Ultrafast Experiments

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Spring 2008
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Abstract: Lasers have caused revolutionary changes in many fields of science and technology. Since 1960, six orders of magnitude, from 10^{-9} to 10^{-15} seconds, have been added to time-resolved observation of fast phenomena, which makes ultrafast research grows rapidly among materials science, biology, and chemistry. The use of these ultra-short high-power pulses went hand-in-hand with their development and already brought some fascinating discoveries. This article provides a brief review of the historical development of short laser pulse along with some important improvements, then gives description of the development of pump-probe technique, and finally presents an application in material science.

1 Introduction

Soon after first lasers were developed in 1960, they revolutionized many fields of science and technology. The inherent features of laser light: long coherence length, very narrow spectral bandwidth, intrinsically well defined polarization and very high irradiance are what makes laser superior to other light sources. [1] There is yet another feature of the laser source - it can be made to produce extremely short pulses. The measurement of short time intervals during the first half of last century was limited to intervals longer than one nanosecond. With the invention of Q-switching, nanosecond pulses became available [2]. Mode-locking techniques in mid 1960s brought pulses to pico-second (pico = 10^{-12}) region [2]. With the application of passive mode-locking and saturable dye absorbers in 1970s, the first femtosecond (femto = 10^{-15}) pulses were made. In early 1990s, Sibbett et al. discovered spontaneous mode-locking without the use of saturable absorber in Ti-sapphire laser [3], which later enabled generation of pulses of only a few femtoseconds in length. The latter made tabletop femtosecond pulse sources rapidly become a (nearly) standard optical laboratory tool [2].

The reason for such fundamental importance of femtosecond time scale is that atomic and molecular dynamics happen on that scale. (Fig. 1) For example, the

frequency of half-oscillation of H_2 atoms is about 4155 cm^{-1}, which has the time scale of 7.6 fs. Therefore, the development of equipments with femtosecond resolution becomes extremely important to the research of ultrafast phenomena. The ultrafast research today is overwhelmingly broad, but it can be divided in two general parts. The first would be the technology of femtosecond pulse generation and manipulation, the second is a common approach to measurement with such pulses, named as “pump-probe”.

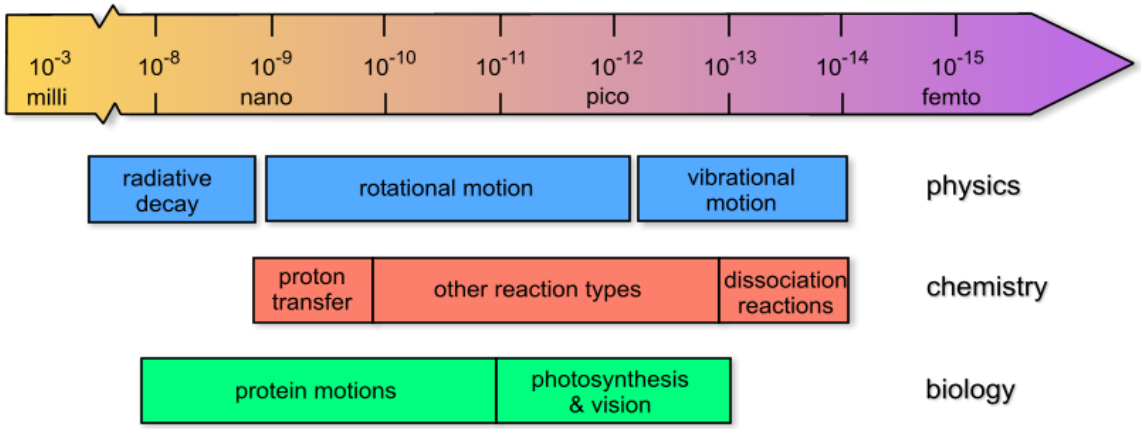

Figure 1: Timescales of some physical, chemical and biological processes.

In this article, fundamental techniques for producing ultra-short pulses are first introduced, followed by the explanation of the pump-probe technique. Finally, one experiment focusing on ultrafast melting is presented.

2 Femtosecond pulse generation

Laser is made of a gain medium enclosed in an optical resonator. Optical resonator is a face-to-face configuration of two mirrors (Fig. 2a), one with reflectivity less than unity in order to pass some output light. Eigen frequencies in 1D resonator of length L are given by the condition

$$\upsilon_n = c\frac{n}{2L} \tag{1}$$

n being a number of modes. Because resonator length L is much larger than the wavelength of light, n is very large. Difference of neighboring resonator frequencies (for n and n + 1) is obtained from (1)

$$\Delta\upsilon_n = \frac{C}{2L} \tag{2}$$

For L = 30 cm resonator length we get 30 eigen-modes in frequency interval of 300 MHz. The simplest has three energy levels: the ground state (having energy of E1) and two excited states (of energies E2 < E3, see Fig. 2b). 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:

$$\upsilon_{12} = \frac{E_2 - E_1}{h} \tag{3}$$

The laser transition is not of a single energy, but rather a band of energies (and corresponding photon frequencies). Depending on the gain spectral, more than one mode can be amplified. Hence, many laser frequencies can be presented on the output - a multimode laser.

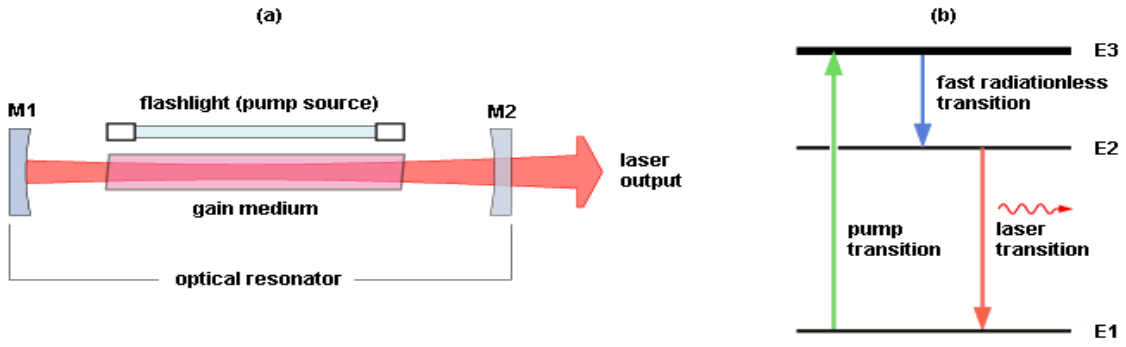

Figure 2: Laser cavity (a) and energy levels (b) of a 3-level laser. (Adopted from [4, 5].)

Ultra-short pulses are generated by mode-locked lasers. To explain "mode-lock", let's consider that the total electric field in laser cavity is the sum of contributions of all amplified modes:

$$E(t) = \sum_{n=1}^{N} E_n e^{i[(2\pi\nu_0 + 2\pi\Delta\nu)t + \Phi_n]} \tag{4}$$

Where υ_0 is some reference frequency and Φn is phase of n-th mode, values which are random. Output power of such a multimode laser is proportional to square of electric field and it's time-dependence looks like noise (Fig. 3a). In a mode-locked laser, phases of all modes are locked to the same value instead. Such laser operates in pulsed mode (Fig. 3b).

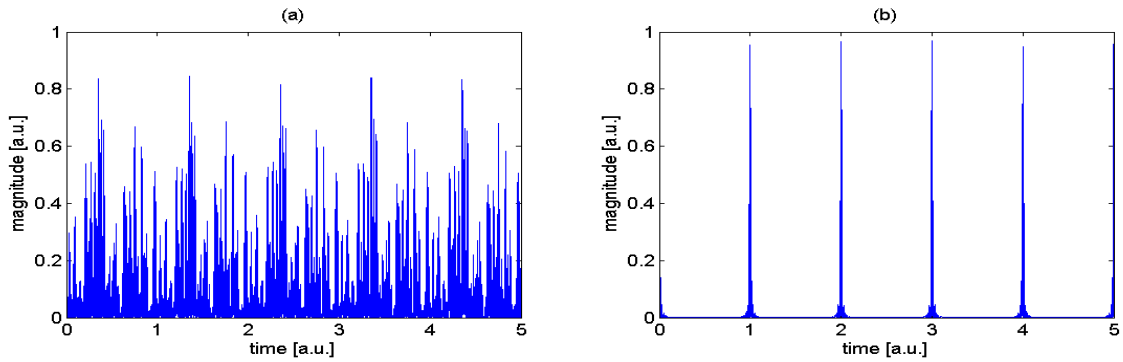

Figure 3: Normalized multimode (a) versus mode-locked (b) laser output power.

Through decades of efforts, nowadays the most commonly used femtosecond laser uses a sapphire (mono-crystalline of Al2O3) crystal doped with Ti3+ ions. Energy required for pumping is that of a green light (around 500 nm) and Ar+-ion or frequency doubled neodymium laser is often used [6]. The gain profile of Ti-sapphire is the broadest yet discovered [6], ranging from 700 nm to 1000 nm and most efficient around 800 nm. Such broadband profile can support approx. 250000 modes in a 30 cm resonator, making it superior material for ultra-short pulse mode-locked laser. Shortest theoretically achievable pulse in Ti-sapphire is of only 3.4 fs. However for pulses below ps, dispersion effects in laser medium itself become significant enough to limit the pulse duration to some tens of fs. [6] Using more than one approach of mode-locking simultaneously, the shortest pulses actually produced in Ti-sapphire lasers are around 5 fs.

However, the problem arises that the pulse energy of femtosecond mode-locked lasers is typically some nJ, which can be too low for some experiments. The problem is solved by the method named “Chirped-pulse amplification” which stretches the pulse in time to hundreds of picoseconds then make the amplification. Using this technique, the intensity of each pulse is remarkably amplified and meets the experimental needs.

3 The Pump-Probe Techniques

Characteristic timescales of electronic and lattice dynamics are in the fs to ps region (see Fig. 1), thus a way beyond any electronic component response time (that being in nanosecond region), thus an all-optical technique is needed for measurement. General principle is as follows (Fig. 4). Sample is first hit by a “pump” pulse, which generates some excitation or modification in the sample (generally photo-excitation changes complex dielectric constant). After an adjustable time delay Δt, usually weaker

“probe” pulse hits the excited spot. Time delay is imposed by optical path difference and controlled mechanically by translational delay stage. Because the speed of light is so high, 1 fs time delay corresponds to only 0.3 µm path difference and stepper-motor must be used to drive the stage. Small relative changes in sample’s transmission or reflection of the probe pulse are then measured (ΔR). By varying time delay, a time-resolved measurement of the pump-induced effect can be recorded. Pump and probe pulse need not be of the same frequency, it is however critical they are synchronized in time.

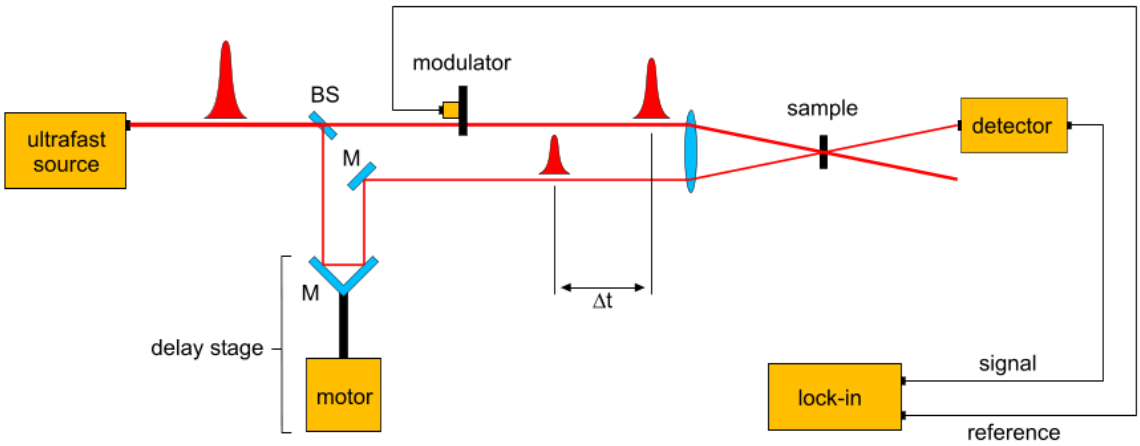

Figure 4: The pump-probe principle. Pulses from ultrafast source are split in two by the Beam splitter (BS). Pump pulse first hits the sample followed by a Δt delayed probe pulse. Delay is produced by varying optical path using mirrors (M) and motor-driven µm delay stage.

Since photo-induced changes are relatively small (10^{-6} to 10^{-3}), therefore conventional lock-in detection techniques are often used to extract the small relative change of ΔR/R. Pump beam is modulated at some kHz frequency. The detector output, being some time-average of the probe pulses, is sent to a lock-in amplifier, which only collects signal at exact frequency and phase of the pump beam modulator. Only changes induced by pump are thus recorded (see Fig. 5). Because of such detection scheme, detector response time only needs to be faster than the pulse period, that being nanoseconds or more. Relative sensitivity up to 10^{-7} can thus be reached.

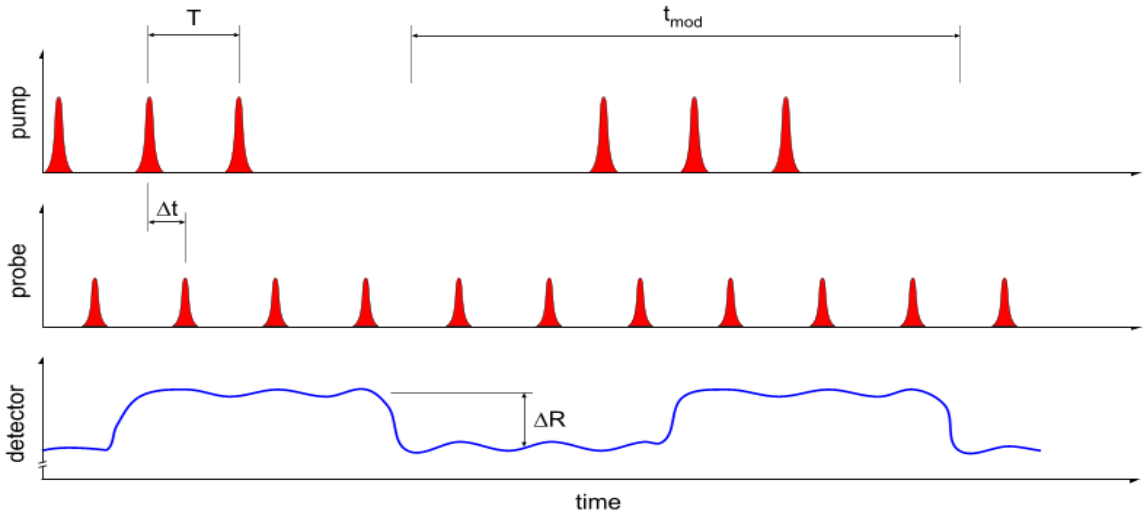

Figure 5: Pump-probe signals (not in scale). Pump beam is modulated (t_{mod}) and only changes corresponding to that frequency (ΔR) are measured as a function of time delay (Δt).

Except the wide use of laser pump with laser probe techniques, the technique named "time-resolved x-ray diffraction" is also well developed in solid state physics. The dynamic properties of solids couple to atomic motion and the relevant time-scale is that of a vibrational period. In contrast with existing, fs laser probe, X-rays have a wavelength approximately equal to the distances between atoms, and hence enable atomic movements to be visualized directly. Thus, the natural technique for studying evolving atomic structures is through X-ray diffraction. In concept, this technique differs from laser pump and laser probe in that it uses x-ray as probe. Previously, the lack of both ultrashort-pulse X-ray sources and ultrafast x-ray detectors has prevented such studies. During recent years a rapid development of pulsed X-ray sources has been achieved, falling into two main categories: Laser-based table top sources; and sources based on partice accelerations (e.g. synchtrotrons). Many excellent papers have been published based on this technique.

In most recent years, instead of using x-ray as probe, people are more interested in using electrons as an ultrafast probe in ultrafast experiments. 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.

Thus, a new technique named “Ultrafast electron Diffraction (UED)” is presented. Fig. 6 shows the femtosecond electron-diffraction apparatus. Conceptually, UED is also similar to an ultrafast optical pump-probe experiment; the only different ingredient is that the probing is done by electron diffraction. Typically, dynamics are initiated with an ultra-short light pulse. Then, at a sequence of delay times, the sample is probed in transmission or reflection with an electron pulse from a photoactivated electron gun. By recording diffraction patterns as a function of delay time, it is possible to follow the various aspects of the real-space atomic configuration of the sample as it evolves. 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 photoactivated electron gun, the other photoexcites the sample. This ensures that the pump and probe pulses can be synchronized at the sample with timing jitter less than 100 fs.

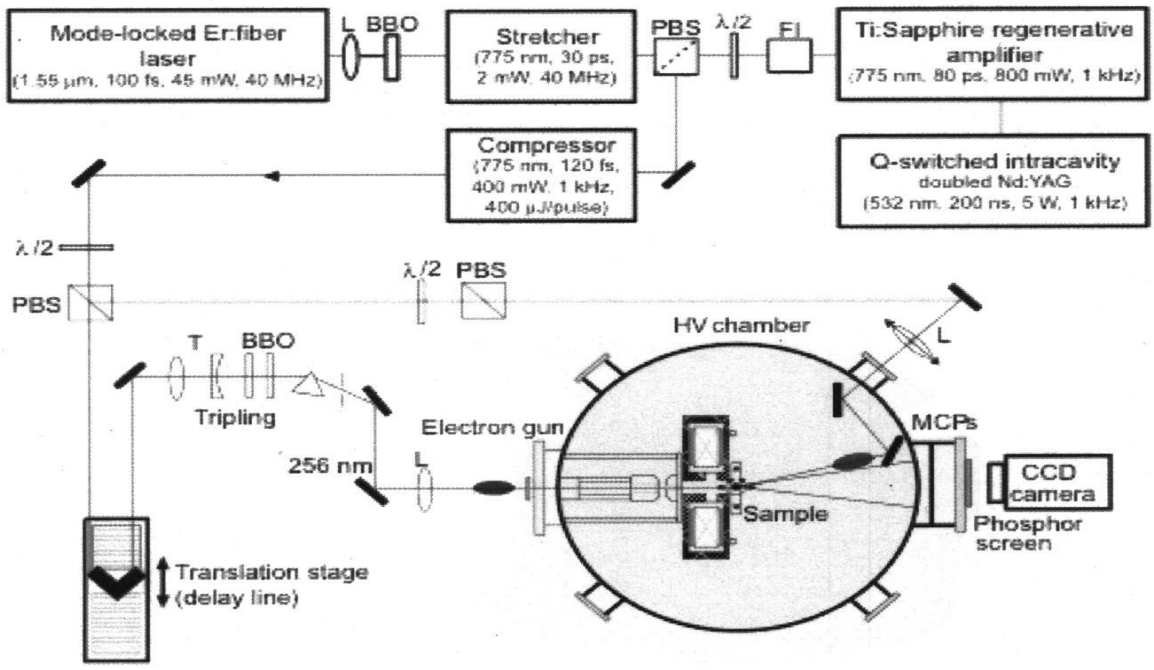

Fig. 6: Femtosecond electron-diffraction apparatus. This system is configured for transmission electron diffraction.

4 Application

The application of ultrafast techniques is remarkably broad. It mainly includes ultrafast phase transitions, above-threshold ionization of atoms, and high-order harmonic generation and acceleration of relativistic electrons by light pulses, and so on. [2] In particular, its application in materials science is most concerned by condensed matter physicists. Here the paradigm of "Ultrafast melting probed by electron diffraction" is demonstrated. The reason of choosing this topic is mainly because: First, the working principle of pump-probe experiment can be demonstrated. Second, the use of UED is highlighted in this experiment so that the continuous development of pump-probe techniques appears obvious.

Melting was one of the primary phenomena studied by developers of the dynamic TEM technique. Melting was induced by irradiation of the specimen with an intense laser. Fundamental aspects of the physical processes involved in the melting of strongly laser-driven materials have been a subject of intense investigation since the development of ultra-short laser systems nearly 30 years ago. The laser-induced melting of polycrystalline Al is first performed in 1984 by Williamson et al. It used the picoseconds electron diffraction to resolve in time and observed that under rapid heating conditions the long-range order of the lattice subsists for lattice temperatures well above the equilibrium point. [7] The advances in ultrafast electron sources and recent spectroscopic investigations on the nature of the ultrafast phase transition prompted a revisiting of this phenomenon with a dramatically improved temporal resolution (600 fs) and structural sensitivity. [1]

The pump-probe experiment works as follows: Referring Fig.6, Ultra-short pulses from Ti-sapphire mode-locked laser were first amplified, the beam was then split in two. Pump beam was delivered straight to a 500 μm spot on a 20 nm thick Al sample. At optical fluence of 70 mJ/cm^2, heat was deposited at a rate faster than the thermal expansion rate, pushing temperature above normal melting point and also causing significant sample vaporization [1]. The second part of laser beam was frequency tripped to UV region and hit a metal plate at the base of the femtosecond electron gun installed in a high-vacuum chamber. Some 6000 photoelectrons produced were accelerated to 30 keV and directed to the target area of 200 μm on the sample.

In the single scattering limit, an atomic pair-correlation function or reduce-density function, G(r), can be computed from a sine transform of the reduced diffracted intensity, $\psi(s)$:

$$G(r) = 4\pi r[\rho(r) - \rho(0)] = 8\pi \int_0^\infty \varphi(s) \sin(2\pi r s)\, ds \quad (5)$$

Where $\varphi(s) = s\left[\frac{I(s)}{f(s)^2} - 1\right]$, $I(s)$ is the coherently diffracted intensity in electron units, s is the electron scattering vector, $f(s)$ is the atomic scattering factor, $\rho(r)$ is the atomic density, and $\rho(0)$ is the average atomic density. The function G(r) describes the deviation of $\rho(r)$ from the average atomic density, $\rho(0)$, as a function of the radial distance from an average atomic origin. Thus, maxima in G(r) give the most probable inter-atomic distances present in the sample. [1]

It is now possible to obtain almost a complete structural record of a material, through snapshots of the time-dependent, reduced density function G(r,t) during laser-induced structural transformations on the sub-picosecond time scale. This is shown in Fig. 7 for the femtosecond laser-induced s70 mJ/cm^{2}d polycrystalline-to-liquid phase transition of a 20-nm-thick Al film. These results indicate that, in this intensity range, the melting of Al by ultrafast laser excitation proceeds through a thermal process with little or no lattice expansion. The information gained in these experiments is extremely useful in developing a more fundamental understanding of melt-zone nucleation and electron–phonon coupling under both highly excited thermal and athermal distributions of electrons.

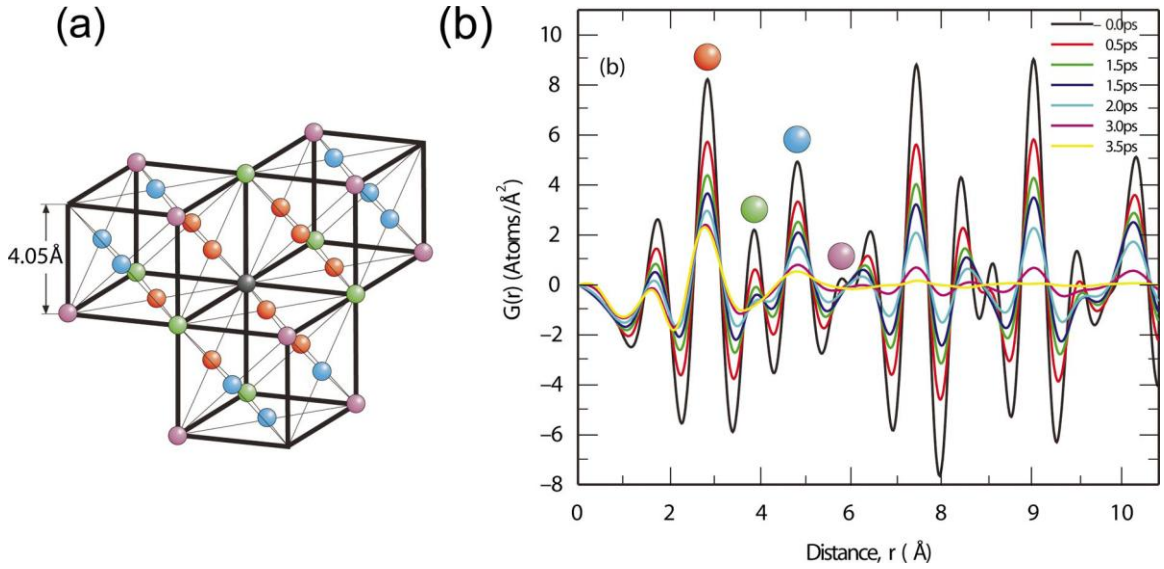

FIG. 7: (Color) An atomic-level view of melting. The correspondence between the peaks in G(r) and the interatomic spacing present in the crystalline FCC Al lattice are shown for the first four peaks by labeling with the same color as in (b). Long-range correlations in atomic position are almost entirely preserved for the first 1.5 ps after laser excitation, but decay after this time such that the material has a disordered liquid-like structure by 3.5 ps.

5 Summary

Ultrafast laser systems have already reached the performance, stability and ease of use to make them part of many optical laboratories around the globe. Many

problems in physics, chemistry, biology and also information technology can greatly benefit from femtosecond time resolution being the “ultimate” time-scale of the dynamics observed. Many new and surprising phenomena are also being discovered along the way. On the other hand, development has not lost its pace as new ways of use of light pulses in the production of hard X-rays and electron pulses are being tested.

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