Ultrafast Experiment

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Abstract

The measurement of short time intervals during the first half of 20 century was limited to intervals longer than one nanosecond. The advent of lasers in 1960 revolutionized the field of time-resolved spectroscopy. The time interval is shortened from 10^{-9} to 10^{-15} seconds, which made ultrafast experiment grows rapidly. New subfields of science, including femtochemistry and femtobiology, have been created. In this paper, a brief review of ultrafast electron microscopy (UEM), ultrafast laser and ultrafast x-ray imaging will be present.

Introduction

Research into ultrafast phenomena, including dynamic processes in material systems and chemical and biochemical reactions, has been largely based on laser pump-probe or laser pump and synchrotron/x-ray radiation probe techniques. Because of the short time scale involved, these are known as ultrafast techniques. The use of electrons as probes has demonstrated effectiveness and great potential to study complex transient

events. The development of lasers capable of producing femtosecond pulses in the 1980s opened the door for a class of spectroscopic experiments observing the dynamics of these processes. Nonimaging optical spectroscopy exclusively measures frequency or time-domain quantities. These data are only indirectly connected to atomic positions (i.e., structure) in condensed matter through complicated and ultimately model-dependent response functions. Significant benefits accrue when structural dynamics can be observed directly, either by x-ray or electron diffraction or by real-space imaging. Figure 1 classifies some phenomena in materials science, biology and chemistry in terms of relevant spatial and temporal resolutions [1].

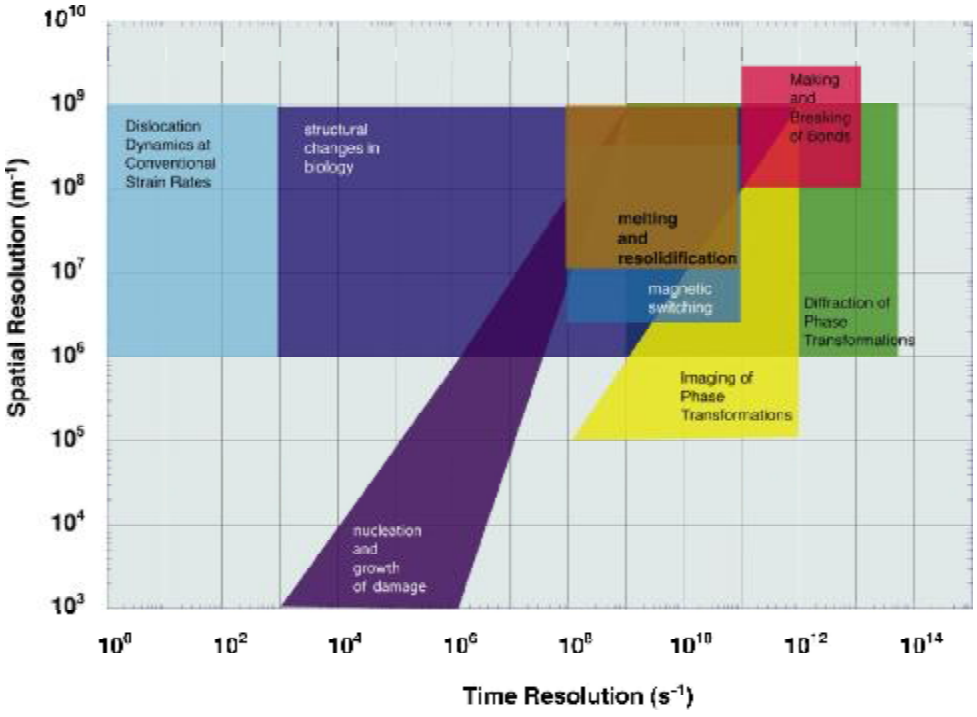

FIG.1. Phenomena classified by spatial and temporal resolutions. Spatial resolution is defined as either (1) if the technique is an imaging method, the low end of the bounding box would be defined by the smallest resolvable feature and the high end by the typical field of view or (2) for a nonimaging technique, the bounding box would be defined by the range of probe or spot sizes. Time resolution is defined as that for a single-shot investigation of irreversible processes. So time resolution is defined as the single-shot exposure time to get data that demonstrate a particular spatial resolution.

Time resolution is perhaps the most important performance criterion for these instruments. Ultrafast x-ray studies have demonstrated time resolution down to 100fs using X-ray free-electron laser [2].

The first pulses shorter than a nanosecond were obtained by DeMaria *et al.* (1966) by passive mode locking of a Nd-glass laser, which has a broader gain profile. The first pulses shorter than 1 ps(10^{-12}s) were obtained by Shank and Ippen (1974) with tunable broad-gain dye laser media in combination with a saturable dye absorber. With compensation of group velocity dispersion by a configuration of glass prisms in the laser cavity, a repetitive train of six femtosecond pulses was reported by Fork ***et al*** (1987). Zhou *et al.* (1994) obtained a pulse of 8-fs duration from a Ti-sapphire laser. An all-solid-state system based on pumping the Ti-sapphire with a semiconductor array to emit a continuous train of short pulses is feasible. Thus femtosecond pulse generators are rapidly becoming a standard laboratory tool [3].

Ultrafast Laser

Laser is made of a gain medium enclosed in an optical resonator. Optical resonator is a face-to-face configuration of two mirrors, one with reflectivity less than unity in order to pass some output light. As shown in Fig 2. If only three energy levels: the ground state (having energy of E1) and two excited states (of energies E2 < E3). Energy is supplied in order to "pump" the E1 → E3 transition. E3 level is then subject to fast non-radiation transition to E2, providing non-equilibrium state known as inverted population (meaning there are more atoms in E2 than E1). The E2 → E1 transition is the "laser transition", providing laser photon of the same frequency.

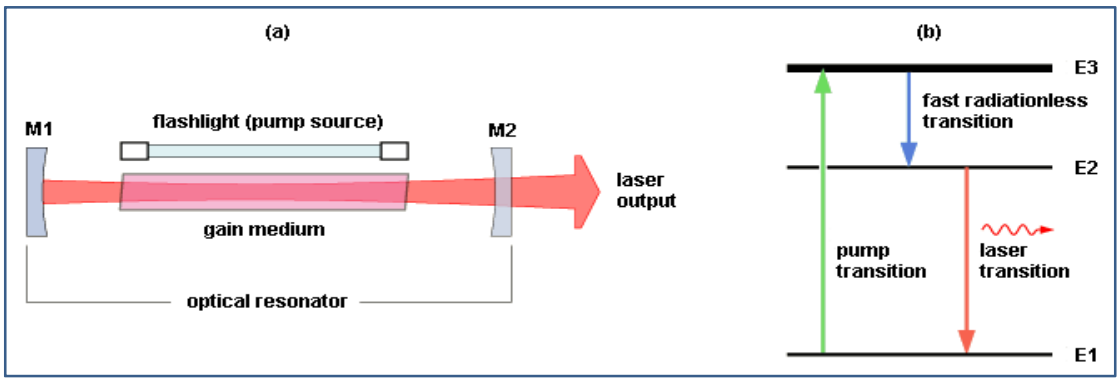

Fig.2. (a) Laser cavity and (b) energy levels of a 3-level laser.

To generate ultrashort pulses from lasers, we need to use modelocking technique. A schematic set-up with a gain and a loss element inside a laser resonator is shown below. An output coupler partially transmits a small fraction of the laser pulse out of the laser resonator equally spaced by the resonator round-trip time. Typically an intracavity loss modulator is used to collect the laser light in short pulses around the minimum of the loss modulation with a period given by the cavity round-trip time $TR = 2L/vg$, where L is the laser cavity length and vg the group velocity (that is, the propagation velocity of the peak of the pulse intensity). There are two type of modelocking: passive and active [6].

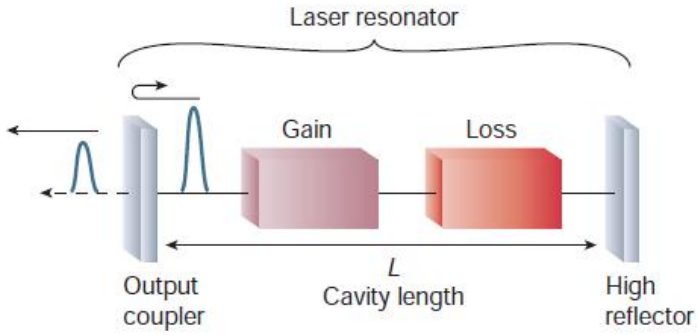

Fig3. Gain and a loss element inside a laser resonator.

For active modelocking, an external signal is applied to an optical loss modulator typically using the acousto-optic or electro-optic effect. Such an electronically driven loss modulation produces a sinusoidal loss modulation with a period given by the

cavity round-trip time *TR*. The saturated gain at steady state then only supports net gain around the minimum of the loss modulation and therefore only supports pulses that are significantly shorter than the cavity round trip time. For passive modelocking, a saturable absorber is used to obtain a self-amplitude modulation of the light inside the laser cavity. Such an absorber introduces some loss to the intracavity laser radiation, which is relatively large for low intensities but significantly smaller for a short pulse with high intensity. Thus, a short pulse then produces a loss modulation because the high intensity at the peak of the pulse saturates the absorber more strongly than its low intensity wings. This results in a loss modulation with a fast initial loss saturation (that is, reduction of the loss) determined by the pulse duration and typically a somewhat slower recovery that depends on the detailed mechanism of the absorption process in the saturable absorber. A saturable absorber behaves differently depending on the intensity of the light passing through it. Ideally a saturable absorber will selectively absorb low-intensity light, and transmit light which is of sufficiently high intensity. Generally, we can obtain much shorter pulses with passive modelocking using a saturable absorber, because the recovery time of the saturable absorber can be very fast, resulting in a fast loss modulation. Modelocked pulses are much shorter than the cavity round-trip time and therefore can produce an ideal fast loss modulation that is inversely proportional to the pulse envelope.

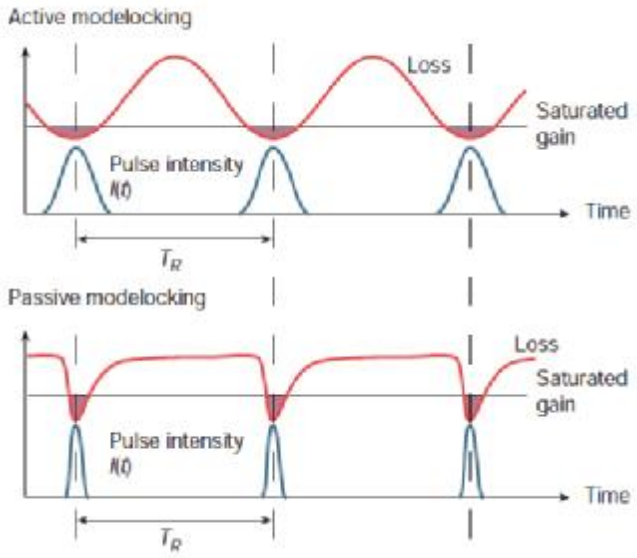

Fig.4. Active modelocking and passive modelocking.

Ultrafast Electron Diffraction

Electrons are found to be complementary (and in some cases advantageous) to x-rays in a number of comparisons [1]:

(1) Electrons are less damaging to specimens. In biological specimens, the ratio of inelastic-/elastic-scattering events is 3 for 80-500-keV electrons and 10 for 0.15nm x rays. The energy deposited per inelastic-scattering event for 0.15nm x-rays is 8 keV compared with 20 eV for electrons. This means the energy deposited per useful elastic scattering event is 1000 times higher for 0.15nm x-rays.

(2) The scattering length of electrons better matches the optical penetration depths "pumped" volume of most samples. Consequently, for thin samples there is less contribution to the diffraction pattern from unpumped materials.

(3) Electrons can be easily focused and the technology for electron-optical imaging at high spatial resolution is well developed.

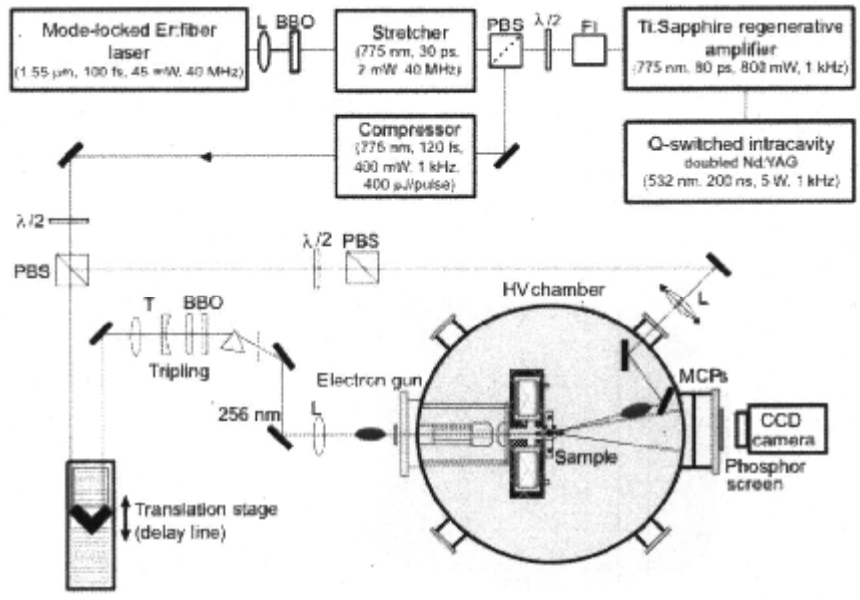

FIG. 5. Femtosecond electron-diffraction apparatus. This system is configured for transmission electron diffraction.

Figure 5 shows a schematic of a typical ultrafast electron diffractometer. The system includes a laser-driven electron source, sample-positioning system, and electron detector. All are contained in one or more vacuum chambers. The light source is normally a conventional Ti:Sapphire chirped pulse amplification laser system. Each laser pulse is divided into two parts: one part drives the photo activated electron gun, the other photo excites the sample. This ensures that the pump and probe pulses can be synchronized at the sample with timing jitter,100 fs. A Faraday cup inserted into the electron beam path can be used to measure the average beam current. This is important because the electron pulse fluence (number of electrons per beam area) is, along with the photocathode-to-sample distance and the acceleration voltage, a primary determinant of the temporal resolution. The key elements of this apparatus are the electron source and detection systems. These elements—along with the particular geometry of the experiment and sample being studied— determine the type and level of structural detail that can be extracted from UED experiments.

X-ray imaging

Measuring atomic-resolution images of materials with x-ray photons during chemical reactions or physical transformations resides at the technological forefront of x-ray science. In Keith A. Nelson's paper, it is reported that their experiment allows the direct measurement of the changes in crystal lattice structure and loss of crystalline order associated with melting. The results represent the early fruits of a worldwide effort to generate ultrashort hard x-ray pulses and use them for the characterization of ultrafast events through time-resolved x-ray diffraction.

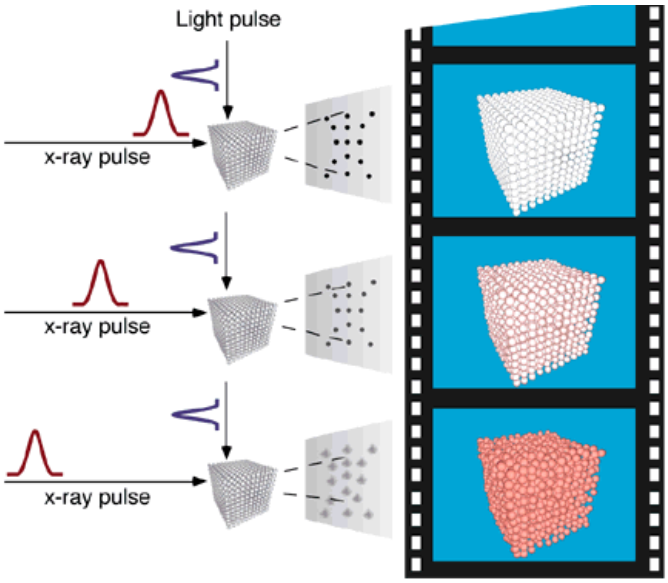

Fig. 6. Molecular movies. Time-resolved x-ray diffraction can record transient structures of materials as they undergo rapid change initiated by an ultrashort optical pulse.

New x-ray–based experimental capabilities have been closely linked with advances in x-ray sources, a trend that will continue with the impending arrival of x-ray–free electron lasers driven by electron accelerators. In K. J. Gaffney's paper, he showed the advantage of the new X-ray free electron laser in the field of coherent x-ray diffractive imaging.

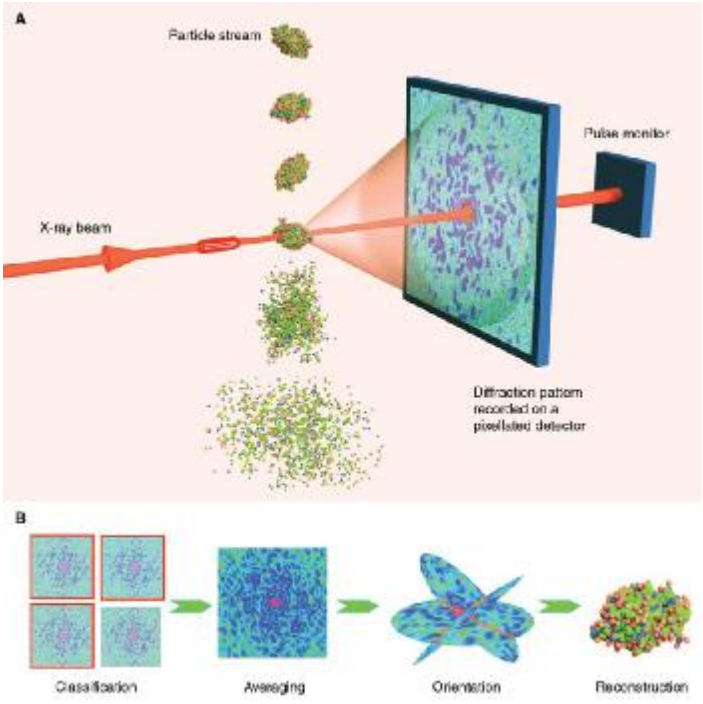

Fig.7. Schematic depiction of single-particle coherent diffractive imaging with an XFEL pulse. (A) The intensity pattern formed from the intense x-ray pulse (incident from left) scattering off the object is recorded on a pixellated detector. The pulse also photo-ionizes the sample. This leads to plasma formation and Coulomb explosion of the highly ionized particle, so only one diffraction pattern [a single two-dimensional (2D) view] can be recorded from the particle. Many individual diffraction patterns are recorded from single particles in a jet (traveling from top to bottom). The particles travel fast enough to clear the beamby the time the next pulse (and particle) arrives. The datamust be read out fromthe detector just as quickly. (B) The full 3D diffraction data set is assembled from noisy diffraction patterns of identical particles in random and unknown orientations. Patterns are classified to group patterns of like orientation, averaged within the groups to increase signal to noise, oriented with respect to one another, and combined into a 3D reciprocal space. The image is then obtained by phase retrieval.[5]

It is reported that in September 2009, the Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC) in California will begin operations [2]. This X-ray free-electron laser (XFEL) will then generate the brightest pulses of X-rays on the planet, leaping almost nine orders of magnitude beyond current synchrotron-based X-ray facilities. The LCLS, and other XFELs of its kind, will produce laser-like pulses of X-rays of about 100 fs duration and 10^{12} photons per pulse, ushering in a new chapter in the field of ultrafast X-ray science. Ultrafast X-ray imaging will enable us to connect these studies with real space motions. Time resolved X-ray crystallography, which is currently bringing new insights into the function of proteins, will be hugely extended with FEL pulses.

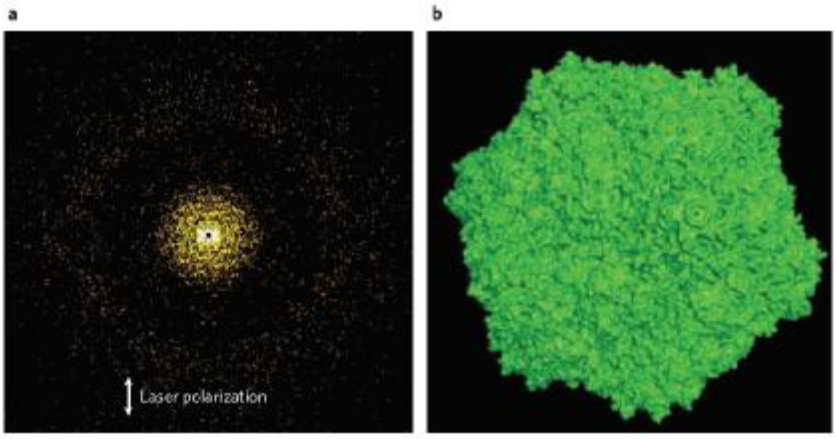

Figure 5 | Diffraction patterns from single objects differ from patterns of crystals consisting of repeats of those objects. The single-object pattern directly accesses the molecular transform without restriction to the crystal's Bragg peaks. a,b, The calculated pattern of the cowpea mosaic virus illuminated by a single LCLS pulse (a), consisting of 10^{12} photons focused to 0.2 μm diameter. In this case the spotty nature of the pattern is due to the very low signal: most of the detector pixels would receive no counts (black) or a single X-ray photon (orange). Even so, new averaging algorithms should allow data to be combined from many patterns of different cowpea mosaic virus particles, recorded at random orientations, to reconstruct the three-dimensional image shown in b.

Summary and Conclusion

The progress in ultrafast lasers and X-rays imaging during the past decade has been simply amazing. The continued development of these ultrafast techniques, and their impact on our understanding of the basic interactions that determine functions in many areas from biology to superconductivity, will be driven by those developments.

Reference

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