INTERACTIONS OF ULTRA-INTENSE LASER LIGHT WITH MATTER

Tabletop pulsed lasers capable of delivering terawatts of peak power are beginning to have a revolutionary effect on atomic physics, plasma physics and accelerator technology.

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When the laser made its debut in 1960, it was often called a solution looking for a problem. Today the laser is hailed as one of the most significant inventions of the 20th century. Lasers are used in almost all fields of science and technology, and they have become commonplace in daily life, from supermarket scanners to CD players. The recent development of ultrahigh-power lasers has opened exciting research opportunities in the field of laser–matter interactions. They range from the interaction of laser light with single atoms to collective effects in plasmas.

Nowadays we can produce laser pulses lasting less than a picosecond that reach intensities exceeding $10^{18}$ W/cm$^2$ at a focus. At such intensities any medium is easily ionized to produce a plasma. Free electrons in the laser field begin to oscillate at relativistic velocities, and radiation pulses can exert gigabar pressures on the medium. Not only do such extreme effects directly present us with new research opportunities; they also hold out promise for many important applications, including compact high-gradient particle accelerators, laser fusion reactors and picosecond x-ray sources.

Ultra-intense laser sources

Since the invention of the pulsed laser, peak power has increased by 12 orders of magnitude. (See figure 1.) In the 1960s free-running laser oscillators were first Q-switched and later mode-locked to provide gigawatts of peak power in picosecond pulses. In later years chains of laser amplifiers boosted gigawatt pulses to still higher peak power.

If gain, bandwidth and energy storage had posed the only limitations on amplifying short laser pulses, terawatt ($10^{12}$ W) pulses would have become common two decades ago. However, in almost all gain media (or in the windows that confine them) nonlinear optical (Kerr) responses that can break up the laser beam imposed a severe limit on the intensity one could get from a laser of given aperture. For example, the nonlinear refractive index of glass restricts the peak intensity that a neodymium–glass laser can tolerate to about $10^{9}$ W/cm$^2$. Therefore the relentless development during the 1970s and early 1980s of ultra-

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short-pulse lasers, which eventually led to the generation of femtosecond (10^{-15} sec) pulses, had little impact on high-intensity lasers.

The situation changed dramatically with the introduction of "chirped" pulse amplification in the mid 1980s. One can chirp an ultrashort pulse by passing it through a dispersive delay line, thereby increasing its duration by a factor of up to 10^4 and reducing instantaneous power correspondingly. In such a dispersively dilated, or chirped, pulse the photon wavelength varies with time. A conjugate delay line can then undo the chirp, returning the pulse to its original duration. The amplifying is done between the two delay lines, when the pulse is in its dispersed state. Thus a long and consequently low-power pulse can be amplified up to the amplifier’s saturation energy, but the amplified pulse is coded in such a way that it can be reconstructed as an ultrashort pulse with very high peak power. Pairs of optical gratings serve as the dispersive delay lines in this technique for making very intense ultrashort light pulses.

Chirping can deliver a joule of energy in a subpicosecond laser pulse. Such incredibly high light intensities have electric fields on the order of 10^{14} V/m. That’s 100 times the Coulomb field binding the ground-state electron in a hydrogen atom. Such a field is strong enough to ionize uranium to U^{82+} in less than a picosecond. When the laser intensity is increased beyond threshold, the ionization rate for the charge state in question increases rapidly. Thus one can now generate a highly ionized plasma containing a narrow range of ionic species simply by choosing the proper laser intensity. That’s something that could not be done before the advent of ultrahigh-power laser pulses. The ability to control the charge state of the plasma has important ramifications for x-ray laser research, as we shall see.

Interacting with an atom
An intense laser pulse can ionize an atom even when the photon energy is much less than the atom’s ionization energy. That’s possible because the atom absorbs many photons at once. Although such multiphoton ionization has been studied extensively, researchers are only now beginning to appreciate the unique properties of media ionized in this way.

Let us first consider the ionization process itself. Figure 2 is a plot, for several noble gases, of the measured threshold intensity for observing a particular ionic state versus the threshold intensity calculated by the so-called barrier-suppression ionization model. (Threshold intensity is defined as that at which 1% of the irradiated atoms are ionized.) In the BSI model the intense electric field of the laser frees a bound electron by effectively suppressing the Coulomb attraction of the nucleus. As shown in the figure, the model predicts threshold intensities for the various ionic states of noble gases rather well.
dimensions of the plasma.

One can also impose initial conditions on the plasma by means of electric currents or magnetic fields. For instance, ionization with circularly polarized light imparts to the plasma angular momentum that results in an induced magnetic field whose magnitude can be as large as a megagauss. The average electron energy of the plasma after the passage of a short laser pulse of intensity \( I \), wavelength \( \lambda \) and polarization \( p \) is given by \( U(p) \), where \( f(p) \) accounts for the polarization dependence and \( U \), the oscillation or "quiver" energy of the electron in the laser field, is proportional to \( \lambda^2 \). Thus one can control the electron energy distribution simply by varying the polarization or the wavelength of the ionizing laser.

Figure 3 illustrates this control over the average electron energy with data on the ionization of xenon. Measured kinetic energy distributions are shown for electrons freed by laser pulses of different wavelength and polarization. For picosecond pulses of 10.6-micron light (shown in red), Corkum and coworkers get a dramatic difference simply by changing the polarization from linear (square data points) to circular (shown as dots). The blue data show the electron energy distribution produced by a 0.4-psec pulse of linearly polarized 625-nm laser light. The differences between the three data sets demonstrate our very considerable ability to control the initial electron distribution transverse to the laser's propagation direction.

These electron distributions are not only nonthermal; they are also anisotropic. In the longitudinal direction the plasmas are cold. Such highly nonthermal plasmas with well-defined initial conditions are the key to extending pulse-probe spectroscopy to plasma physics. One can now design experiments to study many relaxation processes in plasmas, for example electron-electron and electron-ion thermalization, just as solid-state physicists have already been studying relaxation processes in semiconductors for a decade.

X-ray lasers and plasma waveguides

X-ray laser schemes involving a laser-produced plasma as the gain medium are already benefiting from the development of high-intensity, short-pulse lasers. One such scheme is known as the recombination x-ray laser: A subpicosecond laser pulse is used to create a dense, cold plasma of the desired ionic species. Recombination preferentially populates the higher-energy states of the ion, because the three-body recombination rate scales as the electron density squared divided by the temperature. This scaling behavior, coupled with the faster radiative decay of the lower levels, produces population inversions among atomic levels with principal quantum numbers up to 4.

In 1993 Yutaka Nagata and his colleagues at the Institute for Physical and Chemical Research in Japan demonstrated lasing at 13.5 nm in the far ultraviolet by recombination in a hydrogen-like lithium plasma produced by an ultrashort laser pulse.

The length of the x-ray lasing medium can be increased by placing the active atoms in a plasma waveguide, thereby producing, in effect, a fiber laser. Wave guiding in a plasma requires that the electron density profile have a minimum along the axis, so that the refractive index will be highest on the axis. Charles Durfee and Howard Milchberg at the University of Maryland have recently demonstrated such a plasma waveguide. To form the waveguide they ionized xenon gas at 30 torr with a focused 100-psec laser pulse, producing a uniform plasma 6 mm long. After several nanoseconds the thermal expansion of the plasma created the desired axially symmetric density profile. A second probe pulse, with an intensity reaching \( 10^{12} \) W/cm\(^2\) and an initial focused (Rayleigh) length of only 300 microns, was then focused on this plasma waveguide. This "plasma fiber" guided the pulse along its entire length, proving the concept of plasma wave guidance for high-intensity laser pulses. (See figure 4.) Plasma waveguides are also desired for particle accelerators (we'll come to that) and for compressing high-power chirped laser pulses.

Higher harmonics

When ultrashort laser pulses were first being focused onto high-density gas-jet targets a few years ago, several groups observed a surprising result: The interact ion was generating extremely high odd-order harmonics of the incident laser wavelength. The harmonic spectrum thus generated has two characteristic features: First of all, the conversion efficiency falls off rapidly with increasing order, but only up to about the tenth harmonic. Then comes a plateau, where the conversion efficiency (roughly \( 10^{-6} \) to \( 10^{-9} \)) falls off rather slowly. Recent experiments with extremely short laser pulses have approached or reached the 135th harmonic of 1.05-micron incident radiation. These very high harmonics show a cutoff at photon energies of about 3 times the electron quiver energy \( U \).

Until recently there was no semiclassical explanation...
of either the high-harmonic generation or its abrupt cutoff at $3U$. High-harmonic generation can be thought of as a kind of bremsstrahlung due to the recollision of freed electrons with the ions. But unlike the ordinary continuous bremsstrahlung spectrum engendered by random collisions, this emission comes in discrete harmonics of the incident laser frequency. That is because, during the ionization process, the electrons tunnel their way out through the atomic barrier only during restricted portions of the laser field oscillation period. Thus the moment of collision with the nucleus is not a random event. The recollision of previously freed electrons oscillating in the laser field is periodic, and so is the consequent bremsstrahlung.

The energy of the harmonic photon can be as high as the instantaneous electron kinetic energy plus the ionization energy $E_i$. It can be shown that the maximum energy available for harmonic emission is approximately $3U + E_i$. These recombination events are responsible for generating the higher harmonics on the plateau. Other electron scattering processes play a role in, and may even dominate, the generation of the lower harmonics.

Figure 5 shows the harmonic power spectrum produced by J. J. Macklin and his colleagues at Stanford with 125-fsec pulses of 800-nm laser light focused to peak intensities of $10^{15}$ W/cm$^2$ in a 2.5-mm-thick neon target at a pressure of 13 torr. They were able to observe extremely high harmonics of the pump radiation. Between the 21st and 111th harmonics, output intensities varied within not much more than a factor of 10.

At extreme ultraviolet wavelengths around 10 nm, harmonic generation by the interaction of ultrashort laser pulses with noble gases turns out to be one of the most cost-effective ways we have of making intense, coherent radiation. In the first application of this technique, K. Budil and colleagues at Livermore have recently measured the photoionization cross section of neon from 20 to 100 eV. In the past, such measurements in the extreme ultraviolet required access to large and expensive synchrotron-radiation facilities.

High-harmonic generation is only one of the things that happens when electrons come back close to the nuclei from whose pull they had just escaped within the first laser oscillation period. The ionized electron might, for example, scatter inelastically upon its return, knocking a second electron free. Such "multiphoton double-ionization" has recently been observed in the ionization of helium by 0.6-micron laser light.

**Plasmas from solids**

Our discussion so far has been restricted to rather dilute plasmas produced by laser ionization of gases. By contrast, when an intense, short laser pulse hits a solid target, the electron–ion collision times are much shorter than the pulse duration. Therefore it is the laser pulse duration that determines the time scale for temperature changes in the plasma formed from the solid. The expansion of such a solid-density plasma, which erases the density discontinuity at the solid surface, occurs on a still longer time scale. Therefore the optical radiation is in effect interacting with the still-solid target. So one sees in such experiments a short burst of continuum radiation as well as discrete x-ray emission lines.

The x-ray burst from a solid target can be shorter than a picosecond. Though the radiation is incoherent, it can have a peak power many orders of magnitude higher than one can get in the 1–10-keV range from synchrotron light sources. Such short, intense x-ray pulses offer the possibility of picosecond resolution for many kinds of experiments in chemistry and biology. Given the terawatt lasers with kilohertz repetition rates that are now being contemplated, the x rays from such high-density plasmas should find commercial applications in x-ray lithography and x-ray imaging.

**Kinetic energy distribution** of the electron population produced by pulsed laser ionization of xenon depends crucially on the wavelength and polarization of the laser light. For picosecond pulses of 10.6-micron light (shown in red), one gets a dramatic difference by changing the polarization from linear (square data points) to circular (shown as dots). (Data of P. Corkum et al., ref. 4.) The blue squares are data points (from ref. 5) for picosecond pulses of linearly polarized 625-nm light. **Figure 3**
At $10^{18}$ W/cm$^2$ the light pressure of an ultrashort laser pulse is 0.3 gigabar. The light pressure can exceed the thermal pressure of the low-temperature plasma, whose density is still close to that of the solid from which it sprang. Thus the light pressure can impede the plasma's expansion. This effect, known as density profile modification, was well known in early laser fusion experiments with longer laser pulses. Recently X. B. Liu and Donald Umstadter at the University of Michigan managed to observe a reduction in the plasma's expansion velocity with increasing intensity of the ultrashort laser pulses. They measured the Doppler shift of the probe pulse reflected from the expanding plasma with femtosecond resolution during the pump pulse that created the plasma. As one can see in figure 6, the Doppler shift of the probe pulse is reduced near the peak of the pump pulse. That result demonstrates the effect of the radiation pressure, which is the highest at the peak, in reducing the expansion velocity of the plasma.

The radiation pressure of an ultra-intense pulse plays a crucial role in the "fast ignitor" scheme, a new concept for achieving energy from laser fusion. In this scheme the radiation pressure of a short laser pulse would bore a hole through the plasma atmosphere of an imploding fusion pellet so that yet another ultrashort pulse could propagate through the hole and reach the solid surface. At the surface of the pellet the energy of the ultrashort pulse would be converted into a beam of MeV electrons. Those hot electrons in turn would deposit their energy in the compressed deuterium-tritium fuel, igniting the fusion reactions.

The first solid-target experiments that show evidence of such hot-electron generation were performed recently by Stephen Harris's group at Stanford. Both MeV electrons and x rays were detected at a laser intensity of $10^{18}$ W/cm$^2$. The escaping electrons set up a strong space-charge field that accelerates the ions to high energies. A. P. Fews and coworkers at the Rutherford Laboratory in England have recently measured energetic ion emission from solid-target plasmas. They found that as much as 10% of the incident laser energy was transferred to ions with energies above 100 keV per nucleon, with a mean ion energy of 1.3 MeV. Such efficient ion production is only possible if a similar fraction of the energy of the incident laser pulse is transferred to the hot electrons. That's a necessary condition for fast-ignitor fusion.

**Collective effects in plasmas**

What happens if an ultra-intense short pulse interacts with a plasma that has already been formed by some other means or by the laser field itself? Not only will such a laser pulse modify the plasma, for example, by exciting plasma waves; the plasma, in turn, will also affect the laser pulse by modifying its spectral content. To see why the physics of the interaction depends on the pulse's length and intensity it is instructive to review the more familiar scenario of the interaction between a long laser pulse and an underdense plasma. A plasma is said to be underdense if the plasma frequency $\omega_p$ is much less than the laser frequency $\omega_l$. The plasma frequency is the frequency with which displaced electrons would oscillate about their equilibrium positions.

A long pulse in such a plasma, say hundreds of plasma periods long, can excite one of many parametric instabilities, for example Raman scattering. (In plasma physics, Raman scattering refers to inelastic scattering of a photon by an electron plasma wave.) In that case the instability grows as a result of the interaction between the scattering light wave and a plasma wave.

But what if the laser pulse lasts less than a plasma period or is so intense that the quiver velocity $v_0$ of the plasma electrons in the laser field approaches the speed of light? Then, with less than one full wavelength of the plasma wave, the analysis based on wave–wave interactions becomes inappropriate. Such laser pulses give rise...
to new phenomena such as plasma wakefield generation, photon frequency upshifting, relativistic guiding and relativistic harmonic generation.

When a short, intense laser pulse traverses an underdense plasma, the ponderomotive force (essentially the force on the electrons due to the exchange of momentum between the laser field and the plasma) disturbs the electrons and leaves behind a "wake" of oscillations, much like the wake of a motorboat. The ponderomotive force that drives the wake is proportional not to the laser intensity but to its gradient. Thus any laser pulse that has a sharp rise or fall on the scale of \( c/\omega_p \) (the plasma's collisionless skin depth) will excite a wake. A laser pulse of half the wavelength of the resultant plasma wave is particularly effective for generating a wake, because its rise and fall both contribute kicks to the plasma electrons in phase. For a given plasma electron density the optimum pulse duration is therefore given by

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\tau = 0.6 \text{ psec} \left( \frac{10^{16}}{n_e (\text{cm}^{-3})} \right)^{1/2}
\]

For plasma densities on the order \( 10^{17} \text{ cm}^{-3} \), the laser pulse duration must be on the order of 100 femtoseconds to excite plasma wakes.

It is interesting that a plasma wakefield's phase velocity is tied to the group velocity of the laser pulse, which is almost \( c \). Furthermore the maximum longitudinal electric field associated with such a wake scales roughly as \( v_e^2 n_0 \). For \( n_0 \) of order \( 10^{17} \text{ cm}^{-3} \) and \( v_e \) near \( c/2 \), the electric fields are on the order of a few GeV per meter—much larger than one can get in a conventional radiofrequency waveguide. That is why Toshiki Tajima and John Dawson, in their classic 1979 paper, proposed that nonlinear optical effect is called self phase modulation. At some point during the intensity rise of the laser pulse a plasma begins to form, lowering the medium's refractive index and consequently blueshifting a portion of the pulse. Such frequency changes are beautifully illustrated in a recent experiment by W. M. Wood and colleagues at the University of Texas. Focusing a millijoule, 620-nm-wavelength laser pulse lasting 100 femtoseconds into a gas cell, they observed up to 30 nm of ionization-induced blueshift. One could actually see the laser pulse change color.

Self phase modulation is central to many nonlinear optical processes. In optical fibers, for example, it makes possible the compression of 30-fsec pulses to 6 fs, the shortest light pulses ever produced. Also, the short-pulse Ti:sapphire lasers that are now commonplace in laboratories are mode-locked by means of Kerr-lens self phase modulation.

If we are to transfer any of the technology of low-power self phase modulation to high power or short wavelengths, the nonlinear media will have to be fully formed plasmas or ionizing gases. The technical implications of phase modulation by a plasma are already being investigated. For example, ionization fronts have been used not only for frequency upshifting but also for compressing intense, ultrashort pulses. In fact, there is now no reason why plasma compression cannot be done in plasma fibers. Joshi and coworkers have also shown that 35-GHz microwaves can be upshifted in a continuous fashion.
to 173 GHz by means of an ionization front created by a picosecond laser.

A further twist comes from the rich nonlinear response of a plasma medium. When a laser pulse ionizes a gas through which it is propagating, a force is exerted on the medium at the ionization front. As distinguished from the usual ponderomotive force, which is proportional to the gradient of the light intensity, this other force is proportional to the gradient of the dielectric constant. It originates from the longitudinal momentum imparted to the newly born electrons by the laser beam during ionization. This force, in turn, can also induce a wake in the plasma medium behind the ionization front.

There is yet another fascinating way to upshift the frequency of the photons. In wakefield excitation a short laser pulse loses its energy to a plasma wave. The reverse is also possible: A short light pulse can absorb energy from a preexisting plasma wave if it is placed at the proper phase of the wave. Instead of amplifying this light pulse, the plasma wave upshifts its frequency. That is to say, the energy of each photon is increased but the number of photons is not. The situation is analogous to the acceleration of charged particles by plasma waves: Each photon gains energy and is accelerated toward c, because the laser frequency increases and the group velocity of the pulse is \( c \left( 1 - \frac{\omega_p^2}{\omega^2} \right)^{1/2} \). This method of frequency upshifting is therefore called "photon accelerator." As is the case with particle accelerators, this kind of photon acceleration cannot be done with long pulses, because the photons would sample both accelerating and decelerating phases, that is to say, regions of increasing and decreasing plasma density. Thus the interaction of a long light pulse with the plasma wave produces a periodic modulation at the plasma frequency \( \omega_p \), resulting in FM sidebands at discrete multiples of \( \omega_p \). These new sidebands are the familiar Stokes and anti-Stokes cascade generated by the forward Raman scattering instability in nonlinear optical systems.

**Instabilities**

Recently a Livermore-UCLA collaboration has observed a short-pulse version of the classic laser–plasma stimulated Raman backscattering instability. The backscattered light has a novel frequency signature that depends on the laser intensity. Furthermore its frequency spectrum spreads to the blue side of the incident wavelength. That’s different from the classic Raman backscatter, which downshifts the incident frequency by approximately \( \omega_p \). It’s just one example of the fascinating new effects that are being observed as we move into a new regime for collective effects with short laser pulses. One very interesting theoretical prediction is that the growth rate of the Raman forward-scattering instability, which increases with intensity below \( 10^{18} \) W/cm\(^2\), will approach zero for ultra-relativistic laser intensities. (By ultrarelativistic we mean that \( cE \) greatly exceeds \( m_e c \omega_0 \).) It should be possible to test many of these ideas with the petawatt class lasers now being built.

When \( cE \) exceeds \( m_e c \omega_0 \), the motion of the plasma electrons in the laser field becomes relativistic, and new effects such as relativistic guiding, harmonic generation, and overdense penetration (\( \omega < \omega_p \)) become important.

In a plasma, as in any nonlinear optical medium, self-focusing of a laser pulse arises because the refractive index becomes lower where the intensity of the radiation is most intense. By contrast with thermal or ponderomotive self-focusing, which occurs because of temperature or transverse density fluctuations, relativistic self-focusing is caused by the drop in plasma frequency due to the relativistic mass increase of plasma electrons oscillating in the laser field. For long laser pulses, the relativistic self-focusing has a power threshold of \( 20 \omega_p^2 / \omega_0^2 \) gigawatts. The change in refractive index depends on the changes both in the electron mass and in the density of electrons.
For short pulses, therefore, the plasma wake also contributes to the refractive index. The result is that the first plasma oscillation period propagates unaffected but later in the laser pulse the wake’s contribution to refraction enhances the focusing.

Relativistic guiding may eventually find an important application in collective plasma acceleration. The theoretical maximum energy gain in a plasma accelerator can only be achieved if the plasma wave exists over a distance much greater than the Rayleigh range. Relativistic guiding is one way of maintaining the high laser intensities needed to excite a large plasma wave.

Another important relativistic effect is the generation of odd harmonics of the incident laser light. The efficiency of relativistic harmonic emission is rather low in underdense plasmas. But the large density oscillations and relativistic electron velocities created by an intense short pulse incident on the sharp edge of an overdense plasma can yield much higher conversion efficiencies. For longer pulses the radiation pressure can steepen the density profile of a plasma at the point of critical density (where $\omega = \omega_p$), because the ions then have time to move. The interaction of an intense laser pulse at a steepened plasma profile offers a very favorable situation for the efficient generation of high harmonics. Experiments at Los Alamos with a $10^{16}$-W/cm² carbon dioxide laser beam hitting a solid plasma have demonstrated the generation of harmonics up to the 46th.  

References