Extreme measurement in **laser**-generated **plasmas**

Extreme temperature and pressure conditions prevail in inertial-confinement thermonuclear fusion installations, such as the Laser Integration Line (LIL), and more emphatically yet in the Megajoule Laser (LMJ), providing major tools in CEA's Defense Program. These require the development of measuring devices providing particularly high performance, in terms of precision, measurement dynamics, and spectral range, and having the ability to operate in a particularly harsh neutron and radiation environment.

P. Stroppa/CEA

Checking diagnostics inside the Laser Integration Line (LIL) experiment chamber, the prototype installation for the Megajoule Laser (LMJ), consisting in one elementary laser line, comprising four beams

Of the four states of matter, the **plasma** state is undoubtedly the most plentiful. Whereas solids, liquids and gases are defined in terms of restrictive temperature, pressure or density conditions, the conditions under which a plasma may exist are far more extensive, making plasmas ubiquitous in our daily life (fluorescent lighting, television sets, lightning…), and right out to the confines of the Universe (stars, nebulae…). In that range of conditions of existence, hot plasmas generated through **laser**–matter interaction, for the purposes of **inertial-confinement fusion** (ICF) (see Box C, ***The principle of thermonuclear fusion***), are those exhibiting the most extreme characteristics, akin to conditions prevailing in star cores (see Figure 1).

The purpose of hot plasma experimentation

Their ability to bring matter to extreme temperature and pressure conditions (10 million degrees, 100 million **bars**) means high-energy laser installations, such as the Laser Integration Line (LIL), or the Megajoule Laser (LMJ: Laser mégajoule), two installations accommodated at CEA's CESTA center, in the Gironde *département* (southwestern France), provide tools allowing a widely diverse range of experiments to be embarked upon. These relate, first of all, to the Simulation Program of the Military Applications Division (DAM) at CEA (radiation hydrodynamics, behavior of matter at high temperatures and/or high pressures), taking in the entire ICF-related approach, but equally to more fundamental investigations (**spectral opacities**, **equations of state**, astrophysics).
ICF consists in effecting the **thermonuclear fusion** of a **deuterium–tritium** (**DT**) mix, held in a capsule, by bringing it to extreme temperature and pressure conditions (see Box, on the following page, and Figure). Achieving such conditions presents a challenge that may only be met through mastery of the

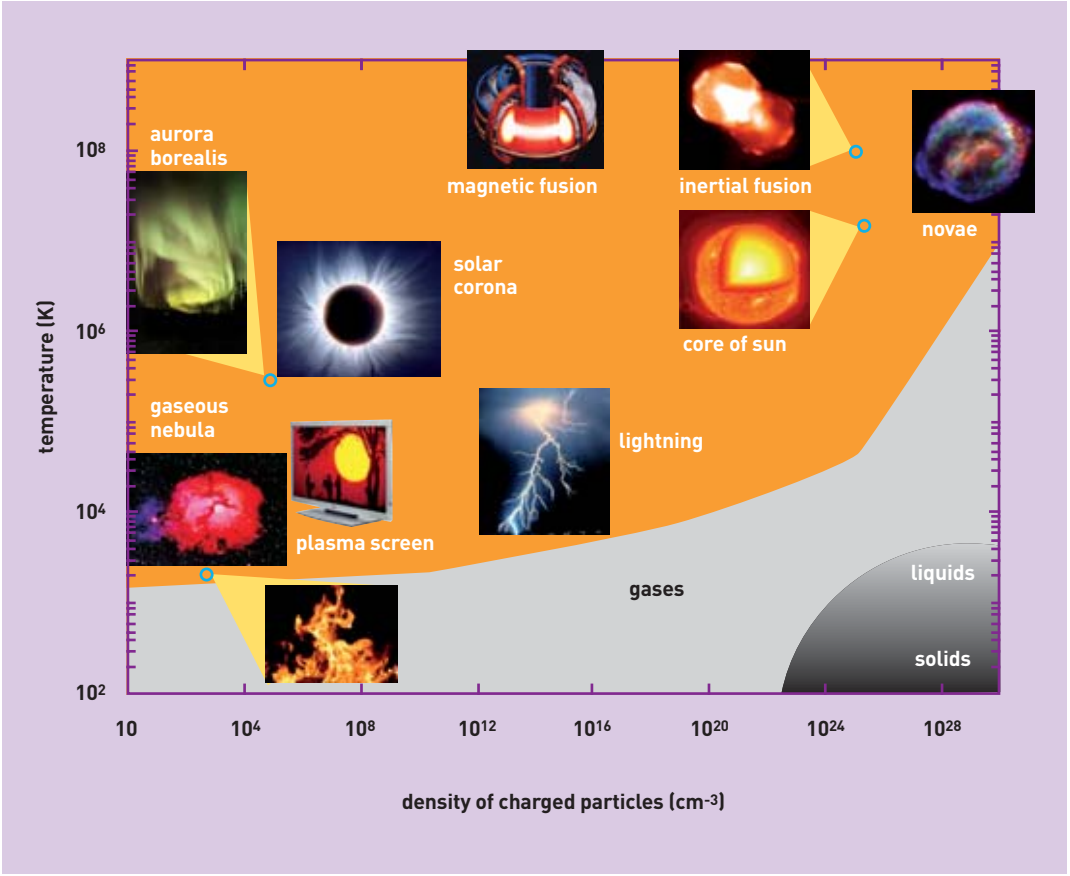

Figure 1.
Existence domains for the various plasmas, as a function of density and temperature. The plasmas found in inertial-confinement fusion exhibit extreme conditions, approximating those encountered in star cores.

multiplicity of physical processes involved in bringing the thermonuclear fuel to the required condition: coupling of the laser wave with the **ion** and **electron** waves in the plasma inside the *hohlraum* (interaction physics); conversion of the laser illumination into **X-radiation**, hydrodynamics of the plasma held in the *hohlraum*, control of capsule illumination symmetry throughout the laser pulse (*hohlraum* physics); capsule ablation by the radiation flux, control of hydrodynamic instabilities, acceleration, and combustion (capsule physics). All of these processes are being addressed by research themes, calling for dedicated experiments, to advance simulation, and understanding of the phenomena.

Inertial-confinement fusion

For the purposes of inertial-confinement fusion (ICF), the deuterium–tritium (DT) mix to be brought to the conditions required (see Box C, *The principle of thermonuclear fusion*) is held in solid form, at –250 °C, in a spherical capsule of millimetric size. This is brought to conditions of temperature and density of some 10 million degrees, and about 100 g/cm^3, by vaporizing suddenly (in a few nanoseconds) the outer shell of the capsule: due to conservation of momentum, the inner capsule implodes, compressing the DT and heating it sufficiently to cause its "inflammation," spreading from a hot spot, and set off fusion reactions (ignition).

Such heating requires considerable power, that only lasers may supply. This is achieved by illuminating the capsule by intense laser beams (direct drive), or by placing the capsule inside a *hohlraum*, used to convert the laser beams into thermal radiation (indirect drive). In the latter case, one constructs, in effect, an "X-radiation oven": it is the radiation generated as the laser energy is deposited onto the *hohlraum* walls that heats the capsule, the inner part of which implodes with a velocity of about 100 km/s, setting up in the DT the conditions for ignition (see Figure).

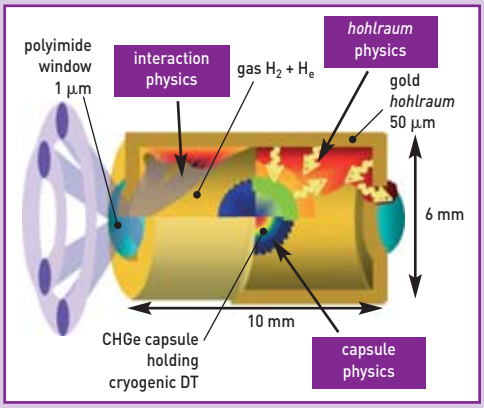

Figure.
ICF target, comprising a gold *hohlraum*, filled with a gas mix and holding a germanium-doped plastic capsule, positioned at its center, holding the fusible mix in cryogenic form. The laser beams, grouped into a number of cones, on either side of the *hohlraum*, enter it through two axial apertures, covered with thin polyimide membranes, to contain the gas. The various physical processes involved may be subsumed under laser–plasma interaction, *hohlraum* radiation hydrodynamics, and capsule hydrodynamics and combustion.

Characteristics of ICF plasmas

In the target designed for ICF, on LMJ (see Figure, in Box), the laser beams, grouped into a number of irradiation cones, on either side of the cylindrical *hohlraum*, enter through the two apertures, impacting the walls of the gold *hohlraum*, after passing through the gas (the pressure of which slows down the expansion of plasma from the *hohlraum* wall) and the polyimide(1)-covered aperture (retaining the gas).

The main characteristics of ICF plasmas (see Table 1) determine the requirements in terms of measurement dynamics, and **resolution**, for the investigation of such plasmas.

Dimensions range over four orders of magnitude, from the *hohlraum* (on the centimeter scale) to the hot spot, a few tens of **micrometers** (**µm**) across, entailing micrometric spatial resolutions for the imaging devices. As for time scales, these range over three orders of magnitude, from the duration of the laser pulse (~ 20 nanoseconds) to the DT burn time (a few tens of **picoseconds** [**ps**]). Power lasers, at the same time, allow pressure levels of several hundred megabars to be achieved, or even up to one gigabar (with LMJ).

P. Stroppa/CEA

LMJ experiment chamber being assembled at the CESTA site. On the outside of the sphere may be seen the square apertures, through which the 240 laser beams will enter (in groups of four), and those, circular as a rule, destined to the many different measuring devices.

Measurements in plasma physics

Controlling the various physical processes involved calls for mastery of the associated measurements, since validation of a simulation is dependent on measurement capabilities: measuring devices define the *observables*, these being the meeting point of experiment and simulation, and the performance of these devices determine measurement *precision*, on which depends the precision of computation **code** validation.

The measurement approach

A good many quantities to be measured, of those most relevant to the description of the state of matter, may not be directly accessed in plasmas. Such is the case for energy, temperature, pressure, or density… At the same time, as a result of the unfeasibility of effecting measurements inside, or in the vicinity of plasma, owing to the extreme conditions prevailing in it, measurement devices are set up at a remove, as instanced – with all due proportion – by measurements in astrophysics.

Measurements that are accessible are those concerning *size*, by way of imaging systems, the *evolution over time of a process*, by means of electrical or optical analyzers, *velocity*, measured by **Doppler effect**, or through a combination of the aforementioned means, finally the *energy* and *number of particles released* (including **photons**), these being measured by means of calibrated detectors, allowing the *spectral density* , or spectrum, to be retrieved.

From these basic measurements, the most relevant quantities may be derived, in some cases with the aid of theoretical **models**: energy or temperature from the spectrum, pressure from velocity readings and the Rankine–Hugoniot relations,(2) density through the broadening of X-radiation **emission lines** (**Stark effect**) or the profile of the **neutron spectrum**…

Most measurements rely on the manifold emissions from the plasma: **visible light** and **ultraviolet** (**UV**) (for the laser energy balance, or emission from a shock front…), X- and **gamma** radiation (from a few tens of **electronvolts** [**eV**] to several **MeV**), particles (**neutrons**, **electrons**, **ions**, **alpha particles**, **protons**). Examples of measurements in the spatial, temporal and spectral domains are given below.

(1) Polyimide: a high-performance engineering polymer, formed from imide monomers, this being a functional group made up of two carbonyl groups and one primary amine.

(2) Rankine–Hugoniot relations: relations (three in all) that relate values for momentum and energy, before and after passage of a shock wave set up in a compressed fluid by a piston moving at constant velocity, while not taking into account the thermodynamic state of the medium considered.

	size	stored energy	duration	temperature	other parameters
hohlraum	1 cm	1 MJ (laser)	15 ns (laser pulse)	3 MK	
capsule	1 mm	100 kJ (X-radiation)	3 ns (implosion phase)	3 MK	implosion velocity ~ 300 km/s
DT prior to fusion	100 µm	10 kJ		3 MK	max. density ~ 300
hot spot	30 µm	1 kJ		30 MK	max. density ~ 100
fusion		10 MJ yield	30 ps (combustion time)	1,000 MK	

Table 1.
Characteristic quantities for ICF plasmas.

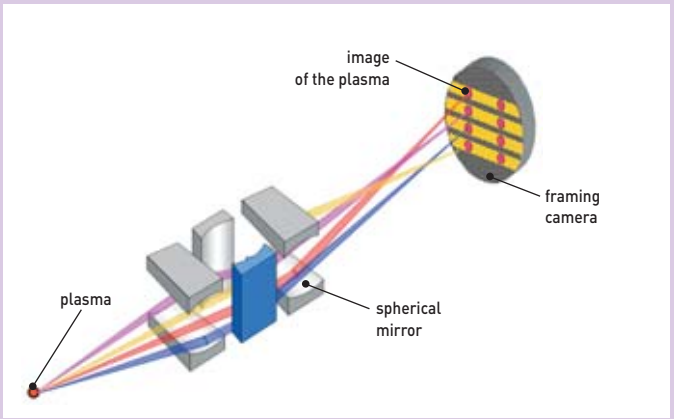

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Figure 2.
Multi-imager device for X-radiation. Use of six grazing-incidence spherical mirrors allows eight images to be produced, by coupling the mirrors two by two (Kirkpatrick and Baez layout). Shown in the figure, the formation of the four images yielded by reflections on the blue vertical mirror, and the four horizontal mirrors. The other four images are yielded by reflections on the other vertical mirror. The eight images are captured at different points in time by means of an framing camera.

Measuring dimensions in the X region

Requirements for imaging systems in the X region are quite plentiful, whether it be to investigate the plasma's own emission or to observe target evolution by radiography. There are various types of imaging devices, functioning by way of projection, reflection, or diffraction. The simple **pinhole** works by projection: its lack of sensitivity may be remedied by means of coded apertures (assembling a number of pinholes according to a variety of geometries), these however requiring **deconvolution algorithms**, in order to yield the object. Reflection is used in X-ray optics involving grazing-incidence mirrors, which may be coupled with *Bragg reflection* (spectral resolution by means of multilayer interference mirrors, or curved crystals). Diffraction is employed by Fresnel zones (circular, variable-pitch diffraction slits: the analogues, in the X region, of Fresnel lenses, as used for marine beacons, in the visible region), which may be coupled to multilayer mirrors. The system using reflection shown in Figure 2, for instance, achieves micrometric resolutions with an operating flexibility that accommodates many different geometries. The basic configuration is that of the optical system devised by Kirkpatrick and Baez, comprising two orthogonal spherical mirrors, operating in grazing incidence. Each mirror carries out focusing in one direction, coupling of the two yielding a two-dimensional image. This device allows a 5-µm resolution to be achieved, over a restricted 100-µm field of view, and 10-µm resolution over a 1-mm field.
Building on this basic structure, a multi-imager system was designed, using six spherical mirrors, two of which are orthogonal to the others. By coupling the mirrors two by two, eight images of the same objects are generated, and may be recorded at different times, to analyze the object's evolution.
By changing the nature of the mirror coating, and altering the angle of incidence, different energy ranges may be worked with: two separate devices were thus developed (see Table 2).

Temporal measurements

The extremely fast processes involved in hot plasma physics call, for their investigation, for temporal analysis at a resolution higher than 100 ps. There is no device available, having the ability to record an image directly at such a high time resolution. Solving this problem entails the use of systems converting photonic signals into electronic signals, which are more readily processed to achieve the required resolutions.

Framing cameras

Framing cameras (FCs; see Figure 3) have the capability to take pictures every 100 ps, with an exposure time lower than 70 ps. Several images of the object to be analyzed are formed simultaneously, on the input side of a microchannel plate, covered with **photocathodes**. The photons generate electrons by **photoelectric effect**, allowing low-intensity electronic images to be obtained. Inside the plate, each microchannel, about 10 µm in diameter – this defining the system's spatial resolution – plays the role of an adjustable-release electron amplifier. The images obtained at the back of the plate are more intense by far (by a factor up to 10,000), and electron fluxes exiting the

multi-imager	angle of incidence	mirror coating	spectral range	magnification
soft X-rays	1.8 °	silicon	100 eV - 1keV	4
hard X-rays	0.8 °	platinum	1 - 5 keV	8

Table 2.
Characteristics of the multi-imager systems developed for the Laser Integration Line (LIL).

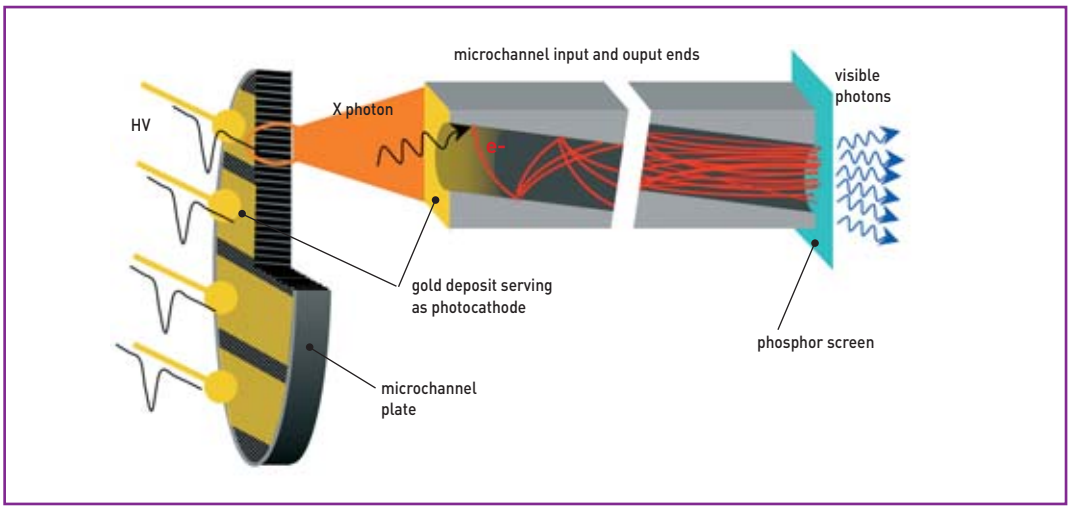
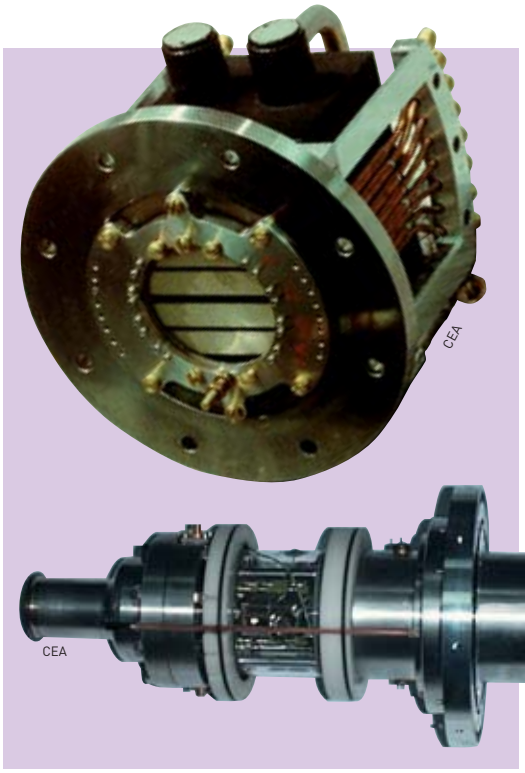


Figure 3.
Schematic of the functioning of a pulsed microchannel plate, in an framing camera (FC). X photons generate photoelectrons at the input end of the microchannels, each of which acts as an electronic amplifier in the brief duration interval of the high-voltage pulse applied to the gold strip deposited on the front side of the plate.

channels impact a phosphor screen, thus forming again a visible image, "recordable" by means of conventional image-capture systems, photographic films or **CCDs**.
The temporal aspect is obtained by triggering the microchannels, by means of a pulsed voltage propagating along gold strips, deposited on the front side of the plate, playing the role of photocathodes. The propagation speed and full width at mid-height of the electric pulse, used to **bias** the plate, determine exposure time for each image. By using several gold strips, triggered with varying delays, images taken at successive points in time appear on the FC's back screen, reproducing the images formed on the front side.



Framing camera, with its four electrode strips at the front, and high-voltage connectors at the back.

Streak camera.

Streak cameras

The streak camera (SC; see Figure 4) is an ultrarapid cinema camera, which has been in widespread use, for a long time, but for which developments in electronics have enabled considerably enhanced performance. A photocathode, forming a slit, converts radiation into electrons by photoelectric effect. These are then accelerated, the electron beam being focused by electronic optics positioned mid-tube, going on to yield in turn

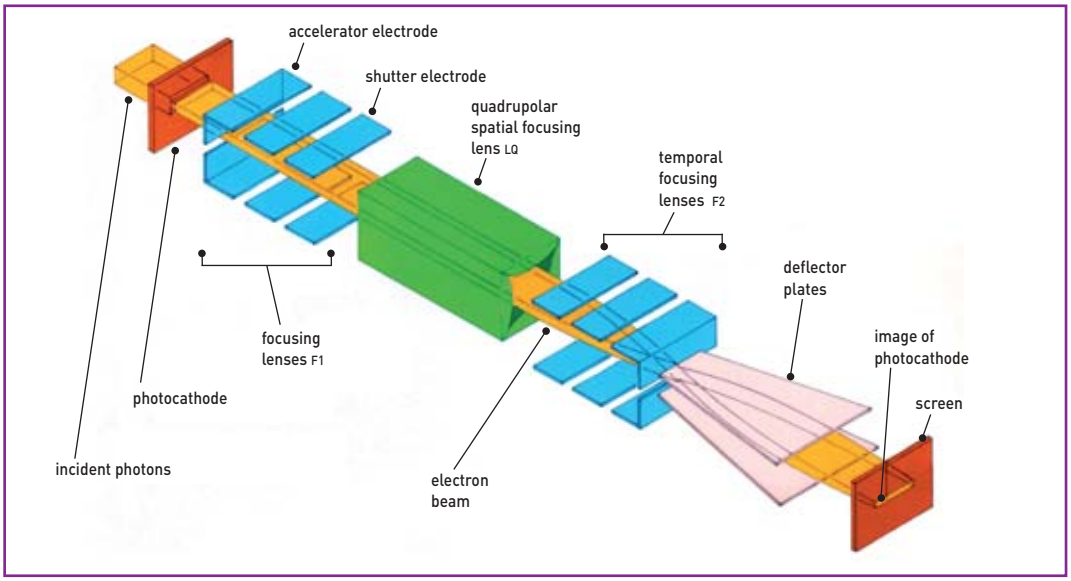


Figure 4.
Streak camera (SC).
The X radiation falling on the photocathode is converted into an electron beam which, after shaping by an electronic optics, is deflected over time by deflector plates, before impacting the output screen, which yields in turn a visible photonic signal.

the image of the photocathode on a phosphor screen at the back. Prior to impacting the screen, the beam travels through a space between two deflector plates. Applying a pulsed voltage ramp to these plates allows the path of these electrons to be deflected as a function of time, the lattermost electrons (yielded in turn by the lattermost photons) undergoing the largest deflection. A two-dimensional space-time image is thus formed, presenting the evolution over time (y-axis) of the intensity of radiation reaching the photocathode slit (x-axis), with a resolution of about 10 ps. Finally, this image is recorded on film by way of a brightness intensifier, or by means of a very-low-readout-noise CCD array detector.

Towards picosecond resolutions

Current limitations, for such devices, are related to power electronics, and the difficulty encountered with the propagation of steep-slope, high voltage pulses. While some devices may achieve resolutions of the order of the picosecond, or even less, this invariably entails a loss in measurement dynamics: the analysis ceases to be quantitative.

While ICF investigations do not require such resolutions, research workers may find they need to use, for investigations of a more fundamental character, short laser pulses (< 1 ps), generating radiation sources that are themselves "sub-ps." Time resolution is then determined by the duration of such sources, rather than by the analytical device.

Measuring temperature

A large proportion of the experiments make use of an **indirect-drive** configuration, whereby the laser interacts inside a *hohlraum* with a wall made of a high-**Z** material (e.g. gold): the X-radiation energy generated by the laser is recycled within the *hohlraum* walls (absorption–emission), and becomes **thermalized**. The X-radiation inside the *hohlraum* approximates that from a **black body**, and may then be characterized by its radiation temperature, which has to be measured.

Determining the temperature of a very small object is no easy task, the more so since the aim is not merely to measure the maximum temperature achieved inside the capsule, but rather to ascertain its evolution over time (with a 100-picosecond resolution).

A number of methods are used, such as measuring the time the radiation flux takes to go through a gold foil (from which temperature is derived, by way of a theoretical model), or measuring the time taken for the shock wave generated by the radiation flux to pass through a material for which the equation of state is know, this allowing the pressure generated within the sample to be worked out, and hence radiation temperature.

The most reliable measurement however, and one not requiring use of theoretical models, remains that obtained by observation of the spectrum emitted by the hohlraum wall, through an aperture. Any body, when heated, undergoes a rise in luminosity, even as its **emission spectrum** shifts to shorter wavelengths. At a temperature of a few million degrees, the emissivity maximum lies in the low-energy X region (100–200 eV). Absolute measurement of this emissivity, by means of a wideband **spectrometer**, yields *hohlraum* temperature.

For that purpose, some 20 measurement channels, comprising filters, grazing-incidence mirrors and coaxial-geometry vacuum diodes (see Figure 5) are set up at a distance of several meters. By changing the nature of the filter, and of the mirror in each channel, a different portion of the emission spectrum is selected, in the 50 eV–20 keV bracket, with spectral resolution standing at $1 < E/\Delta E < 6$. The signal captured by each diode is recorded on a high-speed oscilloscope,(3) to achieve the required time resolution (100 ps). A specific software program readjusts the signals relative to one another, reconstructs the radiation spectrum from the *hohlraum* (typically lying in a range from 10^{12} W/sr/cm^2 to a few 10^{15} W/sr/cm^2), and yields its temperature at every point in time, in the 50–300 eV bracket, with an uncertainty of less than 5%.

One step further into the extreme: the LMJ experimental environment

The devices presented in the above are in use on various laser installations, in experiments serving as preparations for those that will be carried out on LMJ, to

(3) Oscilloscope: an electronic apparatus allowing real-time visualization of the instantaneous value, and evolution over time, of signals varying as a function of time.

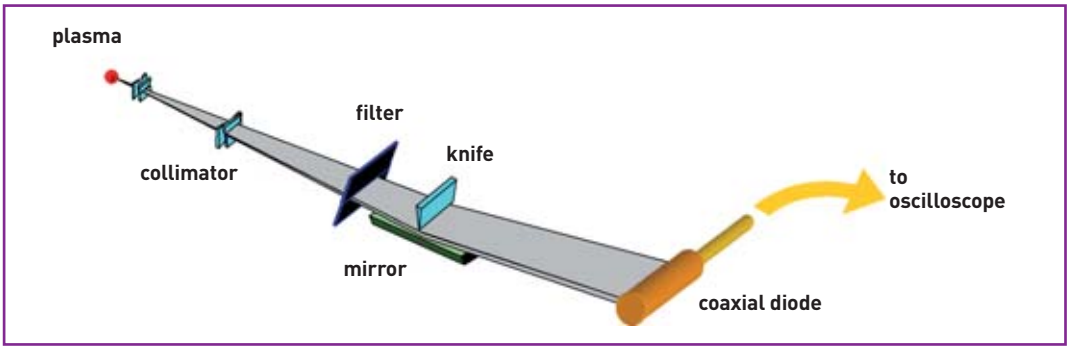


Figure 5.
Principle of wideband spectrometer, for the measurement of *hohlraum* radiation temperature. The spectrometer comprises some 20 measurement channels, each comprising in turn various components, serving to effect the selection, in spatial and spectral terms, of a segment of radiated spectrum. The spectral range of a given measurement channel depends on the nature of the filter, of the mirror, and of the photocathode of the coaxial diode. Through judicious modification of these components, a variety of spectral ranges may be defined, sampling the spectrum in the 50 eV–20keV bracket. This device is fitted to the Omega installation, at the University of Rochester (New York State).

Rochester University

Wideband spectrometer, to measure hohlraum radiation temperature, fitted to the Omega installation at the University of Rochester (New York State), in the United States.

achieve **ignition**. While these do involve extreme characteristics, in terms of precision, measurement dynamics or spectral range, they do not, as yet, take in the constraints involved in the LMJ experimental environment.

Indeed, as and when an ICF target functions, the intense neutron and gamma-radiation flux this will yield will generate severe perturbations, which may go so far as to result in destruction of part of the measuring devices. Developing measuring equipment for such an environment sets a challenge that is all the more ambitious since bringing matter to the required conditions is never an ideal process, and the causes of ICF target malfunction must be meticulously identified, by effecting the measurements appropriate for that purpose. Design of complete systems, from detection to signal recording, more robust with regard to perturbation-inducing radiation, is one direction for research that has already proved fruitful, but which yet provides a vast area to be explored over the coming years, in anticipation of the first LMJ firings.

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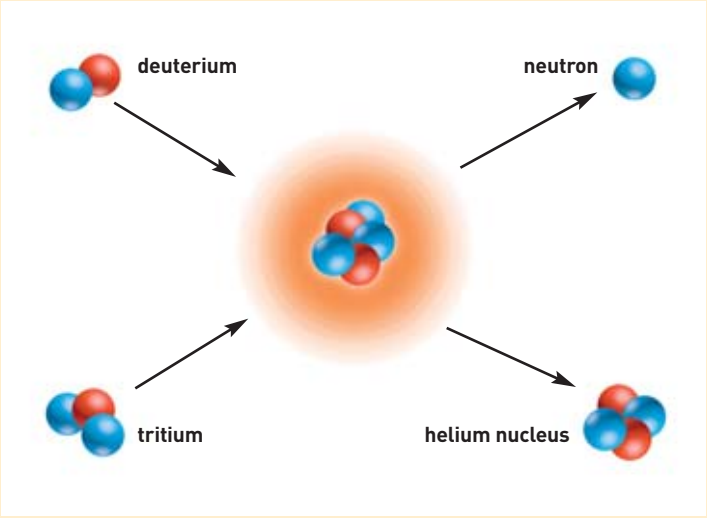
C The principle of thermonuclear fusion

The fusion reaction that is most readily effected is that between deuterium (D) and tritium (T), two isotopes of hydrogen (see Figure). This reaction requires that the DT mix be brought to a temperature of 100 million degrees, and that it remain confined for a time meeting the *Lawson criterion*: the product of density by confinement time must be greater than 10^{20} s/m^3. To set up the conditions for fusion of a light-element plasma, two confinement methods have been developed: magnetic confinement, in tokamaks, corresponding to a stationary regime, where density of the order of 10^{20} m^{-3} is maintained for several seconds by means of a magnetic field; and inertial confinement by laser beams, or particle beams, an explosive regime where density reaches 10^{31} m^{-3} for some 10^{-11} s.



For further information: see Clefs CEA *No. 49, pp. 45–76.*

D Spectroscopy and spectrometry

Spectrometric methods are subdivided, as a whole, into two main categories, radiation spectrometry – itself comprising absorption spectrometry, emission spectrometry, Raman scattering spectrometry, and nuclear magnetic resonance spectrometry – and mass spectrometry.

Radiation spectroscopy and spectrometry[1] cover a ensemble of analytical methods allowing the composition and structure of matter to be ascertained, based on investigation of the spectra yielded by the interaction between atoms and molecules, and various types of electromagnetic radiation, emitted, absorbed, or scattered by the former.

Depending on their energy, photons interact selectively with the various electron shells, or levels, making up the electronic structure of the atom, or molecule. The electrons involved are *core electrons* (close to the atom's nucleus), for X-rays,[2] *peripheral electrons* (furthest from the nucleus, and involved in chemical bonds) for light absorbed, or emitted, in the near ultraviolet and visible region. In the infrared radiation region, it is the leap from one *molecular vibration* level to another that is involved, the switch from one molecular *rotation* level to another for microwave radiation, and atomic nucleus spin for NMR.

Absorption spectrometry

Those spectroscopy methods that rely on absorption make use of the Beer–Lambert law, setting out the proportional relation between the intensity of light absorbed, and the amount of absorbing matter:

$$A = \log (I_0/I) = \varepsilon \, l \, C,$$

where A stands for the absorbance of the medium traversed, I_0 for incident light intensity, I for transmitted light intensity, ε is the characteristic molar extinction coefficient, for a given wavelength, for the substance investigated – expressed in L mol^{-1} cm^{-1} – while l stands for the thickness passed through, expressed in centimeters, and C is the concentration, in moles per liter.

By measuring the medium's absorbance, for a given wavelength, the concentration of a substance, in a sample, may thus be determined.

In an absorption spectrum, as recorded by means of a spectrometer, *absorption peaks* correspond to the wavelengths the medium is able to absorb. Just as the spectrum from the Sun's light is obtained by making it pass through a prism, which breaks it up, spectrometers analyze the spectral distribution of the whole range of electromagnetic radiations, separating them out according to wavelength, by means of a reflection diffraction grating. Spectra exhibit peaks, each one corresponding to a specific wavelength.

Depending of the type of sample to be analyzed, and the performance level being sought, in the laboratory, absorption spectrometry is used either on molecules in liquid or gaseous phase, or on atomic vapor, obtained through thermal breakdown of liquid or solid samples.

Molecular absorption spectroscopy, in the UV–visible region, affords simplicity of use, however it is only applicable to samples of moderate complexity, since, owing to the width of *molecular absorption bands*, absorption spectra, as a rule, do not allow specific discrimination of every constituent, in a complex mixture.

In infrared (IR) spectrometry, absorption is the outcome of molecular vibration and rotation processes. Infrared absorption spectra thus allow the nature of chemical bonds to be determined, that make up a molecule, by ascertaining the bond's elasticity constant (influencing vibration frequency, as for a spring), thus confirming structural hypotheses.

As the number of atoms increases, the spectrum rapidly exhibits growing complexity, and interpretation becomes highly problematical, especially for organic compounds.

Atomic absorption spectrometry, in this respect, brings higher performance, since absorption by atoms yields very narrow absorption lines. Very precise measurements are thus feasible, even when the sample consists in a complex assembly of chemical elements. Atomic absorption is a reference technique for the analysis of trace elements in a wide variety of samples, in particular for biological samples.

Emission spectrometry

Atoms or molecules brought to an excited state may deexcite by emitting radiation, known as *emission radiation*. When the excitation is caused by selective absorption, by the atoms or molecules to be analyzed, of electromagnetic radiation, this represents a fluorescence emission (or a phosphorescence emission, depending on the electron excitation state involved).

As with absorption, fluorescence may be applied, in the UV–visible radiation region, to molecules, or atoms. X-ray fluorescence spectrometry, on the other hand, refers to the X radiation emitted by atoms excited by absorption of X-radiation. Fluorescence techniques are more complex to implement than is the case for absorption techniques, since they entail that the particle subjected to analysis be selectively excited by a monochromatic radiation. On the other hand, since the radiation emitted is likewise specific to the particle, fluorescence spectrometry involves a double selectivity, resulting in very low background noise, thus making it peculiarly well suited for the measurement of very low concentrations.

Emission of radiation may also occur when atoms are thermally excited, in an environment brought to high temperatures. Emission spectroscopy is based on the fact that atoms, or molecules excited to high energy levels deexcite to lower levels, by emitting radiation (emission, or luminescence). This differs from fluorescence spectrometry in that excitation is not applied selectively, rather it involves indiscriminately all of the particles making up the medium. Emission lines thus correspond to radiation directly emitted by a body brought to a high temperature, and the emission spectrum allows the detection, and quantification, of all atoms present in the emission source.

Raman spectrometry

Interactions between matter and electromagnetic radiation also give rise to scattering processes, such as elastic scattering, and inelastic scattering. Scattering may occur when the interface between

(1) The term "spectrometry," initially used only to refer to recording and measurement techniques, has tended to become synonymous with "spectroscopy," as the eye was supplanted, for observation purposes, by other receptors and instruments, while the visible region now only formed one special region, in analytical terms.

(2) It should be noted, at the same time, that X-ray crystallography is not deemed to be a spectroscopy method, in the strict sense of the term.

two media is encountered, or as a medium is passed through. This process, in most cases, is an "elastic" one, in other words it takes place with no change in frequency for the radiation forming the beam involved. Elastic scattering of solar radiation by the atmosphere is, for instance, responsible for the blueness of the sky, observed when the eye is not directed towards the Sun (*Tyndall effect*). Indeed, scattered intensity is all the greater, the shorter the radiation wavelength, which, in the case of the solar spectrum, corresponds to the color blue.

As regards spectrometry, the main use of scattering concerns *Raman spectrometry*. This involves the inelastic scattering of incident radiation by the molecules making up the sample. The difference between scattered radiation frequency, and incident radiation frequency allows the identification of the chemical bonds involved. Raman spectrometry is a technique that is widely used for structural analysis, to complement infrared spectrometry, and mass spectrometry.

Nuclear magnetic resonance spectrometry

The principle of nuclear magnetic resonance (NMR) is based on the fact that an atom has a *magnetic moment*, just like a spinning charge acting as a tiny magnet, governed by quantum mechanics, aligning in a magnetic field as the needle of a compass in the Earth's magnetic field. The principle of NMR consists in inducing, and detecting, the transition, for the nuclear magnetic moment, from the lowest energy level to the highest energy level, through absorption of electromagnetic radiation of a wavelength lying in the radiofrequency region: when the energy of the photon precisely matches the energy difference between the two levels, absorption occurs. Nuclei having numbers of protons, and neutrons that are both even exhibit zero spin. Carbon 12 and oxygen 16 atoms, which are very widespread in nature, thus have zero spin. On the other hand, hydrogen only has one single proton, and its nuclear magnetic moment equals 1/2: it may thus take on two possible energy states, corresponding to the two orientation states of its spin, relative to the magnetic field. Measuring the resonance frequency in the electromagnetic field allowing transition from one of these energy states to the other enables the molecules to be analyzed. This frequency is fixed, however the various nuclei in a molecule do not all resonate at the same frequency, since their magnetic environment is modified by their chemical (electronic) environment.

Many NMR spectra exhibit more peaks than there are protons in the nucleus, owing to the interactions between protons and their neighbors. Two nuclei may interact within the molecule, though they are separated by several chemical bonds: this is known as interatomic coupling. This interaction endows the NMR spectrum with a fine structure.

C. Dupont/CEA

Spectromètre de masse d'ions secondaires utilisé au CEA pour réaliser des mesures isotopiques rapides sur un échantillon par exemple prélevé sur une installation aux activités nucléaires suspectes.

Mass spectrometry

Mass spectrometry is a highly sensitive *detection* and *identification* technique, allowing determination of molecular structures, and thus of a sample's composition. This is not, strictly speaking, a form of spectrometry, since it is not concerned with discrete energy levels. What is its principle? A compound introduced into the device is vaporized, and subsequently ionized by an electron bombardment source (at 70 eV). The ion thus obtained, termed a molecular ion, allows the compound's molar mass to be determined. Breaking chemical bonds within the compound may yield characteristic fragment ions. These are then sorted according to their mass/charge ratio in an *analyzer*, through application of a magnetic and/or electric field, then collected by a *detector*, which amplifies the signal associated to the ions, which arrive with varying delays. A data processing system converts the information from the detector into a mass spectrum, readout of which, by comparing it with reference spectra, allows the identity details of the molecule to be drawn up. Through use of a high-resolution mass spectrometer, the exact mass of the compound may be determined, together with isotope percentages for each constituent atom.

Choice of ionization method is directly related to the nature of the sample, and the type of analysis. If mass spectrometry has gradually adapted to meet the growing demands from chemists, and biologists (separation of increasingly complex, highly polarized mixtures, determination of ever higher molecular masses on samples of ever more constricted sizes), this is essentially due to advances in *ionization techniques*, these including secondary ion mass spectrometry (SIMS), chemical ionization, thermospray ionization, and fast atom bombardment (FAB) sources, further comprising, from the 1980s, matrix-assisted laser desorption ionization (MALDI), and electrospray ionization (ESI), together with advances in *detection techniques*, from time-of-flight (TOF) measurement to "ion traps" (ITs), through quadrupoles (MS or Q).

In proteomics, for instance, only MALDI, ESI and SELDI (surface-enhanced laser desorption ionization) are employed.

Ion mobility spectrometry (IMS) is a chemical analysis technique in the gaseous phase, which consists in subjecting a gas to an electric field. Ionized molecules acquire a velocity that is characteristic for the ion, since this depends on mass, and charge. Arrival of the ions on one of the plates generating the field results in a current, which is recorded. The length of time after which a peak occurs can be related to the nature of the ion causing it.

Scientists often make use of a coupling of devices each belonging to one of the two main families of analytical techniques (see Box E, *What is chromatography?*), e.g. of a chromatograph with a mass spectrometer (or an electron-capture detector [ECD]), particularly for the investigation of trace complex mixtures.

B Fundamental interactions and elementary particles

The standard model of particle physics is the reference theoretical framework describing all known elementary particles (see Table 1) and the fundamental interactions these particles are involved in (see Table 2). The basic constituents of matter, known as fermions, are partitioned into two main categories, as determined by their participation in the fundamental interactions, or forces (the gravitational, electromagnetic, weak, and strong forces), which are mediated by vector bosons, the fundamental particles which carry out the transmission of the forces of nature[1] (see Table 2). Whether a particle belongs to the category of fermions, or to that of bosons depends on its spin (i.e. its intrinsic angular moment, or internal rotation moment), depending on whether it exhibits half-integer spin (fermions) or integer spin (bosons).

At the same time, to every constituent of matter is associated its antiparticle, a particle having the same *mass*, but the opposite *charge*. The positron is thus the positively charged antiparticle of the electron, which exhibits a negative charge.

Leptons and quarks

Fermions include, on the one hand, leptons, which may travel freely and do not participate in the *strong interaction*, which ensures the cohesion of atomic nuclei (it is consequently termed a *nuclear* interaction), and, on the other hand, quarks, which participate in all interactions but are not individually observed, enmeshed and confined as they are within hadrons, the particles susceptible to strong interaction, of which they are the constituents.[2]

In the lepton category, charged leptons participate in the *electromagnetic interaction* (which ensures the cohesion of atoms and molecules, and in the *weak interaction* (which underlies decay processes, in particular β radioactivity). Neutral leptons, or neutrinos, for their part, participate in the weak interaction only. Exhibiting very low mass, there is one type of neutrino for each type of charged lepton.

Independently from their involvement in interactions, the basic constituents of matter are classified into three *generations*, or *families*, of particles. From one family to the next, quarks and leptons having the same charges only differ by their mass, each family being heavier than the preceding one.

The electron, up quark (symbolized *u*) and down quark (symbol *d*), which belong to the first generation, are the lightest massive particles, and are stable. These are the sole constituents of normal matter, so-called baryonic matter (a baryon is an assembly of quarks), which is made up of protons and neutrons, this however only accounting for 4% of the Universe's energy content!

Particles in the other two families are heavier, and are unstable, except for neutrinos, which on the other hand exhibit non-zero mass, but are stable.

These latter particles may only be observed or detected in the final states resulting from collisions effected in accelerators, or in cosmic radiation, and rapidly decay into stable first-generation particles. This is why all the stable matter in the Universe is made up from constituents from the first family.

According to quantum mechanics, for an interaction to take place between particles of normal matter, at least one elementary particle, a boson, must be emitted, absorbed, or exchanged. The photon is the intermediate (or vector) boson for the electromagnetic interaction, the W^+, W^- and Z are the intermediate bosons for the weak interaction, and gluons are those of the strong interaction, acting at quark level.

As to the graviton, the putative vector for the gravitational interaction, it has not so far been empirically discovered. The gravitational force, which acts on all fermions in proportion to their mass, is not included in the standard model, due in particular to the fact that quantum field theory, when applied to gravitation, does not yield a viable scheme, as it stands. While gravitational effects are negligible in particle physics measurements, they become predominant on astronomical scales.

Interaction ranges

Quarks and charged leptons exchange photons. The photon having no electric charge, these particles conserve their electric charge after the exchange. Since the photon's mass is zero, the electromagnetic interaction has an infinite range. Having no electric charge, neutrinos are the only elementary fermions that are not subject to electromagnetic interaction.

In the electroweak theory (a unification of the weak and electromagnetic interactions), the weak interaction has two aspects: charged-current weak interaction, for which the interaction vectors are the W^+ and W^-; and neutral-current weak interaction, for which the mediator is Z^0. These two forms of weak interaction are active between all elementary fermions (quarks, charged leptons and neutrinos). The mass of these bosons being very large (about 80 GeV/c^2 for $W^\pm$, 91 GeV/c^2 for Z^0), the range of the weak interaction is tiny – of the order of 10^{-18} m. Since $W^\pm$ bosons have a non-zero electric charge, fermions exchanging such bosons undergo a change in electric charge, as of nature (*flavor*). Conversely, since the Z^0 boson has no electric charge, fermions exchanging one undergo no change in nature. In effect, neutral-current weak interaction is somewhat akin to exchanging a photon. As a general rule, if two fermions are able to exchange a photon, they can also exchange a Z^0. On the other hand, a neutrino has the ability to exchange a Z^0 with another particle, though not a photon.

Only those quarks that have a color charge[1] exchange gluons, these in turn being bearers of a color charge. Thus,

(1) The participation of basic constituents in fundamental interactions is governed by their *interaction charges* (electric charge, color charge), or "conserved quantum numbers." *Color charge*, a quantum number that determines participation in strong interactions, may take one of three values: "red," "green," or "blue" (these colors bearing no relation to visible colors). Every quark bears one of these color charges, every antiquark one of the three anticolor charges. Gluons are endowed with double color–anticolor charges (eight combinations being possible).

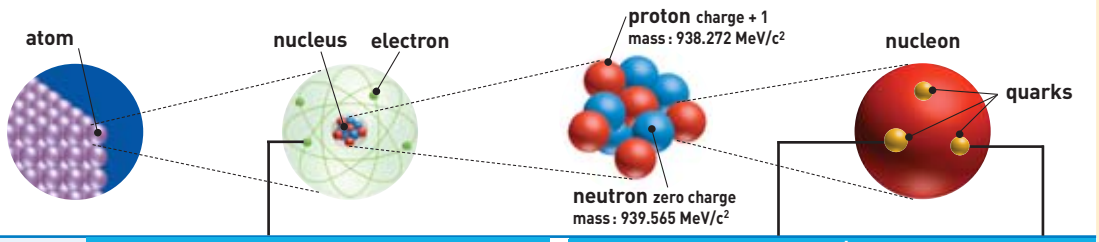
(2) To take e.g. nucleons: the proton holds two up quarks and one down quark, the neutron two down quarks and one up quark. A meson is made up of just two quarks (one quark and one antiquark).

B (cont'd)

when a gluon exchange takes place between quarks, the latter exchange their respective colors. Gluons have zero mass, however, since they do bear a color charge, they are able to interact together, which greatly complicates theoretical treatment of this interaction. The range of the strong interaction is consequently very restricted – of the order of 10^{-15} m.

The quest for unification

The theoretical framework for the standard model is quantum field theory, which allows a quantitative description to be made of the fundamental interac-



		leptons (able to move freely)		quarks (assembled into triplets, or quark–antiquark pairs, to form the many subatomic particles)	
Fermions Normal matter is made up of particles from this group. Most of these particles were around just after the Big Bang. Presently only to be found in cosmic rays, and around accelerators.	First family	**electron (e)** responsible for electricity and chemical reactions charge: - 1 mass : 0.511 MeV/c²	**electron neutrino (ν_e)** has no electric charge, and interacts very seldom with the ambient medium.	**down (*d*)** electric charge: - 1/3 the proton holds one, the neutron two mass : 4 – 8 MeV/c²	**up (*u*)** electric charge: + 2/3 the proton holds two, the neutron one mass : 1.5 – 4 MeV/c²
	Second family	**muon (μ)** a more massive companion to the electron. mass : 105.658 MeV/c²	**muon neutrino (ν_μ)** properties similar to those of the electron neutrino.	**strange (*s*)** a heavier companion to "up" mass : 80 – 130 MeV/c²	**charmé (*c*)** a heavier companion to "down" mass : 1.15 – 1.35 GeV/c²
	Third family	**tau particle (τ)** heavier still. masse : 1,776.99 ± 0.29 MeV/c²	**tau neutrino (ν_τ)** properties similar to those of the electron neutrino.	**beauty (*b*)** tau particle. mass : 4.1 – 4.4 GeV/c²	**top (*t*)** heaviest in the family (observed in 1995) mass : 171.4 ± 2.1 GeV/c²
Vector bosons Fundamental particles carrying out transmission of natural forces.		**photon** elementary grain of light, vector for the electromagnetic force	**gluon** bearer of the strong force between quarks	**$W^{\pm}$, Z^0** bearers of the weak force, responsible for some forms of radioactive decay	
Higgs boson?		responsible for "electroweak symmetry breaking"			

Tableau 1.
Table showing the twelve elementary constituents for which the standard model describes the interactions involved. The three charged leptons (electron e-, muon, μ^-, tau particle τ^-) are subject to electromagnetic and weak interactions, neutrinos (ν_e, ν_μ, ν_τ) are only affected by weak interaction, and the six quarks (up, charm, top – or *u*, *c*, *t* – bearing a charge of 2/3; and down, strange, bottom – *d*, *s*, *b* – bearing a charge of – 1/3) are subject to all three interactions. Every elementary constituent has its antiparticle, having the same mass, and algebraic quantum numbers (such as electric charge) of the opposite sign.

tions between elementary particles, while respecting the principles of *special relativity*, as those of quantum mechanics. According to the latter theory, if one seeks to observe a microscopic structure at high temporal and spatial resolution, this entails transferring to it an amount of energy–momentum, the greater, the higher the resolution being sought. However, according to the theory of relativity, such an energy–momentum transfer is liable to undergo transformation, yielding particles not present in the initial state: fermions may be generated, or annihilated, in particle–antiparticle pairs, while bosons may be so in any arbitrary number.

All processes involving one and the same fundamental interaction are interrelated. The quantum field theory approach, in which properties of **symmetry** play a fundamental part, seeks to describe all of the processes relating to each fundamental interaction, within overarching theoretical constructions.

The strong and electromagnetic interactions are formalized, respectively, in the theories of **quantum chromodynamics**, and **quantum electrodynamics**. The weak interaction, for its part, is not subject to a separate description, being described jointly with the electromagnetic interaction, in the unified formalism of **electroweak theory**. Theories of the *grand unification* of all fundamental interactions do exist, however they remain as yet lacking any experimental validation.

All the predictions of the standard model have been corroborated by experiment, except for just one, to wit, the existence of the **Higgs boson(s)**, which particle (particles?), it is hoped, will be discovered with LHC. The **Higgs mechanism** is thought to be responsible for the mass exhibited by elementary particles, the eponymous boson making it possible for zero-mass fermions interacting with it to be endowed with mass. This would allow the unification, at high energies, of the weak and electromagnetic interactions within the electroweak theory, while effectively accounting for the **breaking** of this **electroweak symmetry** at low energies, taking the form of two interactions, which may be seen as distinct at that energy level (see *The electroweak interaction from one accelerator to the next: the LHC roadmap and the yardstick of LEP measurements*, p. 23).

Going beyond, or completing the standard model?

The standard model features a set of parameters (such as the masses of elementary particles, or the intensities of fundamental forces) which are "anchored" in experimental findings. It is, in any event, a theory that is liable to be improved, or further elaborated, or even surpassed and left behind. It does not account in any way for the classification of the constituents of matter into three generations of particles, whereas it is precisely the existence of these three generations which makes it possible to account for **CP** (charge–parity) **invariance violation** (meaning that a physical process involving the weak interaction is not equivalent to its own mirror image), a violation that is in all likelihood the source of the matter–**antimatter** imbalance, running in favor of the former, in the primordial Universe. The model neither allows quantum treatment of gravitation, nor does it fully account for the fundamental property of *confinement*, which prevents quarks from propagating freely outside hadrons.

To go beyond, or to complete the standard model, research workers are mainly exploring two avenues:

– **supersymmetry** (widely known as SUSY) would associate, to every particle (whether a boson or a fermion) in the standard model, a partner from the other series, respectively a fermion or a boson. Supersymmetric partners would, at first blush, be highly massive, the lightest of them being a particle interacting very weakly only. This would be an ideal candidate to account for the **hidden matter** (or **dark matter**) in the Universe, accounting as it does for some 21% of the Universe's energy content, the remainder (close to 75%) consisting in a **dark energy**, the nature of which likewise remains to be determined. These WIMPs (acronym for "weakly interacting massive particles") are actively being sought (see *EDELWEISS II, the quest for dark matter particles*);

– the **substructure** path assumes there could be a new level of elementarity, underlying the particles in the standard model (or some of them). This would lead to a veritable blossoming of new, composite particles, analogous to hadrons, but exhibiting masses two to three thousand times heavier.

It should be noted that, whereas supersymmetry theories yield predictions that agree with the precision measurements carried out at LEP, the theories propounding substructures (or their simpler variants, at any rate) fail to do so. As for the more complex variants, these are encountering difficulties at the theoretical level.

fundamental interaction	associated particles (messengers)	actions
gravitation	graviton?	having an infinite range responsible for the mutual attraction of any two masses and for the law of falling bodies
electromagnetic interaction	photon	having an infinite range responsible for the attraction between electrons and atomic nuclei, hence for the cohesion of atoms and molecules
weak interaction	W^+, W^-, Z^0	responsible for β^- and β^+ radioactivity, reactions involving particles as neutrinos
strong interaction	gluons (there are 8 gluons)	ensures the cohesion of the atomic nucleus

Tableau 2.
Fundamental interactions, their vectors, and effects.

E What is chromatography?

Chromatography, together with the various forms of spectroscopy and spectrometry (see Box D, *Spectroscopy and spectrometry*), represent the two major basic analytical techniques, the former serving for the separation, the latter for the identification of the constituents of a substance.

Chromatography (from the Greek *khrôma*, "color," and *graphein*, "to write"), allows the *separation* of the constituents of a mixture in a homogeneous liquid or gaseous phase, as blotting paper might spread out in concentric rings a liquid poured onto it.

A chromatograph comprises a sample injection device, a *column*, a detector, and a recording and analysis system. Its principle is based on the equilibrium of compound concentrations, between two phases coming into contact: the *stationary phase*, in the column, and the *mobile phase*, which moves across it. Separation relies on the differential displacement of constituents inside the column, passing through in times that are proportional to their size, or depending on their structure, or affinity for the stationary phase (polarity...). As they reach the far end of the column, a *detector* measures, on a continuous basis, the quantities of each constituent.

The most common form of chromatography is **gas chromatography**, carried out on gaseous samples, or samples that may be vaporized without incurring breakdown. The mobile phase is a gas (helium, nitrogen, argon, or hydrogen), constantly sweeping through the column, which is placed in a thermostat oven. Detectors allow the selective analysis and identification of highly complex mixtures.

If the stationary phase is a nonvolatile, or not highly volatile liquid, exhibiting solvent properties for the compounds to be separated, the process is termed **gas–liquid chromatography**, or *partition chromatography*. If the stationary phase is an **adsorbent** solid (silica, alumina, zeolites, or **polymers**), this is **gas–solid chromatography**. Within this same family, of **adsorption** chromatography processes, **liquid–solid chromatography** is characterized by its stationary phase, this being a polar solid.

In **high-performance liquid chromatography** (**HPLC**), the sample must be wholly soluble in the mobile phase (elution solvent). The latter must be kept at high pressure (hence the alternative name of *high-pressure* liquid chromatography), to ensure a constant flow rate inside the column, and preclude any loss of head. HPLC involves solute–mobile phase–stationary phase exchange mechanisms, based on partition or adsorption coefficients, depending on the nature of the phases in contact.(1)

A chromatographic analysis yields a **chromatogram**, this being a graphical representation of the evolution of a parameter (intensity of the detector signal), related to instantaneous solute concentration, as function of time. This exhibits *peaks*, rising above the *baseline*, which obtains in the absence of any compounds (see Figure).

(1) There are two further types of liquid chromatography, *ion chromatography*, and *exclusion chromatography*.

N.B: This Box reproduces a number of excerpts from a presentation by Pascale Richardin, head of the Datation Group at the Research and Restoration Center of the French National Museums Administration (Musées de France), taken from the pages dealing with analytical methods, as posted on the site : ttp://www.culture.gouv.fr/culture/conservation/fr/biblioth/biblioth.htm

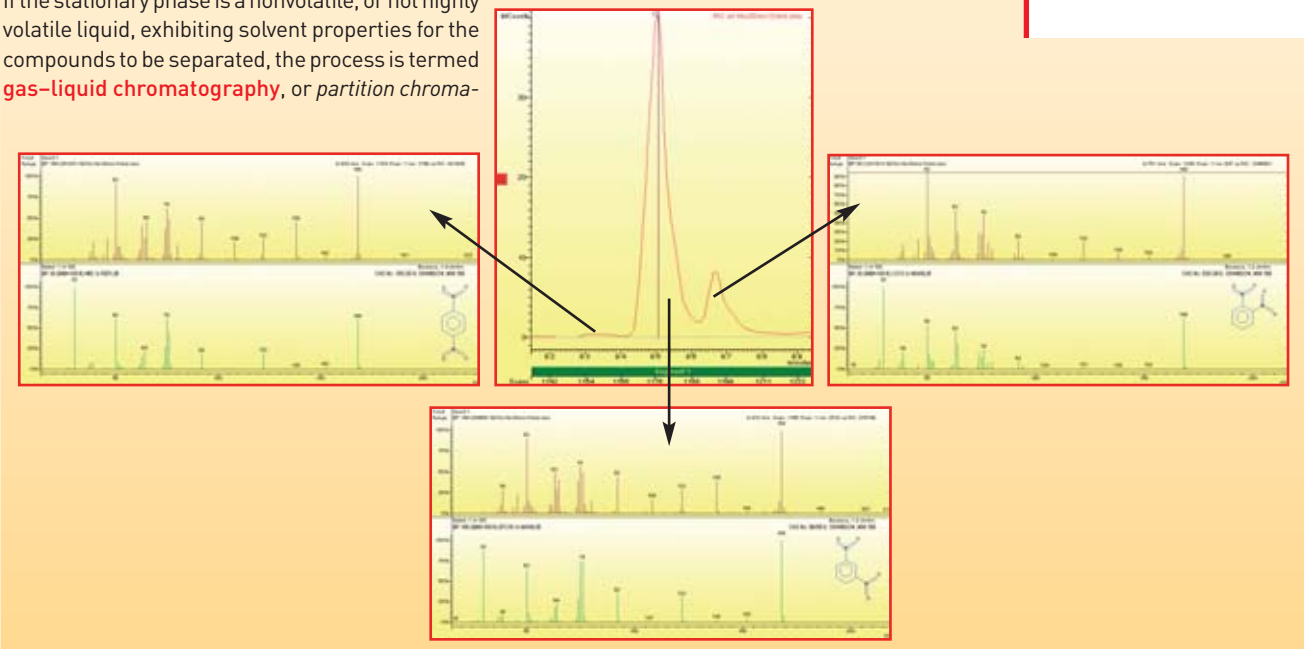


Figure.
An example of the combined use of mass spectrometry and chromatography: the separation of isomers ("sister molecules") of an explosive molecule (dinitrobenzene [DNB]), after solid-phase microextraction sampling, by gas chromatography, and their detection by mass spectrometry (SPME–GC–MS).