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Lund Reports on Atomic Physics
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Introduction

The Division of Atomic Physics, Lund Institute of Technology (LTH), is responsible for the basic physics teaching in all subjects at LTH and for specialised teaching in Optics, Atomic Physics, Atomic and Molecular Spectroscopy and Laser Physics. The Division has research activities in basic and applied optical spectroscopy, to a large extent based on lasers. It is also part of the Physics Department, Lund University, where it forms one of eight divisions. Since the beginning of 1980 the research activities of our division have been centred around the use of lasers. The activities during the period 1980 - 1990 have been described in previous Biennial Progress Reports (Lund Reports on Atomic Physics LRAP-20, LRAP-43, LRAP-85, LRAP-90 and LRAP-119). During the last two years the research programme has been further developed.

A number of important developments have occurred during the last two years. Within the concept of the "Lund Laser Centre" the activities within basic and applied laser physics and spectroscopy are expanding, with the recent establishment of a new professorship and research division in Combustion Physics. This division, under the leadership of Prof. Marcus Aldén, also forms the core of the Lund Combustion Centre (FTC). The Centre suffered a great loss with the death of its former director, Prof. Thure Högberg on August 11, 1992. Till the end he remained very active and was a great source of inspiration and knowledge. With the successful transfer of personnel and projects to the new division, the Atomic Physics Division has terminated its long-standing active research in the combustion field, and activities in this field are now reported separately. We wish the new division all success and look forward to continuing good collaboration.

The Lund University Medical Laser Centre was officially established in October 1991 as an organisation for the co-ordination of research and teaching in the medical laser field at 4 faculties. This is the third centre of a collaborative nature formed with the active engagement of the Atomic Physics Division. It follows the formation of the Combustion Centre (FTC) in 1986 and the Environmental Measurement Technology Centre (CENTEC) in 1990. The medical laser physics group at our division is now expanding to match the many collaborative projects emerging.

During the last 2 years our contacts with the Chemical Centre at Lund University have also developed, partly because of a graduate course on Laser Chemistry with about 10 students also completing special projects and an additional number of chemists following the lecture course on a non-credit basis. This has resulted in joint research projects with the Division of Applied Biochemistry and the Division of Physical Chemistry 1. We are also involved in a fruitful collaboration with the Division of Inorganic Chemistry 2 in the field of crystal growth for novel optical storage materials.

The most important achievement during the period of the present Progress Report is the completion of the Lund High-Power Laser Facility, which is operated by the Atomic Physics Division. This project was funded by 7 MSEK from the Knut and Alice Wallenberg Foundation and 5 MSEK from the Swedish Council for Planning and Co-ordination of Research (FRN). Further, the university provided internal funding for a thorough reconstruction of premises in the Physics Department. The facility was inaugu-

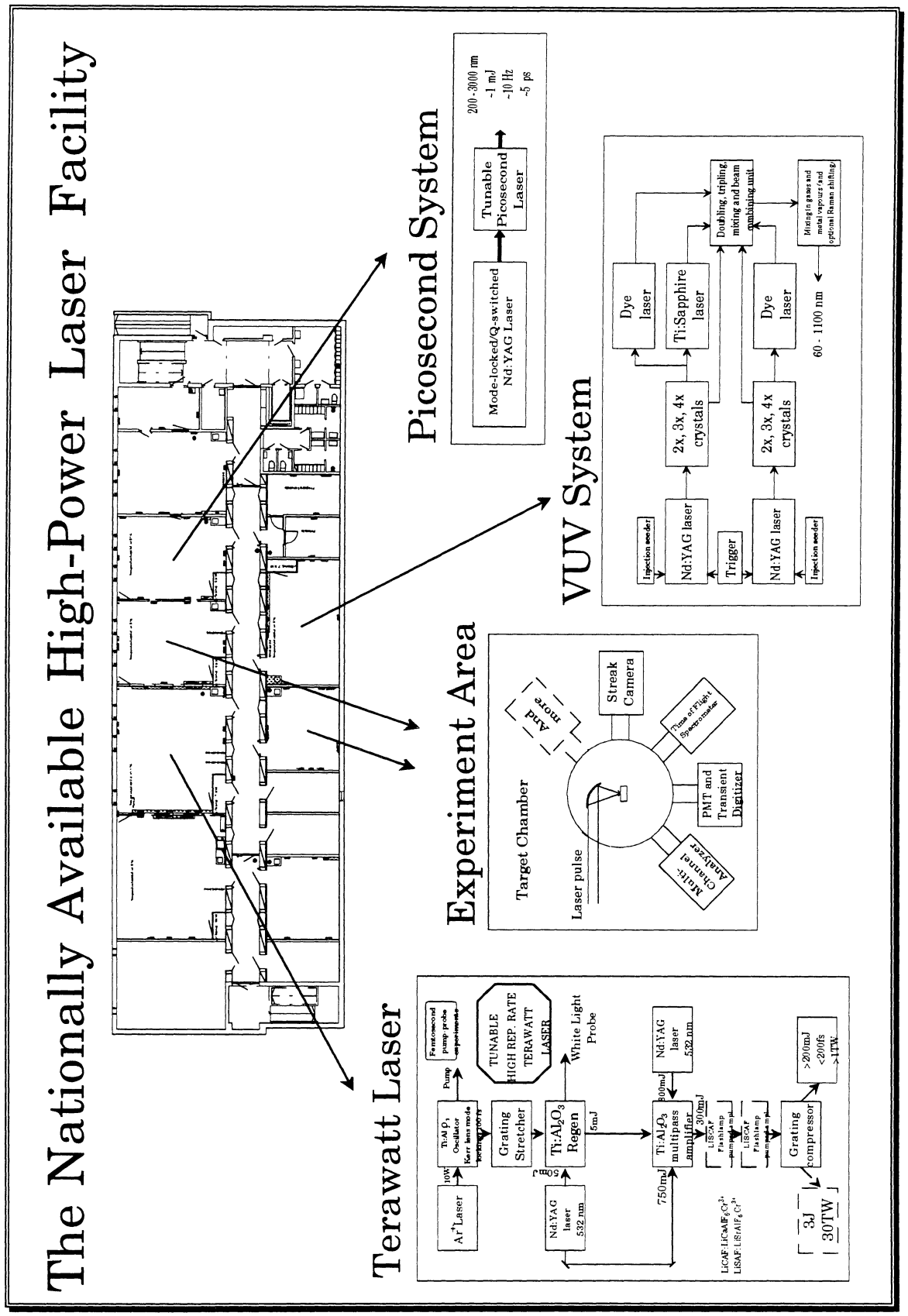

Fig. 1.
Layout of the high-power laser facility

rated on October 22, 1992 in conjunction with a two-day Symposium, on "High-Power Lasers and their Applications". The layout of the facility, which is nationally available, is shown in Fig. 1. The new facility provides excellent research opportunities primarily in basic atomic and molecular physics, but also in chemistry, biology and medicine.

The most exciting of the new laser systems is a terawatt laser based on chirped pulse amplification in titanium sapphire. The system provides 150 fs pulses of terawatt power, delivered at 10 Hz, with a tunability from 760 to 840 nm. A general layout of the system, which was developed by Continuum Inc. in close collaboration with the Atomic Physics Division, is shown in Fig. 2. An argon-ion-laser-pumped femtosecond titanium sapphire laser featuring self-mode-locking provides initial 100 fs long pulses which are stretched by a factor of about 2500 in a grating stretcher with a temporal coding of the different colour components in the Fourier-broadened pulse. One of the stretched pulses is injected into a regenerative titanium sapphire amplifier, which is pumped by 532 nm radiation from a 10 Hz Nd:YAG laser. After 12 double passes and an amplification of about 10^6 the pulse is electro-optically ejected from the cavity and enters the final amplifier, which is pumped by two high-energy frequency-doubled Nd:YAG lasers. After 4 passes of the final amplifier a stretched pulse with a pulse energy of about 400 mJ has been attained. The beam is expanded to 50 mm diameter and is passed through a grating compressor, where the pulse is compressed to almost its initial pulse length. Typically 200 mJ in a

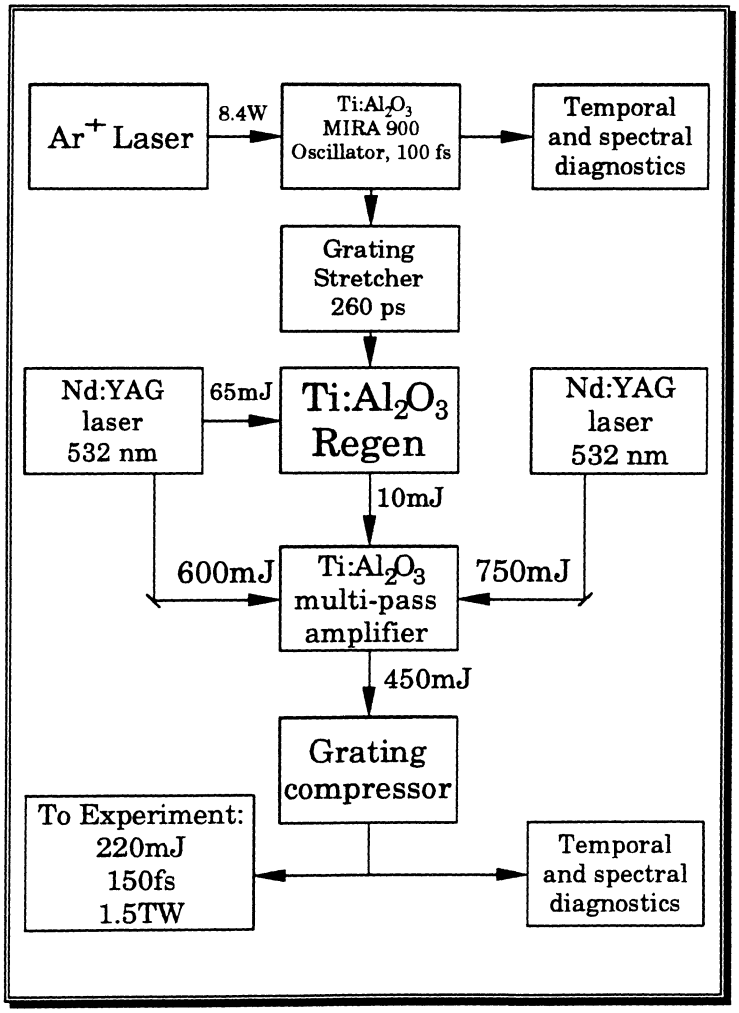

Fig. 2.
Layout of the terawatt system

pulse of about 150 fs duration can be achieved. Two target chambers for terawatt laser experiments have been constructed. Apart from using the high-power pulse, the output from the oscillator alone is very useful for applications, for example, in biology and medicine.

Another laser system is available consisting of two large-scale Nd:YAG lasers pumping dye and titanium sapphire lasers, providing ns widely tuneable pulses. A VUV generator based on resonant four-wave mixing in krypton is used to provide tuneable radiation in the 110-200 nm range. The spectral brightness of these pulses exceeds that attainable from synchrotron radiation sources by many orders of magnitude providing new possibilities for atomic and molecular spectroscopy in a little-studied spectral region.

The third laser system at the new facility provides widely tuneable pulses of 10's of ps duration at a repetition rate of 10 Hz. This is a very versatile system, especially in conjunction with a new streak camera with a 2 ps time resolution.

After this brief sketch of the developmental trends related to the Atomic Physics Division, our ongoing research activities will be briefly outlined.

With the availability of the new terawatt laser system our basic atomic physics research programme is now being focused on high-power laser physics. Experiments on high harmonic generation in inert gases are being pursued in collaboration with CEA Saclay, and are aimed at an understanding of the atomic response as well as the collective radiation with phase-matching considerations. Also, the picosecond laser system is being used for studies of the temporal behaviour of the harmonic generation process. Dense plasmas are being formed by focusing terawatt laser radiation to small spots on solid materials such as tantalum. The plasma produces intense X-rays with energies extending towards the MeV region. The radiation is now being characterised with nuclear physics techniques. At the same time, the small X-ray source is being used for medical imaging with the potential of creating extremely sharp images. This research is being pursued in collaboration with the Department for Diagnostic Radiology at the Lund University Hospital.

Preliminary experiments on plasma spectroscopy and X-ray laser pumping have also been initiated. Much of the classical emission spectroscopy and term analysis work which has long been performed within the Division is expected to be related to the new plasma light source. The basic atomic physics group in Lund is a member of three newly proposed European networks on high-power laser physics; High Harmonic Generation, Broad-band X-ray Generation and X-ray Laser Pumping.

During the last two years, the basic atomic laser spectroscopy group has been much involved in spectroscopy at short UV and VUV wavelengths. Radiative properties of several atomic and ionic species have been measured. Of particular interest are oscillator strengths of transitions in arsenic and ruthenium ions recently observed in celestial objects using the Hubble Space Telescope. The new VUV laser source, giving access to shorter wavelengths, is presently being used in studies of Rydberg sequences in copper and magnesium. A new technique using the pulsed Hanle effect has been developed and provides new means of measuring short excited-state lifetimes. High-resolution laser spectroscopy using continuous wave single-mode laser sources has led to the first

experimental demonstration of tandem high-contrast transmission spectroscopy and to increased experience in using diode laser sources in atomic physics.

In the field of theoretical atomic physics, work has been focused on the application of the multi-configuration Hartree-Fock method for the calculation of radiative properties and hyperfine structure. The theoretical work is performed in collaboration with Prof. C. Froese Fischer, Vanderbilt University and with the Quantum Chemistry Group, Lund University.

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 concept for optical storage and processing. Theoretical calculations performed so far demonstrate that storage densities of tens of Gbits/cm^2 with THz reading and writing rates are possible from a signal-to-noise point of view. All optical, logic 'and' operations and optical pulse compression have been demonstrated using photon echoes.

Novel methods for high-resolution microscopy are being developed. The methods are primarily intended for living biological samples but materials science applications are also important. A compact soft X-ray microscope based on a new debris-free laser-produced plasma X-ray source is being designed. Using optically trapped particles a non-intrusive scanning probe optical microscope, which has potentially better resolution than the diffraction limit, is being developed. Both methods share the common goal of developing small, reasonably priced microscopes suitable for use in research.

Environmental remote sensing using optical techniques is another applied spectroscopy project in progress. The programme includes the development of new optical measurement techniques for air pollution monitoring and field experiments to facilitate the transfer of technology to practical applications in industry and air quality management. Our mobile lidar system has been extensively used for range-resolved mapping of atomic mercury, sulphur dioxide and nitrogen dioxide. The technique has been used in operational measurements jointly with the Swedish Environmental Protection Agency at major Swedish industries. Other aspects of the work are the geophysical ones. Extensive measurements on the Italian volcanoes Etna, Stromboli and Vulcano have been performed in collaboration with Italian researchers. The Swedish lidar system was used onboard the Italian research vessel "Urania", to quantify the total flux of SO_2 from the volcanoes. Geothermal emissions of atomic mercury have also been successfully studied. Differential absorption spectroscopy (doas) using diffuse sky radiation was also used to quantify volcanic fluxes. The doas technique using artificial continuous wave light sources is being used for urban monitoring over the city of Lund. The spin-off company OPSIS AB, operated by two former graduate students from our division, has successfully commercialised the doas technique.

A fluorescence lidar system has been used for remote monitoring of vegetation and water. Our vegetation project is part of the LASFLEUR co-operation within the European EUREKA frame. A new technique for remote multi-colour fluorescence imaging of vegetation has been developed. Field tests were performed in the Botanical Garden of the Karlsruhe University and at DLR, Oberpfaffenhofen. Marine fluorescence monitoring has been performed in collaboration with Italian research institutes with field tests at Trieste and the Venice Lagoon.

During the last two years our activities in the medical field have further increased and are now part of the research activities of the Lund Medical Laser Centre. Tumour fluorescence diagnostics utilising the haemoglobin precursor δ-amino levulinic acid (ALA) has been developed and has been shown to be very powerful. By using the bleaching properties of ALA a new type of dosimeter seems possible. Clinical tumour detection has been carried out jointly with the departments of Oncology, Urology, Neurosurgery and Lung medicine. The group has also participated in extensive tumour therapy using photo-dynamic techniques. Fluorescence diagnostics has also been applied to the diagnostics of heart disease. A spin-off company (SPECTRAPHOS AB) is bringing the technique into the clinical praxis.

Time-gated transillumination imaging through tissue has also been performed. By using the light travelling in a straight path through the tissue it is possible to suppress scattered light which normally blurs the image. Extensive laboratory scattering experiments using picosecond techniques have been performed and theoretical calculation codes have been developed to provide a better understanding of tissue scattering. Further, successful measurements with a pulsed diode laser have been performed. The technique will soon be used in clinical tests in optical mammography in collaboration with the Department of Diagnostic Radiology, Lund University. Time-resolved photon migration studies have also been performed on leaves in collaboration with the Department of Plant Physiology, Lund University, and on paper in collaboration with TFL, the Swedish Newsprint Research Centre, Stockholm.

Further aspects of the optical characterisation of paper are being studied in an industrial collaboration with TFL. In a further spectroscopic project of industrial interest optical diagnostics techniques in connection with ore smelting and processing are being developed. The techniques could have a major impact on the metal industry. Geogas studies including mercury vapour monitoring are also being pursued with an interesting potential for mineral prospecting. Through a spin-off company (SEMTECH AB) in which division members are participating, the new techniques can be made practically available.

In a new industrial collaborative project jointly with ABB Västerås, insulation properties of oils are being studied. Laser-induced breakdown in the insulating fluid is used to trigger discharges in high-voltage devices. Various optical techniques will be used to increase the understanding of the origins of electric breakdown.

In our report series "Lund Reports on Atomic Physics" (LRAP) material which is not published in international journals is presented. The reports include dissertations, diploma papers and special investigations. So far about 140 papers have appeared in the LRAP series. At the end of the period covered by this progress report the staff of the Division totalled about 50. This number includes 17 graduate students and 13 supporting personnel. In addition a number of diploma students perform their projects within our division every year. It is through the dedicated work of these people that the research and teaching accomplishments reported here have been made possible.

We are very grateful for the support of a large number of funding agencies, in particular the Swedish Natural Science Research Council (NFR), the Swedish Research Council for Engineering Sciences (TFR), the Swedish National Board for Industrial and Technical Development (NUTEK), the Swedish Space Board (RS), the Swedish Environmental

Protection Board (SNV), the Swedish Cancer Society (RmC), the Swedish Medical Research Council (MFR) and the Knut and Alice Wallenberg Foundation.

Special thanks are due to Doc. Stefan Kröll, 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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Mr Stefan Andersson (ABB Corporate Research, Västerås)
Dr Viatcheslav Avetisov, General Physics Institute, Moscow
Mr Philippe Balcou (CEA Saclay)
Dr Uldis Berzinsh (University of Latvia, Riga)
Dr Adam Dubik (Institute of Plasma Physics and Laser Microfusion, Warszawa)
Mr Knut Hansen (Kiel University)
Dr Anne L'Huillier (CEA Saclay)
Mr Luo Caiyan (Wuhan University)

Prof. Ma Baozhang (Shanghai Medical University)
Ms Joanna Muffet (Imperial College, London)
Mr Pascal Salières (CEA Saclay)
Dr Wolfgang Schade (Kiel University)
Dr Wang Dadi (Academy of Sciences, Beijing)
MK Ingrid Wang (University of Bergen)
Prof. Yang Yanlong (Fudan University, Shanghai)

PhD Theses

Håkan Bergström	1991-11-08	Time-resolved and Doppler-reduced laser spectroscopy on atoms, Lund Reports on Atomic Physics, LRAP-125
Lennart Sturesson	1992-06-05	The hyperfine structure - a message from the inner circle, Lund Reports on Atomic Physics, LRAP-129

Licenciate Degrees

Jörgen Larsson	1991-12-12	High-resolution spectroscopy in the UV/VUV spectral region, Lund Reports on Atomic Physics, LRAP-127
Per Jönsson	1992-11-06	Theoretical and experimental investigation of atomic radiative lifetimes and hyperfine structures, Lund Reports on Atomic Physics, LRAP-134
Jonas Bengtsson	1992-11-06	Time-resolved laser spectroscopy in the UV/VUV spectral region, Lund Reports on Atomic Physics, LRAP-135

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

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P. Jönsson	L. Malmqvist	(J. Johansson)	J. Johansson	(H. Hertz)
(S. Kröll)	L. Rymell	P. Ragnarson	A. Nilsson	P. Kauranen
J. Larsson	*U. Elman*	E. Wallinder	I. Wang	(S. Kröll)
H. Lundberg	*H. Stattin*	S. Wallin	Yang Yuanlong	Le. Malmqvist
Luo Caiyan	*P. Tidlund*	*K. Ljunggren*	*U. Gustavsson*	W. Persson
A. Persson		*G. Sandberg*	*C. af Klinteberg*	A. Sunesson
(W. Persson)		*K. Svensson*		(C.G. Wahlström)
S.G. Pettersson				W. Wendt
T. Starczewski				*P. Bårmann*
L. Sturesson				
C. Tillman				
C.G. Wahlström				
R. Zerne				

Names in parenthesis are also given under their main activity heading

35 Research Personnel (14 Ph.D., 18 Grad. Student, 3 Long term Visitors)
16 External Positions
9 Diploma Workers
Total Personnel, Div. of Atomic Physics: 51

I Basic Atomic Physics

During the past two years, a great deal of time has been devoted by members of the Basic Atomic Physics Group to the establishment of the new, nationally available, High Power Laser Facility. This facility, including a high repetition-rate, tunable terawatt laser, has been described in the general Introduction. The first funding was obtained in December 1990, and the facility was inaugurated in October 1992. Two symposia on high-power lasers and their applications have been arranged. The first symposium was held during the planning stage of the facility (May 1991) and the second in connection with the inauguration. In section A some of the planned research activities using the very high-power, ultra-short pulses are described. Some of the first results are also presented.

In spite of the time consuming work associated with the new facility, the existing activities in time-resolved and high-resolution laser spectroscopy as well as emission spectroscopy have continued. The laser spectroscopic programme has been increasingly focused on measurements of astrophysical interest. Spectroscopic data from the Hubble Space Telescope have given rise to a renewed need for laboratory data, in particular time-resolved information on various ions. In general, this means short excitation wavelengths and short radiative lifetimes. New techniques for ion production as well as spectroscopic measurements have been developed and will be presented in section B.

The research programme in theoretical atomic physics has continued undisturbed by the establishment of the new facility. Major computer program developments have been carried out and important computational results obtained. In June 1992 a workshop on theoretical atomic physics was arranged to highlight the current activities and trends. The ultra-high intensities now available with the terawatt laser, produce the kind of interactions between laser radiation and matter that cannot be treated by conventional perturbation theory. New theoretical methods and computational techniques need to be developed to treat effects such as high harmonic generation and multi-photon ionisation. During the coming two-year period we can therefore expect a gradual change in emphasis in the theoretical activities of the group.

The work in the basic atomic physics group has resulted in two PhD theses [1,2] and three Licentiate theses [3,4,5] during the past two-year period. Much of the work has been presented at international conferences on atomic physics and spectroscopy [6-17]. To gather more information and expertise related to the new activities we are involved in, or about to start with the new facility, international collaborations have been encouraged. Members of the group have spent periods abroad, in the USA, England, Italy, France and Belgium. Scientists from England, Poland, France, China and Germany have visited the group and some have spent extended periods of time at our labs.

A Atomic physics with high-power laser radiation

The establishment of the Lund High Power Laser Facility has opened the doors for new areas of research. Access to extremely high optical field strengths allows investigations of multi-photon ionisation, above-threshold ionisation, high harmonic generation and photo fragmentation of molecules, for example. The very short (<200 fs) pulses from the terawatt laser also permit the formation and study of super-dense plasmas, incoherent X-ray radiation from such plasmas and new schemes for X-ray laser pumping. In parallel with the establishment of the experimental facility, preparations have been made to enter the new fields of research being planned. In particular, a large number of international contacts and scientific collaborations have been established. Three networks regarding X-ray lasers, Harmonic generation and X-rays from laser-produced plasmas have been formed. Two of these have just been accepted as European networks with financial support from the EC under the *Human Capital and Mobility Programme.*

A1 High harmonic generation

Philippe Balcou, Anne L'Huillier*, Jörgen Larsson, Anders Persson, Pascal Salières* Tomas Starczewski and Claes-Göran Wahlström*

**Visiting scientists*

If laser radiation is focused to a sufficiently high intensity in a noble gas, odd harmonics of the laser frequency are generated. When the incident laser is intense and of a short pulse duration, the harmonics are found to be approximately constant in intensity up to a very high order. This "plateau" behaviour is characteristic of a strong (non-perturbative) laser-atom interaction. Consequently, high-order harmonics (short-wavelength coherent radiation) can be generated with reasonable efficiencies. This radiation emission in the vacuum ultraviolet and soft X-ray region has unique properties of coherence and brightness. Its pulse width follows that of the pump field [A1] and can therefore be very short (e.g. <200 fs with our titanium sapphire system). With a tuneable pump laser, and by the selection of different harmonic orders, the entire UV, VUV, XUV and soft X-ray regions can be continuously spanned.

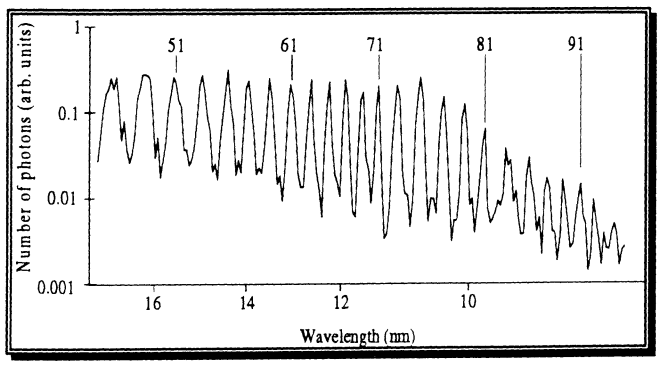

Fig. A1
Harmonic spectrum generated in Ne using the new terawatt laser. The spectrum shown is not corrected for detector sensitivity.

Many aspects of the fundamental physical processes involved are still not fully understood and need to be further investigated. Our aim is to reach a good understanding of these processes, to characterise and to optimise the generated radiation and to explore some of the applications.

Using our new picosecond laser, we have studied up to the 35th harmonic (30.4nm) in Kr. The limit in harmonic order in this case was set by the resolution of the normal

incidence spectrometer used. With the terawatt laser, much higher intensities are obtainable due to the much shorter pulse length. In a collaboration with a group from Saclay, France, using their specially designed grazing incidence spectrometer and our terawatt laser, we have investigated generation of harmonics up to the 93rd (8.5 nm, 145 eV), and their characteristics. In particular, the intensity dependence of the harmonics, their blue shifts due to the ionisation of the non-linear medium and contributions from the produced ions were studied [A2]. The limit in harmonic order, in this study, was set by the short-wavelength reflectivity cut-off for the grating used.

A2 Incoherent X-rays from laser-produced plasmas

Stig Borgström, Sune Svanberg, Carl Tillman and Claes-Göran Wahlström

Focusing the pulses from the terawatt laser onto a solid target can produce extremely dense, hot plasmas. We have performed experiments where the beam has been focused with an f/1 off-axis parabolic mirror onto a rotating Ta target. The focal spot in this case is only a few micrometres in diameter, and the intensity extremely high, ~10^{18} W/cm^2. The plasma produced in this interaction becomes very dense since there is no time for it to expand during the laser pulse. An essentially solid density plasma is created. This plasma radiates incoherent radiation with photon energies up to several hundred keV, in the form of a short pulse. The short duration, and the point-like dimensions makes this an extremely interesting source of hard X-rays. Imaging, including extreme X-ray enlargements, and pump probe experiments are examples of possible applications. In our first set of experiments we have obtained sharp, magnified, X-ray images of various objects. At the same time, the energy distribution of the emitted radiation is being studied as functions of target material, laser intensity, etc. In this project we are collaborating with experts from the Department of Nuclear Physics and the Department of Diagnostic Radiology, Lund University.

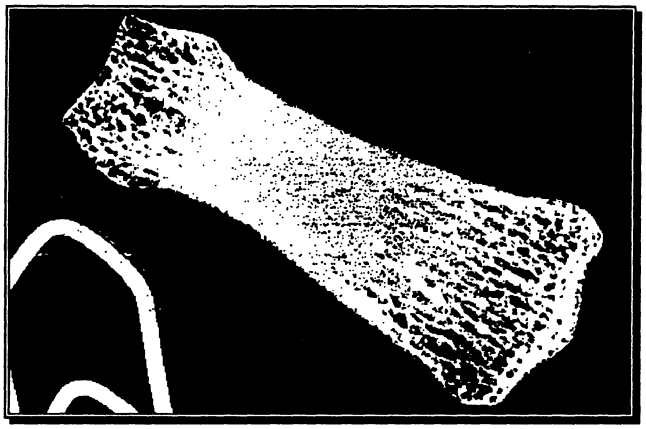

Fig. A2.
X-ray image of a composite target consisting of a human finger bone and a metallic paper clip embedded in 2cm of animal tissue.

A3 X-ray laser pumping

Stig Borgström, Willy Persson, Tomas Starczewski, Sune Svanberg and Carl Tillman

The study and development of soft X-ray lasers is a very active field of research worldwide. This effort can be divided into three major approaches: collisional, recombination, and photo-pumped lasers. The first approach has, so far, been the most successful of the three, and consequently the most frequently used. However, this approach normally requires a large amount of energy in the pump laser pulses and hence large-scale facilities with normally very low repetition rates.

Recombination pumped schemes, on the other hand, may be feasible with substantially less energy. Recent theoretical calculations show that sub-picosecond lasers, such as our terawatt laser, may be able to pump a new class of recombination X-ray laser where the initial plasma is produced by multi-photon ionisation.

The third approach, photo-pumping, has not been much studied. However, with access to ultra-high intensities in the 100 fs regime, this may become a feasible approach to high-repetition-rate X-ray lasers.

We are currently performing the first experiments on laser-produced plasmas from solid as well as gas targets. As soon as all the diagnostics equipment has been tested we intend to study different recombination and photo-pumped schemes with various excitation geometries.

We also intend to continue the long Lund tradition in atomic spectroscopy on highly ionised atoms, focusing on transitions and recombination schemes relevant to X-ray laser pumping. These spectroscopic studies are performed in collaboration with members of the Department of Atomic Spectroscopy here in Lund.

B Laser-spectroscopic investigations of atomic and ionic excited-state lifetimes 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 natural lifetimes of ions, atoms and molecules. Such data can, together with branching ratio measurements 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. Natural lifetimes are also used to verify theoretical models of atoms and ions. A very direct way of measuring lifetimes is to perform laser excitation using a short laser pulse and subsequently study the fluorescence light decay as the excited ensemble of atoms/ions relax to a lower lying state.

In 1990 the Hubble Space Telescope (HST) was launched. Using the Echelle spectrometer mounted on the HST, spectrally resolved observations of UV/VUV light, which cannot penetrate the atmosphere and reach earth-bound telescopes, have been made possible. 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 UV/VUV radiation must be generated, the duration of the laser pulse should be shorter than the excited state lifetime of the studied atom/ion. The strong lines observed in stellar spectra are often lines connecting a short-lived ionic excited state to the ground state. To measure the lifetimes of such states, either laser pulses of short duration must be produced or methods which do not rely on short-pulse excitation can be employed. Lifetimes much shorter than the duration of the laser pulse can be deduced by measuring the line-profile width using the pulsed Hanle method and then evaluate the lifetime from the Heisenberg uncertainty principle. Methods that use both the above mentioned strategies have been demonstrated and applied to produce data of astrophysical interest.

Another issue which has been addressed is finding general-purpose methods of producing the free ions and atoms to be studied. To obtain oscillator strengths both lifetime values and branching ratios must be known. At the Atomic Spectroscopy Division branching ratio measurements can be performed with high accuracy using a Fourier Transform Spectrometer (FTS). A collaboration between our two divisions has been established.

B1 Lifetime measurements on atomic nitrogen using a DFDL laser

Jonas Bengtsson, Knut Hansen, Jörgen Larsson, Wolfgang Schade* and Sune Svanberg*

**Visiting scientists*

Light non-metallic elements are important in numerous atmospheric, combustion and plasma processes and in astrophysics. They are also of considerable interest for comparisons between experiment and theoretical calculations, which tend to be particularly accurate for light atoms. Many of the elements of interest are not readily available as free atoms.

In order to obtain free atoms of light species such as nitrogen and oxygen some dissociation scheme for molecules must be used. Photo dissociation and two-photon excitation of an excited atomic state can be performed with the same intense laser pulse. Following this three-photon process, another laser will allow studies of higher-lying states. This scheme was first used by Kröll et al. (*Phys. Rev. Lett. 55, 284 (1985)*) for studies of neutral oxygen and has recently been used for determinations of lifetimes in nitrogen [B1]. The measured lifetimes are shortened by collisions with neighbouring molecules and atoms in the interaction region. By varying the gas pressure, the true lifetime and quenching rates can be determined from a Stern-Vollmer plot (zero pressure axis intercept and line slope, respectively). Some of the states in these atoms are quite short-lived. The collision-quenched lifetimes may be even shorter than the output pulse of a conventional dye laser. In a recent study [B2], we therefore used a short-pulse distributed feedback dye laser (DFDL) for the second-step excitation. Such lasers have been shown to produce pulses shorter than 1 ns and a spectral width close to the Fourier limit, matching typical linewidths of free atoms The atomic production and excitation scheme used for nitrogen in this experiment is applicable for many other light atoms, such as C, F, Cl, P and S.

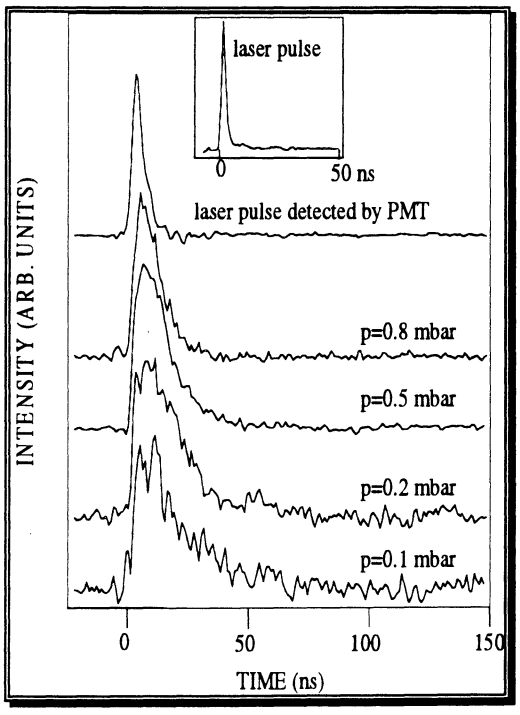

Fig. B1.
Recording of the fluorescence light decay from the $2p^2\ 5s\ {}^4P_{5/2}$ state of neutral nitrogen at different residual N_2O pressure. The laser pulse as detected by a fast diode and the detection system response to this pulse are also shown.

B2 Lifetime measurements on atoms and ions using short-pulse VUV radiation

Jonas Bengtsson, Uldis Berzinsh, Jörgen Larsson, Hans Lundberg, Anders Persson, Sune Svanberg, Claes-Göran Wahlström and Raoul Zerne

There is currently a special interest for accurate measurement of the transition probabilities of the 194 nm lines in ruthenium ions (Ru II) and atomic arsenic (As I), since these lines have just been observed by the Hubble Space Telescope. Methods of measuring short lifetimes were demonstrated on the resonance lines of atomic selenium (Se I) and tellurium (Te I) [B3, B4] and later applied to As and Ru II [B5, B6]. No previous measurements of the Ru II lifetimes exist. For As, Se and Te, beam-foil measurements have been reported previously. All states in these studies have lifetimes of about 3-5 ns. The excitation wavelengths are in most cases in the VUV region. To generate the short pulses, with a peak power high enough to allow for frequency mixing, two pump sources

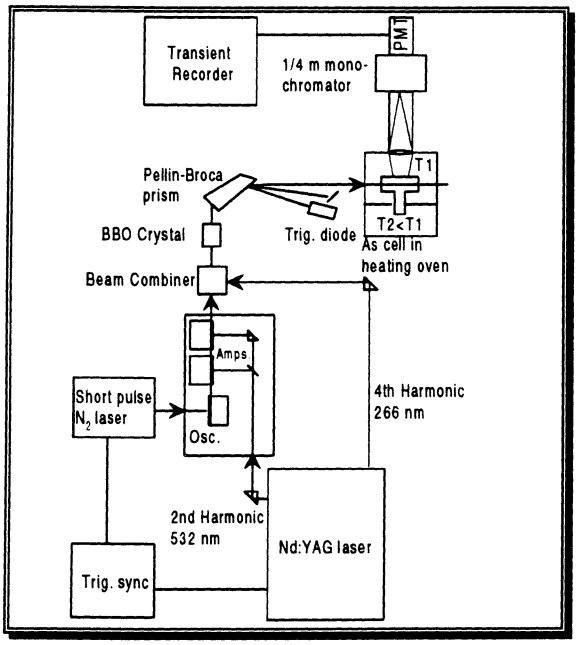

Fig. B2.
With two similar set-ups the natural lifetimes of short-lived excited states in arsenic, selenium and tellurium were measured.

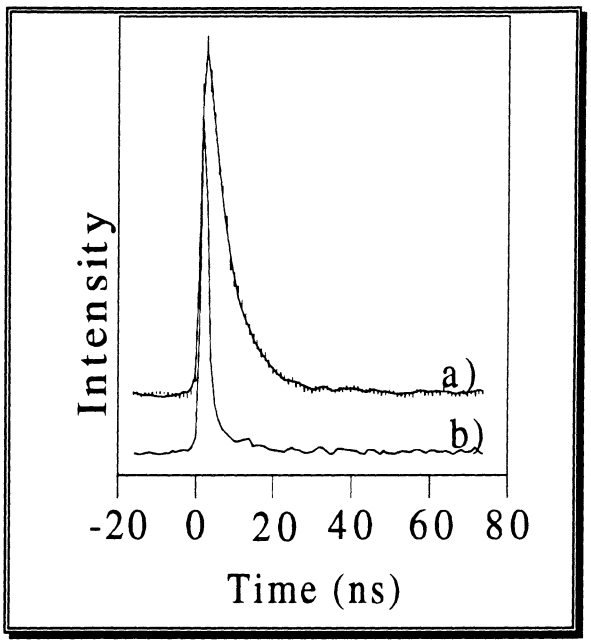

Fig. B3.
The natural lifetime of the $4p^2 5s\ ^4P_{1/2}$ state of arsenic was obtained by fitting a convolution of the recorded laser pulse and an exponential to the recorded fluorescence.

were used. A short-pulse nitrogen laser pumped the dye laser oscillator. The low-power output was then sent through dye amplifiers pumped with the second harmonic of a powerful Nd:YAG laser.

In order to produce free atoms and ions, two different methods were used. Ruthenium is a brittle metal which evaporates at 2300 K. We thus chose to generate Ru II in a laser-produced plasma. As a target we used a powder, confined in a metallic container. To maintain a smooth target surface, the metallic container was positioned on a loudspeaker in a vacuum system. By applying the resonance frequency of the loudspeaker, the powder was shaken and the crater created by the focused Nd:YAG laser was smoothed out before

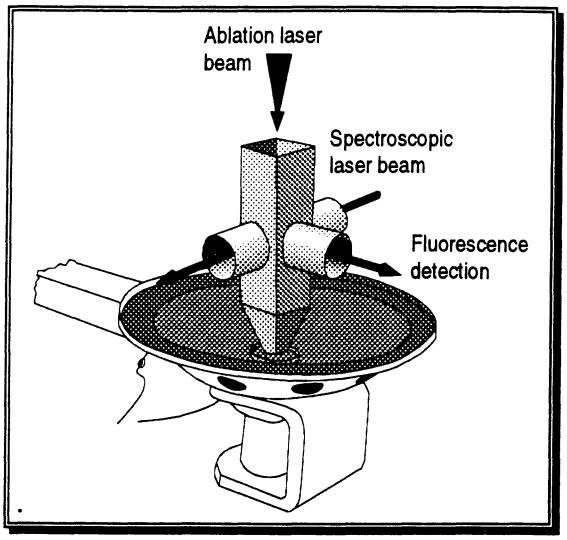

Fig. B4.
Set-up for generating free Ru II ions in a laser-produced plasma. A powder target was confined in a container mounted on a vibrating loudspeaker to keep the surface smooth.

the next laser pulse was applied 100 ms later. Laser excitation of Ru II on the resonance lines in the expanding plasma was performed.

The other elements studied evaporate at considerably lower temperature, but as molecules. In these investigations, a small quartz cell containing the element to be studied was differentially heated to obtain thermal dissociation of the molecules at low vapour pressure. The free atoms or ions were selectively excited using pulsed laser radiation. Pulsed laser radiation in the VUV/UV region was generated by different schemes of frequency mixing. Radiative lifetime values were evaluated from fluorescence decay curves. The measured lifetimes were combined with branching ratios to obtain oscillator strengths for the transitions. Branching ratios in Ru II have been measured by Litzén and co-workers at the Atomic Spectroscopy Division and are under evaluation. For the other elements branching ratios from the literature were used. Investigations on silver and ytterbium using more established experimental techniques have been reported [B7, B8].

B3 Lifetime measurements on atomic magnesium using the pulsed Hanle effect

Jörgen Larsson and Sune Svanberg

Radiative properties of the neutral magnesium atom have been extensively studied during recent years. The motivation behind these studies has been its prominent astrophysical abundance and the possibility of comparing experimental data with theoretical calculations on this relatively light and tractable atom. The resonance line at 285.2 nm has been studied in many investigations. The aim of a recent study was to extend data for this sequence by making an accurate Hanle effect determination of the lifetime for the 3s4p $^1P_1$ state [B9]. Similar measurements were also made on the shorter-lived 3s3p $^1P_1$ state. This work was performed within the framework of extending high-resolution laser spectroscopy to short UV and VUV wavelengths, where only pulsed and rather broadband laser radiation is readily available [B10]. As illustrated, the level-crossing method with pulsed laser excitation is a very useful technique for reliable and accurate determinations of short lifetimes. It is particularly valuable for short excitation wavelengths, where the precision method of time-resolved spectroscopy combining a synchronously pumped picosecond dye laser with delayed-coincidence detection is not easily applicable. Standard pulselength (5-10 ns) dye laser pulses, which can readily be shifted to short UV or VUV wavelengths by non-linear techniques, can be used to measure lifetimes down to 1 ns or below.

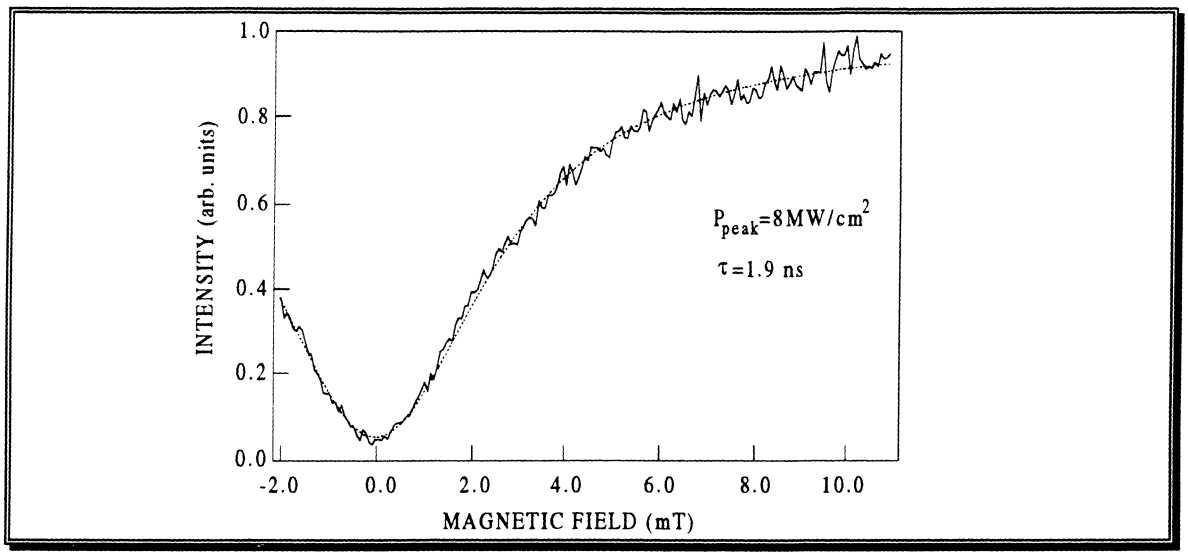

Fig. B5.
Hanle-effect data for the 3s3p $^1P_1$ state of magnesium. A Lorenzian is fitted to the experimental curve

B4 A new VUV laser spectroscopy set-up

Jörgen Larsson, Anders Persson, Sune Svanberg, Claes-Göran Wahlström and Raoul Zerne

Vacuum-ultraviolet (VUV) radiation can be generated in different ways. Efficient frequency mixing in crystals cannot be achieved at wavelengths shorter than 190 nm. To reach shorter VUV wavelengths stimulated Raman shifting is a feasible method. However,

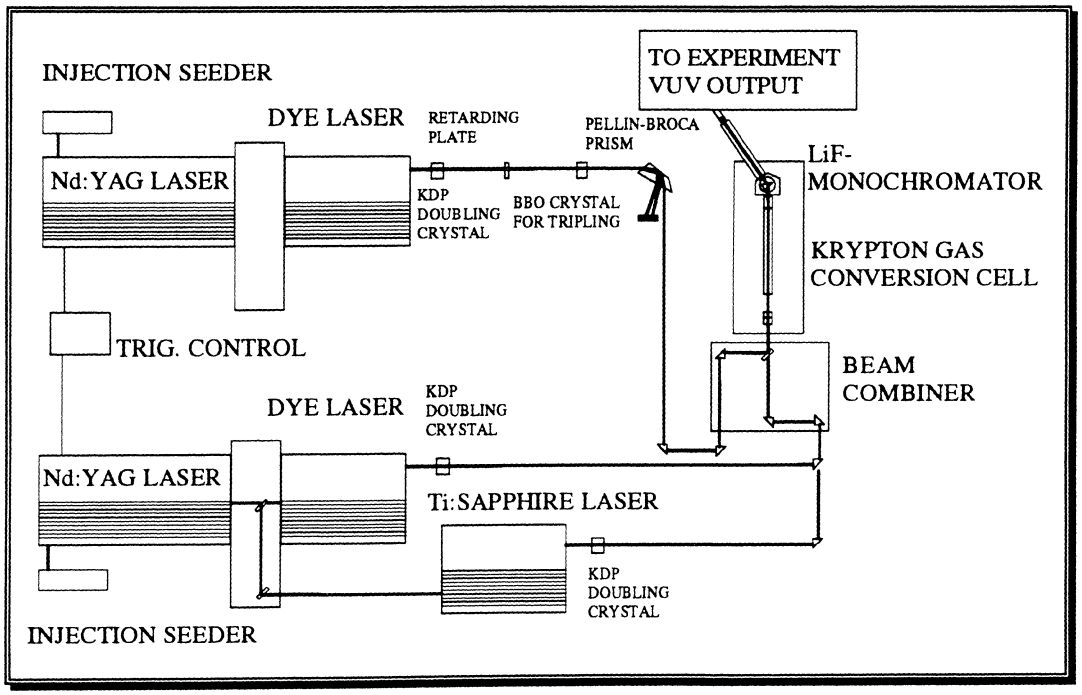

Fig. B6.
Set up for generation of VUV radiation using resonant sum-difference frequency four-wave mixing in krypton

the efficiency of the Raman generation will decrease with shorter wavelengths since higher anti-Stokes components have to be used.

A second possibility to generate tuneable VUV radiation is through resonant sum-difference frequency four-wave mixing in krypton (or xenon or metal vapours) with the scheme $\omega_{VUV}=2\omega_R-\omega_T$. Sum-difference mixing is the most favourable four-wave mixing scheme for producing VUV radiation since there are no restrictions on the sign of the wave-vector mismatch Δk. By tuning the sum frequency $2\omega_R$ to a two-photon resonance, a large enhancement in conversion efficiency can be achieved.

Our system primarily uses the transition 4p-5p[1/2,0] in krypton with the wavelength 212.55 nm for the two-photon resonance. To produce tuneable VUV radiation the UV output is mixed with tuneable visible or near-UV radiation, ω_T. With sum-difference resonant four-wave mixing in krypton, it is then possible to generate VUV radiation in the range 120-200 nm. The VUV radiation produced has a pulse energy of the order of a few μJ. First successful measurements on natural radiative lifetimes of Mg and Cu have been performed using excitation wavelengths down to 166 nm.

C Laser Spectroscopy in the Visible

C1 High-contrast transmission spectroscopy

Luo Caiyan, Stefan Kröll, Lennart Sturesson and Sune Svanberg*

**Visiting scientist*

High-contrast transmission spectroscopy (*Svanberg et al., JOSA B4, 462 (1987)*) is a saturation spectroscopy technique performed on a sample of high optical density. The probe beam will see a Lorentzian hole in the absorption profile burned by the pump beam. The transmission T(ν) of the probe beam as a function of ν, the detuning from line centre frequency will then be, for a cell of length L, proportional to

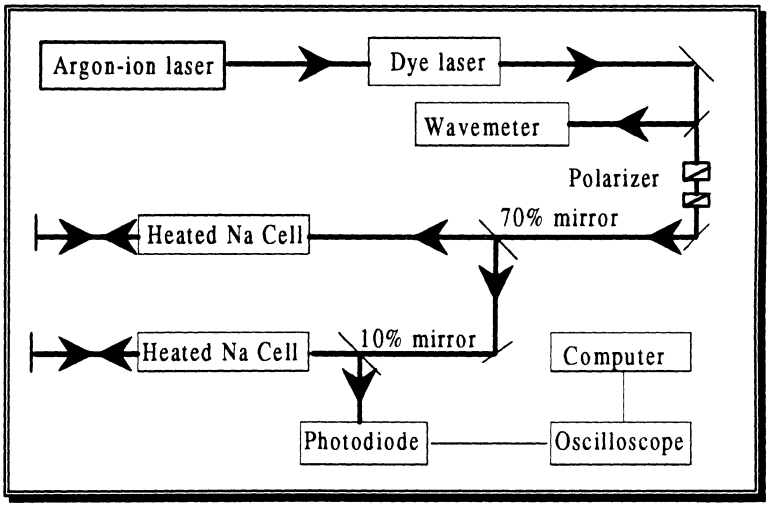

Fig. C1.
Experimental set-up for the tandem version of high-contrast transmission spectroscopy.

$$T(\nu) \propto e^{-\alpha(\nu)L} \qquad \text{C1}$$

where $\alpha(\nu)$ is the frequency-dependent absorption experienced by the probe beam.

For $\alpha(\nu)$ with a Lorentzian-shaped hole

$$\alpha(\nu) = \frac{\alpha_0}{1+\left(\frac{\Gamma}{\nu}\right)^2} \qquad \text{C2}$$

and high values of $\alpha_0 L$ a transmission profile considerably narrower than 2Γ, the FWHM of the hole in $\alpha(\nu)$, will be obtained.

It has been proposed, that a so-called 'tandem' configuration (see Fig. C1), where the output from a saturated absorption experiment is the input to a second identical set-up, would enhance such a line narrowing effect. Earlier experiments have shown that even with a single-cell configuration, high-contrast transmission spectroscopy could yield linewidths below the natural linewidth. The present experiments on the 3s $^2S_{1/2}$ - 3p $^2P_{1/2}$ transition in sodium have shown that the line narrowing effect is indeed even stronger in the tandem set-up [C1]. For the crossover between the $F_{gr}=1\rightarrow F_{exc}=1$ and $F_{gr}=1\rightarrow F_{exc}=2$ transition (F is the quantum number including nuclear spin for ground and excited levels) linewidths down to 5 MHz were obtained, Fig. C2. **This is considerably lower than the natural linewidth for the transition** which is 9.7 MHz. For simplicity, the linewidth was evaluated in terms of the $F_{gr}=1\rightarrow F_{exc}=1$ and $F_{gr}=1\rightarrow F_{exc}=2$ splitting. Earlier investigations [C2] have shown that the frequency shifts of these components in high-contrast transmission experiments are often negligible compared with their separation.

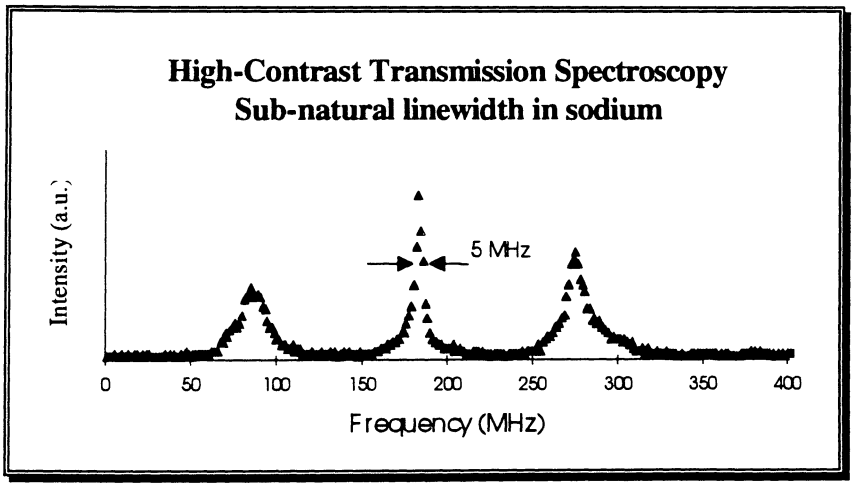

Fig. C2.
$F_{gr}=1\rightarrow$ $F_{exc}=1$ and $F_{gr}=1\rightarrow$ $F_{exc}=2$ transitions together with line-narrowed crossover resonance with FWHM = 0.55(natural linewidth)*

Several theoretical investigations of the line narrowing effect in high-contrast transmission spectroscopy have been published. However, we believe optical pumping, which has not been included in the earlier theoretical analyses, is one of the key elements in obtaining the line narrowing effect in the sodium transition above. To also address the issue of high-contrast transmission spectroscopy theoretically we are therefore presently modelling the line narrowing effect in sodium using theoretical calculations which consist of a rate equation approach where optical pumping between the hyperfine levels is included [C1].

C2 Accurate time-resolved experiments

Jörgen Carlsson, Per Jönsson and Lennart Sturesson

By exciting atoms with light from a mode-locked continuous laser and detecting light by time-correlated single photon counting, lifetime values can be obtained with high accuracy. Such precision values are particularly valuable for atoms which allow accurate theoretical calculations, as a test of the computational methods.

Time-resolved experiments on atoms can give values not only for lifetimes but also, through quantum beats, for the hyperfine structures of the excited states. In connection with the MCHF calculations of hyperfine structures in sodium, an accurate experimental determination of the hyperfine structure of the 3p $^2P_{1/2}$ state was made [C3]. A similar experiment has previously been performed on the 3p $^2P_{3/2}$ state *(J. Carlsson and L. Sturesson, Z. Phys. **D14**, 281 (1989))*. Compared with this the study of the J=1/2 state is somewhat more complicated since it requires circularly polarized light and a non-90° geometry for the quantum beats to appear. A decay curve is shown in Fig. C3.

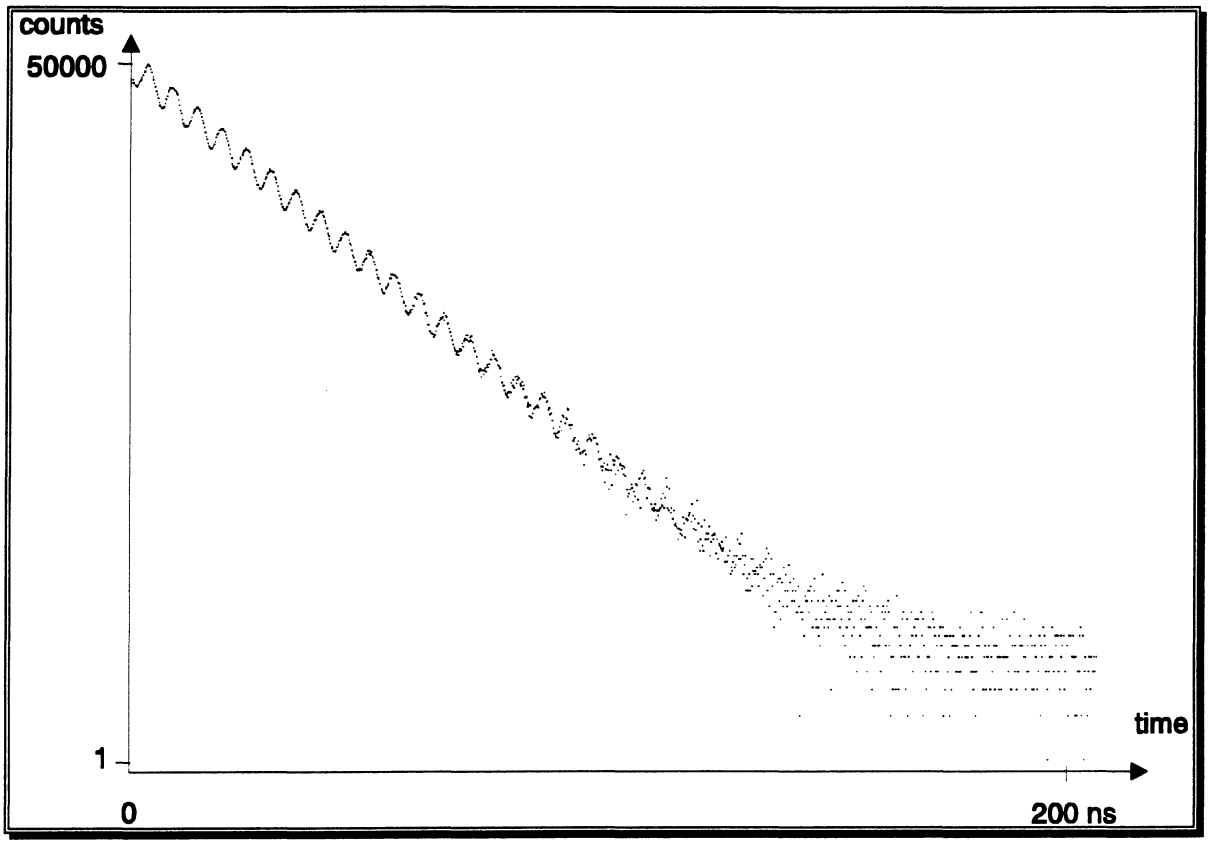

Fig. C3.
A decay curve for the 3p $^2P_{1/2}$ state of sodium on a logarithmic scale.

D Emission spectroscopy

Jorge G Reyna Almandos, Willy Persson and Sven-Göran Pettersson*

**Visiting scientist*

Investigations of the spectra of low and intermediate ionization stages of the rare gases have been continued, as has also the cooperation with the spectroscopy groups in La Plata, Argentina, and Campinas, Brazil. Dr J. G. Reyna Almandos has spent two months in Lund, working mainly on the analysis of the Kr IV spectrum. Sven-Göran Pettersson has spent three months with Dr Antonio Trigueiros in Campinas, where he was involved in the construction of a new theta-pinch light source for atomic spectroscopy.

In Kr IV some 85 energy levels have been established, belonging to the configurations $4s^24p^3$, $4s4p^4$, $4s^24p^25s$, 6s, 5p, 4d and 5d. The report on the spectrum of doubly ionized neon has now been published [D1].

During the last year the spectrograph laboratories have been totally remodelled and are now housing the high-power laser facility.

E Theoretical atomic physics

Much of the theoretical work in our division has been done in close cooperation with Professor Charlotte Froese Fischer's theory group in Nashville, where Per Jönsson has spent another 6 months. Work has also been done together with Dr Michel Godefroid and the theory group at Université Libre de Bruxelles, where Per Jönsson has been working for a period of time, mainly with computer program development.

The activities of the theory group have been concerned with computer program development and calculations of hyperfine structures and lifetimes as well as extremely accurate calculations of different parameters in lithium. The work has mainly been done with the non-relativistic multi-configuration Hartree Fock method, but lately the group has written a hyperfine-structure program for the new GRASP2 code (General-purpose Relativistic Atomic Structure Program) and performed large-scale relativistic calculations on the lithium-like sequence.

The computational resources of the group have improved dramatically since a new DEC5000 workstation was installed, and it is now possible to carry out large-scale atomic calculations. The group also has access to a CRAY2 supercomputer through the Nashville group and a CRAY Y-MP in Brussels. New computational methods adapted for supercomputing have been devised and the computer codes have been optimised.

E1 Computer program development

Per Jönsson and Lennart Sturesson

The new configuration interaction (CI) program of Froese Fischer allows large configuration expansions. A very general configuration generation program has been designed [E1] to be able to use the CI capacity for systematic studies of different atomic parameters. The program has adopted ideas from quantum chemistry and uses different active sets of radial orbitals. The non-relativistic hyperfine structure program [E2] has been optimized and adapted for large-scale calculations by using dynamic memory allocation [E3]. It is now also possible, in a systematic way, to sum contributions to the hyperfine structure from different types of configurations, making the physical interpretation easier.

A general CI program has been developed [E4], in which it is possible to add hyperfine, specific mass and volume operators to the Breit-Pauli Hamiltonian. Together with a transition probability program [E5] it is then possible to study hyperfine-dependent and hyperfine-induced transitions, which are of great astrophysical interest.

With today's powerful computers the possibility of performing large-scale relativistic calculations has increased. A configuration generation program for GRASP2 [E6] has been written, using output from the non-relativistic generation program. In this way the systematic study of parameters can be extended to the relativistic domain. To be able to perform relativistic hyperfine structure calculations a hyperfine program for GRASP2 has been written and tested [E7].

E2 Large-scale calculations

Jörgen Carlsson, Per Jönsson and Lennart Sturesson

Theoretical calculations have now come to a point where they can, in certain cases, compete with experiments when it comes to accuracy. Lithium, for which accurate measurements as well as extremely accurate Hylleraas calculations exist, has been a test case for the new MCHF computer codes and computational strategies. A number of different parameters have been studied in a systematic way with respect to convergence [E8,E9]. As an example values for the hyperfine structure of the ^{7}Li ground state are presented in Table E1.

The methods have been extended to more complicated systems such as sodium, where a comparison with accurate experimental hyperfine structures, determined with the delayed coincidence method [C3], has been performed. Recently, the effect of three- and four-particle excitations on the hyperfine structure has been investigated in a very large calculation [E10]. Magnesium [E11] and aluminium are other atoms for which hyperfine structures and radiative lifetimes have been calculated with large-scale MCHF methods. The purpose of this research is to finally be able to tackle even very complex systems, such as iron and cobalt, where so far little progress has been made.

Table E1.
Comparison of the Fermi contact term for the ^2S state in Li calculated with different methods

Authors	Method	a_c (a.u.)
King	Hylleraas	2.906 359[a]
King and Bergsbaken	Hylleraas	2.907 051[a]
Lindgren	MBPT	2.918 9[b]
Mårtensson-Pendrill and Ynnerman	CCSD	2.899 9[b]
Sundholm and Olsen	FE MCHF	2.904 9[b]
This work [D8]	MCHF	2.905 7[b]
This work [D9]	MCHF, partial wave expansion	
	l-extrapolated	2.905 1[a]
	corrected	2.906 1[b]
This work [D9]	MCHF, n-expansion	
	n-extrapolated	2.905 3[a]
	corrected	2.906 3[b]
Beckmann *et al.*	(experiment, atomic beam magnetic resonance)	2.906 02[c]

[a] No corrections included.
[b] Corrected for relativistic, finite nuclear size and finite nuclear mass effects.
[c] Recalculated from the experimental value $A_{1/2}$ = 401.752 043 3(5) MHz with a nuclear magnetic moment μ_I = 3.256 426 8(17) μ_N.

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II Quantum Electronics

In this chapter two recently started projects are presented. The first project concerns solid state spectroscopy and the investigation of a new concept for optical storage and optical signal processing and the second project concerns the development of new techniques for high-resolution microscopy on living biological matter.

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

The photon echo is a four-wave mixing technique where the excitation pulses do not overlap in time. To generate an output signal from the process under these conditions, the sample must "remember" the previous pulses when each new pulse arrives. For example, at the arrival of the second pulse the interacting atoms must still have a phase memory from the first pulse. The second pulse will then interfere with the atomic polarization remaining from the first pulse and for an inhomogeneously broadened transition a population grating will be formed in the form of frequency-dependent upper and lower state populations. Assuming at least part of the ground state population grating remains after the excited state decays, this modulated population distribution can be excited by, e.g. a brief excitation pulse to the excited state. These excited atoms will then radiate the temporal Fourier transform of whatever frequency pattern that has been stored. Within the photon echo project investigations of fundamental interactions in rare-earth-ion-doped systems are being performed as well as studies of applications related to optical storage and processing.

A1 Photon echo storage and processing

Ulf Elman, Per Tidlund and Stefan Kröll

Optical storage of multiple bits (~50) of information at a single spatial point has been performed in collaboration with SRI International [A1-A3]. The storage energy per bit was 200 pJ. To lower the energy/bit significantly, materials in which the oscillator strength of the active transition is higher are needed. The transitions in the 4f shell of rare-earth ions doped into inorganic crystals generally have a low oscillator strength because the 4f electrons are quite efficiently shielded from the internal crystal field by the outer electrons. The crystal field therefore does not perturb the symmetry felt by the electron in the 4f orbit more than to make the 4f transitions weakly allowed. Stronger transitions can, however, be obtained in the 3d shell transitions in the transition elements, as these 3d electrons are more exposed to the crystal field. To develop suitable materials co-operation with the crystal growth group under Dr Göran Svensson at the Dept of Inorganic Chemistry 2 was initiated and cobalt-doped $LiGa_5O_8$, in which the active transition is at a wavelength reachable for diode laser excitation, has now been grown at the Inorganic Chemistry Department.

Table A1.
Comparison between frequency-domain optical storage (FDOS) and time-domain optical storage (photon echoes). FDOS values are taken from JOSA B9, 998 (1992).

	FDOS	Photon echoes	
Concentration of active centres (cm^{-3})	6×10^{18}	10^{20}	
Optical density	0.43	0.13	
Number of bits/point	1000	1000	10^4
Hole depth, branching ratio	0.2	0.2	1
Storage density ($Gbits/cm^2$)	26	6	30
Writing speed	20 MHz	10 GHz	1 THz
Reading speed	100 MHz	10 GHz	1 THz

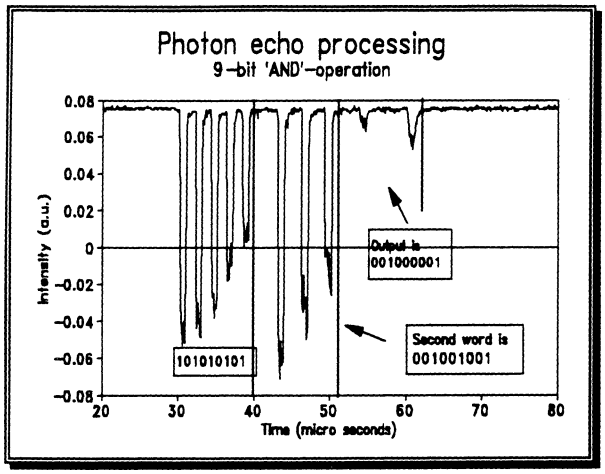

Fig. A1.
Photon echo AND operation

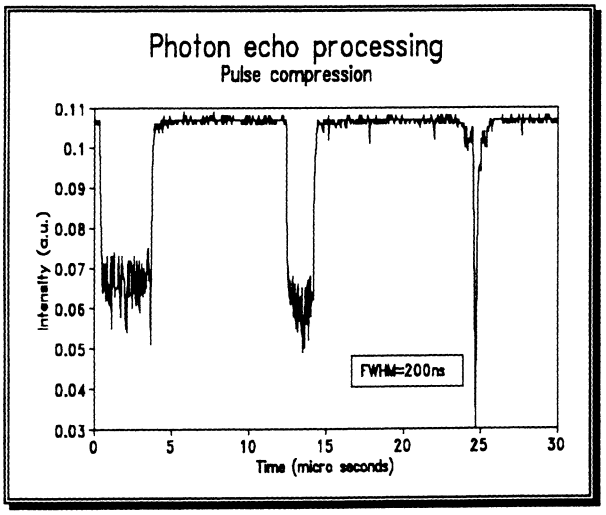

Fig. A2.
Photon echo pulse compression from 4 μs to 200 ns.

An analysis of the theoretical storage density, mainly based on signal-to-noise considerations, has been performed [A4-A6]. This study demonstrated that the maximum storage density was approximately 100 times the maximum storage density of conventional optical storage media. A comparison of the performance of the photon echo approach with that of the closely related spectral hole-burning technique (FDOS) is shown in Table A1.

From the above work it can be concluded that one of the prime advantages of the photon echo approach is its speed. We have therefore looked at approaches for optical data and optical signal processing using the photon echo technique [A7-A10]. Fig. A1 shows the implementation of an all optical photon-echo AND operation on a 9-bit word and Fig. A2 shows photon-echo optical pulse compression by a factor of 20.

A2 Relaxation processes in rare-earth-ion-doped crystals

Stefan Kröll

Photon echo techniques can resolve the (sub) kHz linewidths that exist in rare-earth-ion-doped crystals at liquid helium temperature. The narrow linewidths enables studies of very weak interactions in these systems. Measurement of the photon echo relaxation time gives information on processes which destroy the phase memory of the atoms. High-intensity excitation pulses have been shown to shorten the phase memory. For some crystals excitation-induced frequency shifts have been identified as the mechanism shortening the phase memory (*e.g. Phys. Rev. Lett.* ***68****, 3216 (1992) and references therein*). We have shown that in Pr-doped $YAlO_3$ the main excitation dependent effect is **not** the excitation induced frequency shift [A11,A2,A3] and the mechanism has recently been shown to be mono-energetic phonons (*R. Kachru, private communication*). Future experiments will focus on unravelling the interaction behind an unexpected resonance we have observed for the relaxation time vs. magnetic field strength [A2, A3, A7, A11].

B High-resolution microscopy

In this project we are developing novel methods for high-resolution microscopy suitable for living biological matter. Several methods (electron microscopy, scanning tunnelling microscopy, atomic force microscopy etc.) yield very high resolution but are not suitable for non-destructive studies of living systems. For such non-intrusive studies researchers are still basically limited to the classical optical microscope. Thus, the resolution is limited by diffraction to a few 100 nanometres. Many structures of considerable interest are smaller, e.g. viruses (≈50 nm) or larger proteins (5-50 nm). We are investigating two potential approaches to obtain a microscope suitable for imaging living matter. In the first method (**soft X-ray microscopy**) resolution is increased by decreasing the operating wavelength, while the other method (**trapped-particle optical microscopy**) aims at the development of an optical microscope with resolution better than the fundamental diffraction limit. Both methods share the common goal to develop compact and reasonably priced microscopes suitable for research as well as practical applications.

B1 Soft X-ray microscopy

Lars Rymell, Hans Stattin and Hans M. Hertz

Introduction

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) (*A.G. Michette, Rep. Prog. Phys.* ***51****, 1525 (1988)*). The use of this wavelength range has the advantage that the limitation in resolution due to diffraction is in principle 20-40 Å. Furthermore, the samples may be studied without staining, sectioning or fixation in an aqueous environment at atmospheric pressure. The disadvantages are that current X-ray microscopes for biological studies are based

on synchrotron sources, which often results in a limited accessibility for the application researcher. Below, our progress in the construction of a table-top soft X-ray microscope is described.

Soft X-ray source

Currently the only potential table-top sources yielding sufficiently high soft X-ray photon fluxes to compete with synchrotron radiation are the plasma focus and the laser-produced plasma (LPP). Of these two, the LPP generates a smaller soft X-ray source which is especially advantageous in scanning microscopy.

We are currently constructing a high-brightness LPP soft X-ray source for a water window microscope. Since the zone plate (diffraction) optics conventionally used for high-resolution soft X-ray imaging require relatively monochromatic radiation to operate properly, we are using light-element targets. Heavy-element targets give a higher conversion efficiency but also more broad-band Brehmstrahlung emission. Figure B1 describes the first experiments with a plastic target. A 50 ps mode-locked frequency-doubled 10 Hz Nd:YAG laser is focused with a f=50 mm lens onto the target. X-ray emission is spectrally analysed by a grazing incidence monochromator and the total X-ray flux is measured with a windowless GaAs diode covered with unbacked metal filters. The spectrum is shown in Fig. B2. Here the linewidth is determined by the resolution of the monochromator making this source potentially a monochromatic source provided suitable filters can be developed (see below). The flux is estimated to be $0.5*10^{12}$ photons/ster*line*pulse, which should be sufficient for soft X-ray microscopy as discussed below assuming the source size is a few tens of μm. We are currently measuring this size using a knife edge method [B1].

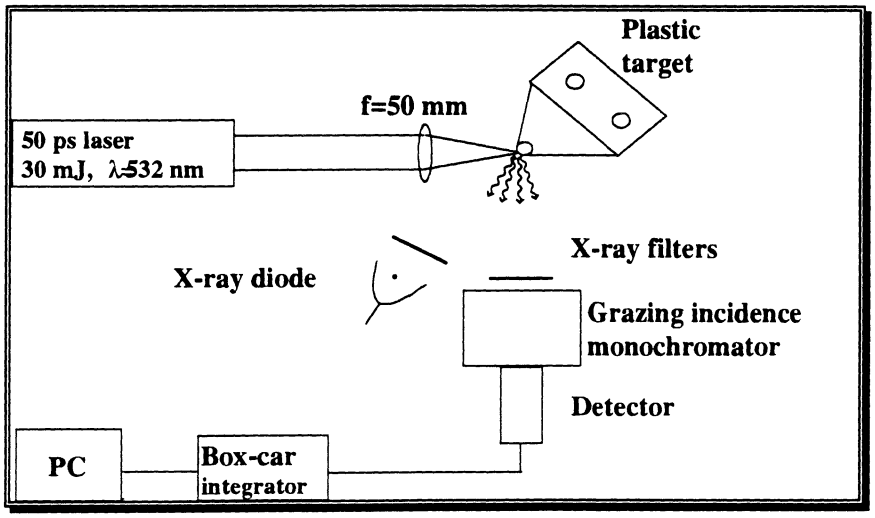

Fig. B1. Experimental arrangement for a high-brightness water-window soft X-ray source.

However, as with any LPP using solid targets the source described above produces debris which contaminates the studied sample or other sensitive components positioned

close to the LPP. We are now investigating the use of small (≈10 μm) liquid droplets as a target. Since this target evaporates upon plasma formation the debris problem is eliminated. Thus, the source can be positioned close to the objective/sample thereby increasing the soft X-ray flux. Using ethanol as target liquid the spectra and flux is very similar the results of the plastic target.

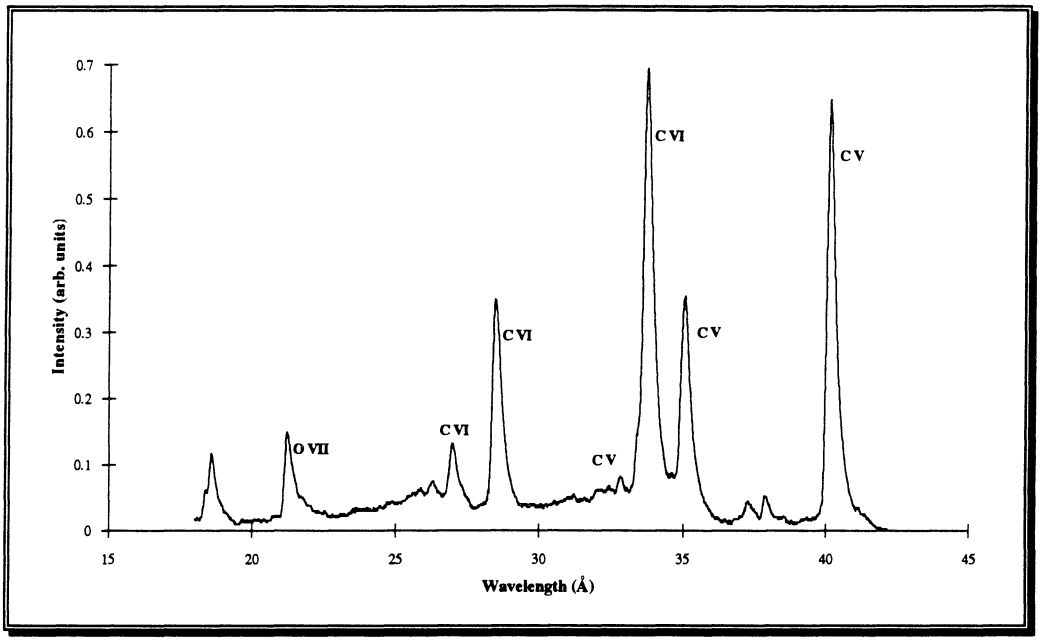

Fig. B2. C and O line emission from a LPP target detected through 0.1 μm Al foil.

Soft X-ray optics

Wavelength-selective free-standing thin-film metal filters are an essential part of the optical system. Specially designed filters have been fabricated [B2]. These filters are tailored to the LPP soft X-ray source in order to yield monochromatic emission.

One major problem in the soft x-ray region is the lack of simple and efficient optics. Currently, Fresnel micro-zone plates (i.e. circular diffraction optics) are the prime choice in microscopy and the throughput calculation discussed below is based on this type of optics. However, alternative methods of imaging are desirable. We are investigating the use of line focusing optics such as strongly astigmatic grazing incidence optics, which may yield higher total efficiency due to their broad band nature. By scanning and rotating such a line focus over the sample and processing the data with Fourier deconvolution and tomographic reconstruction, an image of the X-ray absorption can be calculated. Preliminary experimental tests of the concept have been performed in the optical wavelength region.

Compact soft X-ray microscope

Using the theoretical and experimental results briefly described above, a soft X-ray flux analysis for different microscope designs has been performed. A conservative estimate of the necessary exposure time for a microscope based on the LPP source and zone

plate optics results is 0.1 s/pixel for 100 nm resolution imaging of living biological samples.

B2 Trapped-Particle Optical Microscopy

Lars Malmqvist and Hans M. Hertz

Introduction

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 (*D.W. Pohl, "Scanning Near-field Optical Microscopy (SNOM)", in Advances in Optical and Electron Microscopy Vol. 12, pp. 243-312 (Academic Press, 1991)*). However, the mechanical positioning of the probe restricts their use to mechanically accessible and smooth surfaces, limiting the method's applicability for studies of, e.g., living biological material with intervening membranes.

We are investigating the use of different types of trapped particles as microscopic light sources for non-intrusive scanned probe optical microscopy - Trapped Particle Optical Microscopy (TPOM)[B3]. In our case we use a single-beam gradient force optical trap [B4] to position and scan the microscopic light source. The optical trap consists of a strongly focused laser beam which traps dielectric particles just below the focus due to radiation forces. The particle will act as a light source due to the scattering of the trapping laser beam by the particle. These particles may be non-intrusively positioned and scanned very accurately in transparent media.

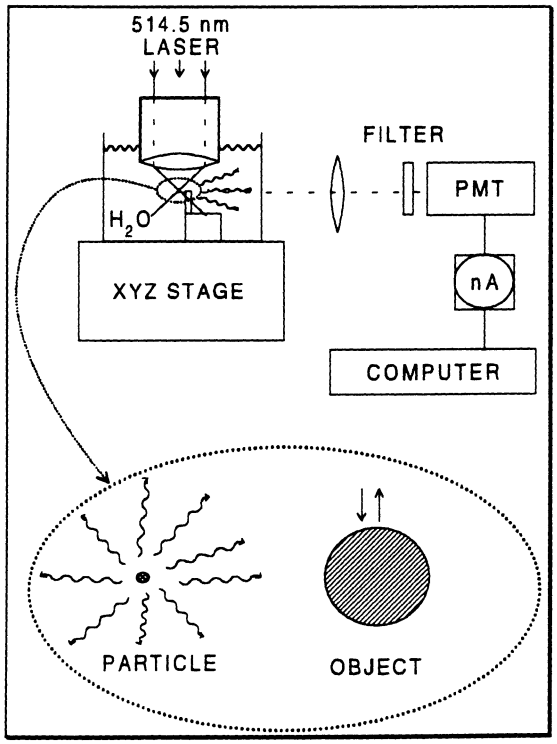

Fig. B3. Experimental arrangement for Trapped-Particle Optical Microscopy.

Experiments

The first experiment with TPOM is shown in Fig. B3. A λ=514 nm Argon-ion laser beam is focused with a NA=1.25 water immersion objective into a water-filled cell where 60 nm or 290 nm diameter SiO_2 particles are trapped. The scattered light from the particles is detected by a photomultiplier and a test object is scanned piezoelectrically close to the trapped particle.

In Fig. B4 290 nm SiO_2 particles were used to image a test object consisting of two crossed 4 μm carbon wires by 2-D scanning. The resolution in this demonstration experiment is approximately 2 μm,

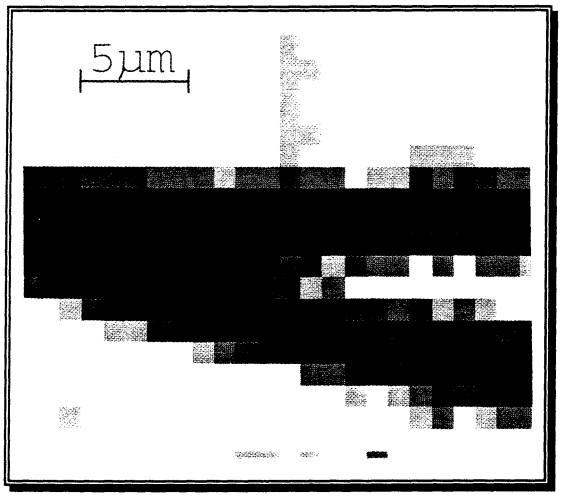

Fig. B4. TPOM image of two crossed 4 μm carbon wires.

i.e. significantly above the resolution of the classical optical microscope. This is due to the difficulty in performing measurements with a sufficiently small particle-object distance when using the experimental arrangement shown in Fig. B3 since laser light from the intensity tails of the laser beam will scatter off the object. Since this scattered light has the same wavelength as the probe light scattered by the trapped particle, the signal-to-noise ratio deteriorates as the object approaches the source, making measurements with a particle-object distance less than 5 μm difficult.

We are currently working on circumventing this restriction by developing two-colour TPOM. In this system different trapping and detection wavelengths are used allowing the object to be positioned very close to the particle. In one implementation of this scheme we use a cw Nd:YAG (λ=1.06 μm) laser to trap $LiNbO_3$ particles [B5]. Due to the non-linear properties of the crystal, green light (λ=532 nm) is generated when the particle is trapped in the focus. As an alternative, fluorescent particles may be used. Microscopy experiments with this source are planned in the near future.

Discussion

If the particle-object distance can be made very small using the above method, the effective size of the trapped particle will ultimately determine the resolution of TPOM. The effective size is the physical size of the particle convolved with its displacement due to Brownian motion in the trap. Assuming that the optical trap is an harmonic potential well, such calculations result in a resolution of approximately 60 nm for a 50 nm diameter high-refractive-index particle in a strongly focused trapping beam.

In summary, we are developing a non-intrusive scanned probe optical microscope using optically trapped particles as probes. Potentially, the method allows near-field optical microscopy to be performed on hitherto inaccessible objects, such as living biological matter with intervening membranes.

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III Environmental Remote Sensing

Research in the field of environmental remote sensing is directed towards the development of spectroscopic techniques, both laser and non-laser, to measure mostly tropospheric gases, but also with applications in terrestrial and aquatic studies. The group is participating in two European projects, EUROTRAC and LASFLEUR, dealing with tropospheric ozone research and vegetation studies, respectively. Furthermore, a project on measurements of gases of geophysical origin is being pursued together with an Italian research group. In Lund some activities are also channelled through the Centre for Environmental Measurement Technology (CENTEC), co-ordinating the research and teaching in environmental sensing technology within the Lund Institute of Technology. The remote sensing work at the department is supported by the Swedish Space Board (RS), the Swedish Environmental Protection Agency (SNV) and the Swedish Natural Science Research Council (NFR).

The method most used in the various projects is the differential absorption lidar technique (DIAL). A review of the DIAL technique emphasing on the work performed by the group over a period of several years will be published shortly [1]. Both a mobile DIAL system and a newly developed fixed system have been used in tropospheric ozone research. Several field campaigns with the mobile DIAL system have been performed during the last two years, including a large campaign in Italy with measurements of gases of geophysical origin. The system has also been used in field measurements of industrial emissions within the regulatory program at SNV. Some improvements have been made to the mobile system, including access to wind data from a wind monitor on a mast fixed to the system. The possibility of remote plume speed determination using the correlation of video images is presently being investigated in a diploma project. In another diploma project new routines for automatic calibration of the dye laser wavelength have been implemented [2].

Another technique being employed is the differential optical absorption spectroscopy (DOAS) method, which is used for long-path absorption measurements in the atmosphere with passive or active non-laser light sources. A description of the DOAS system developed at the department and earlier measurements are currently being published [3], and have also been presented elsewhere [4-7]. The DOAS system has been employed together with the DIAL systems for comparative and complementary measurements in the research projects.

Finally, laser-induced fluorescence (LIF) is being investigated as a tool in vegetation and water quality studies. Special emphasis has been placed on remote multi-colour imaging of vegetation fluorescence following laser excitation. Field campaigns in Germany and Italy have been carried out with the mobile lidar system, where an optical multichannel analyser and a multi-colour imaging system were adapted to fit to the receiving telescope.

A Tropospheric ozone lidar

Hans Edner, Sune Svanberg and Eva Wallinder

Within the European project EUROTRAC the group is participating in the subproject TESLAS, which is an acronym for Tropospheric Environmental Studies by Laser Sounding. Within the project the DIAL system specially dedicated for ozone measurements has been further developed [A1,A2]. The KrF excimer laser (Lambda Physik LPX 210iF) has been upgraded with a different type of unstable resonator with a 50 % higher output energy at low divergence (<0.4 mrad). The efficiency of the stimulated Raman scattering with the new resonator has been investigated, and a considerable increase in the second Stokes component was found, especially with deuterium as the Raman medium. Special studies have been made of rotational Raman components in the generated spectrum under different conditions. These components were found to be negligible at the Raman cell pressures used. Comparisons with a narrow-band tuneable KrF excimer laser (Lambda Physik EMG 150) and quadrupled Nd:YAG laser (Continuum YG 682-20) have been made in this context. Of these only the narrow-band KrF laser produced some rotational lines at low pressure. At pressures around 1 bar of H_2 sidelines with energies of up to 40 % of the first Stokes vibrational Raman component could be generated. However, with increasing pressure the rotational lines decreased rapidly. The photomultipliers in the detection system have been modified to cope with higher peak currents. A number of test measurements have been made. So far the wavelength pair 277/313 nm has been used. Simultaneous measurements with 278.7/286.3 nm from a Nd:YAG-pumped dye laser in the mobile DIAL system have also been performed.

The mobile DIAL system was employed during the TROLIX'91 (Tropospheric Ozone Lidar Intercomparison Experiment) campaign in Bilthoven, The Netherlands, June 10-28, 1991 [A2,A3]. Three other DIAL systems were included in the campaign, and the data were supported by ground- and airborne sensors. The major goals of the experiments were to compare the performances of the systems and to determine the accuracy that can be achieved under realistic conditions. Although the mobile DIAL system with the Nd:YAG and dye laser is not optimised for monitoring vertical ozone profiles, it added two more wavelengths to the number of UV wavelengths used for data retrieval. Since a tuneable system was used, the wavelengths could be chosen to minimise the interference due to SO_2 absorption. An extra folding mirror was installed in the system to facilitate vertical measurements, which was the normal measurement mode throughout the campaign. Another type of measurement was made, a vertical scan from ground upwards. Fig. A1 shows the results of such a scan up to the cloud base. From this scan a vertical profile, essentially from the ground, can be determined with a vertical projection, as can be seen in the left part of the figure.

A DOAS system developed in the TOPAS (Tropospheric Optical Absorption Spectroscopy) project was also included in the campaign. Our DIAL system and one of the others could measure horizontally, and these data could thus be compared with the data from the DOAS system. Fig. A2 shows the results of such an intercomparison. The interesting fact in comparisons between these two techniques is that the measurements can be made in the same air volume - no assumption of the homogeneity of the air over the path is needed. Both techniques utilise the optical absorption, but whereas DIAL only employs two wavelengths with a rather large separation, the DOAS technique uses small, higher-order structures in the ozone absorption profile in a wavelength interval. DOAS

will normally not be affected by extinction due to aerosol scattering. A study of aerosol effects and validation of correct procedures in DIAL measurements can thus be made. DOAS also has the advantage that a number of other gases can be detected and quantified.

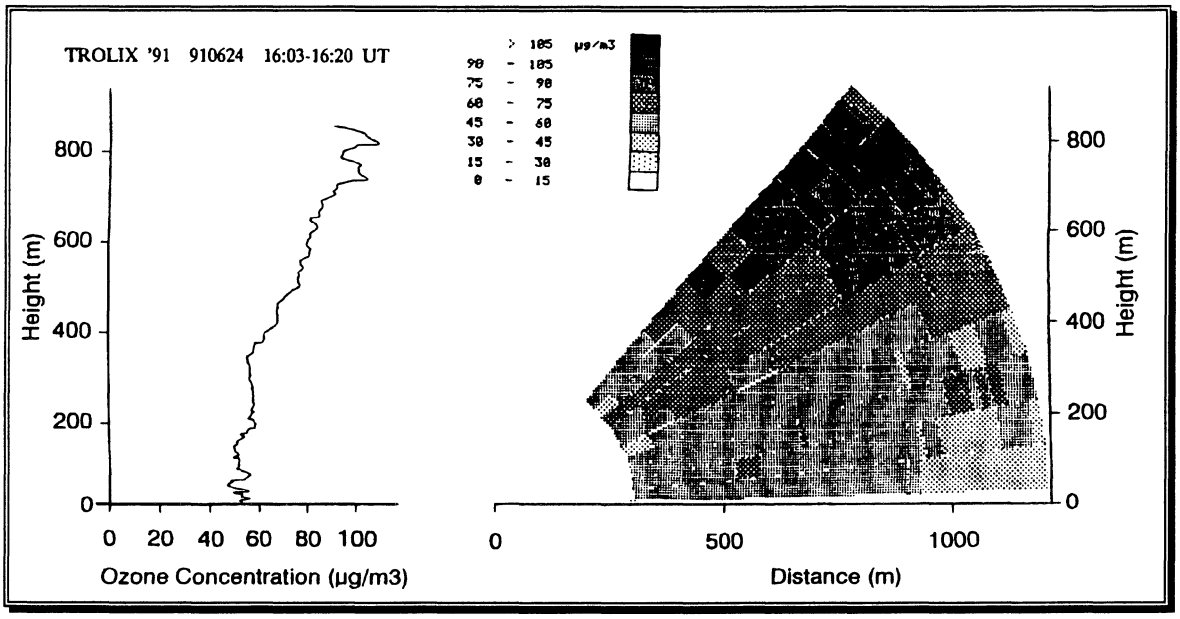

Fig. A1.
Ozone distribution deduced from a vertical scan

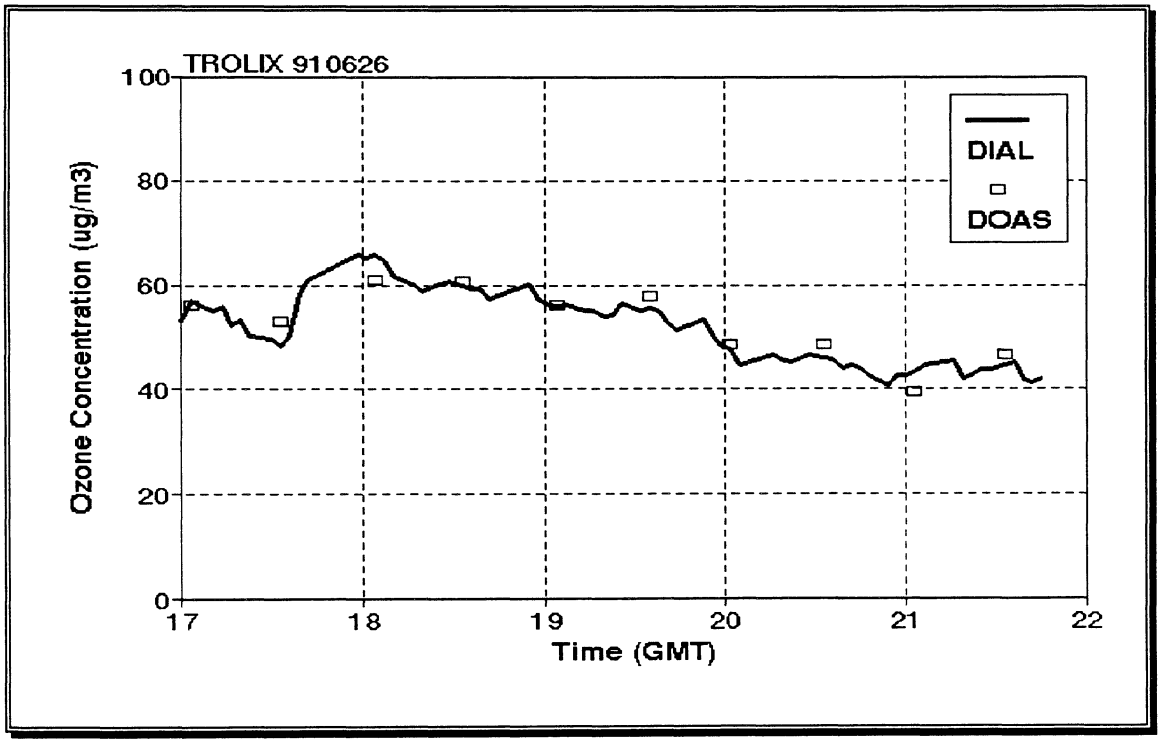

Fig. A2.
Intercomparison between DIAL and DOAS measurements of ozone along a 1 km horizontal path.

B Lidar and DOAS measurements of gases of geophysical origin

Hans Edner, Pär Ragnarson, Sune Svanberg and Eva Wallinder

Geophysical anomalies are usually accompanied by the emission of a number of gases. It has been suggested that remote sensing of these gases could be useful in prospecting for natural resources such as minerals or geothermal energy. It is also important to have knowledge concerning the contribution from natural sources in order to understand chemical reactions in the atmosphere and the potential influence from anthropogenic activities, e.g. pollutive emissions from industry. Mercury is one example that is especially interesting for lidar measurements since most of the atmospheric mercury of geophysical origin is in atomic form. The atomic resonance line at 253.65 nm is strong enough to enable measurements down to the normal background values of a few ng/m^3.

The successful measurement campaigns in the Tuscany region, Italy, 1990, in collaboration with R. Ferrara and co-workers at the CNR Istituto di Biofisica in Pisa, resulted in a number of articles. Extensive intercomparison between the two measurement techniques used by the two groups, lidar versus gold amalgamation combined with flameless atomic absorption, has showed very good agreement. This indicates that most of the atmospheric mercury measured during the campaign was in the atomic form [B1]. Measurements in geothermal fields, at several geothermal power plants as well as in unexploited areas, showed highly elevated concentrations of atomic mercury [B2]. This was in strong contrast to earlier measurements in Iceland [B3,B4]. The annual atmospheric mercury discharge from a power plant in a mineralised area was found to be 160-210 kg. An interesting result of the measurements is that the atomic mercury seemed to emanate from the base of the cooling towers rather than from the top. Measurements at the abandoned mercury mine at Abbadia San Salvatore showed very high concentrations around the distillation plant [B5]. Fig. B1 shows the results from a horizontal scan over the area. Plumes containing mercury concentrations higher than 1 μg/m^3 still emerged from the smoke stacks as they ventilated the contaminated interior of the plant. Also the degassing from the deposits of roasted cinnabar (HgS) resulted in highly elevated concentrations. Vertical profiles above the deposits showed a very pronounced fall-off a few metres over the ground. Results from the 1990 campaign have been further presented at a number of conferences [B6-B13].

Italian plans to further expand geothermal power production have initiated interest in our measurement of atomic mercury fluxes from geothermal power plants. The investigation of a plant in the Mt. Amiata region was followed up by an extensive campaign in August 1992, with complementary measurements of the wind speed and direction at the relevant height. Data from the new measurements confirmed the earlier results. The total flux was found to be 200 kg/year.

In the 1992 campaign the interest in gases of geophysical origin was extended to the study of volcanically active areas. Volcanoes normally emit large amounts of gases, such as carbon dioxide and sulphur dioxide. Interest in monitoring the emission of gases is directed not only at estimations of the environmental impact, but it has also been suggested that monitoring may be used in the prediction of earthquakes and volcanic eruptions. In September, 1992 the mobile lidar system was placed on board the new research vessel Urania, chartered by CNR. The ship made repeated traverses below the

plumes from the volcanoes Etna, Stromboli and Vulcano. Measurements of sulphur dioxide and atomic mercury have been performed to establish the total fluxes. The sulphur dioxide measurements were supported by the DOAS technique in the passive mode using scattered sunlight. Examples of data from Etna are shown in Fig. B2. At Vulcano it was also possible to measure the plume in the normal scanning mode from a fixed position. The emissions of sulphur dioxide and atomic mercury have also been investigated at the volcano Solfatara near Naples. The emission rates of sulphur dioxide from the four volcanoes are: Solfatara 10 kg/h, Vulcano 1 tonne/h, Stromboli 6 tonnes/h and Etna 100 tonnes/h.

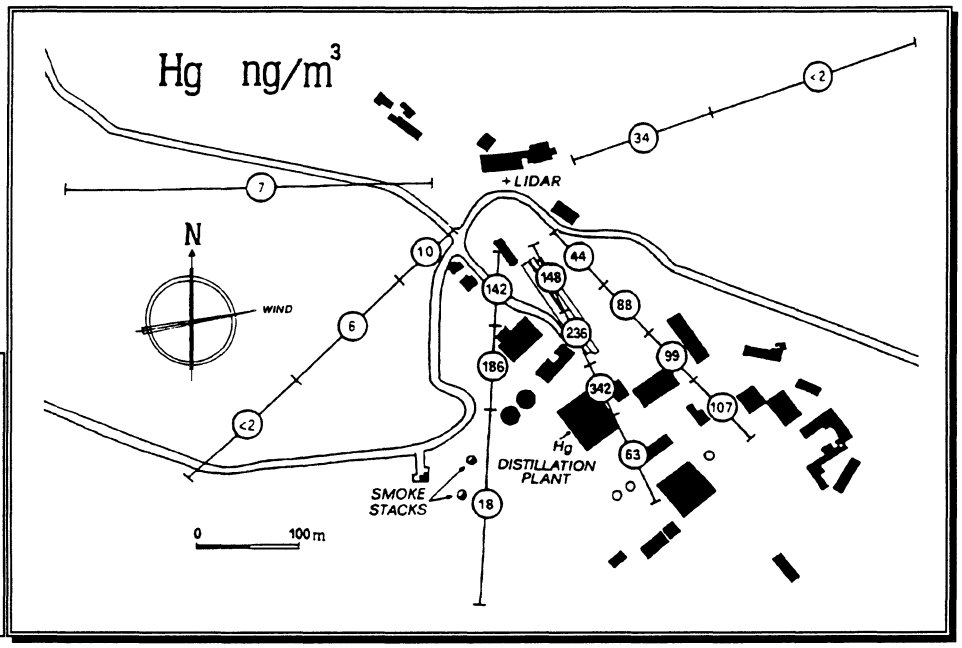

Fig. B1.
Map of atomic mercury concentrations over the central parts of a mining area.

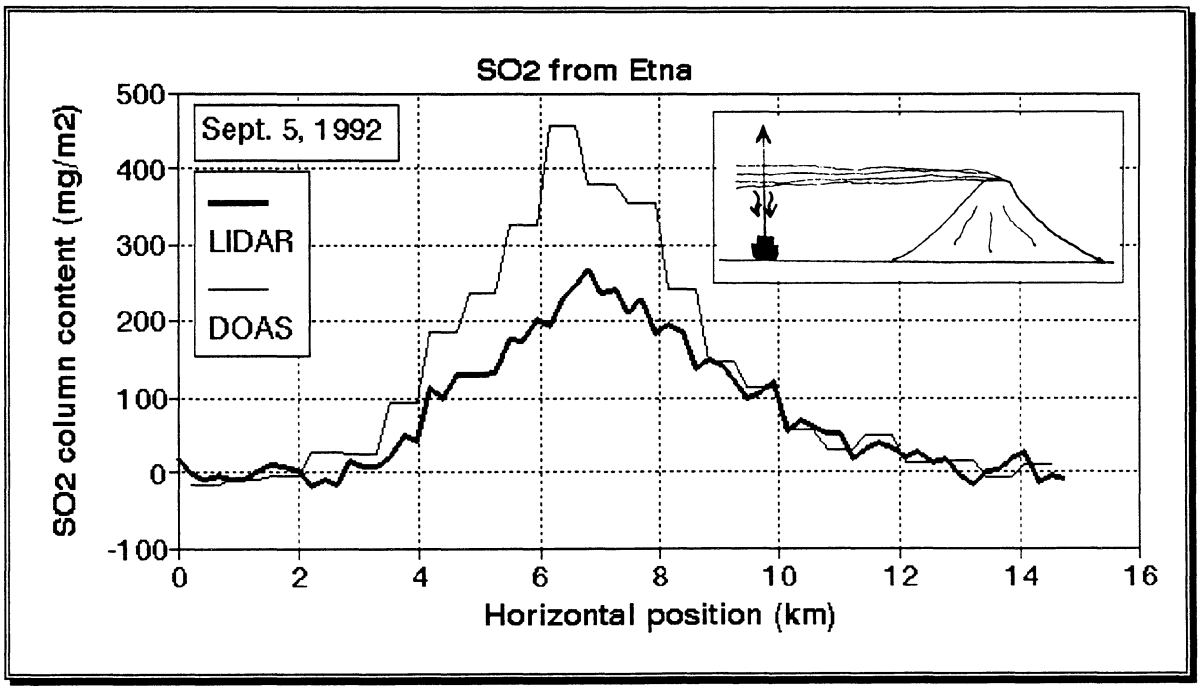

Fig. B2.
Vertical SO_2 column content during a traverse under the plume of Etna, determined from DIAL and passive DOAS measurements.

C Lidar measurements of industrial emissions

Hans Edner, Pär Ragnarson and Eva Wallinder

With the growing awareness of the serious environmental impact of industrial activity and traffic, and more stringent regulations on emission values, the need for powerful measurement techniques for air pollutants is increasing. Optical remote sensing techniques are particularly advantageous allowing large-area monitoring and avoiding sample extraction and preparation difficulties. The mobile DIAL system has been employed in several measurements of different gases in industrial emissions as well as in ambient air. The ability to measure fluxes of a particular gas from different sources has been particularly useful. This is achieved by combining wind data with a mapping of the concentration distribution in a vertical section downwind from the source. In this way diffuse emissions can also be studied and the total pollutant flux can be measured.

Mercury and its compounds are widely used for industrial and agricultural applications due to the unusual physico-chemical properties of these materials. One of the principal uses of mercury (which accounts for about 20% of the total consumption) has been in the production of caustic soda and chlorine. The chlor-alkali processes makes use of mercury in the simultaneous production of chlorine and caustic soda by electrolysis of brine solution using a flowing cathode of metallic mercury. Most of the mercury loss is into the atmosphere, making chlor-alkali plants one of the largest anthropogenic sources of atmospheric mercury. The results of earlier DIAL measurements on mercury emissions from a chlor-alkali complex located in central Italy have now been published [C1]. The mercury flux monitoring was supplemented with measurements made with a point monitoring technique, showing good correlation in the concentration levels. More recent measurements have been performed on two Swedish chlor-alkali industries. An example from one of these studies is shown in Fig. C1, where the upper part displays the results from a vertical scan through the spreading plume. As can be seen, two distinct mercury plumes are detected, one from the major cell house and one from a smaller source closer to the lidar system. This latter source was not considered by the company to be important with regards to mercury emissions and no continuous measurements were performed at this location. However, as is shown in the lower part of the figure, it contributes substantially to the total mercury flux.

The mercury measurement at the Swedish chlor-alkali plant is an example from one of several field campaigns that have been carried out under a contract with SNV. A total of 9 different sites have been visited by the mobile DIAL system. The emissions of mercury, sulphur dioxide and nitrogen dioxide from various types of industries have been monitored, including refuse incineration, coal-fired heating, chemical, paper pulp, petrochemical and metallurgical plants [C2-C4]. The measurements have been used in the regulatory control programme at SNV, and the measured values have been compared with data from the companies. Fig. C2 shows such a comparison of the sulphur dioxide flux from a pulp industry. A rather good correlation can be seen, with the lidar giving somewhat higher values at certain times, which could be attributed to diffuse emissions.

Further measurements, outside the SNV contract, have been performed on the diffuse emission of mercury from deposits of fibre sediments from earlier pulp industries [C5]. Only low mercury emissions could be detected at these sites.

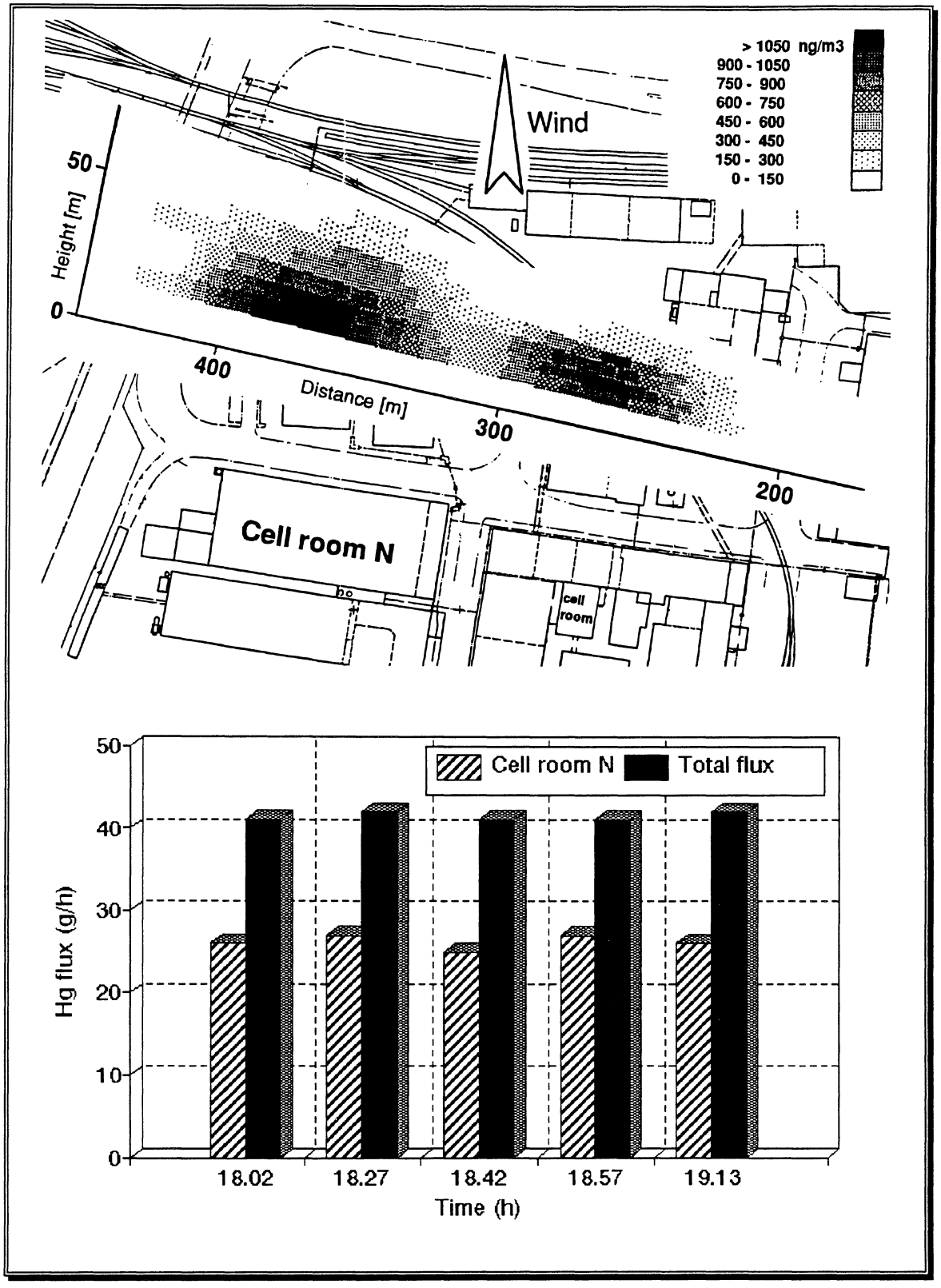

Fig. C1.
Vertical scan through the spreading mercury plume from a chlor-alkali plant and the resulting mercury flux determination.

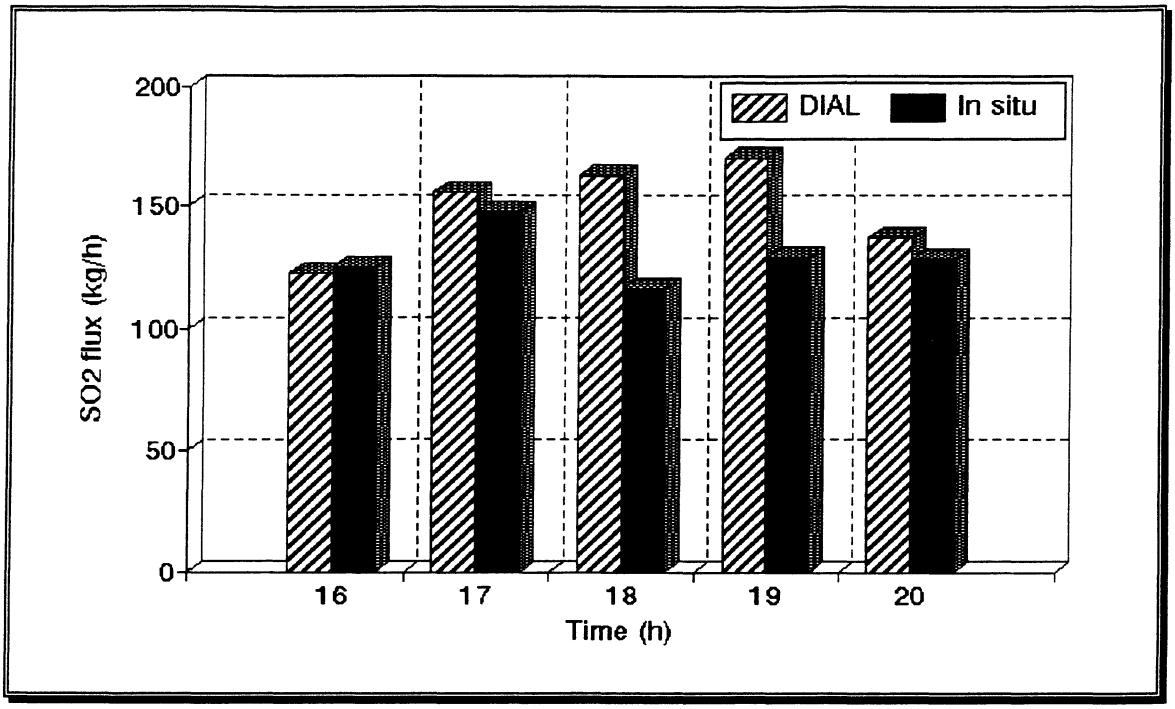

Fig. C2.
Comparison of the SO_2 flux from a pulp industry, determined from remote DIAL measurements and in situ monitoring at the main source.

D Laser-induced fluorescence of vegetation and water

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

Together with the medical laser group, the remote sensing group is participating in the two European projects LASFLEUR and EUROMAR aimed at measurements of vegetation and sea-water, respectively. The joint group has participated in two field campaigns on these subjects, one in the summer of 1990 and one in 1992. The results from the Italian campaign of 1990 have now been fully evaluated. Within the LASFLEUR project a paper has been published on the measurements performed in the beech forests of Pian di Novello [D1]. Further, water measurements on the Arno river has been reported [D2].

Within LASFLEUR further measurements have been done. The optimum wavelength for exciting the fluorescence from primarily chlorophyll has been investigated in a diploma project [D3]. It was found that the wavelength should be about 400 nm or longer to be able to activate the chlorophyll systems. From eye safety considerations it should be below 400 nm since the eye exhibits a dramatic reduction in sensitivity below this limit. By using the tripled Nd:YAG laser combined with a Raman cell, a suitable wavelength of 397 nm could be achieved as the first Stokes component in deuterium. Two different modes have been used for the laser-induced fluorescence: the point monitoring mode and the imaging mode. When using the point monitoring mode, an OMA (Optical multi-channel analyser) system connected to an optical fibre was employed. A spectrum from a specific point on the object is thus achieved. The object can be placed directly under the

tip of the fibre, or it can be far away, seen through the telescope of the mobile lidar system. In the latter case the fibre is placed in the focal plane of the telescope where the image of the object is found. When using the imaging mode a split-mirror Cassegrainian telescope with an image-intensified CCD detector is employed together with the telescope of the lidar system [D4-D6]. Four different optical filters are mounted in front of the Cassegrainian telescope allowing four images in four different colours to be detected on the CCD. For vegetation monitoring interference filters are used which cover the two chlorophyll peaks at 685 nm and 740 nm. A broad-band blue filter is also used for the blue fluorescence, and the fourth filter is, for example, for the total red fluorescence. These images can then be further processed. By dividing the two images of 685 nm and 740 nm for example, the health status of the vegetation in different parts of the picture can be monitored.

The three-week field campaign of 1992 took place in Germany and Italy. The first week was spent in Karlsruhe with the plant physiology group of H. Lichtenthaler. Measurements were performed with the point monitoring system and the imaging system on different kind of leaves. The same samples were also investigated by plant physiologists. The second week was spent near DLR in Oberpfaffenhofen, Germany, together with several other groups in a LASFLEUR campaign. Remote fluorescence was recorded for spruce, maple and potted maize plants as shown in Fig. D1. Measurements were performed on maize plants that had been grown in different kinds of soil, with and without nutrients. Daily cycles of a single maize plant were also investigated as well as the leaf area index of a maple. An example of fluorescence multi-colour imaging is shown in Fig. D2 for a maize plant. Some measurements on a single branch of spruce were performed by the different groups. Since the various groups employed different laser sources, different wavelengths were used. With our system it was easy to change wavelength, using different Stokes and anti-Stokes components from the Raman shifter. We thus had the opportunity to try most of the wavelengths used by the other

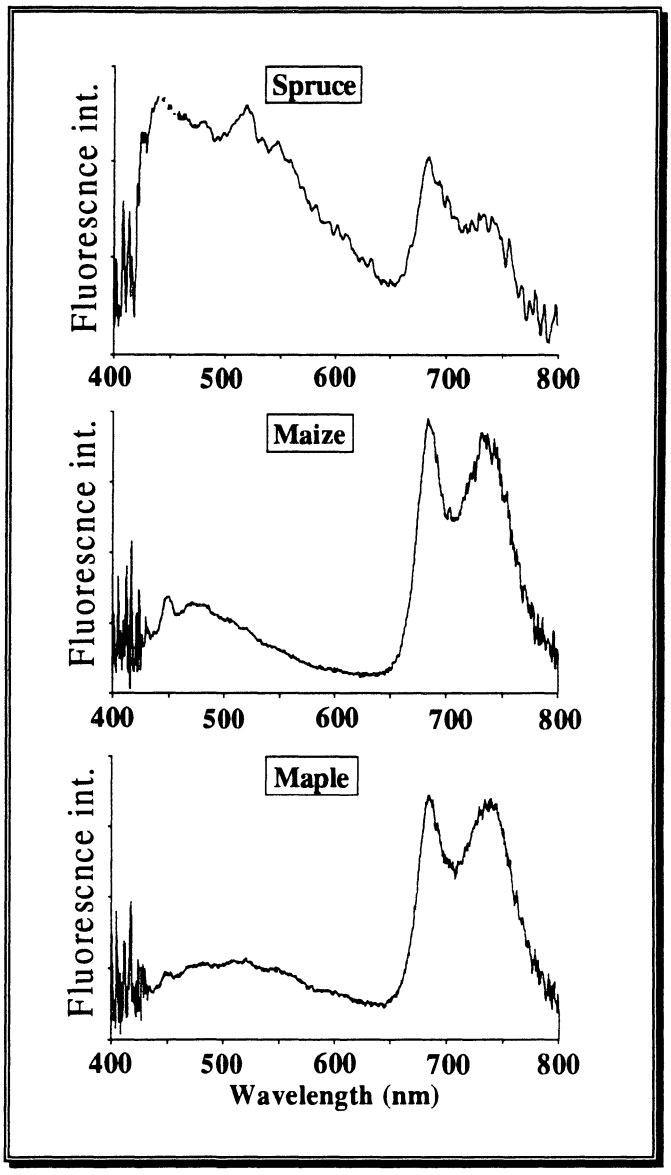

Fig. D1.
Remote fluorescence spectra of spruce, maize and maple. The excitation wavelength was 397 nm and the target distance was 40 m.

groups. In conclusion, the fluorescence ratio I(685)/I(740) was found by all groups to be sensitive to the chlorophyll concentration and to some extent also to ambient light conditions. The blue fluorescence seems to give additional information but is, however, very complex and at the moment not very well understood.

The third week of the campaign was spent in Italy measuring water fluorescence together with an Italian group from IROE, Florence (G. Cecchi, L. Pantani). The first measurements took place at a pier in Trieste from where the Mediterranean could easily be reached. Measurements on the contaminated water close to the harbour were performed. The last few days were spent at Punta Sabioni outside Venice, on the opposite side of the lagoon. The water flows in and out with the tide, and the difference between the clear water from the Mediterranean and the water from inside the lagoon was quite clear. A full daily cycle of the water was measured. Some imaging measurements were also performed on different algae.

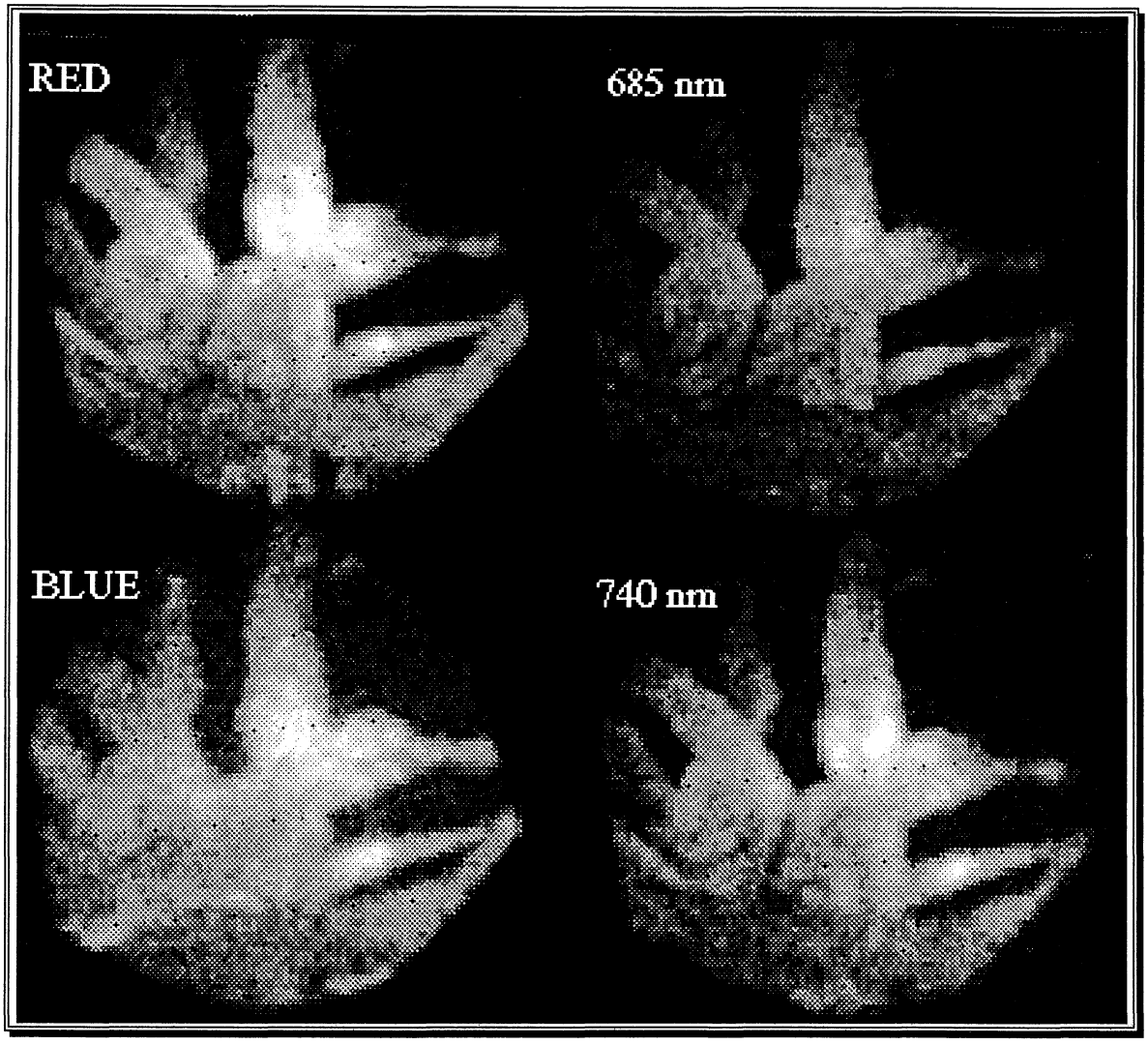

Fig. D2.
Image of maize at 40 m distance. Interference filters at 685 and 740 nm were used as well as a broadband blue and red glass filter, respectively.

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IV Laser Applications in Medicine and Biology

Since the introduction of medical laser applications at the Division of Atomic Physics in 1982 work has been carried out in close collaboration with the Lund University Hospital. This collaboration has now taken on a more organised form as it resulted in the formation of the Lund University Medical Laser Centre in 1991. The aim of this organisation is to support interdisciplinary projects using lasers in medical research between the Medical, Natural Sciences and Engineering faculties 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 Division of Atomic Physics has been very central in this collaboration and in the formation of the Lund University Medical Laser Centre.

The first technique for tissue diagnostics that we investigated in this project was laser-induced fluorescence (LIF) to identify malignant tissue. Both the native tissue autofluorescence originating from endogenous fluorophores as well as fluorescence from administered tumour markers, mostly porphyrins, have been included in the studies. Photodynamic therapy (PDT) of tumours using a photochemical reaction involving administered porphyrin and tissue-bound molecular oxygen has also been investigated. Later studies involved the characterization of the fluorescence of lesions of various heart diseases and potential applications have been identified. More recently, a technique for time-resolved tissue transillumination has been suggested for early detection of malignant breast tumours. Now our research is focused on clinical implementation and evaluation of the techniques developed, to improve the basic understanding of light transport within tissue, including tissue-simulating phantom studies and modelling of light distribution, as well as the study of new alternative laser-based diagnostic techniques, e.g. phase-modulation and Raman spectroscopy and to develop instrumentation for spectroscopy in combination with confocal microscopy.

Our recent work has been presented in a large number of invited talks and review articles [1-8]. One PhD thesis has been presented within the frame of the project during 1991/92 [9], and a member of the group has spent one year in Canada as a post-doctoral fellow.

A *In vivo* and *in vitro* tissue characteristics using LIF

The potential of fluorescence in tissue diagnostics has been investigated in clinical studies including the spectral features of tissue autofluorescence as well as porphyrin signals from low-dose Photofrin-injected patients. Astrocytomas in patients undergoing brain surgery and malignant lesions in the bronchial tree and urinary bladder have been investigated. Furthermore, the potential of various new tumour marking agents/photosensitizers in fluorescence diagnostics and photodynamic therapy has been studied in experimental tumour models in rodent tissues.

A1 Clinical measurements

Stefan Andersson-Engels, Roger Berg, Jonas Johansson and Sune Svanberg

Using a fluorosensor based on optical-fibre light guiding, 20 patients with brain tumours were examined for tissue fluorescence characteristics during neurosurgery, 25 patients with endobronchial tumours were examined during bronchoscopy and about 30 patients with tumours in the urinary bladder were studied during cystoscopy. The aims of the studies were to investigate the potential of fluorescence as a tool to diagnose tissue as malignant or normal in real time, and to investigate the affinity of Photofrin to malignant tissue. The latter is of interest both for tissue diagnostics using fluorescence and for photodynamic therapy. Two excitation wavelengths were used, 337 nm from a nitrogen laser giving rise to strong tissue autofluorescence and 405 nm from a dye laser matching the peak excitation of Photofrin. Five or more spectra at various locations were recorded from each patient. During a recording, which takes about 5 seconds, the tip of the optical fibre was placed in contact with the tissue being examined. The tissue volume sampled with this method is very small, due to the limited penetration depth of the excitation light (of the order of hundreds of micrometres, depending on wavelength and tissue type). When possible, a small biopsy sample was taken for every fluorescence recording, exactly at the position of the tip of the optical fibre. This made it possible to correlate the shape of the spectra with histopathological findings. On several occasions, the lung lobe containing the tumour of a patient having undergone bronchoscopy or the urinary bladder of a patient examined through a cystoscope was removed by surgery the day after the endoscopical examination. In these cases it was possible to study the excised tissue more carefully by means of fluorescence before fixation for histopathological examination.

It was found that the fluorescence from brain tissue is generally very weak compared with other organs. The shape of the autofluorescence spectrum varied with measurement site but no clear correlation with malignancy was found. The overall intensity of the autofluorescence was, however, weaker in tumour than in normal brain tissue [A1-3]. Porphyrin was found to accumulate selectively in the tumour region, but normal tissue close to the tumour border also showed an elevated porphyrin signal. This is probably due to leakage of the porphyrins through the extra-cellular fluid. This effect may be reduced by using a shorter time interval between administration and examination (in these studies 24 h). In the case of malignant lesions in the urinary bladder the autofluorescence was drastically reduced compared with the surrounding normal bladder wall, as can be seen in Fig. A1 [A4-7]. Dysplasia could also be detected by autofluorescence diagnostics alone. Porphyrins were accumulated selectively in malignant lesions. The same conclusions are valid for the bronchial tumours [A1-3, A8].

Studies of tumours in the oesophagus, similar to those described above, have recently been started. Furthermore, studies of excised tumour and surrounding normal tissues are in progress, mainly from breast tumours, tumours in the prostate gland and thyroidea.

Other clinically related studies are being focused on the identification of lesions in various heart diseases. Atherosclerotic plaques have been studied in both *in vitro* [A9-11] and *in vivo* [A11-12] studies. Also the main fluorophores of such lesions and normal artery wall have been identified and characterised in a separate study. Both fibrotic and fatty lesions can be identified with this method. A commercial prototype of an instrument for clinical identification of atherosclerotic plaque has been constructed by a spin-off company from the Division - Spectrophos AB. This instrument is now being clinically evaluated at the NIH hospital in Bethesda, USA.

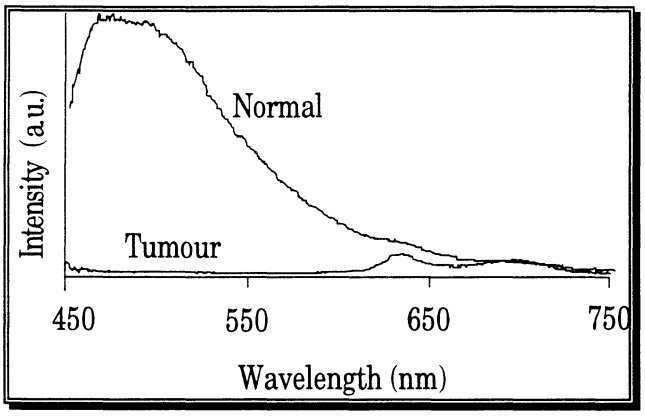

Fig. A1. *Fluorescence spectra excited at 405 nm from a urinary bladder cancer stage IV and normal surrounding mucosa. The patient received 0.35 mg/kg Photofrin 48 h prior to the cystoscopic examination.*

Laser-induced fluorescence is also a potential tool for the identification of damaged heart muscle in clinical cardiology. It would be helpful to have a tool that could identify morphological structures in the heart muscle in real-time, which are critical to the existence of arrhythmia. Another potential application is the identification of allograft rejection in heart transplants. The idea that such abnormal scar lesions could be identified by laser-induced fluorescence initiated a pilot project to investigate the potential of the method [A13]. Heart tissue samples collected during open-heart surgery were examined by fluorescence spectroscopy and the results were correlated with histopathological findings. The results showed that scar tissue as well as fatty lesions could be identified by this method. *In vivo* studies during open-heart surgery are now being planned.

A2 Experimental diagnostics and therapy

Stefan Andersson-Engels, Roger Berg, Jonas Johansson and Sune Svanberg

Fluorescence studies have been carried out on various rodent tissues to investigate tissue distribution of several new substances, which are potential candidates as tumour markers for fluorescence diagnostics and photosensitizers for photodynamic therapy. The studies have recently been focused on two very interesting substances - benzoporphyrin derivative mono acid (BpD-MA) and delta aminolevulinic acid (ALA). The fluorescence signals obtained with these new substances have been compared with those from Photofrin, the standard drug used clinically today. BpD-MA is a true second generation drug for PDT,

with a well-defined chemical structure and an absorption peak at 690 nm. The absorption peak in the far red makes it possible to treat thicker lesions than is possible with the 630 nm peak for Photofrin. This is due to better penetration of the 690 nm light in soft tissue. Another advantage of BpD-MA is that it is quickly metabolised in the body and 24 h after administration no BpD-MA fluorescence can be detected and the patient is no longer photosensitized. The results of this investigation show that the selectivity for malignant tissue of BpD-MA is only half that of Photofrin. This suggests that BpD-MA is better suited as a photosensitizer in PDT, for which a high selectivity of the drug is an advantage but not an necessity, than as a tumour marker for fluorescence diagnostics, which relies on a high selectivity [A14]. The other drug tested, ALA, is a precursor to heme in the production of heme in the cell mitochondria. The administration of ALA to living tissue results in an enhanced production of heme. The last step in the reaction chain which ends with heme, that from protoporphyrin to heme, is much slower than the previous steps. This means that protoporphyrin will be accumulated in the tissue. Protoporphyrin is photodynamically active and gives rise to strong fluorescence, and thus ALA can be used both for PDT and fluorescence diagnostics. Topical application of ALA is possible for superficial lesions. Systemic administration is being tested in animal experiments by our group [A15]. Results so far have shown that protoporphyrin is selectively accumulated in malignant tissue 30 minutes after *i.v.* injection of ALA. In conjunction with these studies a programme for chemical extraction of porphyrins from tissue samples was initiated [A16]. Studies of PDT effects of the drugs in a rat liver tumour model have also been initiated.

B Photodynamic therapy

Stefan Andersson-Engels, Roger Berg, Jonas Johansson and Sune Svanberg

Photodynamic therapy (PDT) is a malignant tumour treatment modality relying on the selective excitation of triplet to singlet oxygen mediated by a laser-excited sensitising drug. The drug is normally injected intravenously and is selectively retained in malignant tissue. Thus, selective necrosis of cancer lesions can be achieved. Laser-induced fluorescence (LIF) can be utilised to localise the tumour based on the specific drug fluorescence and native chromophore fluorescence. The only agent presently in common clinical use is a haematoporphyrin derivative, available under the trade mark of Photofrin. The drug is normally injected 48 hours prior to the laser treatment which follows at a wavelength of 630 nm. The only side-effect of Photofrin is skin sensitisation lasting about four weeks. Recently, a different PDT procedure for treating superficial tumours was been introduced. δ-aminolevulinic acid (ALA) prepared in solution is topically applied to the lesions 3-6 hours prior to the treatment. As described above, the ALA molecules pass the cell walls of the tumour cells and are intra-cellularly transformed to protoporphyrin following the normal heme cycle in the mitochondria. The photodynamic action is initiated following laser irradiation at 630 nm. This procedure does not cause any skin sensitisation. The present PDT activities at the Lund Medical Laser Centre involve the use of both Photofrin and ALA [B1-B5]. The treatment laser is a Technomed Multilase 2100 frequency-doubled Nd:YAG laser pumping a dye laser (Technomed Dye 600). The maximum output power at 630 nm is 2 W. The treatment procedure also includes recordings of LIF spectra before and after treatment. This is achieved with a PARC OMA III system employing a nitrogen-pumped dye laser at 405 nm as a light source. Fig. B1 shows a scan over an ALA-treated basalioma *pre* and *post* PDT. As can be seen, there is

significant fluorescence at 630 nm due to the protoporphyrin in the tumour. After the treatment this signal drops significantly due to the bleaching of the drug. The spectra also show that there is very little porphyrin in the normal tissue, i.e. the selective accumulation of protoporphyrin is very good.

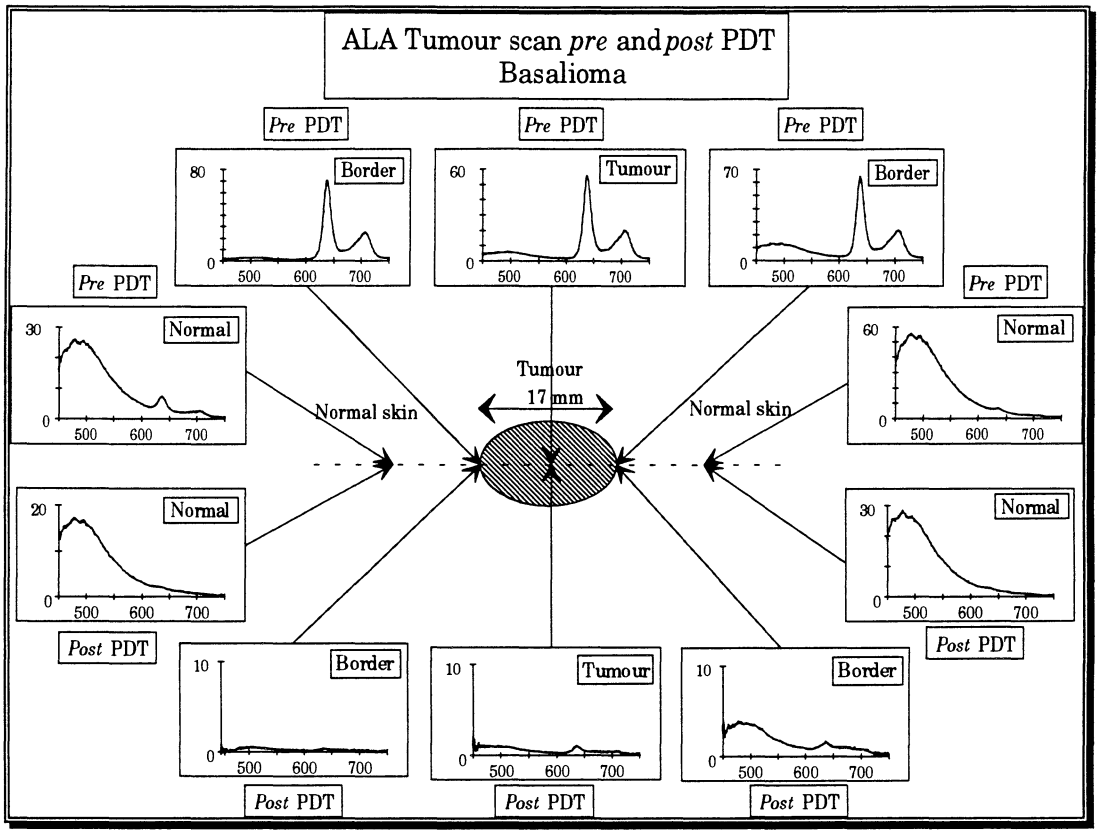

Fig. B1. *LIF spectra of an ALA-treated basalioma lesion. The upper curves show a scan over the lesion obtained pre PDT and the lower curves a similar scan obtained post PDT. The excitation wavelength was 405 nm.*

Some superficial lesions have also been investigated using a multi-colour fluorescence imaging system developed at the Division. This system uses a Cassegrainian telescope with a split mirror and can produce four images at different fluorescence wavelengths on an image-intensified CCD camera. Fig. B2 shows a multicolour fluorescence image obtained from an ALA-treated T-cell lymphoma. The upper images show the fluorescence at 630, 470, 670 and 600 nm respectively. The lower image is a processed image in which the fluorescence at 600 nm has been subtracted from the fluorescence at 630 nm and the result divided by the fluorescence at 470 nm. In doing this we obtain a "dimensionless" image where artefacts due to e.g. geometry and difference in illumination are eliminated. The tumour is located in the lower part of the image which can be seen clearly as a light area in the processed image.

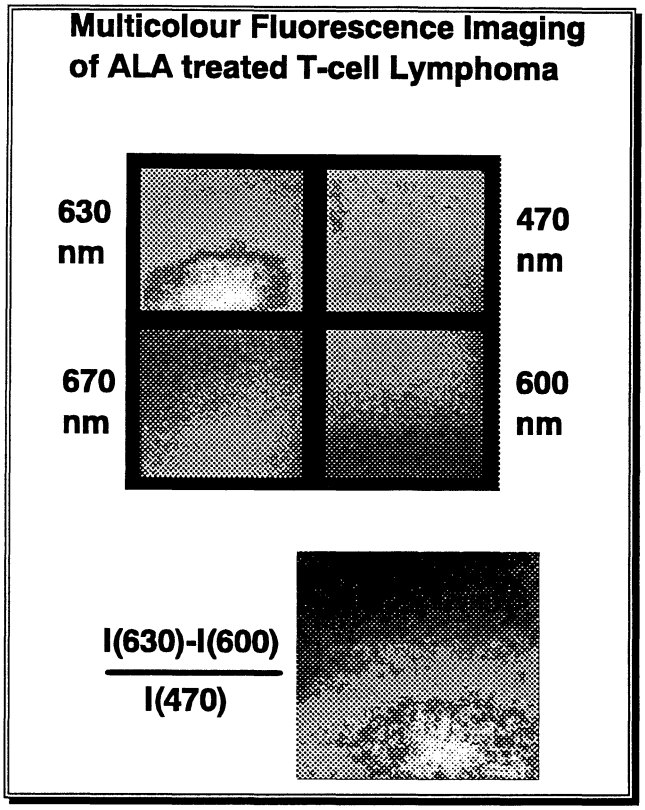

Fig. B2. Multicolour fluorescence image of ALA-treated T-cell lymphoma. The upper images show the fluorescence at 630, 470, 670 and 600 nm, respectively. The lower image is a processed image. The tumour is located in the lower part of the frame. The excitation wavelength was 405 nm.

To date, more than 30 patients with about 140 lesions have been treated in Lund, with ALA and Photofrin PDT. The superficial lesions that have been treated are basalioma, squamous cell carcinoma, cutaneous T-cell lymphoma and breast cancer metastasis. Two patients with psoriasis plaque have also been treated with ALA. Two patients with lung cancer have been treated during bronchoscopy.

C Tissue transillumination

Stefan-Andersson Engels, Roger Berg and Sune Svanberg

When light enters tissue it undergoes a very high degree of multiple scattering. This effect prohibits detailed visual observation inside the tissue. Tissue is characterised by a high scattering coefficient ($\mu_s \approx 10$ mm^{-1}) and low absorption coefficient ($\mu_a \approx 0.01$ mm^{-1}) in the wavelength region of interest, i.e. 600-1000 nm. Traditional light transillumination (diaphanography) suffers from difficulties in detecting tumours located deep inside the tissue. Recently, several different optical techniques have been suggested to improve the detection of features inside turbid media, such as tissue. We have used a time-gated viewing technique in order to suppress multiply-scattered light and thus improve spatial resolution [C1-C4]. The tissue is irradiated with short laser pulses and the transmitted light is detected with time resolution. By gating out the light with the shortest transit time, the multiply-scattered light can be suppressed. We have demonstrated that in the case of scattering-dominated attenuation (scattering coefficient >> absorption coefficient) detection of early transmitted light is very sensitive to variations in the scattering coefficient.

As light sources we have used a mode-locked Ar-ion-pumped dye laser (6 psec) or a near-infrared (815 nm), pulsed (30 psec, 10 MHz) diode laser [C5-6]. The laser light irradiated the sample and the light was collected on the opposite side, guided through an optical fibre, and detected by an MCP-PMT. To achieve time-resolved detection, standard delayed-coincidence techniques were used. The temporal resolution of the system was approximately 60 psec (FWHM). A small part of the laser light was bypassed the sample and was led directly to the detector in order to create a short reference pulse. This reference pulse was added to the temporal dispersion curves to allow compensation for temporal drifts in the electronics. The laser and the detector fibre can be scanned over the sample in two dimensions using computer-controlled stepping motors. Fig. C1 shows a typical temporal dispersion curve obtained when transilluminating a 35 mm thick slab of female breast *in vitro*. The impulse response of the system is also shown in the figure. As can be seen, the transmitted light pulse has been broadened due to considerable multiple scattering in the tissue. A typical time gate of 120 psec is also indicated in the figure.

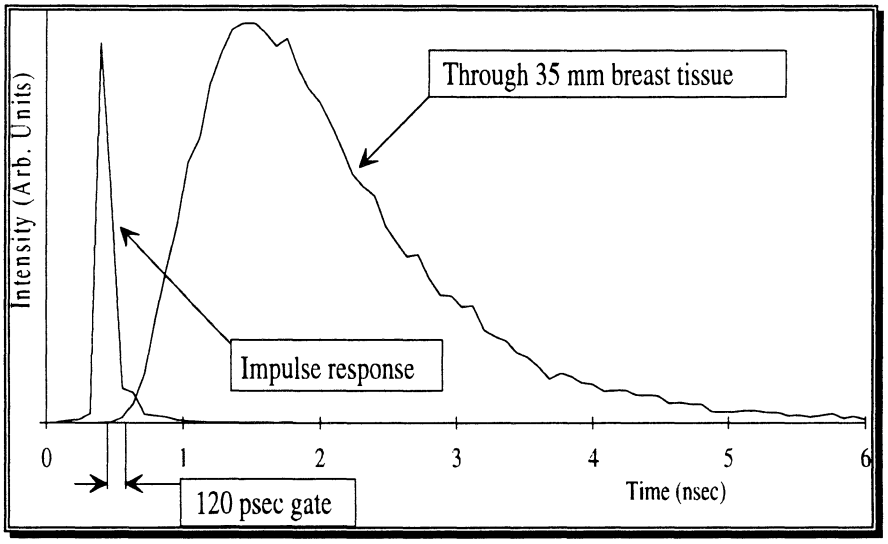

Fig. C1. *Temporal dispersion curve and impulse response obtained when transilluminating a 35 mm thick sample of female breast in vitro. The two curves are normalised.*

Fig. C2 shows a scan along the surface of a 16 mm thick sample of a female breast *in vitro* containing a tumour with a horizontal diameter of 18 mm (ductal invasive carcinoma). The sample was gently compressed between two glass plates to an even thickness. A temporal dispersion curve was recorded every 2 mm. The solid curve in the figure shows the light detected during the first 120 psec and the dashed curve shows the total light intensity at every point. As can be seen, there is a distinct demarcation of the tumour in the gated light signal but nothing can be seen in the total light intensity. The optical properties of the tissue can be estimated by fitting a model based on the diffusion equation to the obtained temporal dispersion curves, and the explanation of the increase in the gated light over the tumour can be found in the lower scattering coefficient in the tumour tissue compared with the surrounding healthy tissue.

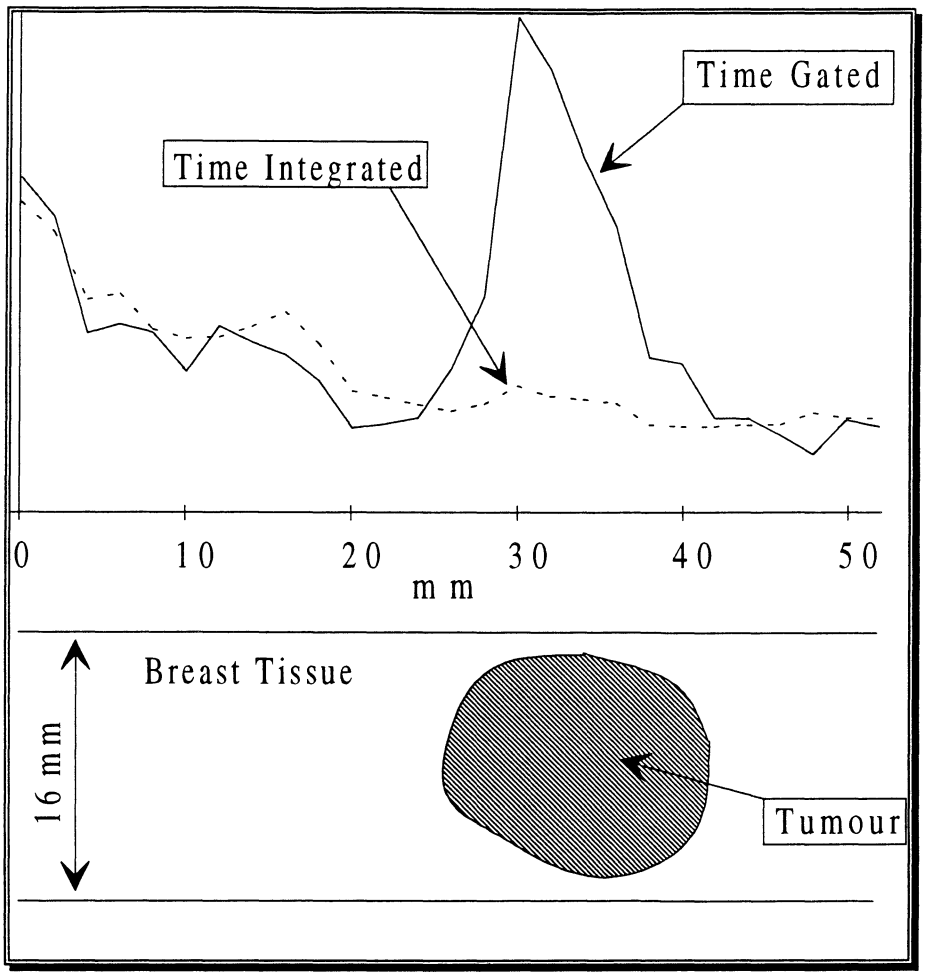

Fig. C2. *A scan across a 16 mm thick sample of breast tissue containing a tumour (18 mm horizontal diameter). The solid curve is the light obtained during the first 120 psec of every temporal dispersion curve. The dashed curve is the total light intensity at every point.*

In order to simulate a tumour with a lower scattering coefficient compared with the surrounding tissue, a phantom was made, consisting of pure animal fat with a transparent plastic disc inserted. The size of the phantom was 100×100×30 mm and the plastic disc was 15 mm in diameter and 4 mm thick. The disc was placed in the middle of the fat. A 2-dimensional scan was performed over a 60×40 mm area in steps of 2 mm. Fig. C3 shows the outline of the phantom (left), the light detected during the first 160 psec (middle) and the total light detected (right). As can be seen in the figure, the plastic disc can easily be detected with the time-gated technique, while there is no sign of the plastic disc at all in the total light image.

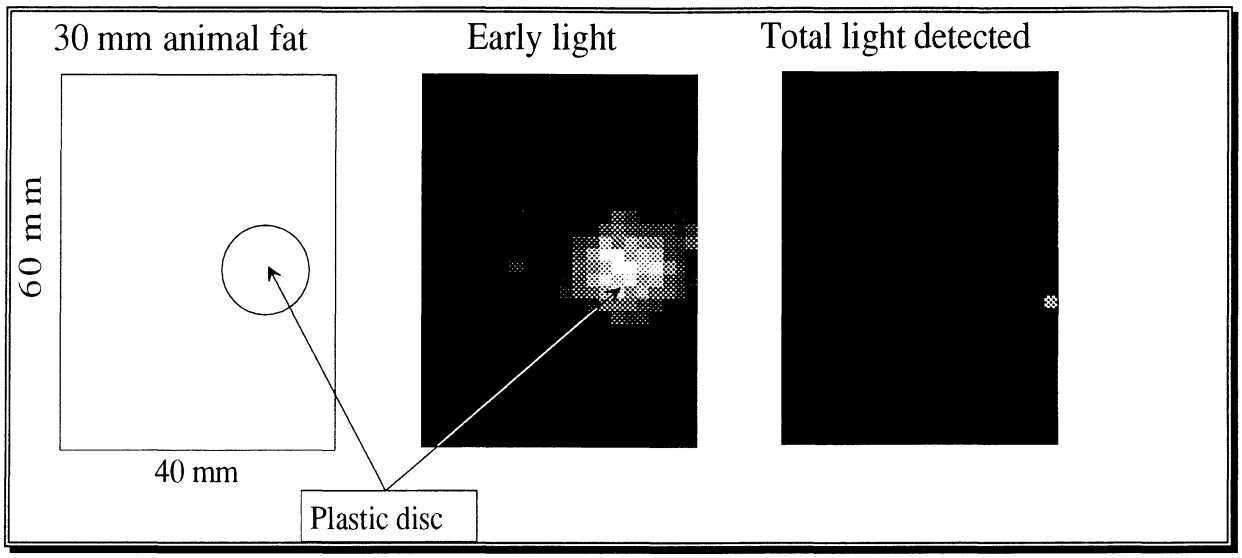

Fig. C3. *Tissue phantom (30 mm thick) consisting of animal fat with a transparent plastic disc inside (15 mm in diameter and 4 mm thick). Left: outlines of the phantom. Middle: light detected during the first 160 psec. Right: total light detected.*

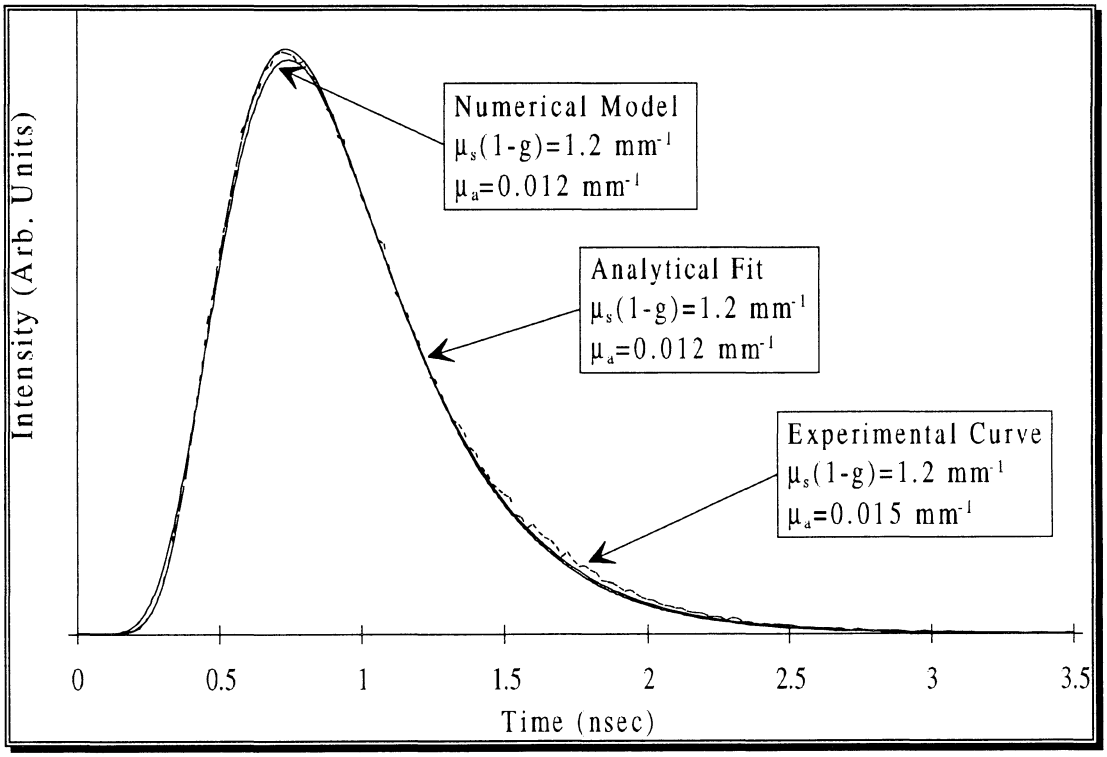

Fig. C4. *A comparison between an experimental curve, the analytical solution to the diffusion equation and a solution obtained with the numerical computer model. The results are from the transillumination of a 30 mm thick homogeneous sample.*

To describe light propagation in turbid media where the scattering coefficient is much larger than the absorption coefficient, the diffusion equation can be used. We have developed a computer model, which solves this partial differential equation numerically as a function of time. By doing this in a discrete form it is possible to solve problems with arbitrary geometrties and boundary conditions. Figure C4 shows a comparison between the numerical solution, obtained with the computer model, an experimental curve and an analytical solution to the diffusion equation. In this case a homogeneous, semi-infinite tissue slab was transilluminated and that is why it was possible to obtain an analytical solution to the problem. The computer model has been used to verify our findings that the "early" light is most sensitive to the scattering coefficient and the "later" light to the absorption coefficient [C7-8].

We have shown that the time-gated technique can be used to detect tissue features that can not be observed with steady-state light transillumination. Normal time-integrated transillumination is much less sensitive to variations in the scattering coefficient.

D New techniques under development

D1 Raman spectroscopy

Stefan Andersson-Engels, Ulf Gustavsson and Sune Svanberg

Raman spectroscopy provides much more information about a tissue sample than fluorescence, since sharp spectral lines result rather than broad fluorescence features. The major limitation with 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 requires limited excitation power. These limitations have made Raman spectroscopy a less attractive tool in tissue diagnostics. However, it has recently been demonstrated that by using diode-laser excitation in the near-infrared wavelength region, only weak fluorescence results and a Raman spectrum with an acceptable signal-to-noise ratio can be acquired with a realistic recording time and excitation power. Since the potential for this method is very good, we have assembled an experimental system based on NIR diode laser excitation and cooled CCD-camera detection to measure Raman signals from tissue [D1]. Preliminary results suggest that we can record Raman spectra from tissue with acceptable quality. Investigations aimed at the study of the discrimination between diseased and normal tissue have been initiated.

D2 Phase-modulation spectroscopy

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

Time-resolved spectroscopy of tissue has proven to be a useful tool both for fluorescence and for diffuse reflection/transmission measurements. Phase-modulation spectroscopy can give similar information, but often requires simpler instrumentation. Therefore, we have started a project aimed at measuring diffuse transmitted light using phase-modulation spectroscopy. An instrument based on a modulated diode laser source and RF-mixer on

the detection side has been assembled [D2]. Modulation of the gain of an image intensifier is being considered to allow direct imaging of phase-modulated light.

D3 Measurements of the optical properties of tissue using diffuse reflection/transmission

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

In many of the applications of interest for lasers in medicine it is crucial to know the optical properties of the tissue being treated. If the light distribution can be approximated with diffusion theory it is in some cases possible to measure the optical coefficients with spatially or temporally resolved diffuse reflection/transmission measurements. However, for all other cases it is not easy to measure these parameters directly. For such measurements it is necessary to collect a small tissue sample. The optical parameters can then be measured with an optically integrating sphere. Monte Carlo simulations of the light distribution for the geometry used then have to be performed for different optical properties to find the best fit to the measured signals. Such an instrument has been assembled at our laboratory and is now being calibrated and tested.

D4 Confocal microscopy

Stefan Andersson-Engels

Confocal laser scanning microscopy is of increasing importance for biomedical research, due to its unique ability to produce sharp images. High-quality fluorescence confocal microscopy of porphyrins and other comparatively weakly fluorescent substances is difficult to perform on commercial units. This is due to the high background from tissue autofluorescence which disturbs the images. These effects can be minimised if spectroscopy is combined with confocal microscopy, making it possible to drastically reduce the influence of other fluorophores. This is fairly simple to achieve, since a confocal microscope works in a scanning mode, *i.e.* is basically a point-monitoring instrument. An instrument based on this idea is being assembled in a collaboration with the Department of Physics at the Royal Institute of Technology, Stockholm. The first measurements are planned for late 1992.

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V Industrial Applications

During the last two decades new sources of light, detectors, fibres, materials, sensors etc. have widened the areas of application of optical measuring techniques. At the same time, industrial processes have grown in speed and complexity, leading to a never-ceasing demand for fast, accurate and detailed on-line process information. Modern optical techniques seem well apt to provide the type of information desired, particularly in often very harsh industrial environments, as they provide an extremely fast, perturbation-insensitive, remote-sensing technique.

At the Division of Atomic Physics close R&D contacts have been established with various process industries within and outside Sweden. During the last five years new methods for optical-spectroscopic production control of smelt-metallurgical processes have been developed in co-operation with copper- and steelmaking companies. The geogas project has been in progress for more than ten years, with partners all over the world, and the understanding of the complex geogas phenomenon is steadily growing.

Two new industry-related projects have been initiated during the last two years. Laser-spectroscopic methods for the study of the physics of electric breakdown in dielectric liquids are being developed in collaboration with ABB. An investigation of the potential of optical-spectroscopic methods for production control in paper-making is being carried out in co-operation with the major Swedish newsprint-producing companies.

Diode lasers in the near infrared region are relatively inexpensive and easy-to-handle. The high sensitivity of two-tone frequency modulated diode laser spectroscopy is taken advantage of in a project aiming at developing methods for the detection of gas species at low concentrations.

A Applications of optical spectroscopy in the metallurgical industry

Lennart Malmqvist, Willy Persson and Wilhelm Wendt

The studies described under this heading have been carried out in collaboration with Semtech Metallurgy AB at the Ideon Research Park.

Spectroscopic observations through the oxygen lance of the SSAB Oxelösund LBE converter have been continued. Recordings have been made during several hundred process cycles in order to verify the relations between the spectral information from the hot spot, created by the injected oxygen, and relevant process parameters. As a result the carbon content of the steel can be determined on-line with a precision of ±0.005% C in the interval 0.120-0.015% C. The spectroscopic observations also indicate the possibility of determining the lance position during blowing.

As reported earlier, the optical-spectroscopic technique for production control is in permanent use in the CLU (Cresot-Loire-Uddeholm) process for alloy steel production at Degerfors Jernverk. The AOD (Argon-Oxygen-decarburization) process for alloy steel production uses argon and oxygen during the various decarburization steps instead of the superheated steam used in the CLU process. Recent measurements at a top-blown AOD converter (i.e. oxygen is injected via a lance ending just above the melt) at ALZ in Belgium show that it is also possible to monitor the light emitted by MnO in the off-gases throughout the full reduction step also at an AOD. As for the CLU there is a good correlation between the emitted MnO intensity and the Cr_2O_3 content of the slag (Fig. A1).

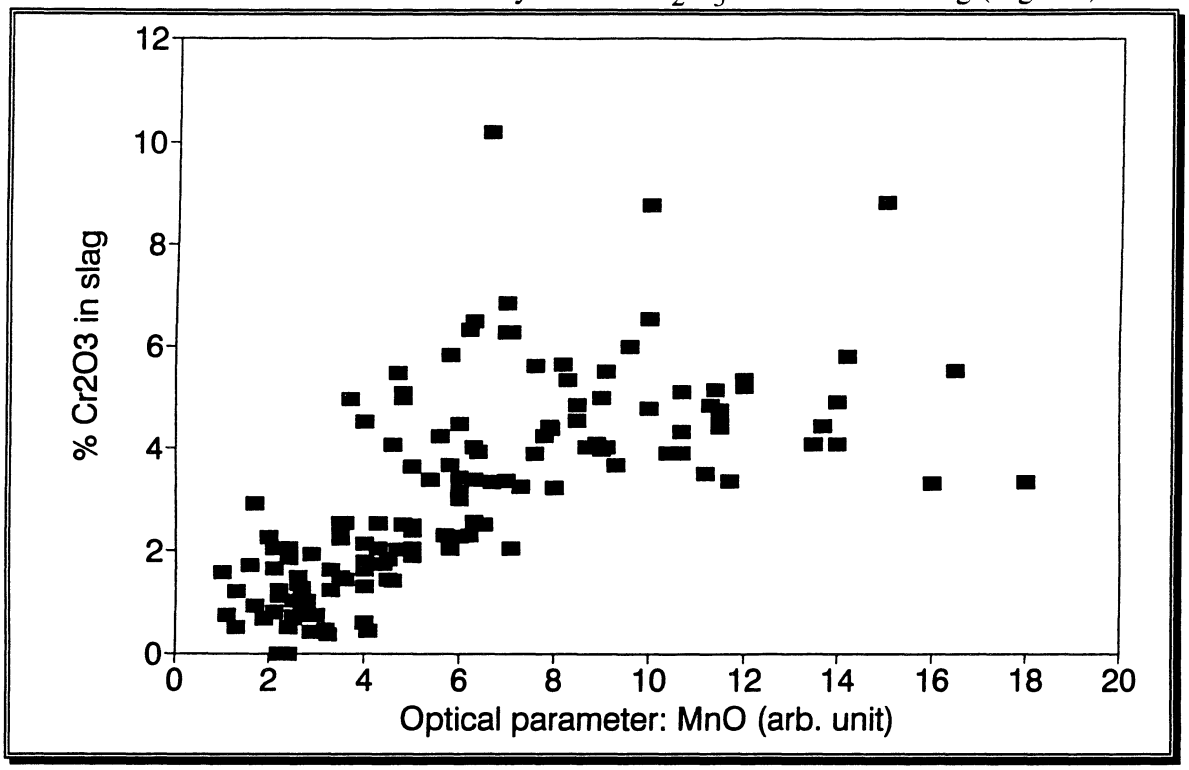


Fig. A1
Chromium oxide content of the converter slag vs. optical parameter.

As long as the optical signal remains above, say, 7 units the Cr_2O_3 content of the slag will, without exception, be above 3% and the reduction is incomplete. The decision which must be taken is to extend the reduction step for a couple of minutes or, if the optical signal is no longer decreasing, to add silica. Thus, by using optical on-line information it is possible not only to make substantial savings in the alloying-metal consumption but also to minimise the content of hazardous heavy metals in the slag, which is of environmental concern.

A new series of investigations of the copper process at Boliden Rönnskärsverken has just been completed. The converter process includes two major steps, namely the slag-forming stage, which aims at raising the copper content of the melt from ≈40% to ≈76-77% Cu, and the copper-making stage, the purpose of which is to produce blister copper (98.5-99.5% Cu).

Five years ago we showed that there is good correlation between the optical spectra of PbO and PbS in the off-gas flame and the copper content in the melt close to the end of

the slag-forming stage. Optimisation of both hardware and software in the optical system has led to improved accuracy in the end-point determination of the slag-forming step as well as additional on-line information on the status of the converter process. At present, it is possible to determine on-line the copper content of the melt with a precision of ±0.2% Cu in the interval 74-78% Cu.

The SiO_2 content of the converter slag determines the impurity capacity of the slag and also its viscosity. These are considered to be critical process parameters. New spectroscopic studies indicate the possibility of making on-line determinations of the slag composition by monitoring the off-gas flame spectrum.

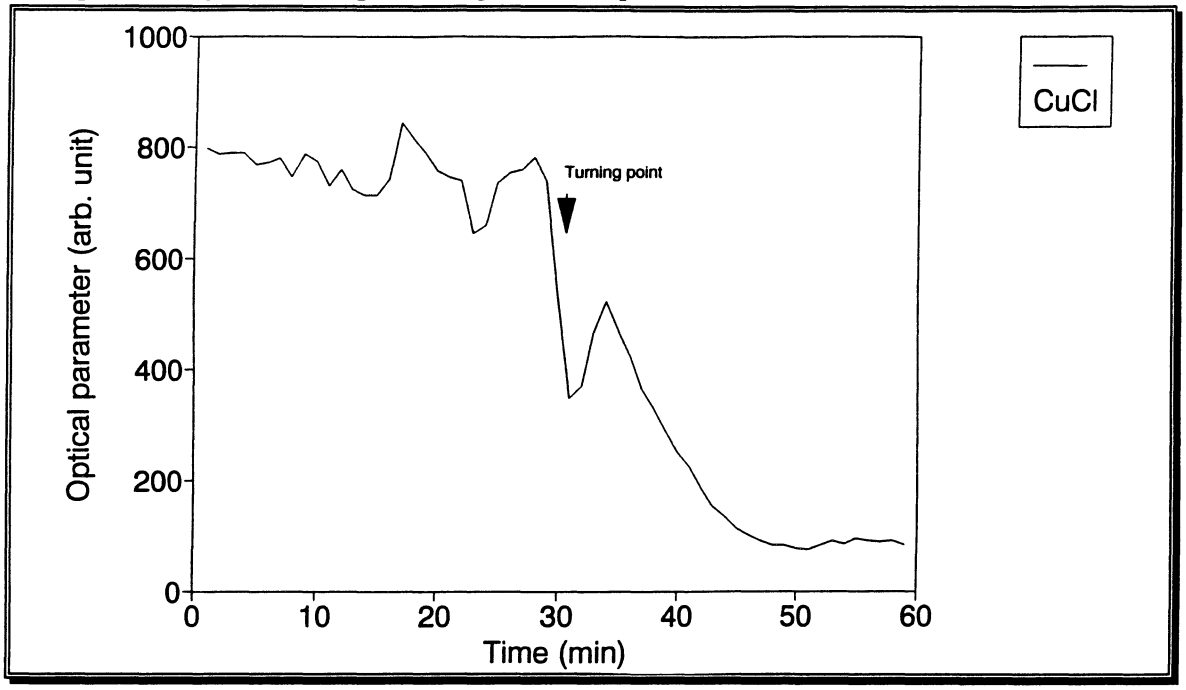


Fig. A2
Intensity of CuCl emission vs. time during the copper-making stage.

During the second converter step, the copper-making stage, the white metal (Cu_2S) produced during the slag-forming step is oxidised to pure copper and large amounts of SO_2 are produced (cf. below C. Mercury monitoring). The sudden disappearance of the white metal phase is generally named the turning point of the converter. From this moment on, CuO is formed, which, of course, represents a loss in production. However, in order to remove the sulphur dissolved in the copper, and impurities such as lead and antimony, it is necessary to continue the blowing for another 15-20 minutes.

During the copper-making stage a number of strong CuCl bands are present in the spectrum of the converter flame. The typical time dependence of the intensity of the CuCl spectrum during the last part of the copper-making stage is shown in Fig. A2. At the turning point there is a sharp decrease in the CuCl emission intensity. This is likely to be due to the difference in solubility of CuCl in white metal and in pure copper. These observations indicate the possibility of estimating on-line the optimum endpoint of the copper-making stage, thereby eliminating unnecessary production of CuO, by monitoring the CuCl molecular spectrum in the off-gas flame of the converter.

B Paper characterisation by optical techniques

Håkan Bergström, Jörgen Carlsson, Lennart Malmqvist, Willy Persson and Claes-Göran Wahlström

The studies described in this section have been carried out in collaboration with the Swedish Newsprint Research Centre (TFL).

The time required for a light pulse to pass straight through a sheet of newsprint paper is 0.3 ps. In reality, this time is somewhat longer due to scattering. To study the scattering of light in newsprint a time resolution in the picosecond range is desirable. Fluorescence from paper has much longer time constants, typically 1 ns.

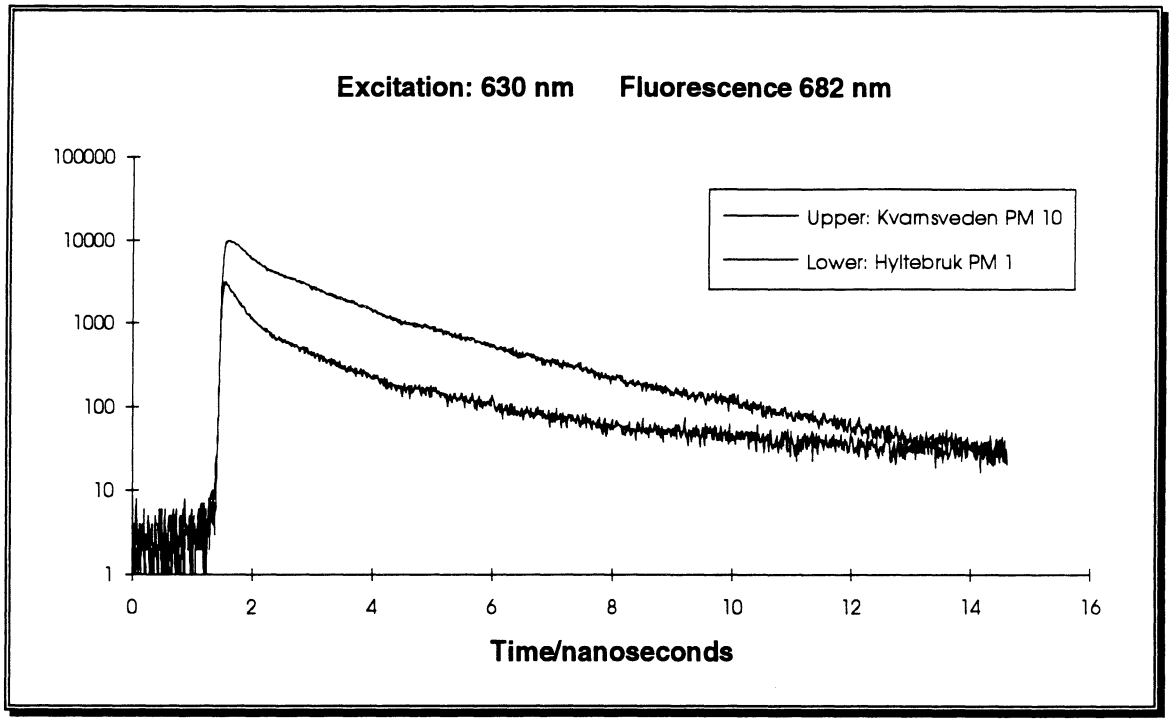


Fig. B1
Time-resolved fluorescence at 682 nm from two different newsprints, following excitation at 630 nm.

The time-resolved fluorescence from newsprint has been studied with the same single-photon counting technique as is used for lifetime measurements on atoms and which is described in Chapter I section C2. In Fig. B1 recordings from two different newsprint papers are shown. The main difference in quality between the two papers is that the one corresponding to the lower curve contains 50% recycled paper while the other has no such contribution. The technique used allows a time resolution of the order of 50 ps.

To study the scattering of light in newsprint a streak-camera is used as detector. The recordings in Fig. B2 were made using a mode-locked continuous Ti:sapphire laser giving 0.1 ps pulses at a 75 MHz pulse repetition rate. By doing single-shot experiments, using the new terawatt laser, a reduction in the width of the response function from 7 to 1-2 ps should be possible. This is desirable since the experiments indicate that the effect of scattering in one sheet of newsprint is of the order of 2-3 ps.

In addition to the time-resolved fluorescence studies we have also performed wavelength-resolved investigations of the fluorescence from newsprint paper. Considerable asymmetry in the fluorescence from the front and the back of the paper have been observed. Variations in the fluorescence properties along the paper web in a paper-making machine may be used as a means of on-line production control.

To examine whether the varying fluorescence properties can be related to surface-density variations or formations we have made simultaneous optical measurements and measurements using a β-densitometer. These measurements are presently being evaluated.

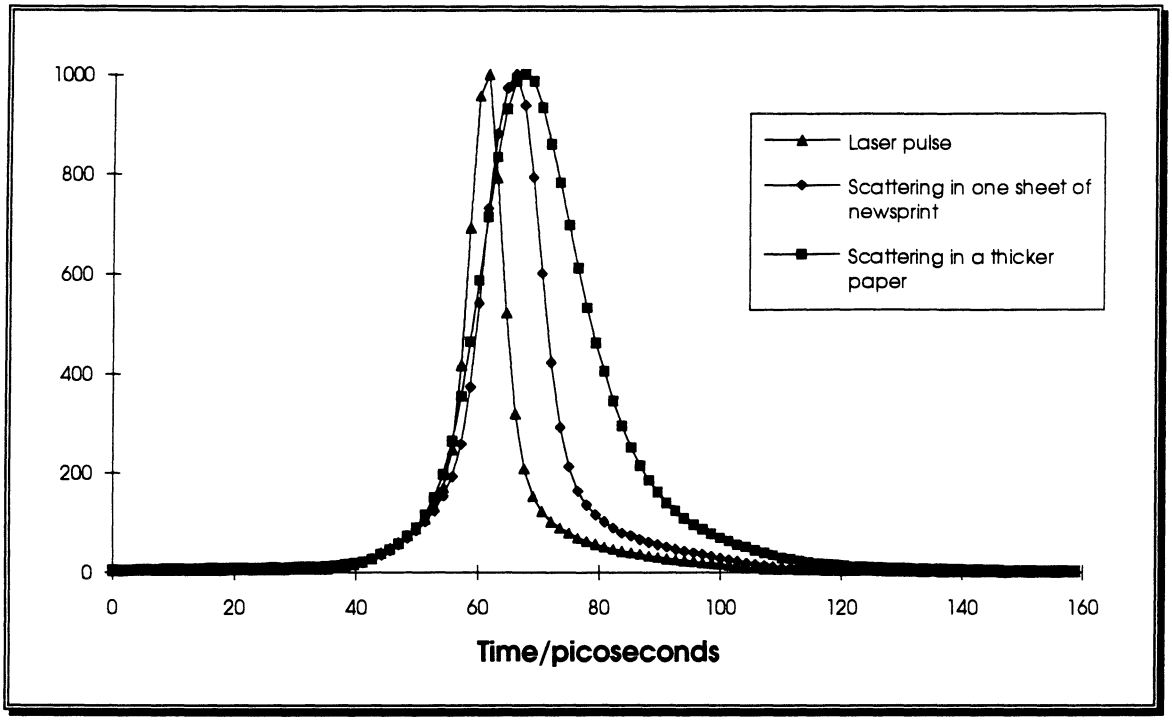


Fig. B2.
Time-resolved recordings of light scattered in one sheet of paper.

The fluorescence was excited by UV pulses from a nitrogen laser. To facilitate the extensive fluorescence measurements PC software has been developed, which controls the OMA via GPIB and also the XY-table carrying the test sheet. A set of data corresponding to a 21*21 matrix of measuring points on both sides of the paper requires a disk space of 3.6 MByte.

A third important aspect of the interaction between light and newsprint is the transmission properties of the paper. To study these an experimental set-up has been constructed and tested at the paper machine winder LINDA at TFL. The system consists of a PC-based A/D card with four channels which is capable of sampling at a rate exceeding 1 MHz. Powerful data analysis techniques are used for on-line analysis of the vast amount of data generated.

The transmission measurements aim at providing two types of information. Analysis of the data in the frequency domain may yield information on periodic variations in the paper

manufacturing process while statistical analysis of the data may provide on-line information on the formation index.

C Mercury monitoring in industrial processes

Willy Persson and Wilhelm Wendt

After a series of functional tests the Zeeman spectrometer for on-line monitoring of Hg^0 emissions, developed in collaboration with Semtech Metallurgy AB at the Ideon Research Park, is now in continuous operational use in a number of different industrial processes and for different purposes. It has been developed into a rugged instrument (Fig. C1.) which can withstand the often harsh environments in process industries. The lower detection limit of the instrument is 1μg Hg^0/m^3. Work is in progress to lower this limit by a factor of approximately 5 in respond to demands from, e.g. the off-shore industry.

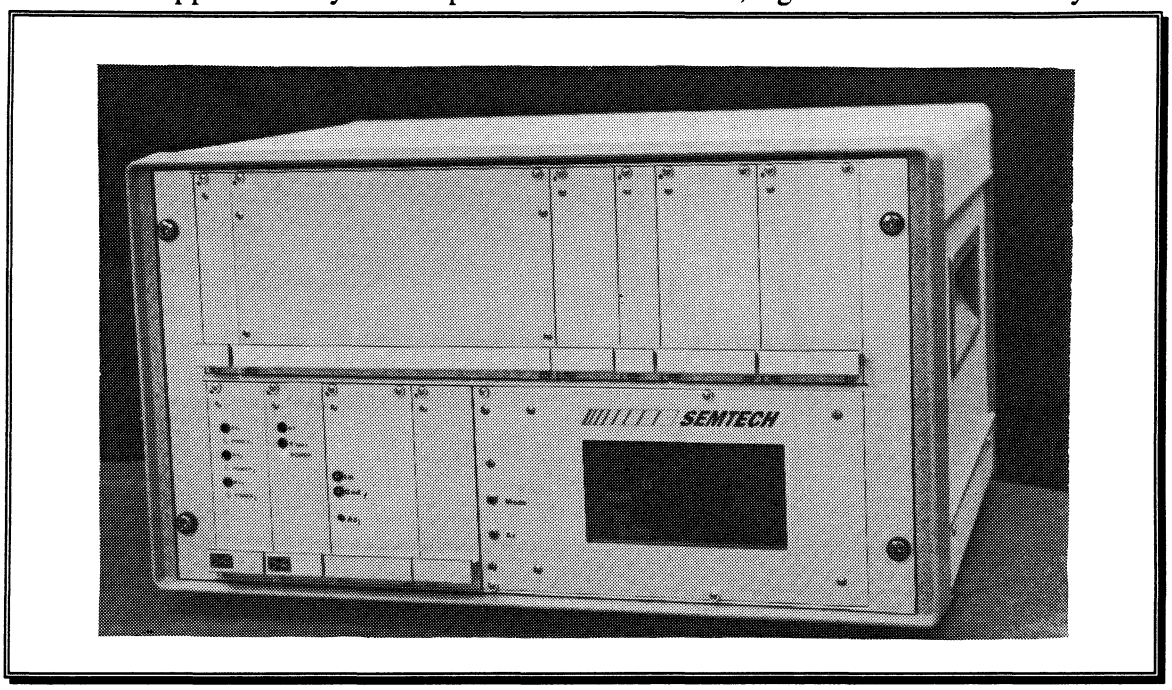


Fig. C1.
Zeeman spectrometer for on-line monitoring of Hg.

In a project supported by the Swedish Environmental Protection Agency aimed at continuous on-line measurements of elemental mercury in off-gases, the first permanent installation was made at a plant (Scandust AB, Sweden) for the recovery of metals from particle filters from the alloy steel industry. The results of the on-line measurements show excellent agreement with data from chemical analyses of samples taken from the process gases.

In a collaboration with Jernkontoret a study of the mercury emission from two electric-arc furnaces for scrap smelting was performed in October 1991. The point measurement was after the filters in the off-gas channel. In general, the emission is low during smelting but rises sharply to very high values (≈500μg/Nm^3) in connection with scrap additions. The mercury peak has a duration of about ten minutes.

Due to the growing concern about our environment, the very large quantities of SO_2 produced in pyrometallurgical activities such as copper, lead and zink production are, to an increasing extent, being converted into sulphuric acid instead of being released into the air. This has led to a saturated market for sulphuric acid and the best strategy for competition in the market is to produce a very pure acid. In particular, there is great concern about the mercury content in the acid. The acceptance level of mercury today is about 1 ppm but will probably decrease in the near future. It is thus becoming increasingly important to continuously monitor the Hg^0 content of the process gases to ensure the product quality. For this purpose Zeeman spectrometers have been installed in, e.g. a copper production plant at Norddeutsche Affinerie, Germany and in a zink production plant at Norzink a/s, Norway.

In some processes the chemical composition of the process gases, e.g. a high chlorine content, results in the appearance of oxidised forms of mercury in addition to Hg^0. This is true, for instance in waste incineration plants. In order to be able to measure the total mercury content of such process gases a unit has been developed for on-line reduction of oxidised mercury to elemental mercury, which can then be measured by the Zeeman spectrometer. This development has been carried out in collaboration with the Department of Inorganic Chemistry at Chalmers University of Technology. The first unit of this type has recently been installed at an incineration plant for hazardous waste (SAKAB). The primary purpose of the system is to monitor the mercury content of the off-gases but it is also used for tracing mercury back to individual "raw" material deliveries and for optimising the carbon injection and the filters used for off-gas purification.

In the off-shore industry, mercury contamination of natural gas deposits is a serious technical problem as mercury causes corrosion, even at low concentrations (~10$\mu g/m^3$), besides being a pollutant. The applicability of the Zeeman spectrometer in this context is presently being evaluated.

D Trace element emissions from concealed mineralizations

Lennart Malmqvist, Willy Persson and Wilhelm Wendt

The programme concerned with the localisation of concealed ore deposits has continued. More than 2000 geogas sensors have been exposed at various sites and analysed using the PIXE technique. This work is carried out in a collaboration between the Divisions of Atomic Physics and Applied Nuclear Physics (Prof. Krister Kristiansson) and Semtech Resources AB at the Ideon Research Park. During the last year attention has been focused on the interpretation of the geogas observations in geological terms.

In Japan, reconnaissance and detailed geogas surveys have been performed in co-operation with the Metal Mining Agency of Japan. This programme has paid particular attention to gold exploration problems related to concealed targets. Continuous efforts are being made to improve the detectability of gold in geogas samples.

The collaboration with DSIR in New Zealand has continued. The geogas results from Mt Erebus and Dry Valley in Antartica have been interpreted. In New Zealand, investigations have been performed of the ongoing deposition of minerals in hydrothermal systems.

In co-operation with the British Geological Survey, a target in central England has been successfully located. This particular type of object cannot be identified with any traditional exploration techniques.

In co-operation with CRA Exploration in Australia a study of the response of the geogas technique to three different targets in Australia has been performed. The geological conditions are very different at the three sites, one of them being situated in an arid area in Western Australia. On some of the sensors from this area uranium has been positively identified for the first time.

An extensive geogas survey was performed in 1991 in Dalsland in central Sweden with the purpose of studying emissions from deep fractures criss-crossing the landscape. One of the areas studied with a characteristic emission pattern of copper is now being used for reproducibility tests of the geogas phenomenon.

Support is being received from NFR to further improve the detection limits of the PIXE analytical technique. This work is being carried out in collaboration with the Division of Applied Nuclear Physics.

E The physics of electric breakdown in dielectric liquids

Anders Sunesson, S. Andersson, Peter Bårmann and Stefan Kröll*
**Visiting scientist*

Electric breakdown of matter is a phenomenon of fundamental scientific interest as well as of great technical/economic value. Breakdown processes in gaseous media are relatively well understood. Breakdown in condensed matter, on the other hand, is much less well understood. In certain cases of well defined solids, breakdown can be modelled, but in dielectric liquids no good theory exists. There is clearly a need for basic studies of this fundamental scientific field. In co-operation with ABB Corporate Research, Västerås, laser spectroscopic methods are being employed to investigate electric breakdown in dielectric liquids. The project started in summer 1991. It has been financed by ABB and, from 1993, by ABB and the Swedish Board for Technical and Industrial Development (NUTEK) jointly. So far, three subjects have been covered: laser initiation of electric breakdown, transmission of high-power laser beams in optical fibres, and construction of an interferometer for the measurement of very small vibration amplitudes.

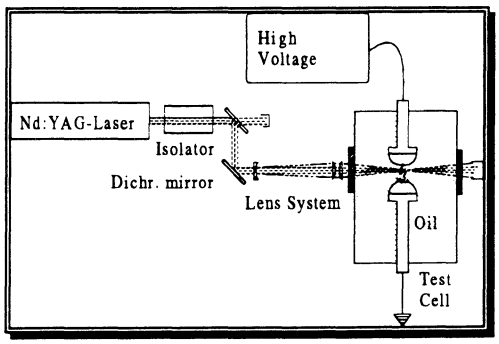


Fig. E1.
Experimental set-up in the breakdown studies

Laser-spectroscopic studies of electric breakdown

An electric breakdown in a dielectric liquid begins as a pre-breakdown channel, or "streamer", in the liquid. From its inception, i.e. through a localised discharge/plasma, the streamer grows between the electrodes with a

speed that varies between ~1 km/s and more than 10 km/s. When the streamer bridges the electrode gap, electric breakdown occurs. To be able to model the electric breakdown process in liquids, laser initiation of electric breakdown [E1] has been employed as a complement to research carried out at ABB Corporate Research. A key element in the present project is that in any dielectric liquid a plasma can be created at any predetermined time and spatial point by laser-induced optical breakdown. The initiated streamer can then be more easily studied as its starting point and time are known.

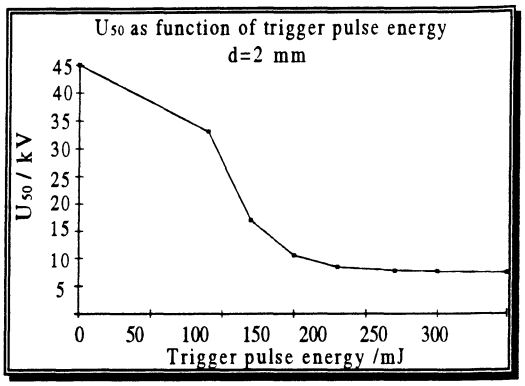


Fig. E2.
U_{50} vs trigger pulse energy.

Using a Q-switched Nd:YAG laser the technique of producing a well-localised plasma has been studied [E2]. Laser pulses are focused in the liquid to cause laser-induced breakdown, and the plasma thus produced serves as a starting point for the streamer. The experimental set-up is shown in Fig. E1. The laser radiation is passed through an isolator to avoid damage to the laser from stimulated Brillouin scattering in the liquid, then expanded to avoid self-focusing in the liquid, which would otherwise make precise control of the initiation point and energy deposition impossible. Most of the work has been dedicated to the development of a suitable focusing system to avoid self-focusing.

Experiments have been carried out using the fundamental 1064 nm line from the laser [E2]. The results (percent probability of breakdown at different voltages) have been fitted to a Weibull distribution.

The voltage needed for breakdown in 50% of the trials, U_{50}, has been determined for different trigger pulse energies; see Fig. E2. The curve shows how the streamer propagation voltage is reached when high enough pulse energies are used. Next, current pulses correlated to the streamer growth will be monitored. This will give hints of the growth mode of the streamers. Experiments measuring light emission and spectral analysis of the streamers are also planned.

Transmission of high power laser beams in optical fibres

To get a flexible tool for laser-initiated breakdown, e.g. to be able to reach deep into a complicated structure with the trigger beam, it would be advantageous if the energy could be transported through an optical fiber. This was investigated in a short project [E3]. The work covered fibers of core diameters 0.60 and 1.0 mm. Both green and IR laser radiation was tested in Q-switched and non-Q-switched mode. It was found that a maximum power of about 4 MW of IR radiation could be transmitted in the 1 mm-fiber. This is not enough to cause laser-induced breakdown in air or liquid, and further work must be spent to determine whether some other optical excitation mode of the liquid could be used to start an electrical breakdown than laser-produced plasmas. If such a low-power mode is found the use of fibers could be considered.

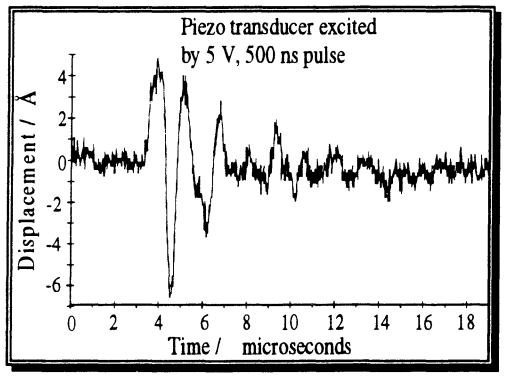


Fig. E3.
Displacement of a piezoelectric transducer excited by a 5 V pulse with a width of 500 ns. The excitation pulse is applied after ca 3 μs.

Construction of an interferometer for small vibrations

A heterodyne Michelson laser interferometer for the measurement of small vibration amplitudes has been constructed [E4]. The interferometer employs modulation of the laser (He-Ne) beam using an acousto-optic modulator and detection at the beat frequency (=80 MHz) to reduce 1/f noise and to ensure optimum contrast in the measurement. The vibration results in a phase modulation in the beat frequency signal, and is demodulated electronically. See Fig. E3 for an example of a measurement on a piezoelectric transducer.

Due to limits in the electronic equipment, true heterodyning was never achieved, however, very encouraging results were found. The sensitivity was determined to be 0.7 Å peak-to-peak vibration magnitude at 100 kHz vibration frequency, mainly limited by electronic noise and digitisation uncertainty. Further developments towards better detection sensitivity and true heterodyning are possible.

F Diode laser spectroscopy

Diode laser spectroscopy based on the two-tone frequency modulation (TTFM) technique is being pursued. The technique combines high detection sensitivity with high detection speed. We have implemented TTFM, at frequencies of several hundred MHz, using different GaAlAs semiconductor lasers, working in the wavelength range of 750 - 840 nm. Diode lasers in the near-infrared region are, due to their application in laser printers, compact disc players, optical communications, etc. relatively inexpensive and are available from manufactures off the shelf. Molecular absorption features in the near-infrared region are due to overtone bands, and are typically two or three orders of magnitude weaker than fundamental band absorption. However, the high sensitivity of TTFM makes the measurement of many gas species possible despite the weak absorption. We have demonstrated an absorption sensitivity better than one part in a million.

F1 Vapour pressure measurements, a tool for water activity determination in solutions

Peter Kauranen

In an experiment carried out in collaboration with the Chemical Center in Lund [F1], a novel technique for water vapour pressure measurements over a solution has been developed. It provides a new way to probe the chemical potential in solutions for studies of intermolecular forces. Fig. F1 shows the peak-to-peak TTFM absorption signal value due to water absorption in a cell at a wavelength around 820 nm. The increase in the recording is due to the change in water vapour pressure, when a 0.5 M NaCl solution is replaced by

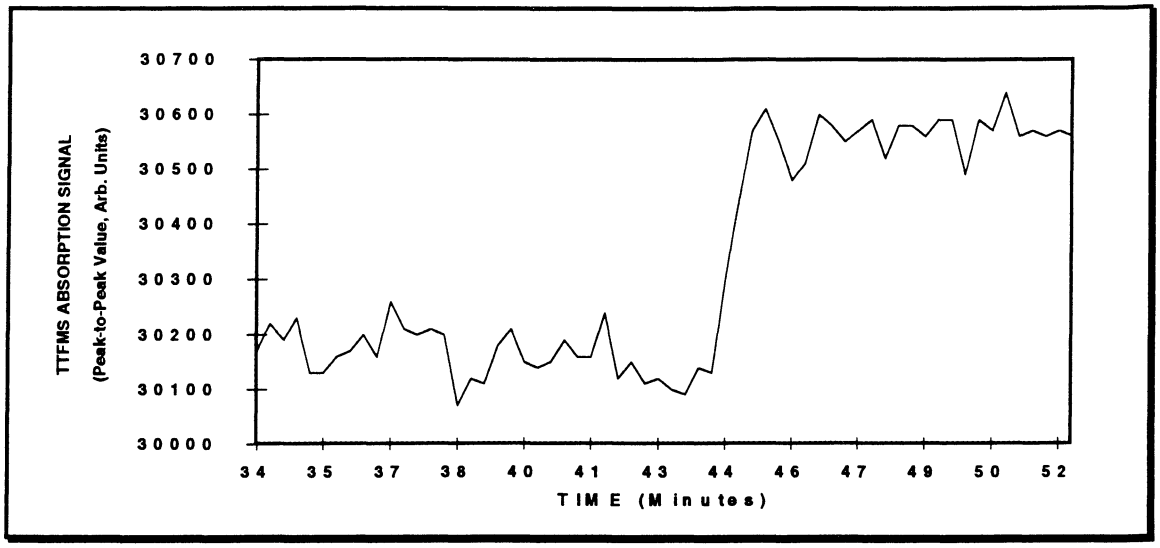
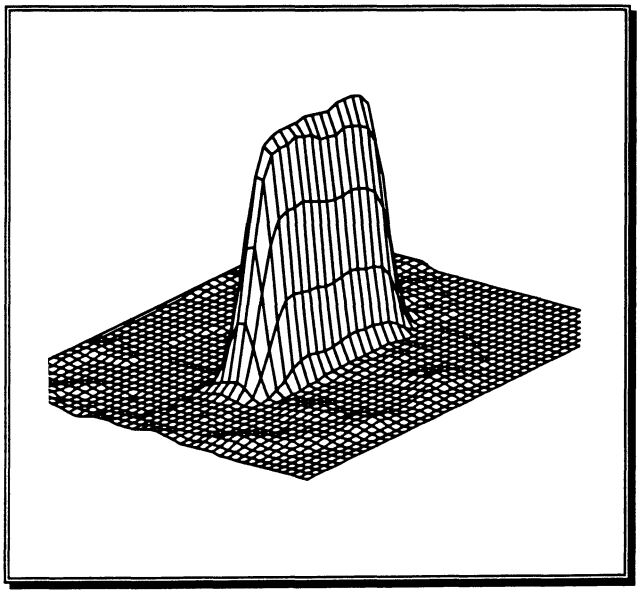


Fig. F1.
Recording of the peak-to-peak TTFM absorption signal value, when a 0.5 M NaCl solution in connection with the absorption cell is replaced by a solution of pure water.

pure water. The recording in Fig. F1 contains a contribution to the absorption from the optical path outside the absorption cell. This contribution is calculated from the known humidity of the atmosphere of 38 % and from the optical path length. Thereafter, the vapour pressure over the salt solution is determined to be 1.8 % below that over pure water.

F2 Tomographic measurements on an oxygen flow using two-tone frequency-modulation spectroscopy

Peter Kauranen, Hans Hertz and Sune Svanberg



Imaging of fluid or gaseous flows is of great importance in, for example, combustion diagnostics. In this project an expanding oxygen flow has been imaged by combining multi-angular TTFM absorption measurements with a tomographic reconstruction technique. Figure F2 shows a reconstruction of the oxygen flow 8 mm above a rectangular 4 × 18 mm laminar flow nozzle. The oxygen absorption line used was one in the A-band around 760 nm. Also, the project involves substantial development of computer routines for steering the experiment [F2].

Fig. F2
Tomographic reconstruction of the concentration of oxygen in an expanding flow.

References

E1. A Sunesson, *"Laser-induced breakdown and laser-induced electric breakdown - literature survey"*, ABB Corporate Research Report SECRC/KJ/TR-92/096 (1992).

E2. A Sunesson, *"Laser focusing in transformer oil"*, ABB Corporate Research Report SECRC/KJ/TR-92/147 (1992).

E3. S Andersson, *"Transportation of high-power laser light through light guides"*, ABB Corporate Research Report SECRC/KJ/TR-92/104 (1992).

E4. P Bårmann, *"Development of a heterodyne laser interferometer for very small high frequency displacement detection"*, Diploma work, **LRAP-137** (1992).

F1. P Kauranen, B Håkansson and I Harwigsson, "*Vapour pressure measurements using a frequency-modulated tuneable diode laser, a tool for water activity determination in solution*", manuscript in preparation.

F2. P Kauranen, H M Hertz and S Svanberg, "*Tomographic measurement on an oxygen flow using two-tone frequency-modulation spectroscopy*", manuscript in preparation.

VI Teaching Programme

Professors and lecturers: *Stig Borgström, Lars Engström, Hans Hertz, Bodil Jönsson, Gilbert Jönsson, Göran Jönsson, Stefan Kröll, Rune Kullberg, Hans Lundberg, Nina Reistad, Rolf Petersson, Sven-Göran Pettersson, Sune Svanberg.*

Teaching assistants: *Jonas Bengtsson, Roger Berg, Jonas Johansson, Per Jönsson, Peter Kauranen, Jörgen Larsson, Lars Malmqvist, Pär Ragnarsson, Lars Rymell, Tomas Starzcewski, Lennart Sturesson, Carl Tillman, Eva Wallinder, 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 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 practices. The number of hours devoted to experimental work is, as a rule, about the same as the number of hours used for theoretical education. During experimental training 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).

The total number of teaching hours is about 7000. Teaching is performed by one professor, six senior lecturers, one junior lecturer, fourteen teaching assistants and about twenty-five other teaching assistants.

Table 1
Courses given by the Division of Atomic Physics

		No. of students	*Hours theory*	*Hours lab.*
Physics course, E	*E1*	192	118	42(8)
Physics course, D	*D1*	92	132	46(8)
Physics course, M	*M1*	128	34	28(8)
Physics course, M	*M3*	123	70	28(8)
Physics basic course, V	*V1*	94	52	18(8)
Physics specialised course, V	*V4*	20	20	36(8)
Physics course, K	*K1*	135	68	28(8)
Physics course, BI	*BI1*	30	58	18(8)
Introductory course	*F1*	77	50	36(8)
Waves	*F2*	66	50	40(6)
Atomic Physics	*F3*	57	42	35(4)
Laser Physics	*F4,E3,M3,D3*	80	32	16(4)
Advanced Optics	*F4*	28	36	16(4)
Atomic and Molecular Spectroscopy	*F3,F4*	28	36	30(4)
Holography		16	10	15(10)
Radon		16	20	12(8)
Physics for Poets		30	20	12(8)
Laser Chemistry	*Graduate*	16	38	-
Computers in Measurement Systems	*Graduate*	16	20	-
Nonlinear Laser Spectroscopy	*Graduate*	8	30	-

A1 Basic courses

For the School of Engineering Physics the basic course *Physics, Extended course* is given. This consists of three parts, Introductory course, Waves and Atomic Physics, coupled with laboratory practices. *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 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, waves and modern physics combined with laboratory work.

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

For 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 general 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 post-graduate studies at the Department.

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 Department. This course, emphasising Fourier optics, interferometry, fibre-optics, holography and phase-conjugation techniques was given for the first time in the autumn of 1985.

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

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

A3 Diploma work

There are several undergraduate students performing their diploma work within the Atomic Physics Division. Below those who finished their diploma work during the present period are listed together with the title of their diploma thesis.

Peter Bårmann	*Development of heterodyne laser interferometer for very small high-frequency displacements detection, LRAP-137*
Ulf Elman	*Photon echo optical processing, LRAP-136*
Ulf Gustafsson	*Near-infrared Raman spectroscopy of tissue using diode laser and CCD detector, LRAP-138*
Claes af Klinteberg	*Studies of intensity modulated light in turbid media, LRAP-139*
Kristofer Ljunggren	*Metod för våglängdskalibrering av färgämneslaser, LRAP-132*
Kerstin Sandell	*Kardiografen CTG - utveckling och vardaglig användning*
Hans Stattin	*Thin film soft X-ray absorption filters, LRAP-140*
Per Tidlund	*Material considerations for time-domain optical storage, LRAP-131*
Raoul Zerne	*Radiative lifetime and Landé-factor measurements of the Se I $4p^3 5s\ ^5S_2$ level using pulsed laser spectroscopy.*

B Graduate teaching

A graduate course in *Laser Chemistry* has been given at the Lund Chemistry Centre in order to increase the interaction between chemists and physicists in the field of lasers. The course treated tuneable laser sources, laser spectroscopic methods, laser-induced chemistry and femto-second laser chemistry. At a concluding symposium 9 graduate students presented special projects. The course was also attended by a number of senior chemists.

A course in *Computers in Measurement Systems* is given for the first time in 1991. The course especially focus on interfacing instrumentation in experimental physics to personal computers. It also gives an introduction to programs for evaluating and presenting the results.

The graduate course in non-linear optics, which has been given biennially, was temporally replaced by a course in *Non-linear Laser Spectroscopy*. The course treated basics of atom-photon interactions. Higher-order corrections of interactions between atoms and narrow band radiation, and the optical Bloch equations for a two-level system. A part of the course was lectured by Stig Stenholm, director at the Research Institute for Theoretical Physics in Helsinki.