Ultrafast Lasers in Materials Research

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Guest Editors

Abstract

With the availability of off-the-shelf commercial ultrafast lasers, a small revolution in materials research is underway, as it is now possible to use these tools without being an expert in the development of the tools themselves. Lasers with short-duration optical pulses—in the sub-picosecond (less than one-trillionth of a second) range—are finding a variety of applications, from basic research on fast processes in materials to new methods for microfabrication by direct writing. A huge range of pulse energies are being used in these applications, from less than 1 nJ (a billionth of a joule) to many joules.

Keywords: *laser, ablation.*

Introduction

Ultrafast lasers—that is, lasers that produce optical pulses with a duration of less than a picosecond—are playing an increasingly important role in many science and technology disciplines. Ultrafast, time-resolved measurements are well established in physical chemistry, where fundamental time scales of chemical reactions become accessible,[1] and in solid-state physics and electrical engineering, where carrier dynamics and transport are probed on picosecond time scales directly relevant to the operation of modern high-speed devices.

Applications in materials research have been slower to emerge, because only within the past decade have commercial instruments reached a level of reliability and sophistication that make them practical tools for scientists and engineers who may not be experts in laser technology and the manipulation of short-duration, high-intensity optical pulses. The rapid development of ultrafast laser technology is a principal motivation for this issue of *MRS Bulletin*: the time is ripe for ultrafast lasers to take on a rapidly expanding role in studies of the science of materials and in materials characterization, materials modification, and microfabrication. Ten years ago, very little materials research was conducted using high-intensity, ultrafast lasers. With the commercial availability of off-the-shelf ultrafast tools, a small revolution in materials research is underway.

Ultrafast optical pulses of <1 ps duration are generated by mode-locked laser oscillators. The phases of the longitudinal optical modes of the laser cavity are locked together by either an active element (e.g., an acousto-optic modulator) or by passive effects such as Kerr-lensing in the gain medium or the use of a saturable absorber. Mode-locking produces short-duration optical pulses with a high repetition rate determined by the length of the optical cavity. The wide bandwidth of optical gain in sapphire doped by Ti enables extremely short-duration pulses.

Ti:sapphire lasers dominate the market, but Ti:sapphire laser oscillators are, of course, actually a set of three lasers. Continuous-wave (cw) diode lasers pump a cw solid-state laser, which is frequency-doubled and used to pump the Ti:sapphire oscillator. Ultrafast laser oscillators and amplifiers that can be directly pumped by diode lasers, such as Er:glass-fiber lasers and Yb:tungstate lasers, are becoming more common and may lead to more compact and less expensive instruments, but they have a more limited range of output wavelengths and a longer pulse duration than Ti:sapphire.

The typical repetition rate of a laser oscillator is 80 MHz. Therefore, a laser oscillator with an average power of 1 W produces optical pulses with an energy of approximately 10 nJ. This pulse energy is sufficient for metrology using picosecond acoustics, for experiments that probe heat transfer or carrier dynamics, and for many forms of optical spectroscopy; it is not generally sufficient for materials modification, except with pulses that are tightly focused by high-numerical-aperture microscope objectives. Higher-energy pulses are available from so-called "extended-cavity oscillators" that operate with a lower repetition rate, on the order of 10 MHz, but this technology is currently limited to <100 nJ in commercial Ti:sapphire lasers and <1 μJ in Yb:tungstate lasers.

Optical pulses from laser oscillators must therefore be amplified to reach energies of >1 μJ. The ability to amplify ultrafast lasers was an elusive goal for 20 years following the development of the ultrafast oscillator in the mid-1960s. In 1985, high-intensity ultrafast lasers emerged with the development of the chirped-pulse amplifier.[2] In a chirped-pulse amplifier, pairs of diffraction gratings are used to temporally stretch the optical pulse prior to amplification and then temporally compress the pulse after it leaves the amplifier. Twenty years later, Ti:sapphire chirped-pulse amplifiers that produce 1–2 mJ optical pulses at 1 kHz repetition rates are available from a number of commercial suppliers. Higher-repetition-rate (>100 kHz) lasers with microjoule energy pulses are desirable for many applications in materials removal and modification. These types of lasers are becoming more common. The relative simplicity of amplifiers that are directly pumped by diode lasers is driving the development of systems based on Er:glass and Yb:tungstate. A remarkable feature of the articles in this issue of *MRS Bulletin* is the huge range of pulse energies that are used in the research: from <1 nJ optical pulses applied in metrology to the 1 J energies used for a relativistic optics phenomenon called *plasma wakefield acceleration of electrons* that may one day replace synchrotrons as bright sources of x-rays[5,6] and γ-rays.[7]

How Fast Is Ultrafast?

A simple illustration of the time scale represented by a femtosecond (one-quadrillionth of a second) is the fact that light travels around the Earth about seven times in a second, but only about 300 nm in a femtosecond (see Figure 1).

The answer to the question "How fast is ultrafast?" in materials research more accurately depends on the characteristic time scale of the application or the science being studied. For example, for the generation of high-frequency longitudinal acoustic waves in metal films, an important time scale is the optical absorption

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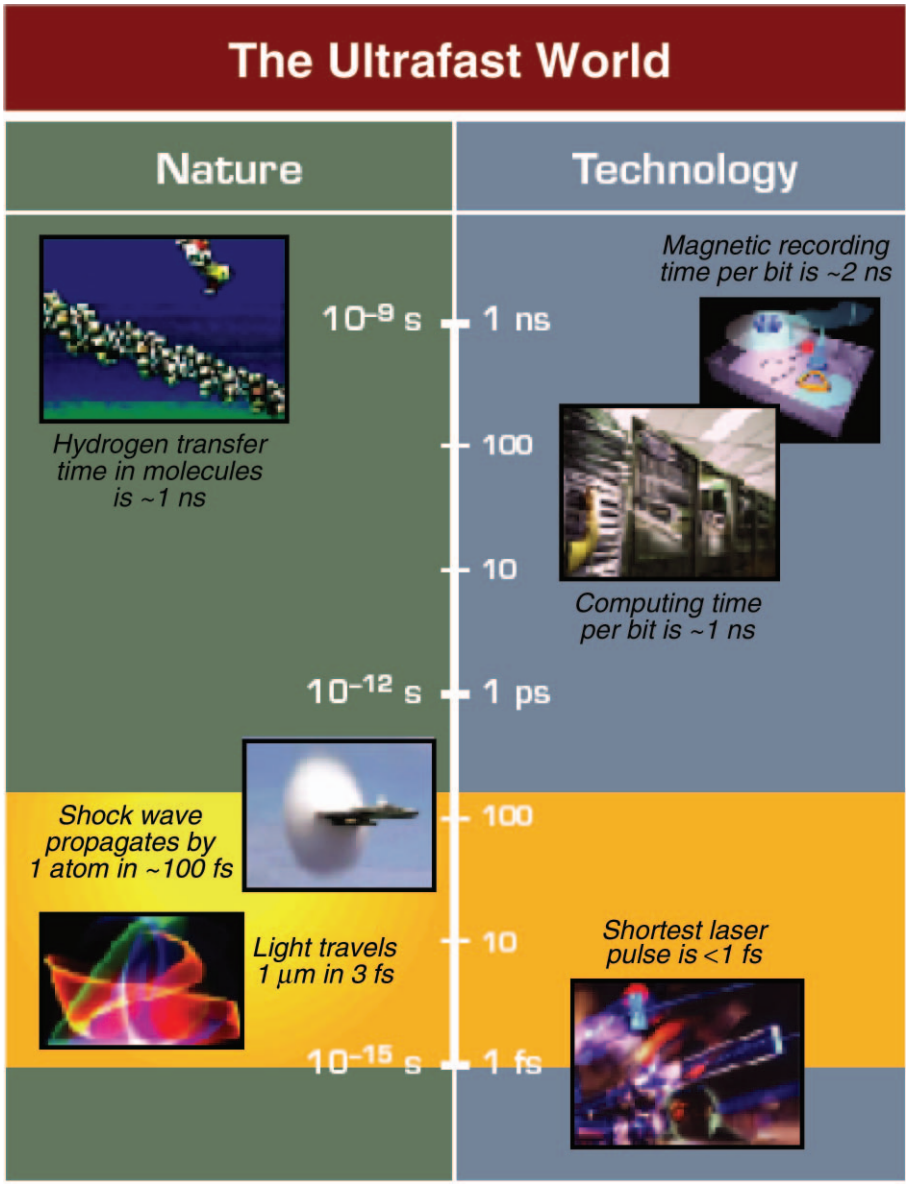

Figure 1. Illustration of ultrafast processes in nature and technology. A femtosecond, the duration of the shortest ultrafast laser pulse, is an extremely short period of time; light can travel around the Earth about seven times in a second, but only about 300 nm in a femtosecond. Adapted from Reference 8.

depth divided by the speed of sound, typically 1–5 ps. Thus, optical pulses with duration of <1 ps are usually sufficient for observing the sharpest possible acoustic signals.

Each of the articles in this issue of *MRS Bulletin* will answer the question differently, and in many cases the answer is complicated by the existence of multiple processes, some of which are not well characterized.

It is often stated that the ultrafast regime is reached when the pulse duration is less than the time scale of electron–phonon coupling. This statement has truth to it but sometimes oversimplifies reality. The laser pulse directly excites the electronic degrees of freedom in a material, and it takes time for the energy absorbed by the electrons to be transferred to the lattice. The complete description of this process is complex, even for the relatively simple case of a laser pulse incident on the surface of a metal. Energetic electrons transfer energy to other electronic excitations and to the motions of the atoms (i.e., the phonons), and despite the popularity of so-called "two-temperature" models that assume the electrons and phonons are independently in thermal equilibrium at two different temperatures, the time scales for these processes are not always well separated.[3] Furthermore, ballistic transport and rapid diffusion of hot electrons can distribute energy in a metal over much longer length scales than the optical absorption depth. Because many scattering events are needed to transfer energy from the highly excited electron system to the motions of the atoms, the time scale for the heating of the lattice can be much greater than 1 ps, depending on the material under study and the density of the energy deposited in the material.

The first article in this issue describes in detail the mechanisms and characteristic time scales of near-threshold laser ablation in metals and semiconductors. Reis and co-authors summarize the x-ray diffraction results obtained last year at the Stanford Linear Accelerator Center. This experiment *tour de force*—the publication in *Science*[4] has 51 authors associated with 18 institutions—represents a diffraction study of ultrafast melting with the fastest time resolution to date. During the initial 200–400-fs period following the laser pulse, a significant fraction (10–15%) of the valence electrons are excited to higher electronic states; this strong excitation greatly reduces the attractive part of the interatomic potential and allows the atoms to freely drift with their room-temperature thermal velocities until the electrons relax, 400–600 fs later. Atoms can drift as much as half of the unit-cell dimension, even though the center of mass, collectively, is still in the original position, without thermal expansion. Yet, once the electrons relax (600 fs to 1 ps), the system evolves in the same way as it would if the lattice of atoms was heated instantaneously by the laser pulse. Large-scale molecular dynamics (MD) and associated hydrodynamics simulations that are described later in the first article bear this out as well.

The articles that follow Reis et al. expand on these ideas and discuss specific applications of ultrafast lasers in the characterization and fabrication of materials.

Antonelli et al. describe how the rapid heating of the near surface of metal films by nanojoule ultrafast optical pulses can be used to generate strain and temperature fields for measurements of the mechanical and thermal properties of thin films and interfaces. Conversely, if the mechanical properties are known, the geometry of thin-film structures can be derived from the spectra of acoustic echoes and oscillations. This approach, termed *picosecond acoustics*, is widely used in the

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microelectronics industry for measuring the thicknesses of metal interconnects and interlayer dielectrics.

King et al. describe their efforts to develop an ultrafast imaging tool to observe the time-dependence of phase transformations and shock-driven mechanical processes related to the interaction of ultrafast light and materials. This article discusses progress on two distinct types of instruments, an ultrafast transmission electron microscope and a laser-produced, relativistic electron beam with MeV energies that is subsequently used as a source of high brightness x-rays and γ-rays.

Itoh et al. illustrate how bulk dielectrics can be modified with ultrafast lasers to produce waveguides, Bragg reflectors, optical devices, and microfluidic channels by direct-writing in three dimensions. Again, a thermal mechanism is identified as the root cause of these materials modification processes. In all cases, the extremely sharp thermal gradients produced by ultrafast laser pulses permit such structures to be created—something that would not be possible for laser pulses of longer than 10 ps.

Tull et al. describe the morphological changes that an ultrafast laser can produce at surfaces and interfaces. They describe the use of high-intensity ultrafast light to produce "black silicon" by taking advantage of laser-induced periodic structures and accelerating the process by machining in a halogen gas environment. This morphological variant of Si strongly adsorbs light in the IR, something flat Si does not do. The authors also describe another morphological modification of a Si–SiO_2 interface. Here, the ultrafast light is absorbed at the Si surface and the softened glass is blown into a bubble by the spallation of a thin layer of ejected molten Si, again a thermal mechanism. These bubbles have highly reproducible heights and can be accurately joined to form tubes capable of transporting fluid.

Finally, Haight et al. describe the use of ultrafast lasers to repair lithographic masks at the 20–200-nm scale. This is, to the best of our knowledge, the first application of ultrafast lasers for nanoscale fabrication in a manufacturing facility. Here, the deterministic threshold is exploited to either photo-dissociate precursor molecules to deposit a thin line or ablate unwanted material below the diffraction limit.

These articles describe but a few of the exciting materials research topics that are actively being studied today with ultrafast lasers. Unfortunately, the number of topics in materials research that we could not include because of space constraints is very large: the much more mature field of ultrafast spectroscopy,[8] new areas of spectral and temporal pulse shaping,[9] ultrafast interactions with organic and biological materials,[10] high-harmonic generation as a source of UV and soft x-rays, ultrafast generation of particle beams,[11] terahertz spectroscopy and imaging,[12,13] and ablation mechanisms in dielectric materials[14] (which are quite different from those in metals and semiconductors). Our hope is that the selection of topics will illustrate the breadth of this new area of ultrafast lasers in materials research and the applications that have already been generated.

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