Laser Physics Centre

Professor Barry Luther-Davies - Head of Laser Physics Centre

The Laser Physics Centre is engaged in laser-based research on topics spanning fundamental and applied physics and engineering.

http://laserspark.anu.edu.au

Introduction

Research within the Centre covers many of the most exciting aspects of contemporary laser-based research. The Centre has strong programs on optical solitons; quantum computing; laser cooling and trapping of atoms; optical materials; and photonics, to mention just a few. Research in photonics is partly supported by the Australian Photonics Cooperative Research Centre, and Professor Luther-Davies remains a Director of that Centre.

This year several members of the Centre worked on a contract to assist RPO Pty Ltd, a spin-off company from the Laser Physics Centre, which has its operations on the ANU campus. RPO makes photonic "chips" using proprietary Inorganic Polymer Glasses (IPG™) originally developed in the Laser Physics Centre. RPO has made excellent technical progress during the year and has employed new staff including Ben Cornish, Graham Gordon and Maureen Brauers. Reiner Friedrich and Dr Anke Freydank returned to Germany in September to undertake research at Potsdam University funded by the Australia-Germany exchange agreement. Unfortunately they are unable to return to Canberra to complete their appointment. Dr Max Lederer, a valued member of our photonics team also left late in the year to join a laser company in Austria. Dr Olivier Uteza, an IREX Fellow from the Lasers, Plasmas and Photonic Processes Laboratory at the Aix-Marseille II University, France, completed his Fellowship in June. He was involved in the research of femtosecond laser induced order-disorder phase transitions with Andrei Rode and Eugene Gamaly.

The Centre congratulates two members of its academic staff on their promotion to level D: Dr Marek Samoc and Dr Andrei Rode; and Dr Wieslaw Krolikowski for his appointment to a continuing position. On general staff matters, Renee Vercoe took over as Departmental Administrator from Kristina Milas during the year whilst Craig McLeod took an extended period of leave to work in Canada for a year.

The activites within the Centre can be broadly divided into seven areas: Laser Matter Interactions, Nonlinear Optical Phenomena, Nonlinear Optical Material development, Solid State Spectroscopy, UV Spectroscopy, Atom Manipulation, and Photonics.

Laser-Matter Interaction Physics

Ultra-Fast Pulsed Laser Deposition (UFPLD) is a process patented by ANU researchers that allows the deposition of exceptional quality thin films or novel nano-structured materials by evaporating solids into either high vacuum or an inert gas using short laser pulses. UFPLD can be optimised for efficient evaporation of almost any solid, but the laser parameters required are beyond those available from commercial systems. As a result the UFPLD project involves both the development of special laser systems for UFPLD, and application of UFPLD to the production of novel materials.

Laser Hardware

Good progress has been made towards the creation of an optimised laser system for UFPLD based on a very low repetition rate (down to 1.5 MHz) mode-locked Nd:YVO_4 laser oscillator and a power amplifier. High-modulation 15-quantum-well InGaAs semiconductor saturable absorber mirrors (SESAMs) have been developed to produce stable passive mode-locking. The very low mode-locking frequencies require an exceptionally long optical resonator and a novel "zero-q-transforming" multi-pass cell was used to provide a variable cavity length up to 100 m long. Currently, the laser oscillator operates at three different repetition rates: 4.1 MHz, 2.6 MHz and 1.5 MHz, output pulses of 13-ps duration, and an average power up to 4 W in a diffraction-limited beam (M^2<1.1). A double pass Nd:YVO_4 amplifier was developed to increase the output power up to 10 W. Frequency conversion to 532 nm with an efficiency >80% was achieved using an external non-critically phase-matched LBO frequency doubler.

This laser has been used to deposit chalcogenide glass (As_2S_3 and $Ge_{33}As_{12}Se_{55}$) films and obtained a three-fold increase in the deposition rate compared to our previous work. This demonstrates that UFPLD can deposit thin films for important technological applications at rates relevant to industrial processes. In collaboration with the Fraunhofer Institute of Laser Technology in Aachen, Germany, a higher-power amplifier to increase the laser power to >40 W is being implemented. This system will allow evaporation of the most challenging materials such as Si or highly reflecting metals.

Magnetic Carbon Nano-Foam

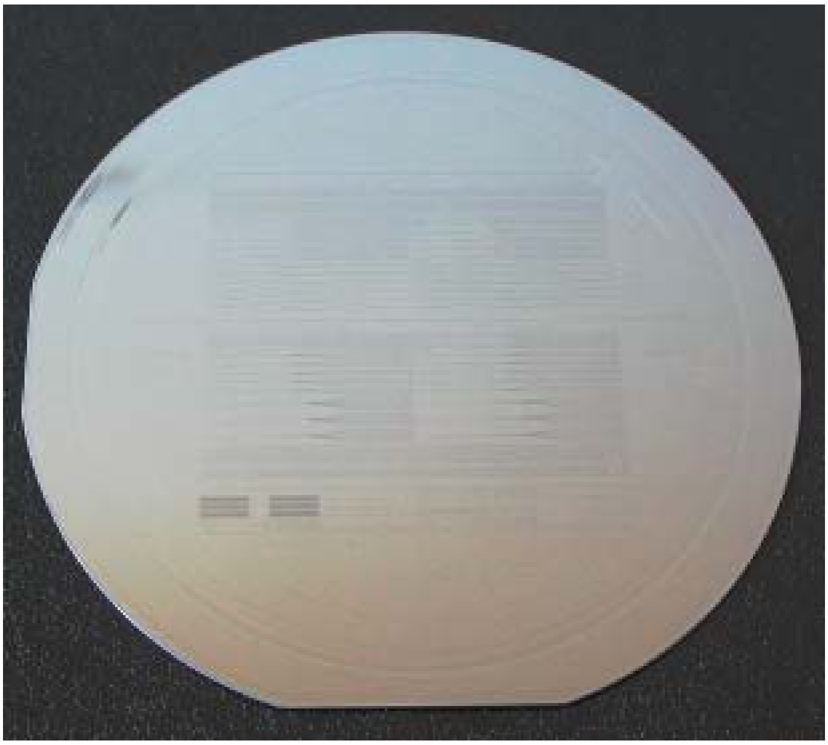
It was reported earlier that the evaporation of C into an Ar atmosphere using UFPLD leads to the production of an unusual magnetic C nano-foam. This year a thorough study of ferromagnetism in these materials was performed indicating a very high density of unpaired spins — up to 1.51×10^{20} per gram. The saturation mass magnetisation of the foam is several times higher than any previously observed all-carbon system. The nano-foam has been shown to be a soft ferromagnetic semiconductor with a narrow hysteresis curve and a Curie temperature exceeding 90 K. This unexpected behaviour, as we have shown, can be quantitatively interpreted using spin-polarised *ab initio* calculations. The results suggest that unpaired spins are introduced by sterically protected carbon radicals, which are immobilised in the non-alternant aromatic system of C sheets with negative Gaussian curvature (so called schwarzites).

BN Nano-Clusters Produced by High-Repetition-Rate Laser Ablation

We synthesised Boron Nitride (BN) nanomaterial *via* UFPLD of a BN target in nitrogen gas. The material was microscopically and chemically analysed by high-resolution transmission electron microscopy; electron energy loss spectroscopy; electron diffraction; and energy-filtered TEM. Nano-rods, multi-layered nano-cages, double-layered "nano-horns", and multi- and particularly single-walled BN nano-tubes, which are extremely rare in BN-nano-structures produced by other means, were discovered in the product. BN nano-tubes with various diameters and number of layers, including single-walled nano-tubes, were frequently found assembled as "ropes".

Laser Induced Phase Transitions

Order-disorder phase transitions induced by femtosecond laser irradiation of G films have been studied using a pump-probe technique with a record 200-fs time resolution. The data obtained allowed us to derive numerically the temporal evolution of the real and imaginary parts of the dielectric function. The time history of the phase transition induced by the femtosecond laser is fully reconstructed at the picosecond time scale and the nanometer space scale on the basis of the experimental data.



IPG™ polymer wafer with characterizing structures

The analysis shows that the first sharp reflectivity rise in a few ps after the excitation coincides with the time required for the transition to a new phase when the heat conduction is negligible, while the second stage of slow change, ~100 ps, is dominated by the heat transfer. It appears that the recovered electron-phonon collision rate and heat conduction coefficients are transient non-linear functions of temperature. The spatial (nanometer-scale) and temporal (picosecond-scale) constraints are dominant in the course of the phase transition that makes it drastically different from the phase transform in a bulk solid in conditions of thermodynamic equilibrium. A comprehensive analysis leads to a proper control of the evolution over the optical properties related to the reversible phase transition in the material induced by the femtosecond laser.

Nonlinear Optical Phenomena

Optical Solitons in Saturable Nonlinear Media

Certain materials modify their optical properties, such as index of refraction, when illuminated by light of sufficiently high intensity. These so called nonlinear media can be used to observe a variety of effects induced by propagating light beams, and in particular formation of spatial solitons. A spatial optical soliton represents an optical beam that propagates in a nonlinear medium without changing its shape. The process of diffraction is prevented by the self-induced change of the refractive index. Spatial solitons are of special interest for applications in all-optical switching or processing. We have studied theoretically and experimentally the formation and interaction of various types of spatial optical solitons in optical systems. We have shown the possibility of formation of complex objects consisting of several mutually trapped higher order soliton beams. We have demonstrated this effect experimentally using photorefractive nonlinear crystals and have observed stable propagation and collisions of three-component vector dipole solitons.

Optical Beams in Nonlocal Nonlinear Media

In another study we investigated theoretically the propagation of solitons in nonlocal nonlinear media where the intensity of a wave (or a beam) at a particular point affects not only the medium at that very point but also the surrounding region. Some important examples of such media include plasmas with non-locality caused by thermal effects, and a Bose-Einstein condensate where non-locality is due to the finite-range of particle interactions. We proved that nonlocality suppresses the instability of wave fronts, prevents the collapse of finite beams and leads to the formation of stable multi-dimensional solitons.

Localisation of Light in Optically-Induced Periodic Structures

The propagation of light beams in a medium whose refractive index varies periodically in space leads to many interesting effects such as discrete diffraction and the appearance of band gaps where the propagation of light is forbidden. We have investigated theoretically and experimentally the propagation of beams in photorefractive crystals containing optically induced index gratings. We showed that the optical beams created various types of localised structures (discrete solitons) when propagation occurs in the presence of nonlinearity.

Nonlinear Optical Materials

Development of new, more efficient nonlinear optical materials, optimised for particular applications in photonic technologies, remains one of the important goals of research in modern optical sciences. LPC researchers have been focusing on both the theoretical understanding of factors influencing microscopic and macroscopic optical nonlinearities and on development of several classes of nonlinear materials, including polymers and glasses suitable for use in waveguide devices.

Nonlinear Properties of Model Molecules and Polymers

Third-order nonlinear optical properties of various organic and organometallic materials were studied to establish structure-property relations to guide the design of high nonlinearity materials for photonics.

A particular focus has been electro-chromic switching of the third-order optical nonlinearity in organometallics, including dendritic structures. We had previously discovered that it is possible to switch the sign of the absorptive and refractive nonlinearities of certain organometallic molecules at 800 nm by electron transfer from an electrode. This study has now been expanded to the longer wavelength region important for telecommunications applications.

Optical Nonlinearity of Chalcogenide Glasses

We have used our tunable fsec optical parametric generator to measure the dispersion of the optical nonlinearity of chalcogenide glasses. The nonlinearity of As-S-Se glasses was found to be enhanced at particular Se concentrations in agreement with earlier work. The glasses had excellent nonlinear figures of merit for optical switching and useful nonlinearities ($\approx 10^{-13}$ cm^2/W). The Verdet constants for these materials were also determined and indicated it should be possible to make chip sized magneto-optic devices in chalcogenide glass.

Nonlinear Optical Chromophores for Polymer Optical Fibres

By incorporating a nonlinear optical chromophore inside the core of an optical fibre, enhanced nonlinear response is expected both because of confinement of the optical field and enhanced interaction lengths. Our interest in chromophores suitable for inclusion in polymer fibre cores has been motivated by an industry-sponsored project to develop an electro-optic fibre for voltage sensing.

We have synthesised soluble derivatives of π-conjugated distyrylbenzene: an oligomer of *p*-phenylenevinylene asymmetrically substituted in the *para* position with electron donating alkyleneoxy group bearing the methacrylic group, and an electron accepting alkylsulphonyl group bearing alkyl chains of different length (MTPV-ORSO, MTPV-SOHE). The molecular second-order and third-order nonlinearities were estimated with quantum chemical calculations (MOPAC). The methylmethacrylate side-chain copolymer (MTPV-ORSO-MMA) could be incorporated into the core of the fibre perform and used to create a single mode polymer fibre.

The nonlinear optical properties of the polymer materials containing the new chromophores were measured with several techniques including second harmonic generation induced by corona poling to determine the second-order nonlinearity and degenerate four-wave mixing to determine the third-order nonlinearity

A comparative study of the molecular second hyperpolarisability was performed for the conjugated molecules of MTPV-SOHE and *p*-bis(o-methylstyryl)-benzene (Bis-MSB). The asymmetric substitution causes a red shift (24 nm) of the absorption maximum and enhancement of third-order optical susceptibility. The second hyperpolarisability of MTPV-SOHE was about six times bigger than that in Bis-MSB.

Solid State Spectroscopy

Quantum Computing

The concept of a quantum computer emerges from the realisation that the laws of quantum mechanics sets the ultimate lower limit for the resources required to perform any calculation. If a computer can be built whose operation fully utilises the laws of quantum mechanics, it is predicted that it will be exponentially faster, for a range of important algorithms, than its classical counterparts.

The Solid-State Spectroscopy Group is developing quantum computer architectures based on nuclear/electron spins that are associated with optically active centres. This allows the spins to be manipulated and measured using purely optical techniques. The use of optical centres neatly sidesteps many of the problems found in other architectures. Two types of centres are currently being investigated, rare-earth ions doped in insulating crystals and colour centres, particularly the nitrogen-vacancy centre in diamond.

Work is being undertaken to determine the feasibility of a quantum computer based on rare-earth doped crystals. The study is concentrating on Eu and Pr doped Y_2SiO_5. This year single qubit gate operations were demonstrated and characterised using quantum state tomography. Fidelities of greater than 90% for the combined preparation and measurement process were achieved. The electric dipole-dipole interactions required for multiple qubit operations were also measured. These results confirm that our existing experimental system can access the parameter space necessary for two qubit operations.

Up until now a major argument against using rare-earth doped crystals for quantum computing was the relative short coherence times of their nuclear spin states. We have developed a new method for extending these coherence times. In the first demonstration of this technique, the nuclear spin coherence time of $Y_2SiO_5{:}Pr^{3+}$ was increased from 0.5 ms to 81 ms. In related work, the robustness of the spin coherence to optical perturbations was also investigated using Raman and Raman heterodyne techniques.

The application of the nitrogen-vacancy centre in diamond for various quantum computing functions is being investigated. A promising application is the use of the centre as a single electron spin detector. The concept is to transfer the spin of the centre to be measured onto the electron spin of the N-V centre and then to optically readout the N-V spin state. In this way the spin state of non-optically active centers can be determined. The theoretical analysis of this technique has been completed and, in collaboration with DSTO, a confocal microscope system is being developed to perform single site spectroscope experiments. Measurements on bulk diamond samples were undertaken to establish the relevant dynamical properties of the N-V centre, in particular the dynamics of the optically induced spin polarisation. Associated fundamental studies investigating perturbation of electromagnetic induced transparencies due to auxiliary driving fields were performed using the nitrogen-vacancy centre.

Optical Signal Processing

The intermediate goal of this project, funded by DSTO and the Australian Photonics Cooperative Research Centre, is to develop a microwave spectrum analyser for radar and communication applications. The analyser which utilises the long optical coherence times of rare-earth doped crystals has the unique feature of being able to perform real time spectral analysis of all frequencies over a potential bandwidth of 10 GHz. In this application, a spectral grating is stored across an inhomogeneous broadened infrared absorption line, which is used to scatter a beam modulated by the unknown microwave frequency. The time dependence of the scattered beam indicates the spectrum of the microwave source. Operation at 1.54 μm, corresponding to the $I_{15/2} \Leftrightarrow I_{13/2}$ absorption line in Er doped Y_2SiO_5, allows us to capitalise on technology developed for optical communications.

The electron spin resonance and Raman heterodyne techniques have been employed to study the structure and dynamics of the electron spin states of the Er site. The results of this work are to be used to develop techniques to reduce the optical dephasing of the $I_{15/2} \Leftrightarrow I_{13/2}$ transition, a critical parameter for the spectrum analyser's operation.

Joanne Harrison working on the electron paramagnetic resonance equipment

UV Laser Spectroscopy

The work in this laboratory is aimed at developing and applying high resolution laser and nonlinear optical techniques for the study of molecules of atmospheric and environmental interest in the vacuum ultraviolet region (below 200 nm). This year, a new high resolution laser source based around a periodically poled KTP crystal in a ring-cavity optical parametric oscillator (OPO) has been developed and characterised. The OPO is injection seeded by a cw diode laser, and pumped by a frequency-doubled single-longitudinal mode Nd:YAG laser.

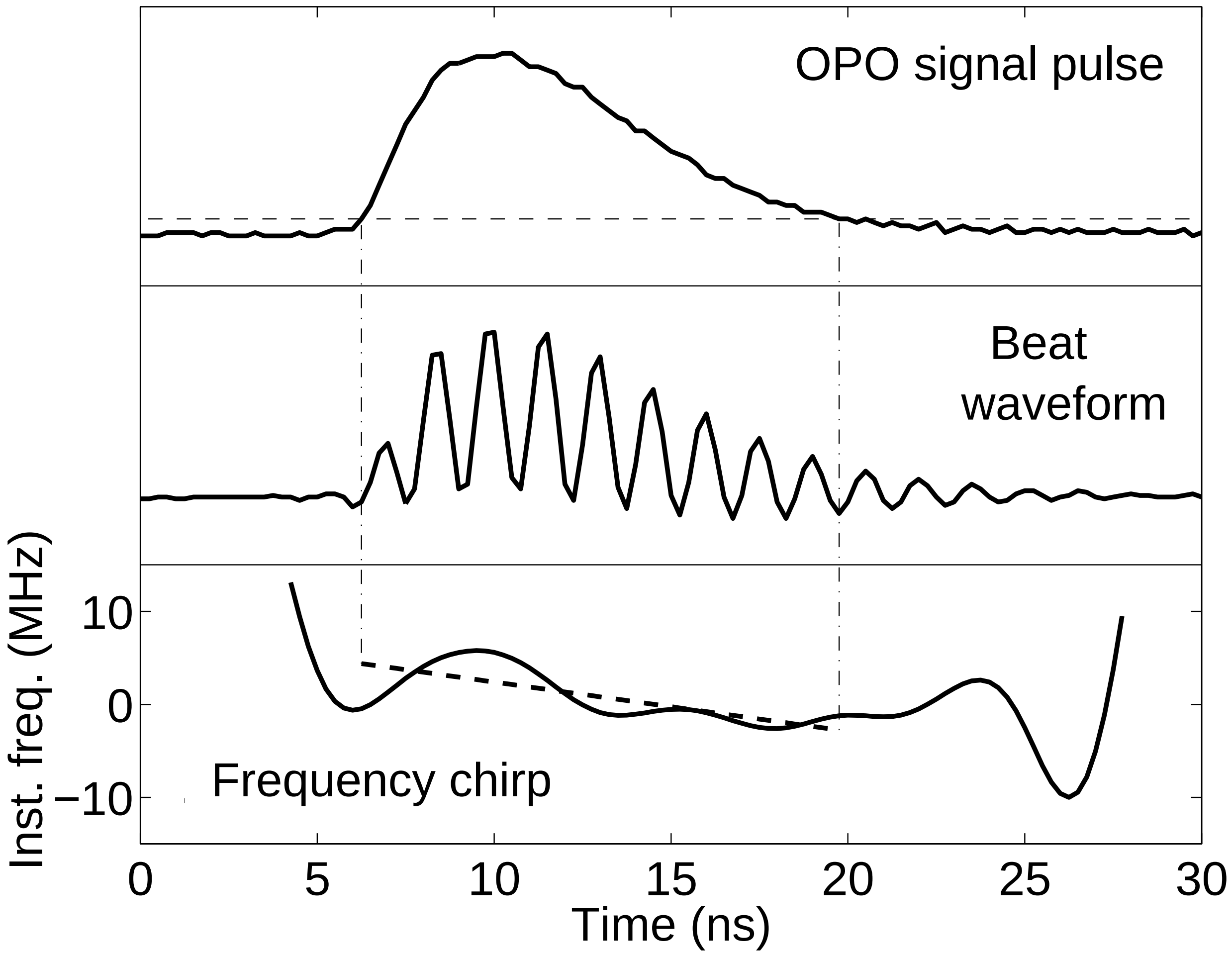
The figure below shows how the optical phase measurements can be used to reconstruct the frequency chirp in the OPO output that arises from phase mismatch and self-phase modulation. The frequency excursions were found to be reproducible, and less than the Fourier transform limit of the pulse duration, making the OPO a promising source for high resolution spectroscopy. Ways to reduce both the frequency chirp and the pulse-to-pulse frequency jitter in the OPO output have been implemented, as a prelude to amplification and upconversion to a wavelength of ~120 nm for high-resolution vacuum-ultraviolet spectroscopy.

Atom Manipulation

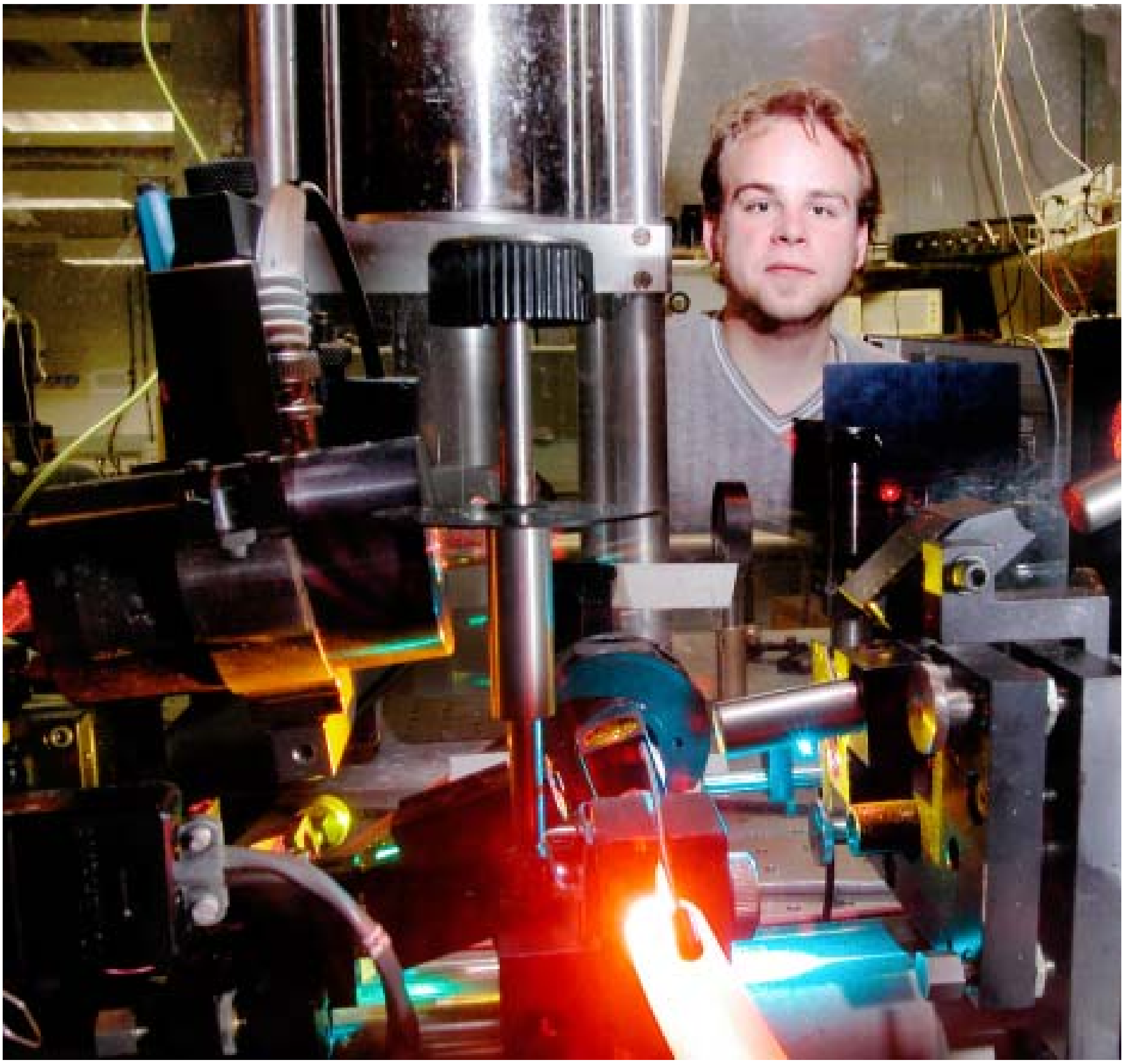
The Atom Manipulation project is a joint program between the Laser Physics Centre and the Atomic and Molecular Physics Laboratories (AMPL) which uses laser cooling and trapping techniques for atom optics and atomic collision physics experiments.

Significant improvements to the metastable helium magneto optic trap (MOT) have been implemented this year, which have increased both the number of trapped atoms, and the sensitivity of detection using both atomic fluorescence and electron multiplier techniques. The atom trap now has a maximum of ~2×10^9 atoms at 1 mK (~3×10^8 after optical molasses cooling to 250 μK), and the trap diameter is now ~5 mm yielding a peak density of ~10^9 atoms/cm^3.

The improvements have been due to an increase in the capture velocity to ~80 m/s and the introduction of a quasi-dark spot MOT. This has been achieved through implementation of a higher magnetic field gradient using an optimised coil design, together with the introduction of a fibre laser enabling larger laser beam diameters. In addition, the magnetic coil switch-off time can be reduced to <300 μs using a novel circuit design to provide a higher duty cycle for experiments.



Beat signal (top), with reconstructed OPO temporal pulse (centre) and instantaneous-frequency curve (bottom) following Fourier transformation



Elliot Fraval making adjustments to the ultra-high resolution ring laser

Photonics

Inorganic Polymer Glasses

With support from the Australian Photonics CRC, we have continued to develop novel organic-inorganic polymer glasses IPG™ for the fabrication of optical chips. Following the transfer our earlier poly-siloxane materials to a spin-off company, RPO Pty Ltd, research has focused on the synthesis of new photo-patternable polysiloxanes with reduced optical loss and improved physical properties. We have developed a new approach to the synthesis of silica-zirconia based IPGs that allows reduction in the OH absorption compared with previous methods.

Because of growing interest in electro-optic polymers, we are working to incorporate electro-optic chromophores into IPG™ materials. Several new donor-bridge-acceptor three-ring acetylenes have been synthesized, employing the diethylene glycol group as the donor, the fluoro substituents and the 2-ethylhexylsulfonyl group as the acceptor. Compared with previous chromophores, the optical loss in the CH overtone region is dramatically decreased due to replacement of hydrogen with fluorine and the replacement of the CH double bonds with C-C triple bonds.

Synthesis of a push-pull disperse red type molecule bearing a methyl methacrylate group on one side and an alkoxy silane group on the other side of the molecule was continued. The attachment of the siloxane group offers the possibility to incorporate the organic molecule into the IPG™ matrix. This work was carried out in collaboration with the group of Dr Mark Humphrey in the Department of Chemistry.

Photonic Modelling and Design

A range of optical waveguide devices for RPO Pty Ltd have been designed. Of particular interest have been designs for particular functionalities which are insensitive to manufacturing tolerances. In addition, we have investigated the use of segmented waveguides to produce up and down tapers to enhance coupling between the waveguide chips and an optical fibre.

Optical Sources

We have continued the development of psec neodymium lasers passively mode-locked using ion implanted, MOCVD grown, Semiconductor Saturable Absorber Mirrors (ii-SESAMs). Long cavity mode-locked lasers producing modest average powers (≈1 W) have sufficiently high peak power to allow efficient conversion to new wavelengths via nonlinear processes. A versatile 20 MHz repetition rate Nd:YVO_4 laser producing ≈1.1 W was shown to produce up to 200 mW of 265 nm light by frequency quadrupling. The same laser has been used to sub-harmonically pump a KTP optical parametric oscillator producing >200 mW of average power at 1573 nm of use for testing telecommunications devices.

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