>>> Béatrice CHATEL, CNRS scientist at Laboratoire Collisions Agrégats, Réactivité (LCAR, UPS/CNRS).

The lasers, 50 years on and still full of promise

The laser, which is short for Light Amplification by Stimulated Emission of Radiation was first demonstrated by Theodor Mainman on May 16th 1960. This pure lab product was initially solely devoted to research. But thanks to its unique properties (such as directivity, coherence, power and monochromaticity), it rapidly became useful for a host of other applications.

The laser is at the heart of CD-readers and printers but also, since 1974, in the barcode scanner you see being used at the supermarket. Laser shows produce visual displays by using beam effects and the laser's ability to be focused to a pinpoint makes it ideal as a precision scalpel in medicine, for example to reshape the cornea or to kill tumors without any risk of damaging nearby healthy tissue. It also serves as an ultra-precise optical tweezer to transport nano-objects and biological molecules.

In 50 years, the laser has become part of our daily lives. Its power has fascinated us since the beginning, like in the movie Goldfinger, where the laser is used as a frightening arm against 007. Its power allows to cut, drill, mark and clean stones, steels, papers and plastics. Today these applications represent 35% of the laser market. Inside fiber optics, analogous to a complicated version of Morse code, the light can carry with it vast amounts of data over long distances. Laser beams are also used to align roads and tunnels and implement 3D reconstruction. The LIDAR technique allows to analyze pollutants in the atmosphere.

The laser's exceptional properties are also useful in fundamental research. At Paul Sabatier, the laser is used to cool atoms to temperatures just barely above absolute zero (a technique known as Bose-Einstein condensation), to factorize numbers, follow the dynamics of chemical reactions in real time, study and manipulate nano-objects, image biological samples in 3D, study their composition, create an artificial star in order to optimize telescope resolution, study turbulence, detect pollutants, and analyze the composition of paintings. This issue of our magazine will present some of these applications.

Since 1960, nine Nobel prices have been awarded to laser scientists (for holography, non-linear optics, spectroscopy, metrology, the observation of chemical reactions and cold atoms).

Future projects, like the simulation of thermonuclear reactions as well as interstellar chemical reactions, biology imaging, probing matter with attosecond pulses and particle beam generation for medical applications, are further opening up a fruitful route for this 50-year old discovery.

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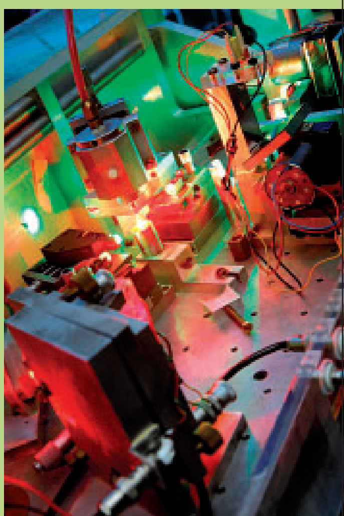


>>> Experimental room with a femtosecond laser chain in the Femto Lab (LCAR).

LCAR: Laboratoire Collisions Agrégats et Réactivité/ Collisions, Aggregates and Reactivity Laboratory

IMFT: Institut de Mécanique des Fluides de Toulouse/ Toulouse Institute of Fluid Mechanics

LMTG: Laboratoire Mécanismes de Transfert en Géologie/ Laboratory for Mechanisms and Transfer in Geology

LAAS: Laboratoire d'Analyse et d'Architecture des Systèmes/ Laboratory for Analysis and Architecture of Systems

The laser revolution in fluid mechanics

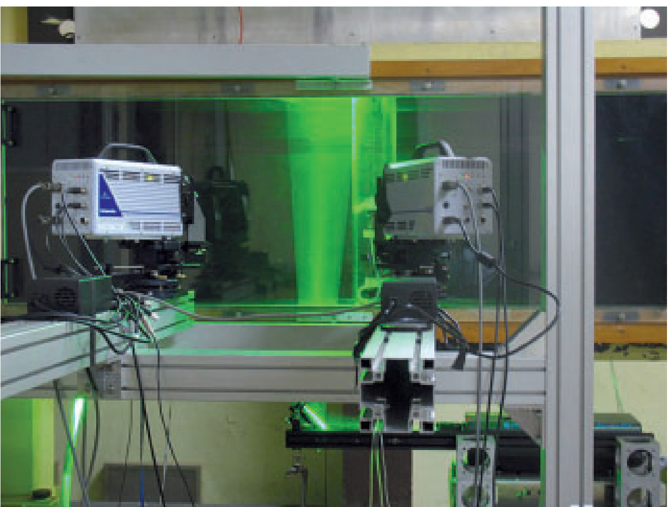
The laser has allowed fluid mechanics researchers to measure movement without interfering with flow. A revolution that continues today.

>>> Emmanuel CID, CNRS research engineer at the Institut de Mécanique des Fluides de Toulouse (IMFT,UPS/INP/CNRS).

Lasers have had a considerable impact on the field of fluid mechanics. Indeed, the laser is now the elementary building block of most modern techniques for measuring gas or liquid flow in which we need to determine, if possible, local and instantaneous characteristic parameters such as velocity, concentration of a species, and the size of droplets or particles.

Laser Anemometry

Several techniques are widely used for measuring the velocity of fluid flow at a single point, such as the Pitot tube or hot wire probes. The main drawbacks of these techniques are their invasiveness and the need for frequent calibrations of the thermal anemometers. In 1964, Yeh & Cummins proposed a Laser Doppler Anemometer (LDA). Here, two coherent collimated laser beams intersect to create a measurement volume whose dimensions are around several tens of microns in diameter and a millimeter in length. The light scattered by the two beams is collected by a photodetector and its frequency is proportional to the velocity of tracers through the measurement volume. This technique, at a single point, is non-intrusive and requires no calibration. The use of a continuous laser source (He-Ne) and the different spectral lines of argon were used to measure multiple velocity components. In the 1980s, this technique was extended to measure the size of particles (Doppler phase) using multiple photodetectors. The diameter of solid or liquid particles, in this case, is proportional to the phase of Doppler signals collected by different photomultipliers.



>>> Time Resolved Stereoscopic PIV measurements in the near wake of a circular cylinder in IMFT's wind tunnel.

Tracer particles

Measuring the concentration of species in a flowing fluid can be achieved by taking samples over periods longer than a millisecond. This measurement can also be made instantaneously by Laser-Induced Fluorescence (LIF). Molecules injected into the flow are excited at the wavelength of a laser source. The light re-emitted by fluorescence is then captured by a photocathode or a sensor matrix. The amplitude of this signal is, in a certain range, proportional to the concentration of the molecular marker.

In the 1990s, another velocimetry technique appeared and was a great success: Particle Image Velocimetry (PIV). Particle "tracers" are illuminated by a laser sheet. Two short pulses then "freeze" the position of the tracers in the flow, which is recorded by a camera. We measure the local displacement of groups of markers that move as the fluid moves. This provides an instant map of the velocity field in any slice of the flow. In the standard configuration, two components of velocity are simultaneously mapped (2D-2C). In two geometry-viewing cameras, three velocity component vector fields (2D-3C) can be simultaneously measured. If you work with at least four cameras, an entire volume is illuminated and you enter the field of Tomographic PIV, which gives access to three velocity components in a volume of small size (currently, the size of a cellular phone for gas flows and the size of a small book for liquid flows). Importantly, recent improvements in pulsed lasers delivering several tens of millijoules at kHz frequencies has increased data rate acquisition for these three techniques and has allowed scientists to explore many transient phenomena.

The use of lasers in fluid mechanics has made non-invasive measurements possible. At the beginning, it was used only for small volume samples but the technique is now is becoming three-dimensional and time-resolved.

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A laser for ultra-precise measurements

A group in Toulouse specialized in precision optical measurements has built a novel laser-based device capable of measuring light velocity with such accuracy that it can detect the deformations generated by magnetic and electric fields in matter.

>>> Cécile ROBILLIARD, CNRS scientist at the Laboratoire Collisions, Agrégats, Réactivité (LCAR, UPS/CNRS)

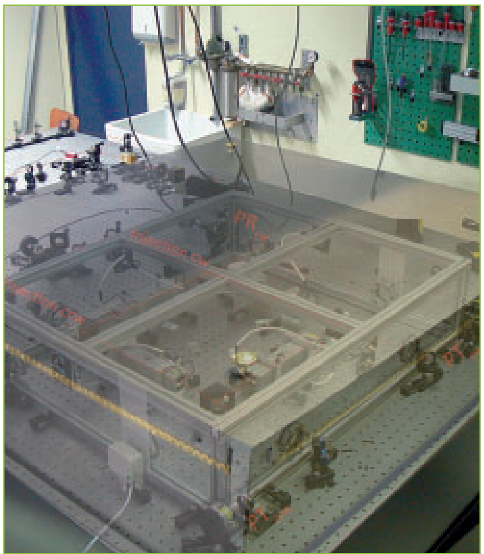
Laser levels, laser rangefinders. The laser is found everywhere, even on construction sites. The laser level takes advantage of the good spatial definition of a laser beam, which allows for precise pointing several tens of meters away. The rangefinder principle is more interesting, since it consists of measuring the dephasing of the laser wave diffracted on the target after a round trip . Standard devices have an accuracy of around one millimeter at distances of 100 m, hence a relative precision of 10^{-5}.

Atomic clocks

Present-day metrology experiments do not use this principle anymore because measuring frequencies yields much higher accuracies. Frequency is indeed the world's best measured quantity, in particular in the optical range thanks to the extremely well-defined wavelength of lasers. Converting any quantity into a frequency and measuring it accurately can be achieved with a high finesse optical resonator, in which light can be confined provided it has the proper frequency. Recent progress in optics and electronics allows to control and measure optical frequencies with relative accuracies below 10^{-18}. These ultra-precise measurements are performed, for example, in atomic clocks, in optomechanical measurements aimed at studying the coupling between radiation and a macroscopic mechanical element, and in gravitational wave interferometers.

Directional anisotropy

Our group specializes in this type of measurement. We have thus developed an experiment aimed at measuring very weak optical anisotropies induced in gases by magnetic and electric fields using frequency metrology. For example, electrodynamics predicts that if crossed transverse electric and magnetic fields are applied, light does not propagate at the same speed in both directions. Although this directional anisotropy was predicted more than 30 years ago, it has never been observed for lack of sensitive enough apparatus. A few weeks ago, our group achieved this for the first time in nitrogen. We detected a refractive index difference between the two propagation directions, hence a light velocity difference, which is on the order of a few picometers par second, compared with the 300 000 m/s velocity of light in vacuum! Other magneto-electro-optical effects are within experimental reach too and measuring these will enrich our knowledge of how light interacts with simple atoms and molecules. Beyond the physico-chemical interest of these studies, our apparatus, once optimized, will be the tool of choice for testing fundamental laws and symmetries, which are the foundation stones of contemporary physics.



>>> Apparatus used at LCAR: the set-up is rather simple but it must be carefully designed to keep perturbations under control.

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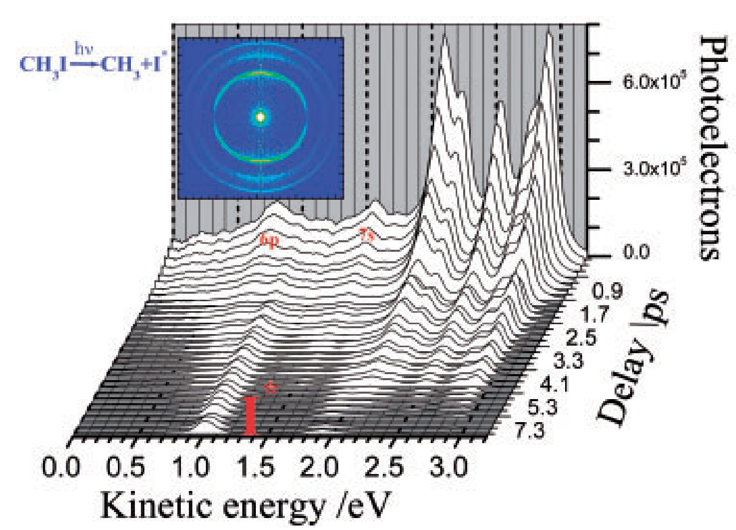
Chemistry at the femtosecond scale

>>> Valérie BLANCHET, CNRS scientist at the Laboratoire de Collisions, Agrégats, Réactivité (LCAR, UPS/CNRS).

By using state-of-the-art femtosecond lasers combined with the latest developments in the imaging of charged particles, it is now possible to follow a chemical reaction in real time at the molecular level.

A chemical bond is electrons shared by several atoms and defines a molecule. The energy shared is called the bonding energy. In a chemical reaction, this shared energy becomes redefined between potential and kinetic energies as the reagents approach each other. The bonding energy depends therefore on the relative position of the atoms. Determining this dependency and understanding what is at the heart of the chemical reaction is thus the main goal of physical chemistry that combines experiments and numerical simulations with quantum mechanics.

Understanding a chemical reaction means, among other things, identifying the set of forces that trigger the relative motion of atoms. The molecules are produced in the gas phase where they are free of interactions. Experiments therefore directly probe the relaxation that takes place within the molecule and these can be directly compared to ab initio calculations. In general, information is delivered by frequency-resolved experiments in which laser interactions occur over nanosecond scales. By comparing the absorption and fluorescence spectra, photoionization and photoelectron spectra as well as by measuring the appearance energy of dissociation, for instance, and branching ratios, the chemical reaction can be reconstructed with all the main parameters. But it is like being in a detective story with only clues and not actually a witnessing what is really going on.



>>> Energy distribution of the photoelectron produced after a pump-probe delay in which the pump triggered the predissociation of CH_3I. Drastic changes appear when predissociation takes place, leading to CH_3+I^* fragments.

Large speed

How can one become a witness then? The atoms inside the molecule can travel as fast as 2 km/s over a distance of a few Angströms and the typical time involved in this atomic movement is in the range of femtoseconds (a millionth of a billionth of a second). Up to now, no electronics alone could resolve such fast dynamics. Femtochemistry is a spectroscopy based on femtosecond laser pulses that allow researchers to follow a reaction triggered by laser excitation in real time.

A first laser pulse initiates the molecular dynamics by exciting its electrons. This laser pulse is called the pump pulse and its main aim is to set t=0 for the reaction. Due to this initial extra energy brought by the pump pulse, the molecule will relax via the movements of the electrons and the nuclei – known as electronic and vibrational relaxation. The molecules might dissociate too. A second laser pulse then interacts with the excited molecules at a variable delay relative to the pump pulse. This delay is the main variable of any femtochemistry experiment. The second pulse is called the probe pulse and its function is to photoionize the excited molecules. Next, the photoelectrons and the photoions are recorded as a function of the delay between the pump and the probe. It is not only the amounts and the masses of these two entities that carry information about molecular dynamics, but their energy distribution and angular distributions as well. These energy and moment balances are recorded by velocity map imaging.

Stroboscopy

Femtochemistry allows us to directly visualize a chemical reaction. It is a stroboscopy technique involving femtosecond laser pulses. What is the time-scale involved in the different steps of the reaction, what are the conditions that enhance or inhibit some reactions? These are some fundamental questions that femtochemistry experiments can now answer.

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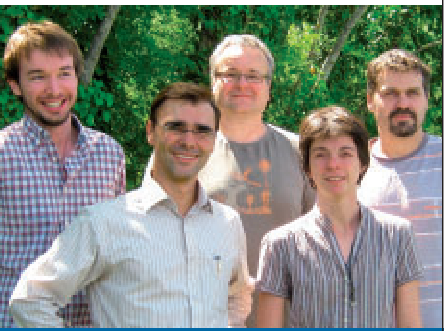
Lasers decipher minerals



>>> Frank POITRASSON, CNRS senior scientist, and colleagues from the Laboratoire Mécanismes de Transfert en Géologie (LMTG, UPS/IRD/CNRS).

Analysis of materials by laser ablation is undergoing a major revolution. It is helping us to better understand the composition of tiny pieces of complex materials.

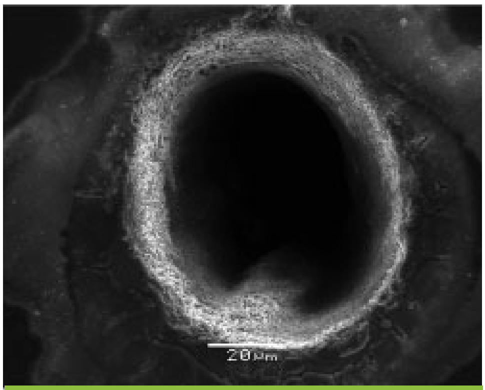
The technique was developed in the mid 80s and can determine the composition of minerals at the scale of a few tenths of millimeters. This analytical technique (called Laser Ablation Inductively Coupled Plasma Mass Spectrometry or LA-ICP-MS) involves using a laser to "shoot" the solid being analyzed. The aerosol product is then introduced into a plasma source mass spectrometer to detect chemical elements. The value of in situ analyses by laser ablation is that it allows us to study complex heterogeneous materials, such as those encountered in nature.

Magmatic rocks and shells

At the Observatoire Midi-Pyrenees (part of Paul Sabatier University), LA-ICP-MS is used to analyze elements at concentrations of around parts per million. These measurements are made on minerals from the Earth's mantle and magmatic rocks, on their melt or fluid inclusions, or on experimental charges produced at temperatures and pressures exceeding 1000°C and several gigapascals. They allow to determine the nature and origin of aqueous or siliceous fluids that affect the depths of the Earth but which do not exist on the surface of our planet. Experimental work on marine shells using LA-ICP-MS have also demonstrated the importance of animal diurnal cycles on environmental records that can be obtained through analysis of the animals' shell.
The first LA-ICP-MS techniques were not without flaws however. They used ruby lasers emitting long duration pulses, exceeding the microsecond in the infrared. This meant that transparent materials, such as quartz or calcite, were impossible to analyze. Ablations generated by this kind of laser produced thermal effects that substantially degraded the accuracy and precision of analyses.

Transparent materials

Major advances were made in the 90s with the use of "Q-switched Nd: YAG" or "excimer" lasers and the use of shorter wavelengths, in the near ultraviolet. Transparent materials could be analyzed and thermal effects were significantly reduced. The lower wavelengths and the evolution of optical systems have also improved the spatial resolution of mineral analyses to the scale of tens of microns, thereby extending the scope of possible studies.



>>> Laser ablation in a sample of Monazite (Brazil). Energy: 100 microjoules/pulse, pulse width: 100fs, rate: 5Hz, duration: 120s. Note the sharp edges and walls of the crater, and the absence of melt.

The 2000s have seen us crossing a new threshold with the implementation of femtosecond laser pulses 100,000 times faster than the nanosecond lasers used previously. This new field continues to evolve and allows a significant improvement in chemical analysis, not least by dramatically reducing the thermal phenomena. Matrix effects, by which the behavior of a material during its ablation greatly varies depending on its nature, are also virtually eliminated.

The Observatoire Midi-Pyrenees, in collaboration with the Laboratoire Collision Aggrégats Réaactivité (LCAR, CNRS-UPS) and with the support of the company Amplitude Technologies, a French manufacturer of ultrafast lasers, is at the international forefront in developing this technique. This group experimentally studies the mechanisms of femtosecond laser-matter interactions and their analytical implications. Crater ablation and generated particles are being studied for the first time by transmission electron microscopy. This work should lead to improved quality and reliability of in situ analyses of solids. They will also help to develop new fields of applications for this technique, such as in situ measurements of stable metal isotopes.

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Smaller and smarter – a new generation of laser diodes

Researchers at the LAAS Photonics Group are overcoming two challenges: inventing laser diodes that can be integrated onto a chip; and functionalizing them for new applications.

>>> Françoise LOZES, CNRS senior scientist and her "Photonique" group at the Laboratoire d'Analyse et d'Architecture des Systèmes (LAAS, CNRS/UPS).

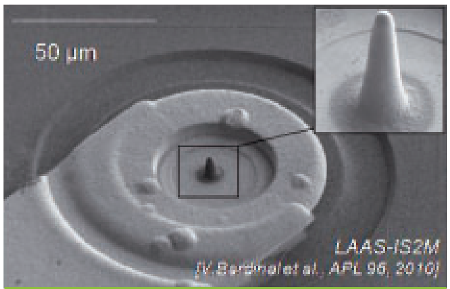
Semiconductor lasers, or laser diodes, are everywhere we look. At the heart of optical telecommunications networks, they continue to become more and more sophisticated as the Internet becomes increasingly complex. Semiconductor lasers are also central to computers and multimedia equipment, such as optical mice, in information storage and retrieval via hard disks, for transmitting data, and for communication between networks. These lasers are also widely used in many industrial processes, as surgical tools, to analyze biological samples and to monitor the environment.

Semiconductor lasers are becoming more elaborate thanks to advances in materials and nano-science, and the fundamental domains of optics and photonics is directly benefiting a diverse variety of other disciplines, such as biology and cosmology.

The Photonics Group at LAAS was a pioneer when it came to understanding different generations of laser diodes, in fields such as visible light emission, increasing laser power and obtaining surface emission. Current challenges include integrated laser diodes and optical functions to make optical "lab-on-a-chips", as has already been done in microelectronics.

Micro-cavities

The Photonics Group exploits quantum phenomena and forbidden band-gap engineering to make lasers diodes that are radically different from existing ones. The LAAS-CNRS technological facility, which is part of the Basic Technological Research Labs (rtb.cnrs.fr), has fabricated gallium arsenide-based laser sources that emit at around 1 micron. The planar photonic crystals used in these devices allow for high quality factor coefficient micro-cavities. The team has also recently integrated single-mode planar laser diodes entirely defined by photonic crystals with a precise control of the wavelength emission to better than 0.3 nm. This new cavity architecture, compatible with planar integration, is a turning point for realizing integrated photonic circuits in on-chip laser systems.



>>> Polymer micro-tip fabricated on the surface of a VCSEL and shaped by the laser beam itself.

Micro-tips

Apart from the challenge of making an integrated laser, researchers also need to broaden the field in which laser diodes are used today. These include all-optical circuits, "smart" systems that associate optics with software, opto-fluids (which combine optics with biology or chemistry), micro- or nano-systems with optical functionalities, electronics, and micro- and nano-mechanical systems (NEMS and MEMS). An important research area is the study of vertical-cavity surface-emitting lasers (VCSELs) that integrate optical filtering and photodetection, as well as micro-optics based on polymers for smart sources with weak divergence. Very recently, the team succeed in integrating self-aligned micro-tips on a VCSEL. This result opens the way to new applications in near-field spectroscopy or optical manipulation for biological analyses.

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