection has been improved significantly in recent years using frequency modulation (FM) techniques. A variety of FM methods exist, but fundamentally they are variations of the same technique. Common to the different approaches is the shifting of the detection band to higher frequencies to avoid laser source (1/f) noise. The absorption signal is homodyne detected, either at the same frequency as the applied modulation frequency, at an overtone or at an intermediate frequency. We have specialised in a certain approach called two-tone frequency modulation spectroscopy (TTFMS), which provides a high dynamic range, a favourable detection bandwidth and shot-noise-limited detection, i.e. ultra-sensitive absorption measurements.

Our experimental arrangements normally have a detection sensitivity of about 10^{-6}, however, sensitivities of 10^{-7}-10^{-8} are possible. The high sensitivity is suitable for work on small volumes of absorbing gas, and for measuring weak molecular transitions in the near-infrared region, where inexpensive diode lasers (0.7-1.7 μm) are commercially available. The near-infrared lasers are simple to operate and can be conveniently frequency modulated by superimposing an a.c. current on the laser injection current. The two closely spaced modulation frequencies we use are selected in the 500-1,000 MHz region and detection is carried out at an intermediate frequency, about 10 MHz. The work has been presented in Refs. [C1-C11].

C1 Vapour pressure measurements, a tool for water activity determination in solutions

Peter Kauranen and Viacheslav Avetisov

The water vapour concentration in a gas in chemical equilibrium with a solution depends on the water activity in the solution. By measuring the water vapour pressure over the solution (p) relative to the vapour pressure over a pure water reference ($p°$), the water activity (a) in the solution can be determined from the relation $a=p/p°$. With laser absorption techniques the relative water vapour concentration can be measured independently of other components present in the gas. This opens up new possibilities in chemical analysis. The water activity depends on the composition and the concentrations of the dissolved molecules in the solution, and is coupled to the interactions between the molecules. We have obtained a minimum detectable pressure change of 0.3%, with a signal-to-noise ratio of one, by measuring water absorption lines at 820 nm [C1]. At the moment, work is in progress to construct a new experimental set-up, with better mechanical stability and with better control of the system thermodynamics. This will improve the absolute accuracy of the water activity determination, and may allow us to study very small pressure changes. The technique will be applied to the study of solution structures of biologically significant molecules, including the evaluation of thermodynamic factors affecting association in solution, e.g. the hydrophobic effect.

C2 Tomographic imaging of an oxygen flow

Peter Kauranen, Hans Hertz and Sune Svanberg

By combining multi-angle absorption measurements with tomographic reconstruction, quantitative and spatially resolved measurements can be performed. The combination of tomography with TTFMS makes measurements on weakly absorbing objects possible. The method is demonstrated by mapping the concentration in a section of a weakly absorbing oxygen gas flow, by measuring the absorption from one of the oxygen A-band

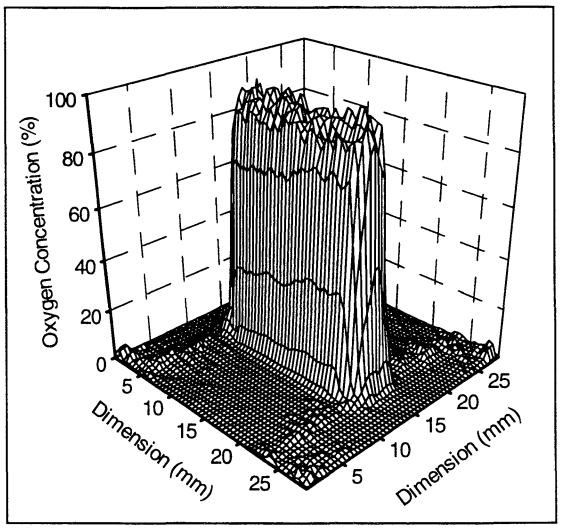

Fig. C1. *Tomographic reconstruction of the concentration of oxygen in an expanding flow, 3.2 mm above a rectangular (4×18 mm) nozzle.*

lines at 760 nm [C2,C3]. Fig. C1 shows a reconstruction of the oxygen concentration in a section 3.2 mm above a nozzle orifice (4×18 mm), using six projection angles 30° apart. Each pixel i 0.5×0.5 mm. At the moment, preparations are being made to reconstruct concentration and temperature profiles in a burning flame.

C3 High-resolution absorption measurements

Viacheslav Avetisov, Peter Kauranen, Ulf Gustavsson and Sune Svanberg

The lineshape of a TTFMS signal depends strongly on the absorption linewidth, modulation frequency and the frequency modulation (FM) index. Employing the peak-to-peak value of the TTFMS signal to measure the concentration of a species in a gas at different compositions and/or pressures, may lead to large uncertainties when the absorption line profile is altered. We have shown that it is possible to determine the line parameters accurately from TTFMS spectra, using a least-squares fitting procedure [C4]. This widens the applicability of TTFMS for quantitative measurements using weak absorptions, and also provides a useful tool in high-resolution spectroscopy, when sufficiently strong absorptions are difficult to obtain, e.g., when studying gases at high temperatures.

We have performed extensive work aimed at a more general understanding of TTFMS lineshapes and how accurately line profile parameters, such as broadening, narrowing, line shift and line intensity, can be obtained from a least-squares fitting procedure under different experimental conditions. The high measurement resolution we can obtain, together with an accurate theoretical description of the TTFMS lineshapes, allows us to resolve collisional (Dicke) narrowing. For this reason, we have employed more elaborate line profiles, including collisional narrowing, such as the Galatry and Rautian-Sobelman profile in our investigations. Both numerical [C5] and experimental [C6] investigations have been performed. The influence of a small dispersion signal, appearing in the detected signal when the detection phase angle is not fully in phase with the absorption signal, and non-linear modulation response of the diode laser are included in the TTFMS theory and have been thoroughly investigated. The influence of the FM index on the lineshape has been evaluated and a method of determining the FM index accurately is demonstrated. The experimental investigation also includes least-squares fittings of the direct transmission profiles for

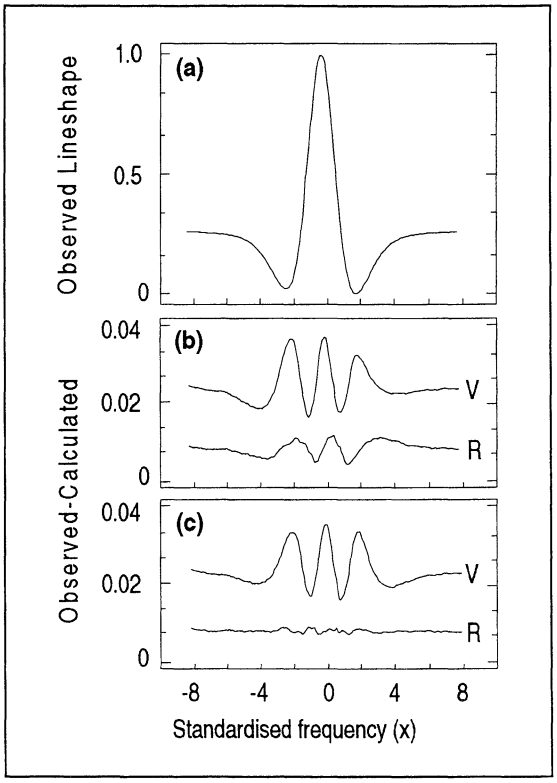

Fig. C2. *Results of least-squares fitting with a Voigt (V) and a Rautian-Sobelman (R) profile, respectively. The upper trace (a) shows a recorded oxygen lineshape. The middle (b) and lower traces (c) show the residual errors of the fittings. In the lower traces non-linear distortions in the frequency modulation response of the diode laser have been included in the TTFMS theory.*

a more complete evaluation of the TTFMS results. The line parameters obtained from the two measurement methods show excellent agreement.

Fig. C2(a) shows a TTFMS recording of the oxygen R15Q16 line at 13156 cm^{-1}. The oxygen pressure in the cell was about 200 torr, giving a peak absorption of about 5%. The upper trace in Fig. C2(b) shows a plot of the residual errors from a Voigt fit to the experimental data. The characteristic signature of the residual errors indicates the presence of collisional (Dicke) narrowing. The lower trace in Fig. C2(b) shows the residual errors when the Rautian-Sobelman profile is used in the fit. The asymmetry in the plots is due to harmonic and intermodulation distortions in the frequency modulation response of the diode laser, which influence the power distribution between the sidebands. We have included non-linear distortions in the TTFMS theory, and the results as demonstrated in Fig. C2(c) show a significant improvement in the fits to the data.

C4 Construction of an extended-cavity diode laser

Jonas Bengtsson

We have tested a diode laser in an external cavity configuration, where a diffraction grating is used for optical feedback and wavelength tuning. An extended cavity increases the wavelength tunability of the diode laser and improves the frequency stability drastically, resulting in linewidths of a few hundred kilohertz. The construction was tested by measuring the hyperfine spectrum of the ^{87}Rb D2 line at 780 nm using Doppler-free saturation spectroscopy [C7].

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III Environmental Remote Sensing

Research projects in the field of environmental remote sensing are directed towards development and applications of optical techniques, both laser and non-laser, to measure mostly tropospheric gases, but also as a tool in vegetation and water quality studies. The group has participated in two European environmental projects, EUROTRAC and LASFLEUR, and also collaborates closely with two Italian research groups, at CNR-Istituto di Biofisica in Pisa and IROE in Florence, in other projects. In Lund, some activities are channelled through the Centre for Environmental Measurement Technology (CENTEC), which coordinates research and teaching in environmental sensing technology within the Lund Institute of Technology. The remote sensing work at the department is financially supported by the Swedish Space Board (RS) and the Swedish Natural Science Research Council (NFR).

The work in the group has resulted in three PhD theses during the past two-year period [1-3]. Some overviews of the research have been presented elsewhere [4-7]. The method most used in the various projects is the differential absorption lidar (DIAL) technique. Reviews of the DIAL technique and measurements performed by the group have also been published [8,9]. The mobile lidar system developed at the Division has been used in several field campaigns during the last two years. Atmospheric mercury pollution has been studied during measurements in a mercury mining area in Almadén, Spain, and volcanic sulphur dioxide fluxes have been measured on Sicily and the Aeolian islands in Italy. A LASFLEUR project was carried out near Avignon, France, where laser-induced fluorescence from vegetation was studied with a multi-colour imaging system adapted to the receiving telescope in the lidar system. Further campaigns with the system have been performed by the newly formed company Lighten AB, located in the Idéon Research Park in Lund. This new spin-off company makes lidar measurements on a commercial basis. One such measurement was made in the Mt. Amiata region in Italy, where mercury concentrations and fluxes were measured in geothermal areas and at cinnabar deposit sites. During the latter part of 1994 two campaigns involving measurements of industrial and urban pollution were performed in different parts of the Czech Republic.

A description of the multi-path DOAS (Differential Optical Absorption Spectroscopy) system developed at the Division has now been published [10]. The system has been employed together with two DIAL systems for comparative and complementary measurements of tropospheric ozone concentrations. The DOAS system has also been used in a CENTEC project together with the Nuclear Physics Division, where DOAS measurements of several gases have been combined with PIXE (Particle-Induced X-ray Emission) analysis of aerosols. The results have been used in a receptor model to identify various sources affecting the air quality in Lund [11]. The DOAS system has been in regular use in the course in Environmental Sensing Technology for a number of years.

A Tropospheric ozone lidar

Hans Edner, Pär Ragnarson, Sune Svanberg and Eva Wallinder

The participation of the group in the subproject TESLAS (Tropospheric Environmental Studies by Laser Sounding) within EUROTRAC has now been concluded. The results of the intercomparison campaign TROLIX (Tropospheric Ozone Lidar Intercomparison Experiment) in The Netherlands have been fully analysed and published [A1]. In Lund, some additional intercomparisons have been performed between the mobile DIAL system and a fixed ozone DIAL system based on a Raman-shifted KrF excimer laser [A2,A3]. Both horizontal and vertical measurements have been performed. In the horizontal measurements the data could also be compared with the path-integrated value from the DOAS system. The DOAS technique uses weak, higher-order structures in the ozone absorption profile in the wavelength interval 277-287 nm. The DOAS values will normally not be affected by extinction due to aerosol scattering, which can be inherent in DIAL measurements with a large separation between the on and off wavelengths. A study of aerosol effects and validation of correct procedures in DIAL measurements can thus be made. Fig. A1 shows an intercomparison for several horizontal measurements made with all three systems, compared pairwise. The on and off wavelengths used were 278.7 nm and 286.4 nm for the mobile DIAL system (frequency-doubled dye laser), and 277 nm and 313 nm for the excimer-based system. A correction for differential extinction due to molecular and aerosol scattering was made on the DIAL values. To calculate the aerosol extinction correction value, which changes from day to day, the slope of the logarithm of the range-corrected off-wavelength signal from the excimer-based system was used. A homogeneous aerosol load for the horizontal measurements was assumed. The differences in ozone concentration levels deduced from the two DIAL systems might well be explained by a small difference in measuring path, since the measurements were performed in an urban area. The values obtained with the DOAS system are frequently slightly higher than the DIAL values; the cause of this is not yet known.

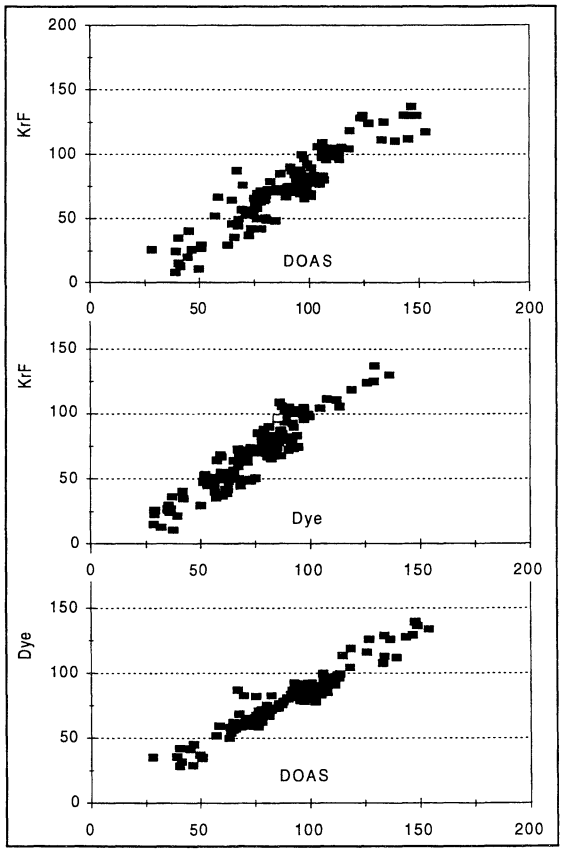

Fig. A1. *Ozone concentrations (μ g/m^3) measured over a horizontal path in Lund. Comparison between the results from the excimer-based lidar system, the dye laser-based lidar system and DOAS.*

Fig. A2 shows a vertical ozone concentration profile recorded simultaneously with the two DIAL systems. Corrections for Rayleigh and aerosol scattering, as a function of height, have been performed. The agreement is normally better than for the horizontal measurements, which is due to the fact that the air volume probed by the two systems is essentially the same.

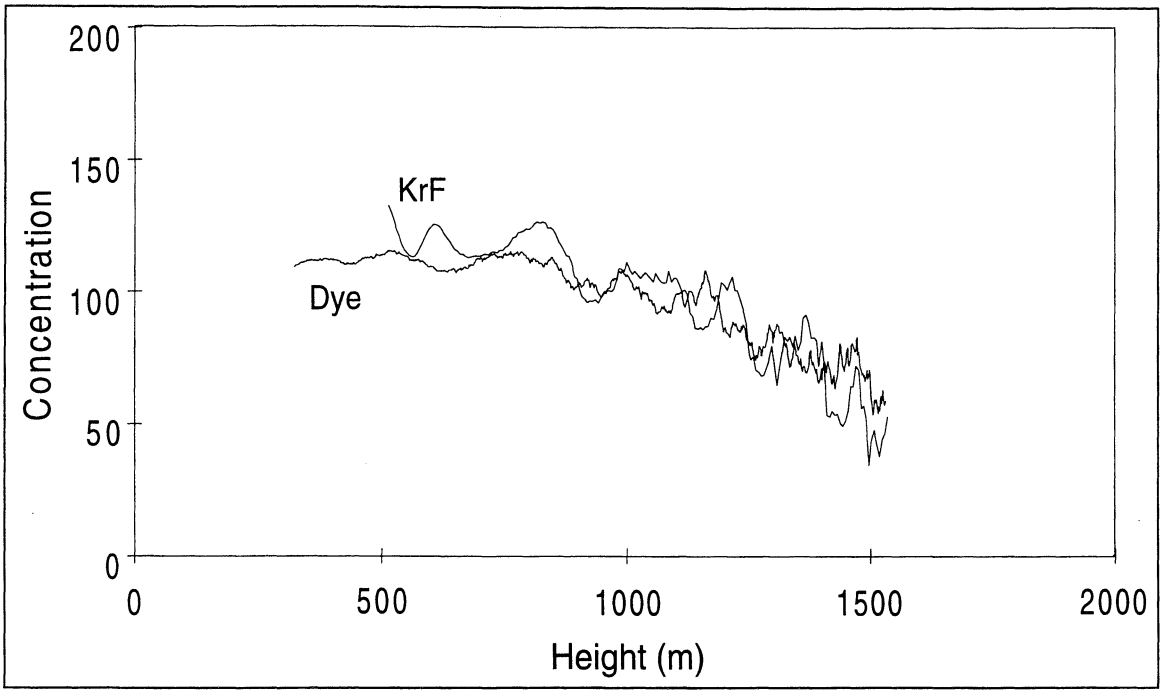

Fig. A2. Vertical ozone concentration ($\mu g/m^3$) profile measured with two lidar systems.

B Measurements of gases of geophysical origin

Mats Andersson, Petter Anderson, Hans Edner,
Pär Ragnarson, Sune Svanberg and Eva Wallinder

The results of the campaign in Italy during September 1992, where gas concentrations and fluxes from the volcanoes Etna, Stromboli, Vulcano, and Solfatara were measured, have now been fully analysed and published [B1,B2]. The measurements have also been presented at several conferences [B3-B6]. Measurements of the composition and flux of emitted gases and particles from active volcanoes are of great importance for the monitoring of volcanic activity, but are also of interest from an environmental point of view regarding possible effects on the earth's climate. Sulphur dioxide is generally the main sulphur-containing species in a high-temperature volcanic gas plume. During the campaign, the total flux of SO_2 from three volcanoes was measured with the mobile DIAL system secured on the aft deck of the Italian CNR research ship "Urania". In the normal measurement mode, scans through the volcanic plume were obtained by traversing under the plume, with the lidar pointing in a fixed vertical direction. Fig. B1 shows an example of a measurement of the SO_2 plume from Etna, captured at a distance of 23 km from the source. Wind data were obtained from observation stations at Etna, at 2500 and 3000 metres above sea-level. By combining the integrated gas concentration over the plume cross-section with wind velocity data, it was possible to determine the total flux of SO_2. We found fluxes of about 1,300, 180, and 25 tonnes/day for Etna, Stromboli, and Vulcano, respectively.

A new measurement campaign on board the Urania was performed during September 1994. The SO_2 fluxes deduced during these measurements were in good agreement with the ones from the earlier campaign. The lidar measurements of volcanic gas fluxes are the

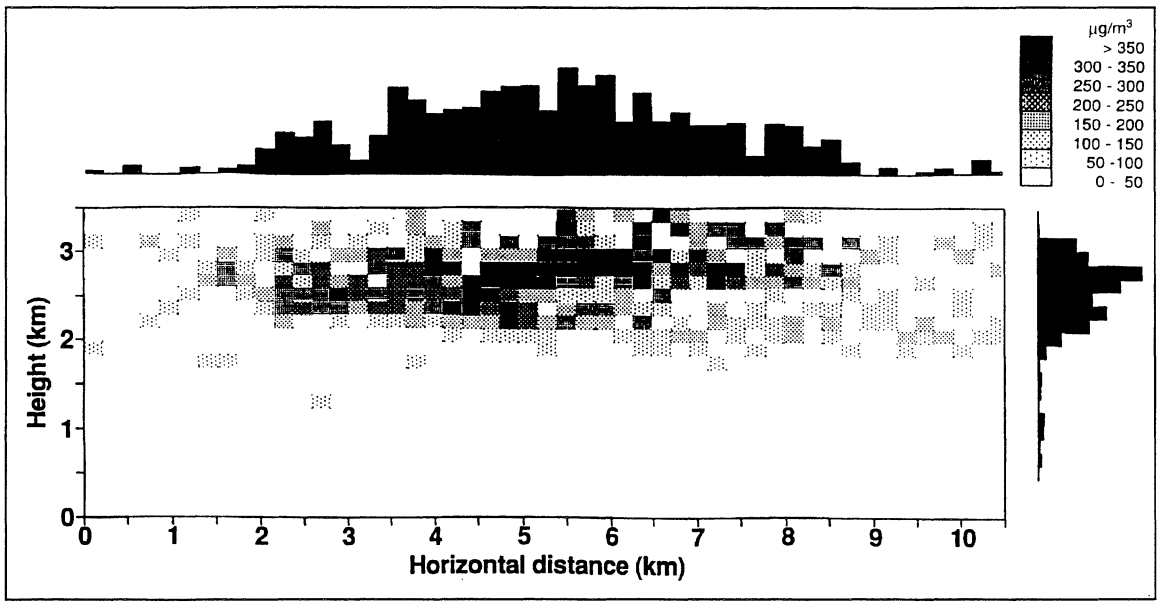

Fig. B1. *SO_2 distribution in the volcanic plume from Etna obtained from range-resolved lidar measurements.*

first, to our knowledge, in which an *active* optical remote sensing technique has been used. DIAL should be able to measure the overhead burden more correctly than passive techniques, which often require correction for scattering within or below the plume. The DIAL technique also gives the height and vertical extension of the plume, which are important parameters in the determination of the appropriate wind velocity. During both campaigns, the lidar data were compared with simultaneous recordings with passive DOAS, using the sky radiation as the light source. In the last campaign, a COSPEC (UV correlation spectroscopy) instrument was also employed. Most volcanic SO_2 flux monitoring performed during the last 20 years has been carried out with COSPEC techniques. A distinct difficulty with passive optical measurements, compared with active ones, is establishing the effective path through the plume. Work is in progress to study and quantify these scattering effects in passive measurements, using the simultaneous backscattering lidar signal from the plume.

Earlier measurements of atmospheric mercury concentrations near the abandoned mercury mine at Abbadia San Salvatore in Italy have now been published [B7]. Similar measurements were performed during September-October 1993 in Almadén, Spain [B8,B9]. The most important mercury mines in the world are located in this district, with mining activity without interruption over the past 2000 years. The area is highly affected by elevated concentrations of mercury in air, soil and vegetation. The mobile DIAL system was used in conjunction with point monitors to study the spatial and temporal distribution of atmospheric atomic mercury in and around the town of Almadén. The use of the remote sensing technique allowed rapid coverage of large areas, both horizontally and vertically, with a good temporal resolution. Atmospheric mercury concentrations up to several $\mu g/m^3$ were measured close to the smelting plant, near the ventilation outlet from the underground

mine and above old deposits of roasted cinnabar. Vertical lidar scans were combined with wind data to measure the total mercury flux into the atmosphere from all sources. The point monitors were used to measure mean concentrations at ground level at several positions in Almadén and the surroundings. Measurements of the degassing rate of the roasted cinnabar deposits and the soil were also performed, as well as determinations of the mercury content in the leaves of vegetables. Both lidar and monitors were also used in measurements at an open pit site 15 km outside Almadén.

C Industrial and urban pollution measurements

Mats Andersson, Hans Edner, Pär Ragnarson, Sune Svanberg and Eva Wallinder

A useful application of the DIAL technique is in mapping the pollution distribution in horizontal or vertical scans in industrial and urban areas. With a vertical scan downwind from one or several sources, the total flux of a certain gas can be monitored, including possible diffuse emissions. The results of several measurements of atmospheric fluxes of SO_2, NO_2, and Hg from Swedish industrial plants, performed within the framework of a control programme commissioned by the Swedish Environmental Protection Agency, are discussed in Refs [C1-C3]. Fig. C1 shows an example of SO_2 lidar measurements at a metallurgical plant, which contains several different sources. The result of a vertical scan downwind from all the sources is displayed in part a) of the figure. As can be seen, at least three plumes contribute to the total flux. Part b) shows another type of representation which can be made from such a measurement. Here the concentrations are vertically integrated and the resulting vertical column content is copied onto a map of the industrial area. The direction of the scan is indicated by the horizontal axis. Together with the wind direction, this type of figure can be useful in tracking the sources, especially if measurements are made in various directions during different wind conditions. A comparison was made of the total SO_2 flux deduced from the lidar measurements and from *in situ* measurements made by the company. The latter value was calculated by summing the emission values from six different locations which are continuously monitored, and values from manual measurements made irregularly at some additional points. The lidar gave on average 30-50% higher values for the total SO_2 emission. This difference is probably due to diffuse emissions, which are not registered in the measurements made by the company. In conclusion, the measurements showed that the lidar technique can be a very useful method for remote surveillance of industrial

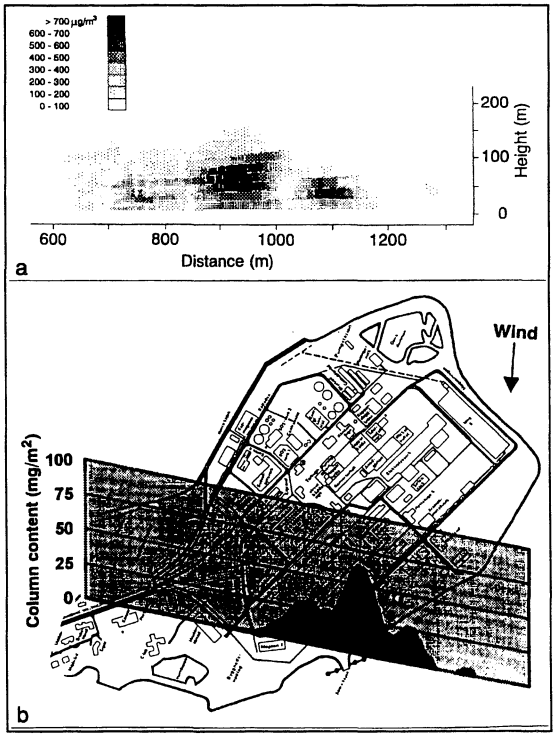

Fig. C1. *SO_2 mapping downwind from a metallurgical plant. a) SO_2 distribution in a vertical plane. b) SO_2 vertical column content, copied onto a map of the industrial area.*

emissions, also at unannounced inspections. During the campaigns, measurements were often performed at different factories on consecutive days, without long set-up times. One main advantage is the possibility of rapidly measuring the total emission from several sources, including diffuse emission, at an industrial site without any preknowledge of the exact position of the different sources. This ability is quite unique and often makes lidar measurements more cost-effective than the numerous measurements needed with point monitors or long-path absorption techniques to achieve the same result.

Collaboration with authorities in the Czech Republic has recently been initiated. A measurement campaign with the mobile lidar system at different industrial, urban and rural areas was performed during September-October 1994. A follow-up campaign is planned for December 1994. This second campaign will be run by the Lighten AB company.

D Laser-induced fluorescence of vegetation and water

Mats Andersson, Hans Edner, Jonas Johansson,
Pär Ragnarson, Sune Svanberg and Eva Wallinder

A project concerned with the remote sensing of laser-induced fluorescence (LIF) from vegetation was initiated in 1990. This is part of two larger European projects, LASFLEUR and EUROMAR. The aim of the project is to construct a remote sensing system which can operate in an airborne-like situation [D1]. In this context two major difficulties must be addressed. Firstly, a useful signal/noise ratio must be achieved in single-shot operation. Secondly, fluorescence must be recorded remotely when there is an interaction between reflected sunlight and fluorescence from objects nearby the target. Furthermore, fluorescence imaging is desirable, and we have continued to pursue the multi-colour imaging technique [D2].

The remote sensing group at the Division of Atomic Physics has participated in five cooperative field campaigns with studies of LIF from terrestrial vegetation and water. The campaigns in Karlsruhe, Oberpfaffenhofen, and Venice during 1992 were discussed in the last progress report, and the results have now been published [D3-D5]. The last field campaign took place in Avignon, France in 1993 [D6]. The detector systems were housed in the remote lidar truck, as in previous measurements. The Nd:YAG laser of the lidar system was frequency tripled and shifted to 397 nm in a D_2 Raman cell. The transmitting and receiving optics of the lidar system were also used. The detection line consisted of two parts, an imaging branch and a spectral branch. For the imaging experiments, a secondary telescope was utilised to divide the image into four identical images, spectrally separated by four different wavelength-selective filters. The sub-images were recorded simultaneously with an image-intensified CCD camera. Computer processing of the images was performed in terms of forming a ratio of two or more of the sub-images. Thus, an image showing, for example, the ratio of the two chlorophyll fluorescence peaks could be displayed. The spectral branch consisted of an optical multichannel analyser (OMA), which is also described in the medical chapter of this progress report. As a development for the Avignon investigation, the sensitivity of the spectral instrumentation was substantially improved by constructing a round-to-line optical fibre bundle for the optical coupling between the telescope and the spectrometer. Furthermore, the same CCD camera was utilised for both imaging and spectral recordings, since the CCD camera has much better signal-to-noise characteristics than the diode array previously used.

Several plant species were available for fluorescence investigation during the Avignon campaign, including maize, cypress, poplar and various citrus trees. In particular, the maize field was well controlled in terms of watering and nutrition supply. In Fig. D1 a set of multi-colour fluorescence images of a maize plant, measured at 40 m distance, is shown. The four upper images show the fluorescence at selected wavelength bands and below these a processed image, displaying the ratio of the 685 nm fluorescence and the broadband blue fluorescence, is shown. Similar images were recorded for maize plants with different degrees of watering and also for maize plants where the herbicide DCMU had been applied. In the latter case, an increased fluorescence was detected indicating blocking of photosynthesis. In Fig. D2 two fluorescence spectra from a maize plant recorded at 40 m are shown. The spectrum to the left was recorded during 100 laser shots, while the one to the right is a single-shot spectrum. It is very interesting to see that the signal-to-noise ratio is still very good for single-shot operation at this distance. Furthermore, this suggests that single-shot operation at 250 m, which is a realistic altitude for an aircraft, might be possible, thereby achieving one of the goals set up for this project. Operation at 250 m distance was unfortunately not possible during the measurements due to the limited electronic delay of the detector controller. This can, however, quite easily be solved in the near future.

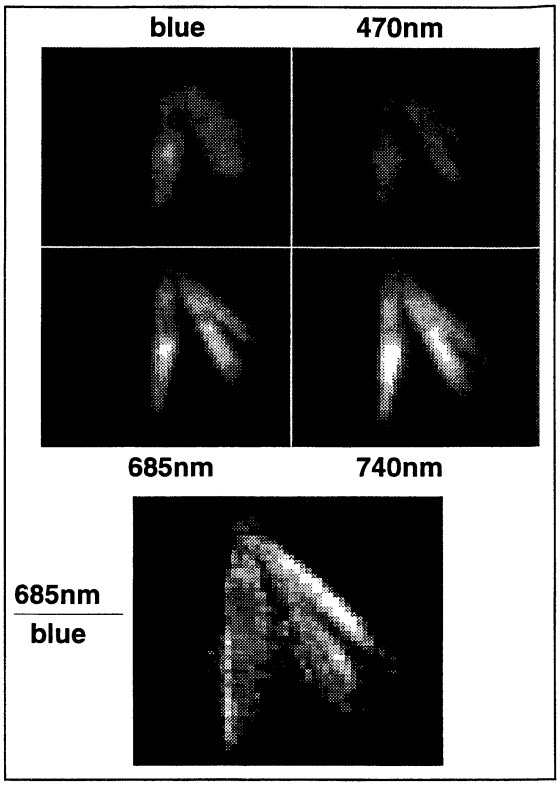

Fig. D1. Example of multi-colour fluorescence imaging for a maize plant. The distance was about 40 m and the target diameter 40 cm.

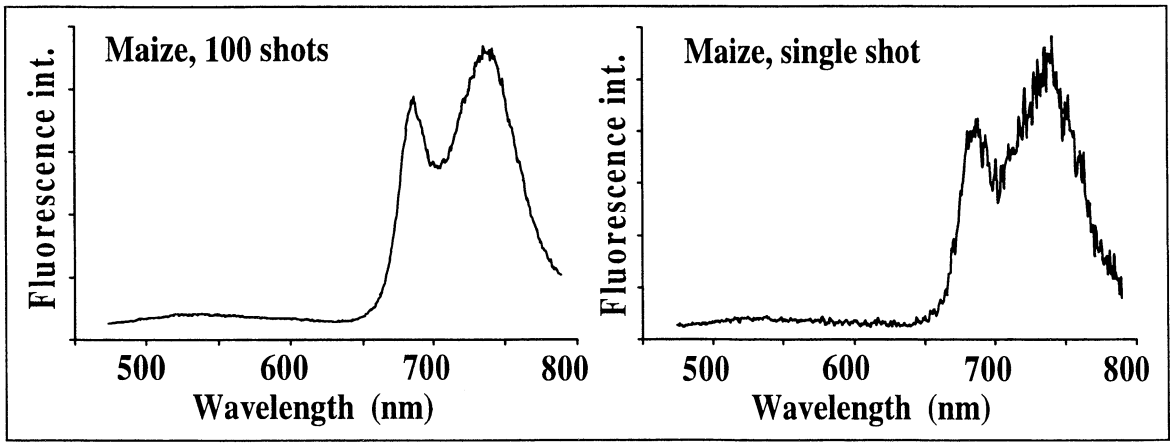

Fig. D2. Fluorescence spectra of a maize plant at a distance of 40 m. The spectum to the left was integrated over 100 laser shots while the spectrum to the right was recorded with a single laser shot only. The laser pulse energy was about 30 mJ.

The next project, which has just started, concerns an alternative approach for remote fluorescence imaging. One difficulty with the present imaging system is the poor signal-to-noise ratio. A possible way of dealing with this problem is to perform one-dimensional imaging, also called the "push broom" technique. By measuring the fluorescence from one line only, the signal-to-noise ratio will probably be sufficient and the full image can be integrated by scanning the target area or, in the future, by the movement of the aircraft.

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IV Laser Applications in Medicine and Biology

The Division of Atomic Physics has continued to play a central role in research at the Lund University Medical Laser Centre. The aim of this organisation is to support interdisciplinary projects using lasers in medical research involving the Faculties of Medicine, Natural Sciences and Engineering within the University, to run courses and seminar series in related subjects, to act as a base for grant applications, and to act as a partner for other groups in multicentre research studies. The research activities within the Centre are focused on wide interdisciplinary research, involving clinicians from various clinical specialities, physicists and chemists. During the past two years, the number of research projects has increased drastically and the group has expanded to include several new PhD students. We have moved into larger premises and 14 scientists and PhD-students from different specialities, working together on a daily basis, now have their offices on the same corridor at the Department of Physics. This greatly simplifies the day-to-day work in the interdisciplinary research conducted. In our presentations of the personnel involved in the different projects, only these people are listed. In addition, a large number of collaborators, within and outside the Medical Laser Centre, also contribute to our work.

Our research activities are focused on tissue diagnostics using laser spectroscopic techniques and photochemical treatment of malignant tumours. Tissue diagnostic research has mainly been directed towards the early detection and identification of malignant tumours. Laser spectroscopic studies have also been conducted on tissues in the circulatory system to identify degenerated areas causing decreased function. The first technique for tissue diagnostics investigated in this project was laser-induced fluorescence to identify diseased tissue. This project started in 1982. The native tissue autofluorescence, originating from endogenous fluorophores, as well as fluorescence from administered tumour markers, mostly porphyrins, was included in the studies. Research in this project during the past two years has been partly directed towards clinical studies of a prototype imaging system to evaluate the potential for the early detection of malignant tumours in various endoscopic applications; and partly to investigating new fluorescent substances regarding their tumour marking capabilities in experimental studies. Photodynamic therapy (PDT) of malignant tumours using a photochemical reaction involving externally administered photosensitising agents and tissue-bound molecular oxygen, has been examined in clinical trials. A large comparative study between PDT, following topical application of δ-amino levulinic acid (ALA) for photosensitisation, and cryo-therapy in the treatment of non-melanoma malignant skin tumours has been initiated. It is interesting to notice that the PDT project has passed the pure scientific stage and about 5 cancer patients are treated every week in Lund using this technique. A potentially very interesting technique under development, for early breast tumour detection, is based on temporally resolved tissue transillumination measurements. At present we aim to fully explore the spectroscopic basis of the technique with *in vivo* and *in vitro* tissue studies. We furthermore aim to improve our basic understanding of light transport in tissue. This work includes tissue-simulating phantom studies and theoretical and numerical modelling of light distribution. Other alternative laser-based diagnostic techniques under investigation include elastic and

Raman scattered light as well as IR tissue spectroscopy. We have also initiated two new projects for laboratory studies: spectroscopy at the microscopic level in two-photon fluorescence microscopy and in a confocal microscope. The concept of fluorescence imaging detection in chemical separations using electrophoresis is also being explored.

Collaborations with more than 10 other research groups have taken place during the past two years. Our recent work has been presented in a large number of invited talks and review papers [1-11]. Last year, Jonas Johansson presented his PhD thesis within the project [12].

A Tissue diagnostics using laser-induced fluorescence

The potential of fluorescence in tissue diagnostics has been evaluated in clinical studies. Both the tissue autofluorescence and fluorescence from externally administered tumour markers have been included in the studies. Spectroscopy, as well as multicolour fluorescence imaging, has been performed. We have also carried out several experimental studies to evaluate the potential of a number of new substances for fluorescence tumour marking.

A1 Experimental diagnostics

Stefan Andersson-Engels, Roger Berg, Jonas Johansson, Liu Lexin, Katarina Svanberg, Sune Svanberg and Ingrid Wang

Several spectroscopic studies have been performed to evaluate the potential of new candidates as fluorescent tumour markers for tissue diagnostics. Photofrin, developed for photodynamic therapy, has been used for a number of years as a tumour marker in the fluorescence diagnosis of tissues. As Photofrin is not a perfect tumour marker and has the side-effect of photosensitising the tissue for several weeks after administration, it would be preferable to find a better agent for marking tumours in fluorescence diagnostics. We have recently examined several candidates. Benzoporphyrin derivative (BPD) is quickly cleared from the body, but our investigations showed that it is not as good as Photofrin in terms of tumour specificity [A1]. Another potential candidate is *meso*-tetra(hydroxyphenyl)chlorin (*m*THPC). Like BPD, this substance is photodynamically active and is fairly quickly cleared from the body. It also showed better tumour demarcation than Photofrin [A2-A3]. In both these studies, the anatomical distribution of the drugs was investigated. As can be seen in Fig. A1, the administration of the endogenous substance δ-amino levulinic acid results in a body production of the photodynamically active substance protoporphyrin IX (PpIX). This process is part of the haem cycle and takes place in the mitochondria. PpIX can be used both for PDT and fluorescence diagnostics of tissue. We have examined the potential of ALA for fluorescence marking of tumours [A4-A5]. High PpIX concentrations, with, in some cases, good tumour selectivity, are observed in the tissue. Other interesting substances for tissue diagnostics using LIF are porphyrins linked to carotene. The carotene quenches the photodynamic activity but not the fluorescence emission. Very interesting results have been obtained regarding fluorescence diagnostics in animal experiments for two such drugs [A6-A7].

The multicolour fluorescence imaging system for tissue diagnostics has been examined in detail and evaluated in clinical studies [A26-A31]. For the diagnostic criterion, this system utilises the differences in tissue autofluorescence (in the wavelength region around 500 nm) and in the porphyrin fluorescence (above 600 nm) between normal and malignant tissue. The system demarcates regions that fulfil the cancer criterion. In the clinically adapted prototype system, suspect regions of malignant growth are displayed on a monitor almost in real time. The image is presented as an overlay on the normal white light image of the tissue. Typical fluorescence spectra for a squamous cell carcinoma and the surrounding vocal cord 4 hours after oral ALA administration, are shown in Fig. A2. A significant difference in blue-green autofluorescence intensity around 500 nm is obvious. Also, the dual-peaked PpIX emission above 600 nm is only seen in the tumour spectrum. A white light image of the same vocal cord is shown in Fig. A3 (upper right). The multicolour imaging system indicates a suspect region on the upper part of the left vocal cord as a whitish overlaid image (lower right). To the left a similar pair of images is presented for a basal cell carcinoma following 6 hours of topical application of ALA in a cream.

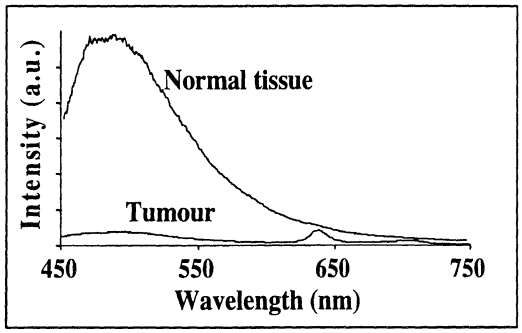

Fig. A2. *Fluorescence spectra from a squamous cell carcinoma and the normal surrounding vocal cord, following 405 nm excitation.*

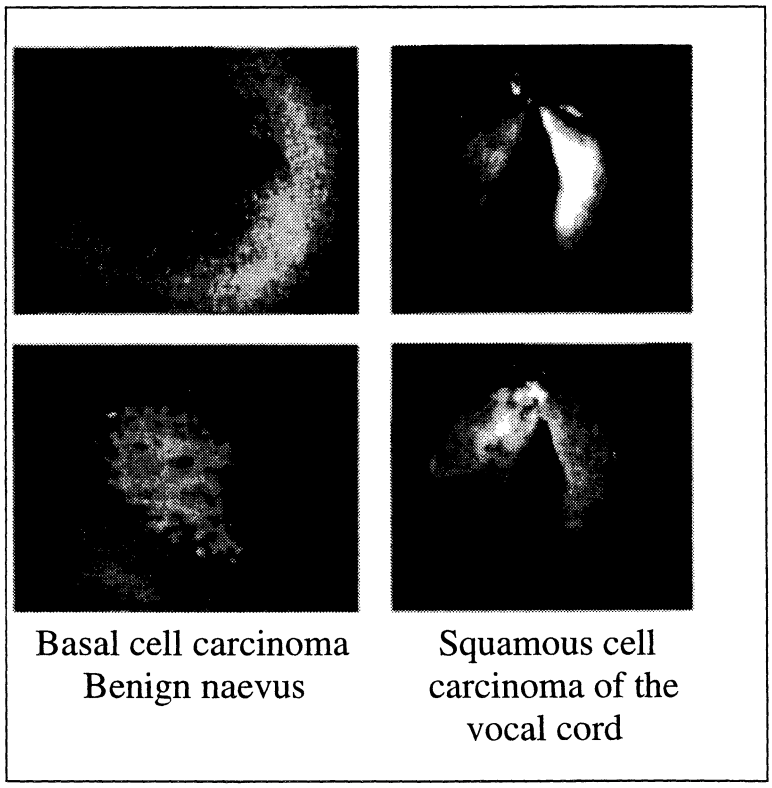

Fig. A3. *White light images (above) and the same images with the multicolour imaging device indicating suspect areas as a bright overlay (below) for a basal cell carcinoma and a benign naevus, as well as for a squamous cell carci-noma on the vocal cord. The patient with the basal cell carcinoma had received ALA topi-cally in a cream before the examination, while ALA dissolved in orange juice was given orally to the other patient 3 hours prior to the examination.*

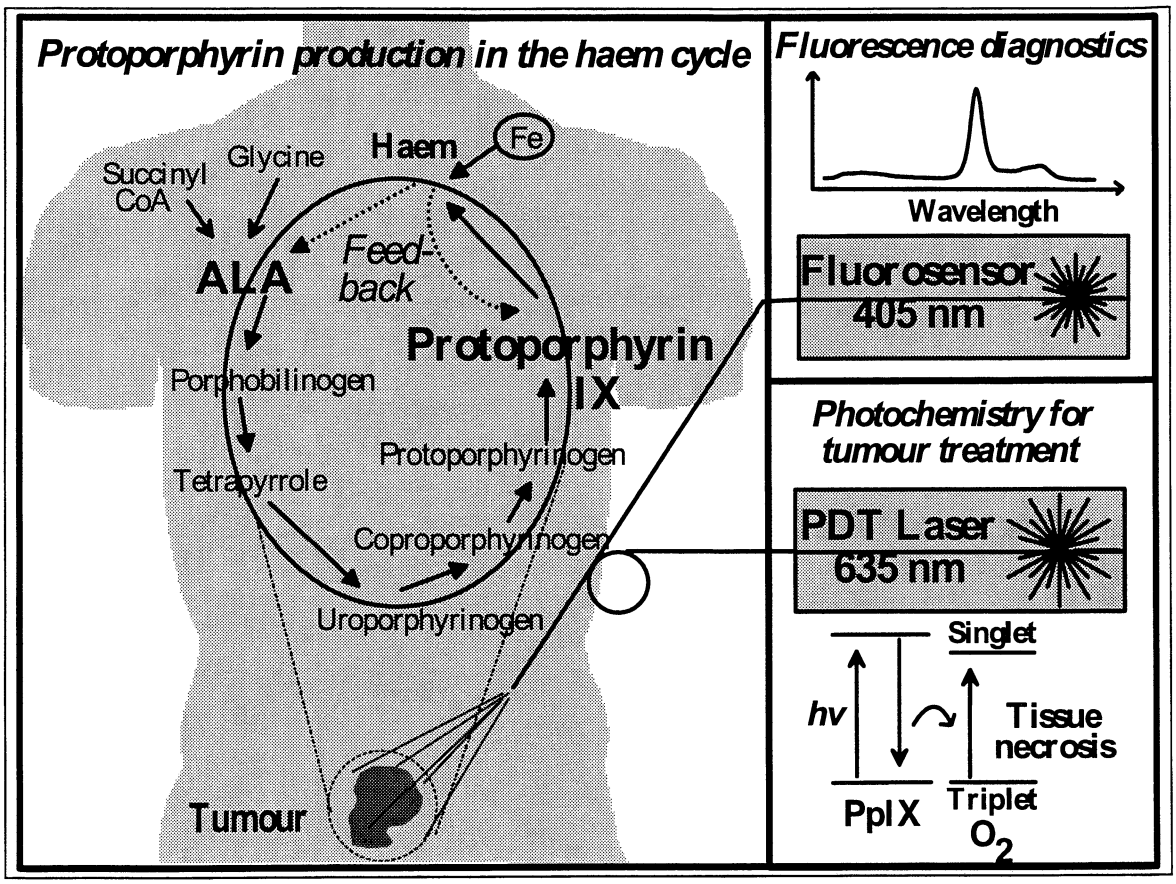

Fig. A1. *Protoporphyrin synthesis in the human body following ALA administration with applications to tumour diagnostics and therapy.*

A2 Human studies

Stefan Andersson-Engels, Roger Berg, Jonas Johansson, Annika Nilsson, Ingrid Rokahr, Katarina Svanberg, Sune Svanberg and Ingrid Wang

Much of our recent fluorescence research on tissue diagnostics has been directed towards the clinical evaluation of the method for various tumour types and tissue localisations. Both *in vivo* and *in vitro* studies of various diseased tissues have been performed. The possibility of being able to exactly correlate the fluorescence reading with histopathology provides strong motivation for *in vitro* investigations in the evaluation of optimal conditions for high contrast between normal and diseased tissue [A8-A12]. In the clinical evaluation, *in vivo* clinical studies are necessary to determine the sensitivity and selectivity of a method. Normal brain tissue and malignant tumours have been studied in tissue autofluorescence following 337 nm and 405 nm excitation. The results of these studies are somewhat difficult to interpret, and no clear demarcation between the tumours and surrounding tissue could be found [A13-A14]. Photofrin in low doses (0.35 mg/kg body weight) has been used as a tumour marker in order to visualise Barrett oesophagus as well as breast and urinary tumours. Tumours could be detected at all these locations using LIF [A9-A12, A15-A24]. Clinical measurements have also been performed using orally administered ALA for tumour marking in the ENT region [A25].

B Photodynamic therapy

Stefan Andersson-Engels, Roger Berg, Jonas Johansson, Claes af Klinteberg, Liu Lexin, Annika Nilsson, Katarina Svanberg, Sune Svanberg and Ingrid Wang

Photodynamic therapy (PDT) is a method used to treat malignant tumours. It relies on the selective transfer of triplet to singlet oxygen and the generation of free radicals, causing tissue necrosis. The drug is normally injected intravenously and is retained to a greater degree in the malignant tissue than in normal tissue. Thus, selective necrosis of cancer lesions can be achieved.

During the last couple of years, a new PDT procedure for treating superficial tumours has been introduced. A solution containing δ-amino levulinic acid (ALA) is applied topically 3-6 hours prior to treatment. The ALA molecules enter the tumour cells, where they are transformed into protoporphyrin IX (Pp IX) following the normal haem cycle in the mitochondria. It is possible to monitor the accumulation of PpIX by means of laser-induced fluorescence (LIF) [B1]. The photodynamic action is initiated, by irradiating the lesions with red light, 630 nm. The efficiency of ALA-PDT in a number of different tumour types has been investigated, including non-melanoma malignant skin tumours [B2-B7], head and neck cancer [B8], basal cell carcinoma (BCC) of the eyelid [B9-B10] and experimental hepatic tumours [B11-B13].

To minimise the variation in tumour growth and size between the treated tumours and those used for control, a new experimental model has been developed [B14]. The model employs two identical tumours in a rat liver - one is used for treatment and the other is left untreated as an internal control. The two tumours are inoculated in the subcapsular region of two liver lobes. This tumour model has been used in ALA-PDT studies.

The superficial blood flow has been monitored immediately before and after the PDT procedures in a number of studies, utilising laser-Doppler imaging equipment [B15-B17]. These studies indicated direct vascular damage due to PDT when the photosensitiser was delivered systemically, while no constriction of the blood flow resulted for PDT following topically applied ALA.

Today, cryo-therapy is the conventional treatment modality for BCC of the skin. To investigate the efficiency of ALA-PDT, the two treatment modalities are being compared in an extensive study. A total of 160 patients with nodular or superficial BCC will be included in the study and will be treated with one of the methods, selected at random. Before treatment, the patients undergo a health control. The lesions are photographed and recorded on video tape and LDI measurements of the blood flow are made. In conjunction with ALA-PDT, the amount of PpIX is monitored, both before and after treatment. The patients return for follow-up visits one week after the treatment and then once every month for twelve months.

As light does not penetrate very far in tissue, PDT is most efficient on thin tumours. To be able to treat thicker tumours, a computerised light delivery and monitoring system is being developed at the Division. The light from the treatment laser is divided along a number of optical fibres. These fibre probes are inserted into the tumour through a hypodermic needle and the positions of the probes are indicated in a co-ordinate system on the computer screen. Each fibre can now be used either to deliver light to the tumour or to collect the

light from any other probe. This makes it possible to treat tumours close to critical tissue structures as the delivery of the light dose can be well controlled.

C Measurements of the optical properties of tissue with applications to medical diagnostics

Stefan Andersson-Engels, Roger Berg, Claes af Klinteberg, Annika Nilsson, and Sune Svanberg

As light interacts with tissue, it is either absorbed or scattered in various proportions depending on the optical properties of the tissue. These tissue characteristics are important to be able to understand the mechanisms of light interaction with tissue in various medical laser applications. Knowledge concerning light transport in tissue is essential for dosimetry in conjunction with photodynamic tumour treatment, transillumination imaging and fluorescence diagnostics. It is therefore important to be able to determine the scattering and absorption coefficients, as well as the mean cosine of the scattering angle for various wavelengths.

This can be done with an *optical integrating sphere technique* using an experimental arrangement as shown in Fig. C1. The method is general and can be used to determine the

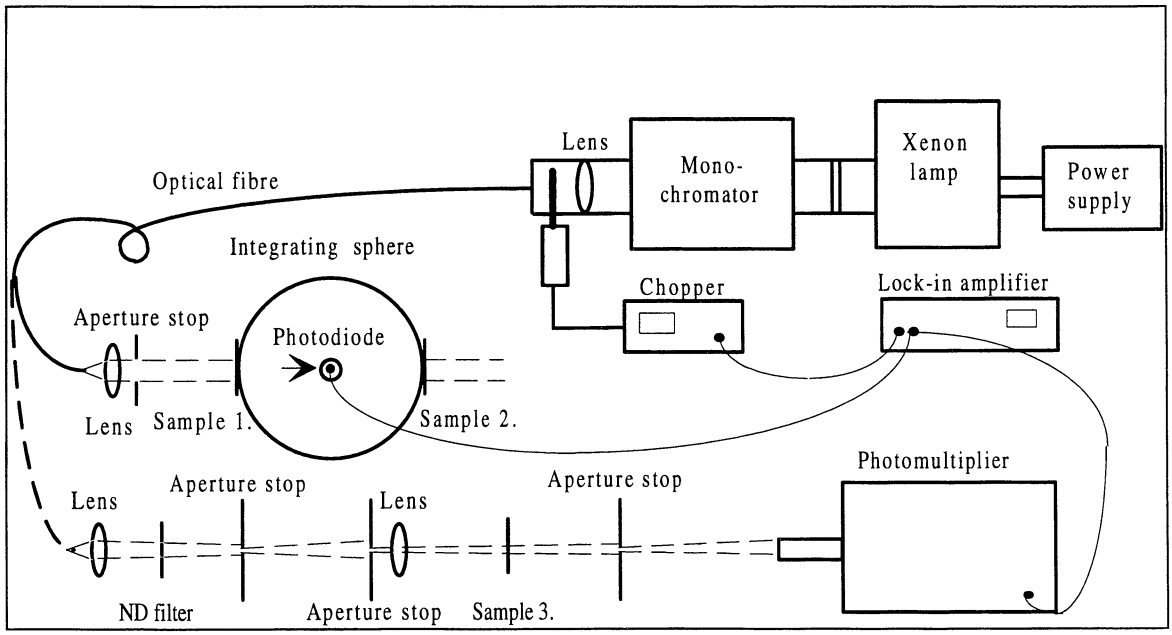

Fig. C1. *The setup for integrating sphere measurements of tissue optical properties.*

optical properties of tissues as well as those of any kind of scattering material. A xenon lamp is used as a light source and the wavelength is selected with a monochromator. The light is guided in an optical fibre to a tissue sample attached to the integrating sphere. The intensity of the light interacting with the sample, is measured with a photodiode. Depending on the position of the tissue specimen, either the reflectance or the total transmittance is recorded. The direct unscattered light transmittance is also measured. The measured values are compared with results from Monte Carlo simulations and the scattering coefficient, absorption coefficient and mean cosine value of the sample can be deter-

mined [C1-C2]. This setup has been used to measure the optical properties of tissue before and after photodynamic therapy, to see if the light penetration changes during treatment [C3-C5]. The results show a major increase in the absorption coefficient, causing a decrease in the treatment light penetration, which most probably reduces the effect of the treatment.

The use of ionising radiation, X-rays, when screening larger populations for mammary malignancies, has been under discussion lately. As ionising radiation may induce cancer, the use of non-ionising radiation (light) has become more and more interesting [C6]. *Time-resolved transillumination* uses red and near-infrared light to transilluminate human tissue. This technique suppresses the influence of the multiply scattered light [C7-C13], which otherwise gives rise to a severe blurring of the images. This is important, as the dominating attenuation effect in this wavelength region is scattering. The samples are irradiated with picosecond laser pulses and the transmitted light is detected time-resolved. By focusing on the first light exiting the sample, features located deep inside the tissue can be enhanced. Different light sources (picosecond diode laser, mode-locked Ar-ion/dye laser, mode-locked Ti:Sapphire laser) and detection techniques (time-correlated photon-counting and streak camera detection) have been used. We have found that for the case of scattering-dominated attenuation (scattering coefficient >> absorption coefficient) detection of early transmitted light will be practically insensitive to variations in the absorption coefficient. It is the scattering properties that determine the amount of early light detected. The time-resolved curves obtained when transilluminating a highly scattering sample contain information on the optical properties of the sample (effective scattering and absorption coefficients). We have developed a new technique for performing multispectral measurements of tissue optical properties [C14-C18]. The terawatt laser system at the Lund High-Power Laser Facility is used as a light source, generating a white continuum of femtosecond pulses. Fig. C2. (left) shows the setup. The sample is irradiated with the white light and the transmitted light is recorded with a polychromator and a streak camera. This gives a two-dimensional image containing the intensity as a function of wavelength and time. The figure also shows a streak camera image obtained when transilluminating a 33 mm thick female breast *in vitro*.

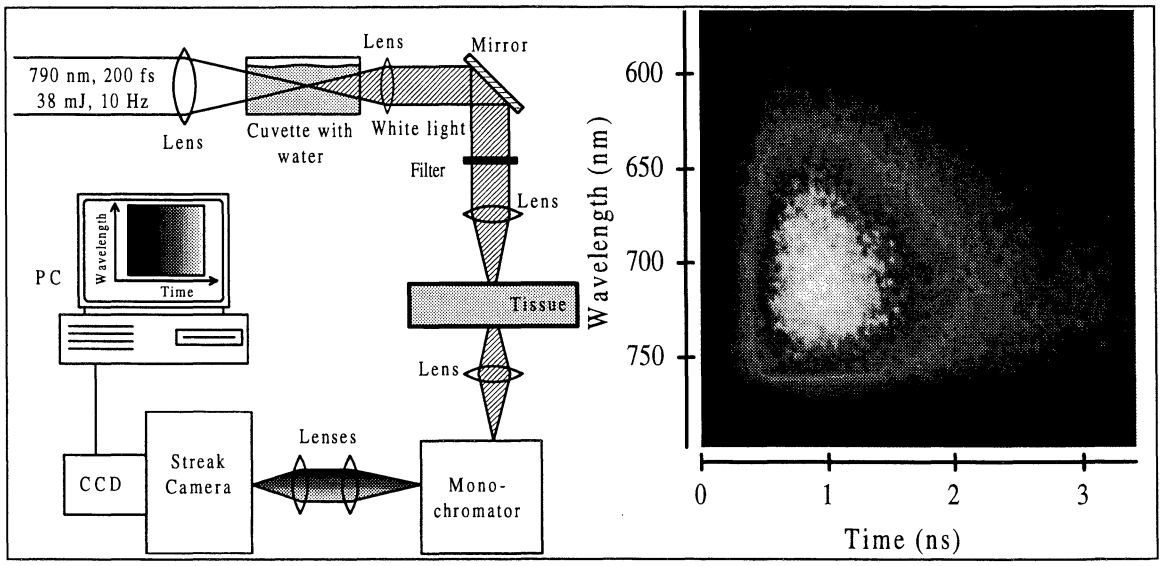

Fig. C2. *Left: The setup for time-resolved white light experiments. Right: The intensity as a function of time and wavelength when transilluminating a 33 mm thick female breast sample in vitro.*

We have developed a computer model for predicting the behaviour of light in tissue. It is based on numerical solution of the diffusion equation. The model permits calculations of the flux as a function of time for inhomogeneous samples. The model has been shown to accurately predict the time-dispersion curves obtained experimentally [C19-C24].
In a collaboration with University of Linköping, we have performed *in vivo* studies of tissue optical properties for various blood perfusions [C25-C26]. These studies could provide useful information for the development of laser-Doppler perfusion measurements.

D Two-photon-excited fluorescence microscopy

Stefan Andersson-Engels, Jörgen Carlsson and Ingrid Rokahr

Two-photon fluorescence excitation is a new concept for scanning microscopy. With this technique, fluorescence is excited only in the focal volume of the microscope lens. This technique has been evaluated for high-resolution fluorescence microscopy and for temporally and spectrally resolved spectroscopy on well-defined sections or single voxels. With this technique it is also possible to perform deep sectioning and in that way obtain three-dimensional images.

The setup used is shown below in Fig. D1. A Ti:Sapphire laser yielding 150 fs long pulses at 76 MHz centred at 792 nm was used as an excitation source. The fluorescence emission between 400 and 700 nm was studied using a photomultiplier tube (imaging mode) or a

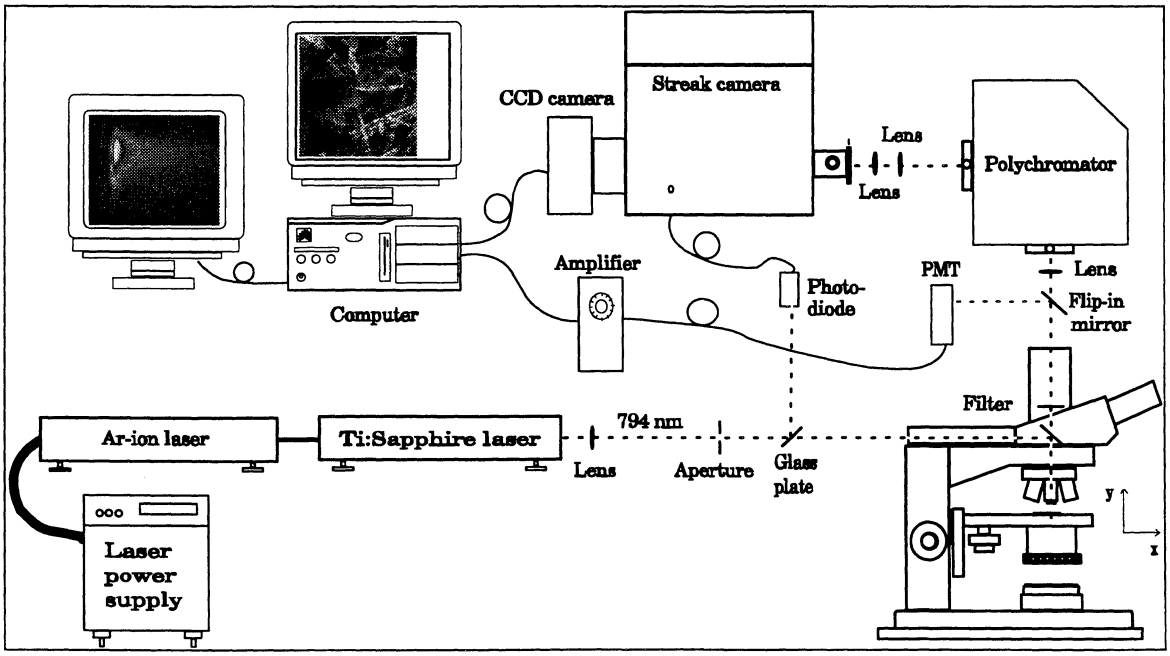

Fig. D1. *Setup for the two-photon-excited fluorescence microscope with spectroscopic detection.*

spectrometer and a streak camera (spectral and temporal spectroscopy mode). The probability of two-photon excitation is quite low and depends on the square of the laser intensity. The peak power must thus be high, while the average power must be kept low in order to avoid sample heating. Due to the quadratic dependence, the excitation is limited to

the focal volume and one intrinsically gets rid of the scattered light from other parts of the sample - an almost background-free method thus results. A further advantage is that lens aberrations are lower and easier to compensate for the longer the wavelength.

With this method we have studied samples of paper (see Section V.B), plant leaves and rodent liver and lung tissue [D1-D7]. It was shown that it is possible to distinguish between different fluorophores using their spectroscopic features and that it is possible to detect protoporphyrin IX in tissue used for photodynamic tumour therapy. We aim to investigate localisation and binding features of various photosensitisers to better understand the mechanisms of tumour selectivity. Similar investigations have been performed with a scanning confocal fluorescence microscope in a collaboration with other groups [D8-D10].

E Capillary electrophoresis

Jonas Johansson

A project on real-time fluorescence imaging of capillary electrophoresis separation has recently been started. The project is a collaboration between the medical group at the Division of Atomic Physics and the Department of Technical Analytical Chemistry at Lund Institute of Technology. Capillary electrophoresis is widely used for analytical separation of various molecular species, from small ions to macromolecules, such as proteins and DNA fragments. The detection principle used has, in most cases been UV absorption, while the use of laser-induced fluorescence has more recently shown superior sensitivity. Point monitoring of a small window at the end of the capillary has been employed in all cases. Our approach is to image a substantial length of the capillary in order to be able to follow the separation process [E1-E3]. Applications for this technique, besides standard capillary electrophoresis, include isotacophoretic preconcentration and isoelectric focusing. Furthermore, with the proper software it may be possible to follow the peaks as they travel across the window and to integrate the signal for several minutes. With this technique, the signal-to-noise ratio may be substantially improved.

The currently used excitation source is an excimer-laser-pumped dye laser with a pulse energy of a few mW at about 500 nm. The light is guided through an optical fibre and beam-shaping optics to form a 7 cm streak of excitation light at the capillary. The fluorescence is recorded by an image-intensified CCD camera equipped with a 35 mm objective. Line profiles of fluorescence intensity as a function of position along the capillary are presented on the screen of a PC. The line profiles are updated on the screen at the repetition rate of the laser or, if integration over several laser shots is employed, with the integration time for each frame (typically 2 sec.). Measurements of the system sensitivity were performed using a fluorescein dye in a buffer solution and the sensitivity found to be about 10^{-9} M with the

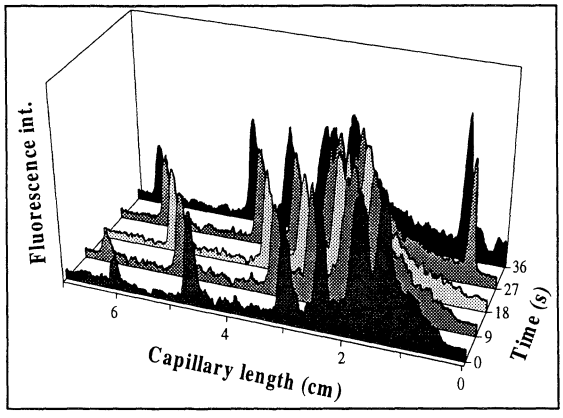

Fig. E1. *Five electropherrograms recorded with a time difference of 9 s between each.*

present set-up. Electrophoretic separation of DNA fragments has also been performed. Fig. E1 shows selected fluorescence line profiles about 5 min post-injection from such a run. It should be pointed out, that this presentation does not illustrate the dynamics of the moving pictures that are actually shown on the screen but gives only a slight impression of peaks moving from the right to the left. Furthermore, successful experiments were performed on isotacophoretic preconcentration of Rhodamin B. The limiting factor of the system sensitivity is probably the pulse-to-pulse fluctuations of the excimer laser. Thus, experiments with a Kr-ion laser as excitation source are planned for the near future. Methods of increasing the collection efficiency, such as fibre optic arrays, will be investigated as well as different methods for computer-assisted signal integration and filtering.

F New techniques under development

Stefan Andersson-Engels, Roger Berg, Joakim Bood, Hugo Carlsson*, Ulf Gustafsson, Claes af Klinteberg, Charlotta Lindquist, Åsa Persson* and Sune Svanberg*

**M.Sc. Students*

In addition to the projects briefly presented above, we have during the last 2 years started a number of new projects related to medical optics. A treatment modality for malignant tumours is *hyperthermia*, implying heating the tissue to 43-47°C for a time period of 30 min to 1 hour. Cell death ensues, in part mediated by detrimental vascular effects caused by the heat. As the cellular response is highly dependent on the local temperature, knowledge of the entire tissue temperature distribution during the treatment is important. For this purpose, a numerical algorithm which calculates the complete temperature distribution in a laser-irradiated tissue has been developed [F1-F3]. According to well-controlled experimental measurements, the model is able to predict the dynamic temperature changes with an accuracy of 95% in liver tissue during laser irradiation. The algorithm includes complex surface heat processes, such as evaporation of water. Hyperthermia may have a clinically significant synergistic effect when combined with photodynamic therapy (PDT). A study has been initiated to investigate this. The vascular damage caused by both PDT and hyperthermia has been studied both with laser Doppler flowmetry studies and in electron microscopy. The electron microscopy study [F4] showed that three hours after PDT, 3 hours following *i.v.* administration of ALA, the tumour cell membranes were deformed and damaged. Three days after PDT the vascular lumen in the treated tissue was partially blocked as a result of endothelial cell oedema and adherence of platelets to the vascular wall. Hyperthermia caused widening and protrusion of the borderlines of endothelial cells, the vascular lumen became small and openings of branch vessels were narrowed. A large number of blood cells, platelets and fibrin networks were accumulated in the vessel and blocked almost the entire vascular lumen.

A number of new concepts for optical tissue diagnostics have been evaluated. An alternative to time-resolved transillumination is the same measurement using *frequency domain spectroscopy*. When light traverses a strongly scattering medium, such as tissue, the propagation of the photon density can be described as a diffusion process. If the incident light is amplitude modulated, the detected light will show a phase shift and a demodulation due to the optical properties of the medium and the modulation frequency. To be able to detect the phase shift, the modulation frequency should be at least 100 MHz. In this frequency region, both the phase shift and the demodulation can be detected

instantaneously, which makes the sampling time short. Experiments with an intensity-modulated laser diode in the near-infrared wavelength region have been performed [F5]. On the detector side a photo-multiplier tube and an RF mixer were used to detect the phase lag and the demodulation.

If two frequency-modulated light sources are used, 180 degrees out of phase, two *interfering diffuse photon density waves* will propagate through the tissue and the interference pattern can yield improved spatial resolution. Theoretical studies show that this could be a useful tool for finding objects in a turbid medium [F6]. Experiments have been performed to confirm the theoretical predictions [F7-F8].

Another less sophisticated technique for non-invasive superficial tissue diagnostics is to measure the diffuse reflected light spectrally resolved. *Elastic scattering spectroscopy* has been tested for human tissue samples *in vitro* [F9]. The small study conducted shows that differences in the spectral shape between malignant and non-malignant tissue could be seen in the wavelength region between 300 and 580 nm. The concept for optical diagnostics is very interesting, as only simple instrumentation is necessary. Further investigations are required to optimise the measurement geometry. *In vivo* studies will then be necessary to obtain the sensitivity and selectivity of this method for the identification of various kinds of malignancies.

Raman spectroscopy provides much more information about a tissue sample than diffuse reflected light or fluorescence, since sharp spectral lines result rather than broad absorption or fluorescence features. The major limitation of Raman spectroscopy is the weak signal generated - typically a thousand times weaker than fluorescence. This makes the Raman signals difficult to detect, as they are superimposed on tissue fluorescence. Tissue diagnostics also require limited excitation power. These limitations have made Raman spectroscopy a less attractive tool in tissue diagnostics. However, in the near-infrared wavelength region only weak fluorescence results and a Raman spectrum with an acceptable signal-to-noise ratio can be acquired [F10-F11]. We have been using an experimental system based on 795 nm excitation wavelength from a Ti:Sapphire laser with cooled CCD camera detection. With this system we have been able to measure Raman signals from tissue constituents, e.g. cholesterol and elastin. In Fig. F1 a recorded spectrum from cholesterol is shown. The aim is to record Raman spectra from human tissue and to discriminate normal tissue from diseased. As with Raman spectroscopy, *infrared spectroscopy* probes the vibrational levels in the molecules studied. Similar results to those from for Raman spectroscopy should thus be expected for infrared signals. Preliminary results using a BOMEM DA 8 FT spectrometer on tissue samples indicate that FTIR spectroscopy can be used for tissue characterisation.

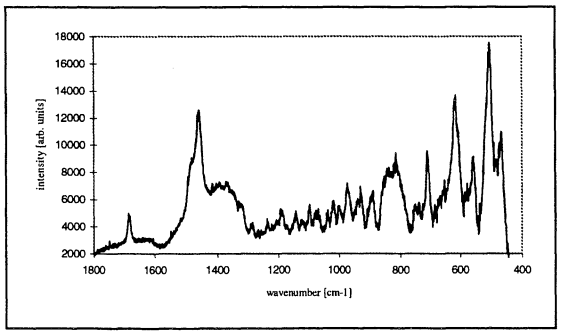

Fig. F1. *Raman spectrum of cholesterol using 795 nm excitation from a Ti: Sapphire laser and a cooled CCD camera detector. The integration time was 150 s.*

In a collaboration with the Department of Radiation Physics at the Lund University Hospital, an *in vivo dosimetry technique for radiation therapy* is under development. Knowledge of the absorbed dose and dose rate is very important for the outcome of the treatment in radiation therapy. Small implantable dosimeters based on thermoluminescence exist but have not proven to be sufficiently accurate. The requirements of an *in vivo* dosimeter include high accuracy, linearity over a large dose interval, small size and ease of use in clinical situations. An *in vivo* dosimeter based on laser-induced fluorescence emission is under development. The sensor consists of a piece of a $BaF(Br,I):Eu^{2+}$ imaging plate attached to an optical fibre. The common use of imaging plates is as replacements for photographic plates in X-ray screening. When the X-rays hit an imaging plate, the Eu-doped crystals are excited to a higher energy level and then relax to a very long-lived excited state. After X-ray screening, the imaging plates are read using a light source at about 600 nm, resulting in an emission at 400 nm in proportion to the absorbed dose. Furthermore, prompt emission at 400 nm can be detected during irradiation, thus yielding information about the dose rate. We are presently using a setup with a gated image-intensified CCD camera and a pulsed dye laser pumped by an N_2 laser. The gated detection facilitates reliable operation at high ambient light conditions. Our measurements so far have shown that this sensor has good linearity over at least 4 orders of magnitude. The signal-to-noise ratio is excellent and the accuracy is mainly limited by pulse-to-pulse fluctuations in the N_2 laser. These fluctuations can probably be compensated for by normalisation on the scattered laser light or by direct measurement of the laser output energy. First measurements on a patient are scheduled for November 1994.

Other work from the Division published in the field of medical laser applications can be found in Refs [F12-F13].

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V Industrial Applications

The industrial applications of techniques for optical measurements are pursued with consideration of the needs of some of the major Swedish industries: metallurgical-, paper- and electrotechnical industry. Methods for controlling smelt-metallurgical processes using optical spectroscopy are developed in cooperation with copper- and steelmaking companies. Studies of the propagation of light in paper and of the distribution of different substances in paper using fluorescence spectroscopy are performed in cooperation with the Swedish Newsprint Research Centre (TFL). Electric breakdown in dielectric liquids, particularly high-voltage insulators, is studied using optical methods in cooperation with ABB.

A Applications of optical spectroscopy in the metallurgical industry

Lennart Malmqvist, Willy Persson and Wilhelm Wendt

During the last 6-7 years new methods for optical-spectroscopic, on-line production control of smelt-metallurgical processes have been developed in collaboration with various copper- and steelmaking companies. The technique involves definition of optical process parameters, based on time- and frequency resolved registrations of the light emitted by a high-temperature process and identification of correlation between these optical parameters and relevant metallurgical data. The growth in speed and complexity of industrial processes in general leads to a never ceasing demand for fast, reliable and detailed on-line information on the instantaneous status of the processes at a reasonable cost. The goal is improved, dynamic process control, eventually leading to high and stable product quality, predictable logistics and reduced fugitive emissions: substitution of 3S-technology for 3D-jobs!

The technology, which is being developed in co-operation with Semtech Metallurgy AB at Ideon Research Park, has been tested at various process steps in copper- and steelmaking. Late 1993 an applicability test was performed at Chuquicamata in Chile. During 1994 the technology has been permanently installed for production control at two coppermaking plants, more precisely at Pierce-Smith converters at Boliden Rönnskärsverken in Sweden and at Norddeutsche Affinerie in Germany. The technique is being used for optimising the end-point determination of the slagforming and coppermaking steps and for dynamic control of the silica content of the slag. These goals are achieved by monitoring continuously the light emission from gas-phase PbO, PbS and CuCl in the off-gas flames of the converters. The vapour pressures of these elements above the melt/slag mixture turn out to be closely related to the oxygen potential in the converter, which, in turn, affects the impurity refinery [A1,A2].

The development of the technology for alloy-steel converting has continued, including field tests in Spain and Italy. An echelle spectrometer under construction will considerably extend the wavelength coverage without deteriorating the spectral resolution, thus increasing the number of tentative optical process parameters.

B Optical spectroscopy of paper

The studies of paper described in this section are being carried out in cooperation with the Swedish Newsprint Research Centre (TFL).

B1 The propagation of light in paper

Hans Busk, Jörgen Carlsson, Lennart Malmqvist, Willy Persson and Claes-Göran Wahlström

The propagation of light in paper affects the possibility of printing with high resolution on the paper or printing on both sides of a thin sheet of paper, as is desirable, for example, when printing newspapers.

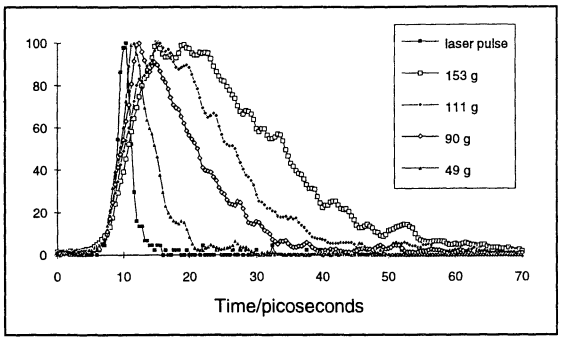

Fig. B1. *Response function of the detection system (2.5 ps FWHM) and single-shot recordings of light pulses at 795 nm which have passed through single sheets of paper with different basis weights.*

Paper consists of a network of cellulose fibres. Most of the fibres are aligned in the plane of the paper and most of the originally cylindrical fibres have collapsed and are flat. In addition to the cellulose fibres, strongly scattering fillers consisting of much smaller particles may be included in the paper. Many types of paper are also coated to provide a white surface. In these studies, however, the main emphasis is on non-bleached, non-coated paper, as is used, for example, for printing newspapers. Attempts are being made to develop a realistic model for the propagation of light in paper, including reflection and scattering by fibres, pores and fillers in the paper. The model is used in Monte Carlo simulations and the simulations are compared with and tested against various experimental results. The optical properties most easily measured for a sheet of paper are reflection and transmission. In addition to such measurements, time-resolved experiments on the propagation of light in paper have also been performed [B1]. These studies were performed using the 0.2 ps high-power pulses from a Ti: sapphire laser system and a streak-camera. To facilitate studies of the dependence of the propagation of light on individual properties of the paper, sheets of paper with well-controlled properties are used. Measurements are being made both on systematic series of such sheets and on ordinary newsprint. Examples of these measurements are given in Fig. B1, which shows time-resolved recordings of a 0.2 ps pulse transmitted through sheets of paper with different basis weight. Other properties of the sheets, such as chemical composition and density, are identical. These measurements show that the average path of the light inside the paper is, for red light, of the order of ten times the thickness of the paper. For shorter wavelengths, which in non-bleached paper have a higher absorption, the average path is shorter. A survey of the possibilities of optical measurements in studying paper was made at a meeting of Swedish paper industry [B2].

In a related project, optical dot gain - the apparent increase in size of a printed dot, due to light-scattering in the paper near the edge of the dot - will be studied.

B2 Paper fluorescence

Håkan Bergström, Jörgen Carlsson, Lennart Malmqvist and Willy Persson

Optical methods offer the possibility of very fast measurements without physical contact with the paper. These properties are particularly attractive as the requirements for on-line production control and product-quality assessment increase. Fluorescence methods have the ability to combine fast, non-intrusive measurements with spectroscopic identification of specific substances used in paper making.

A study has been made of the spectral and temporal behaviour of fluorescence from newsprint and from substances present in newsprint [B3, B4]. Short laser pulses of different wavelengths were used to induce the fluorescence. The spectral distribution of the fluorescence light was analysed with a spectrometer and the time-dependence of the fluorescence was studied using time-correlated single-photon counting.

The only isolated paper constituent emitting a noteworthy fluorescence after irradiation by visible light is lignin. After excitation with visible light, newsprint samples emit fluorescence with spectra and lifetimes similar to those from lignin. With UV excitation, copy paper exhibits a strong fluorescence with a spectrum and lifetime similar to those found for fine paper whitening agents. Among other paper samples, to which no fluorescent substance has been deliberately added, those containing recycled fibres give the strongest fluorescence. They have spectra and lifetimes similar to those of whitening agents and copy paper and this fluorescence may be assumed to originate from recycled fine-paper. Newsprint without recycled fibres exhibits much weaker fluorescence, at longer wavelengths and with other lifetimes.

Knowledge concerning the spectral characteristics of the fluorescence from individual paper constituents can be used to determine the spatial distribution of each fluorescent constituent; in the case of newsprint, the lignin and the recycled fibres. A portable "lignin-meter" has been constructed for this purpose. By recording the fluorescence intensity with a sampling frequency of 20 kHz, a 1 mm resolution is obtained for paper moving at 20 m/s, whish is the typical speed in a newsprint-producing paper machine. Examples of recordings of the lignin fluorescence signal of paper from two different paper machines are shown in Fig. B2.

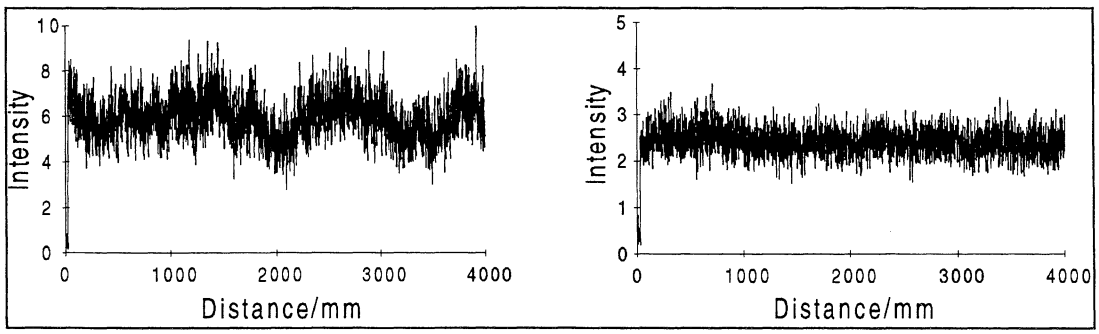

Fig. B2. *Fluorescence recordings made on moving sheets of newsprint revealing the lignin concentration in the paper with 1 mm resolution.*

The distribution of fluorescent substances in paper can also be obtained with much higher spatial resolution using two-photon-excited fluorescence microscopy [B5]. The development of this new microscopic method is described in section IV-E. By combining this form of microscopy with spectroscopic detection techniques, individual substances, such as lignin and fluorescent whitening agents, can be identified. This technique is used both for performing spectroscopic studies on these substances and for recording the distribution of the substance in the paper, in both cases with a three-dimensional resolution of the order of 1 to 2 μm.

C The physics of electric breakdown in dielectric liquids

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Electric breakdown is a phenomenon of great scientific and economic interest. In liquids, the understanding of the processes preceding breakdown is at best incomplete, although optical analysis methods are revealing more and more of the processes underlying the observations. The power industry has a great interest in knowing how best to avoid electric breakdown. The insulation system constitutes a large part of the cost in, for example, a power transformer. Providing better insulation is an important goal for industrial research and development departments.

Research in insulation systems is performed at the Department for High Voltage Insulation Systems at ABB Corporate Research in Västerås (a research centre within ABB). ABB is a leading manufacturer of electrotechnical equipment in the world. Scientists at the Division of Atomic Physics have vast experience of optical diagnostic methods. ABB Corporate Research and the Division of Atomic Physics have been cooperating in a project to study the physics of liquid electric breakdown since 1991. The aim of the project is to initiate a new area of research in Lund, and to yield new methods of analysis relevant for ABB, as well as new results for the scientific community.

So far, laser-induced initiation of breakdown in liquids, transient currents and broadband light emission from pre-breakdown phenomena, and spectral analysis of the pre-breakdown phenomena to deduce the electron density in the pre-breakdown channel have been studied. The laser-initiation technique and the broadband light emission measurement technique developed at the Division have been adopted by ABB Corporate Research and are now routinely used in Västerås.

C1 Spectroscopic investigations of pre-breakdown phenomena in liquids

Each breakdown event is preceded by an electrical connection between the two electrodes. The connection is termed a *streamer*. Some measurements indicate two distinctive parts in the streamers: the *head* and the *stem*. The properties of streamers, such as charge density, liquid/solid phase, diameter, and temperature, are not well known. The goal of our measurements is to deduce the electron density and the temperature in streamers from spectral measurements on Stark-broadened hydrogen Balmer series lines.

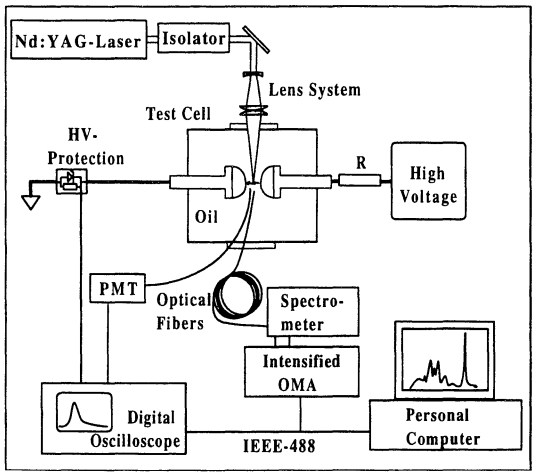

Fig. C1. *Experimental set-up.*

Emission spectra of pre-breakdown events in dielectric liquids were recorded employing a spectrograph and a sensitive intensified diode array or CCD detector. This permits detection of the very faint light emission from the pre-breakdown events. Together with the spectra, current and broadband light emission are recorded for analysis and synchronisation of the measurements. The experimental set-up is shown in Fig. C1. The pre-breakdown events were generated by a laser-produced plasma in the electrode gap. This technique has been described previously [C1,C2].

The spectra, which contain lines from the hydrogen Balmer series and the C_2 radical, were then analysed by fitting the hydrogen Balmer α line to tabulated theoretical line shapes calculated using temperature and electron density as parameters. The linewidth is strongly dependent on electron density, and less strongly dependent on temperature. The

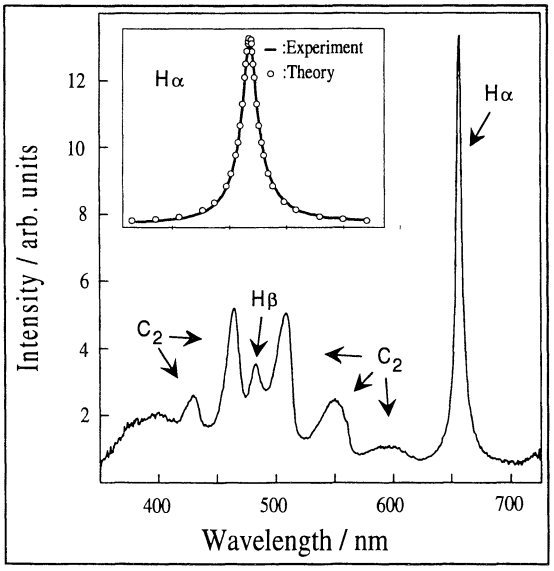

Fig. C2. *Spectrum of breakdown arc. C_2 lines and hydrogen Balmer series lines identified. A fit of a tabulated Balmer α line spectrum to the experimental Balmer α line is inserted. The fit yields an electron density of 10^{18} cm^{-3}.*

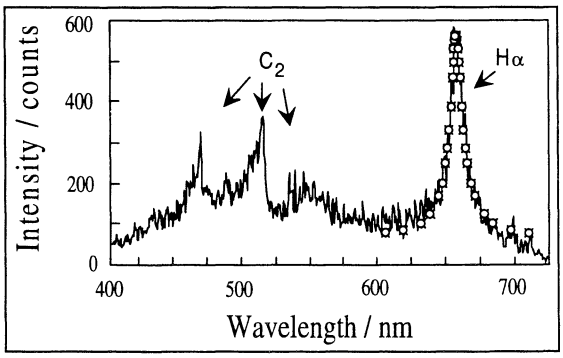

Fig. C3. *Pre-breakdown spectrum of event with positive polarity. C_2 lines and hydrogen Balmer α line identified. A fit of a tabulated Balmer α line spectrum to the experimental Balmer α line yields an electron concentration of $5x10^{18}$ cm^{-3}.*

temperature could be inferred from the relative intensity of the Balmer α and β lines. The electron density was obtained from the best fit. Measurements on breakdown arcs yielded electron densities of about 10^{18} cm^{-3} (see for example Fig C2), and measurements on pre-breakdown events of positive and negative polarity yielded electron densities varying from $5x10^{18}$ cm^{-3} (positive polarity) to below 10^{17} cm^{-3} (negative polarity). An example of a pre-breakdown measurement is shown in Fig. C3. This is the first time these parameters have been measured in a streamer, to our knowledge [C3].

C2 Correlation of light and current during the pre-breakdown phase

In this project, which was an Msc project, the pre-breakdown currents and the broadband light emission from the pre-breakdown process were studied [C4]. A propagating streamer will induce currents in the electrode gap, due to the movement of charges during propagation. Simultaneously, light emission occurs. The aim was to ascertain whether there is any correlation between current and light emission, and to try to explain the observations.

The current and light signatures were recorded for different voltages and positions of the triggering laser pulse. A very good correlation was found in most cases, see Fig. C4. From the time delays between laser plasma and breakdown, and by detailed analysis of the signatures, a simple model of the triggered breakdown process was conceived. Although the model explains some of the observations, it still needs further refinement.

C3 Imaging of the breakdown process

This Msc project is aimed at developing the technology necessary to image the breakdown process and employs a laser-shadow system and a video camera [C5]. Imaging of the process is the best way to verify a model and to understand what is actually happening. The object to be imaged was illuminated by a pulsed laser with a pulse length of about 10 ns, thereby effectively freezing the pre-breakdown events. A simple video camera was then used to capture the shadow from the object. Imaging could be achieved with a resolution of 20x20 μm. The first results indicated, as expected, that bubbles are formed after the laser plasma. A MSc project in this area will focus further on imaging.

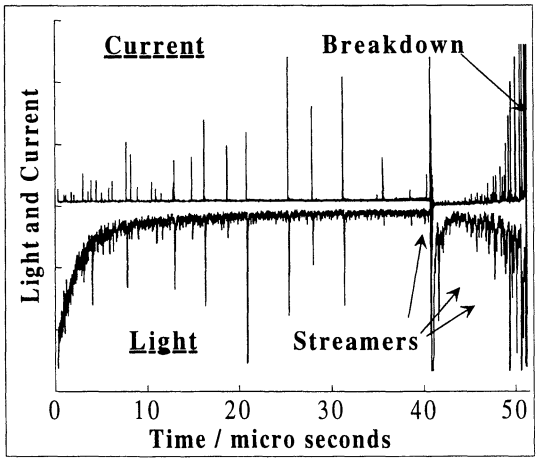

Fig. C4. Current and light emission recording from a laser-initiated breakdown. Positive polarity, 35 kV high voltage.

References

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[A2] W. Persson and W. Wendt, *"Optical spectroscopy for process monitoring and production control in metallurgical industry"*, Mining in Century XXI, Copiapó, August 1993.

[B1] J. Carlsson, P. Hellentin, L. Malmqvist, A. Persson, W. Persson and C-G. Wahlström, *"Time-resolved studies of light-propagation in paper"*, accepted for publication in Applied Optics.

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[C1] A. Sunesson, P. Bårmann, S. Kröll, and L. Walfridsson, "*Laser Triggering of Electric Breakdown in Liquids*", Proc. of the Eleventh Int. Conf. on Conduction and Breakdown in Diel. Liquids (11th ICDL), Baden-Dättwil, IEEE 93CH3204-5, pp.572-576 (1993).

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[C4] J. Bäckman, "*Light and Current Measurements from Prebreakdown Events in Laser-Triggered Electrical Breakdown in Transformer Oil*", Master's project, **LRAP-150** (1994).

[C5] D. Stålhandske, "*Imaging of Laser-Initiated Pre-breakdown Events in Transformer Oil using Laser-Shadow Method*", **LRAP-171**, to appear.

VI Teaching Programme

Professors and lecturers: *Stig Borgström, Lars Engström, Hans Hertz, Gilbert Jönsson, Göran Jönsson, Stefan Kröll, Rune Kullberg, Hans Lundberg, Willy Persson, Rolf Petersson, Sven-Göran Pettersson, Nina Reistad, Sune Svanberg*
Teaching assistants: *Roger Berg, Peter Bårmann, Ulf Elman, Jonas Johansson, Per Jönsson, Peter Kauranen, Claes af Klinteberg, Jörgen Larsson, Lars Malmqvist, Annika Nilsson, Pär Ragnarsson, Lars Rymell, Eva Wallinder, Tomas Starzcewski, Carl Tillman, Raoul Zerne*

A Undergraduate teaching

At the Department of Physics, basic physics teaching is provided for the Schools of Engineering Physics (F), Electrical Engineering (E), Computer Science and Technology (D), Mechanical Engineering (M), Civil Engineering (V), Fire Protection Engineering (BI) and Chemical Engineering (K). Furthermore, specialised courses in *Atomic Physics*, *Laser Physics*, *Advanced Optics* and *Atomic and Molecular Spectroscopy* are given. Courses not included in the regular study programmes are *Holography*, *Radon*, *Medical Laser Techniques* and *Physics for Poets*.

The purpose of the courses in physics is to provide knowledge about fundamental physical concepts, laws, principles, models and their application. Experimental training is an important part of the courses.

The courses in physics are based on lectures, problem-solving sessions and laboratory work. Lectures and problem-solving sessions provide a good basis in fundamental physical principles as well as an introduction to the application of these principles. Laboratory work provides experience in the design of experiments, implementation of theory, experimental methodology and the evaluation of results.

The courses contain both theory (lectures and problem solving) and laboratory practicals. The number of hours devoted to experimental work is, as a rule, about the same as the number of hours spent on theory. During experimental sessions in basic courses, the students generally work in groups of two, and each supervisor teaches four such groups, i.e. 8 students at a time. For the specialised courses, each supervisor can teach only 4 or 6 students at a time, since the amount of equipment is limited. In the courses on atomic and molecular spectroscopy and advanced optics, research equipment is used by the students in their experimental work.

A brief survey of the courses available and the attendance is given in the Table 1. In the first column, the name of the course is given, in the second the school and year, in the third the number of students, in the fourth the number of teaching hours (not including experimental instruction) and in the fifth, the number of hours of experimental work (the figures in parentheses give the number of students in each experimental group.)

Table 1
Courses given by the Division of Atomic Physics, 93/94

Course	*School/year*	*No. of students*	*Hours theory*	*Hours lab.*
Physics course, E	*E1*	215	118	42(8)
Physics course, D	*D1*	92	132	46(8)
Physics course, M	*M1*	161	34	28(8)
Physics course , M	*M3*	118	70	28(8)
Physics, basic course, V	*V1*	98	52	18(8)
Physics, specialised course, V	*V4*	10	20	36(8)
Physics course, K	*K1*	124	68	28(8)
Physics course, BI	*BI1*	30	58	18(8)
Introductory course	*F1*	90	50	36(8)
Waves	*F2*	71	50	40(6)
Atomic Physics	*F3*	58	42	35(4)
Laser Physics	*F4,E3,M3,D3*	60	32	16(4)
Nonlinear Optics	*F3,F4*	13	42	-
Advanced Optics	*F4*	30	36	16(4)
Atomic and Molecular Spectroscopy	*F3,F4*	32	36	30(4)
Holography		20	10	15(10)
Radon		13	20	12(8)
Physics for Poets		38	20	12(8)
Laser Chemistry	*Graduate*	16	38	-
Computers in Measurement Systems	*Graduate*	16	20	-
Nonlinear Laser Spectroscopy	*Graduate*	8	30	-
Medical Laser Techniques		19	24	8

The total number of teaching hours is about 7000. Teaching is performed by two professors, eleven senior lecturers, one junior lecturer, sixteen graduate teaching assistants from the Division of Atomic Physics and about thirty other graduate teaching assistants from other departments at LTH. Most lecturers divide their time between teaching and research.

A1 Basic courses

For students in the School of Engineering Physics, the course *Physics, Extended course* is given. This consists of three parts, Introductory course, Waves, and Atomic Physics, coupled with laboratory practicals. *The Introductory course* comprises experimental methods, general physics, thermodynamics and geometrical optics. The course *Waves* makes the students well acquainted with phenomena in physical optics and acoustics. *Atomic Physics* provides the students with basic knowledge on the structure of atoms and molecules and their properties. The course also gives some orientation in spectroscopic methods in different energy ranges.

For students in the Schools of Electrical Engineering and Computer Science and Technology the basic course *Physics course for E and D* is given. This comprises general physics, thermodynamics, optics, waves and modern physics combined with laboratory work.

For students in the School of Mechanical Engineering two basic courses are given. These consist of general physics with thermodynamics, optics, waves and atomic physics combined with laboratory practices.

For students in the School of Civil Engineering (course V1) and Fire Protection Engineering (course BI1) the basic courses *Physics basic course for V* and B*asic course for BI* are given. These consist of general physics with thermodynamics and fundamental electricity combined with laboratory practices. For students in their fourth year, V4, the *Specialised course* in physics is given, which is directed towards physical measuring techniques.

For students in the School of Chemical Engineering the basic course *Physics course for K* is given. This consists of electricity, wave physics, geometrical optics and nuclear physics combined with laboratory work.

A2 Specialised courses

The specialised course *Laser Physics* is designed to provide the students with knowledge concerning the physical principles of laser physics and to teach them about the most common types of lasers and their most important fields of application. In laboratory practicals the students learn to make simple adjustments and measurements with different types of lasers. This year, the course will be followed by about 80 students from the Schools of Engineering Physics (F), Electrical Engineering (E), Mechanical Engineering (M) and Computer Science and Technology (D).

The specialised course *Atomic and Molecular Spectroscopy* is intended to provide knowledge about modern atomic and molecular spectroscopy with special emphasis on technical applications. About 20 students follow this course. Together with the laser physics course, this course forms the natural introduction to graduate studies at the Division.

A course in *Holography* is also available to those interested in photography, imaging techniques and optical measurements. The course starts with lectures in geometrical optics and wave optics and, together with laboratory sessions, the fundamentals of holography and related topics are discussed and different types of holograms are made.

A specialised course in *Advanced Optics* has been established at the Division. This course, emphasising Fourier optics, interferometry, fibre optics, holography and phase-conjugation techniques, was given for the first time in the autumn of 1985.

The specialised course Medical Laser Techniques was also given in the academic year 1993/94.

The graduate course in *Non-linear Optics* was in the spring 1994 for the first time also open for the undergraduate students as an elective course. It will in the future be jointly given biannually for undergraduate and graduate student. This theoretical course is based on the first five chapters in R.W. Boyd's book "Nonlinear Optics" and treats non-linear susceptibilities, its symmetry properties and quantum-mechanical derivation, the wave-

equation description of non-linear optical interactions and non-linear optics in two-level systems.

A course in *Radon* was given for the first time in 1992. Measuring techniques as well as physiological aspects are included in the course.

The course *Physics for Poets* was given for the first time in 1992, in collaboration with the Department of Mathematical Physics.

A3 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.

Johan Anderberg	*Scanning Knife-edge Method for Laser Plasma Size Measurements, LRAP-165*
Anders Arvidsson	*Transmission of Light into Packs*
Magnus Berglund	*Distance Control in Trapped Particle Optical Microscopy, LRAP-161*
Joakim Bood **Hugo Karlsson**	*Raman and Infrared Spectroscopy for Tissue Diagnostics, LRAP-168*
Jonas Bäckman	*Light and Current Measurements from Prebreakdown Events in Laser-triggered Electrical Breakdown in Transformer Oil, LRAP-150*
Carina Ekelius **Jan Svensson**	*An Optical Transmission Method for Continuous Measurement of Blood Volume Changes during Haemodialysis, LRAP-166*
Magnus Johansson	*Fibre Amplifiers in Digital Point-to-Point Links*
Charlotta Lindquist	*Numerical Diffusion Modelling of Light Propagation in Turbid Media for Medical Diagnostics, LRAP-157*
Peter Lindskog	*Detection of Light Distribution in a Package*
Richard Lloyd **Sven Gunnarsson**	*Erbium-Doped Fibre Lasers*
Claire Lyngå	*Studies of a Laser-produced Carbon Plasma with Spatial, Temporal and Spectral Resolution, LRAP-164*
Åsa Persson	*Optical Diagnostics of Tissue Pathologies Based on Elastic Scattering Properties, LRAP-169*
Jonas Sandsten	*Optical Methods for Detection of Very Small Vibrations and Design of a Laser Diode System using Optical Feedback, LRAP-163*
Christian Sturesson	*Theoretical Modelling of the Temperature Distribution in Laser-induced Hyperthermia, LRAP-159*
Dag Tyllered	*Theoretical and Experimental Investigation of Hydrogen Peroxide Condensation on Packaging Materials*

B Graduate teaching

The graduate *Non-linear Optics* course was given in the spring 1993. It was then in 1994 open also for undergraduate students, see section A2 in this chapter.

A course in *Optical Quantum Electronics*, based on the book "Lasers" by A.E. Siegman was given for the first time. This course attracted 16 graduate students in its first version. Starting in spring 1995 it will be given biannually for graduate and undergraduate students.

A new course on *Medical Laser Physics* was given in collaboration with the Lund University Medical Laser Centre. A number of graduate physics students and clinically active researchers attended the course, which reviewed basic physical and medical aspects of lasers in medicine. Two laboratory exercises and study visits were included in the programme.