

Fundamentals of ultrafast laser–material interaction

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Short pulse laser irradiation has the ability to bring a material into a state of strong electronic, thermal, phase, and mechanical nonequilibrium and trigger a sequence of structural transformations leading to the generation of complex multiscale surface morphologies, unusual metastable phases, and microstructures that cannot be produced by any other means. In this article, we provide an overview of recent advancements and existing challenges in the understanding of the fundamental mechanisms of short pulse laser interaction with materials, including the material response to strong electronic excitation, ultrafast redistribution and partitioning of the deposited laser energy, the peculiarities of phase transformations occurring under conditions of strong superheating/undercooling, as well as laser-induced generation of crystal defects and modification of surface microstructure.

Introduction

The emergence of ultrashort pulse lasers has opened up new opportunities for high-precision processing of a wide range of materials, including dielectrics, semiconductors, and metals, enabling numerous applications in fields ranging from micromachining to photonics and life sciences.^{1–3} The ability of ultrashort lasers to confine energy deposition in small regions of irradiated targets makes it possible to perform selective material modification and, at the same time, provides unique opportunities for investigation of material behavior and properties under extreme conditions of strong electronic excitation, rapid heating and cooling, and ultrafast mechanical deformation. A hierarchy of intertwined processes triggered by ultrashort laser pulses in metals, dielectrics, and semiconductors^{2,4} is schematically illustrated in **Figure 1**. The laser-induced processes can be roughly separated into three groups, highlighted by different colors in the figure, namely, laser excitation of optically active electronic states in the irradiated

material and the initial ultrafast processes occurring in response to the electronic excitation (yellow), rapid nonequilibrium phase transformations triggered by the energy transfer from the excited electrons to atomic vibrations (green), and the subsequent cooling and solidification of the surface region of the irradiated target (turquoise).

In semiconductors and dielectrics, a high concentration of free electrons promoted across the bandgap by laser excitation may lead to non-thermal phase transformations induced by transient modification of interatomic bonding.^{5,6} Meanwhile, the excited electrons in semiconductors and dielectrics, as well as conduction-band electrons in metals, are capable of effective laser energy absorption, which may bring the material to plasma states or even produce a Coulomb explosion due to the electron emission and charging of a surface region of a dielectric target.⁷ The energy transfer from electrons to lattice vibrations can lead to rapid heating with rates exceeding 10^{14} K/s, resulting in homogeneous melting^{8,9} or even formation of a supercritical fluid.¹⁰

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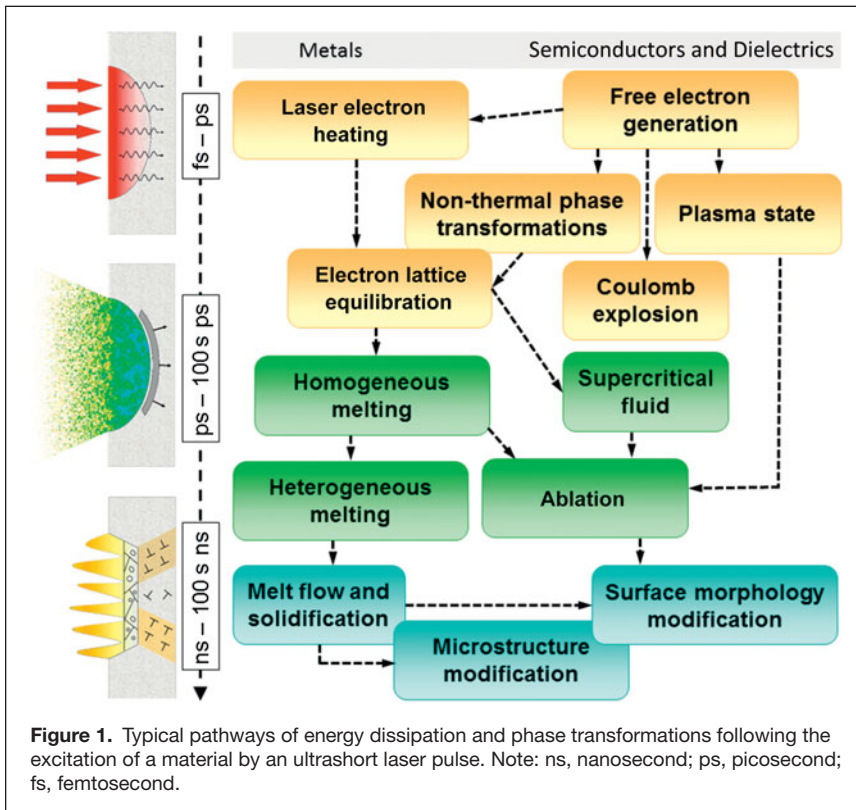
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The extreme temperature and pressure conditions created by the rapid laser energy deposition may cause generation of unusual metastable phases,^{11–13} as well as complex surface morphology^{3,14–16} produced in the course of photomechanical spallation^{17,18} or explosive decomposition of superheated material into a vapor–droplet mixture.^{19–21} When the temperature drops below the equilibrium melting point, resolidification starts. In metals, strong laser energy localization combined with large thermal conductivity result in high cooling rates of up to 10^{12} K/s,^{22,23} which bring the material into a state of strong undercooling. The solidification of highly undercooled liquid may lead to the formation of unusual defect configurations,^{24,25} as well as nanocrystalline²³ and amorphous^{22,26} structures.

In this article, we provide a brief overview of the processes previously outlined, starting from the discussion of the laser excitation and non-thermal processes occurring in the electronically excited state of the matter and following with laser-induced structural/phase transformations and laser ablation.

Laser excitation and non-thermal processes

The energy of a short laser pulse couples mainly to electrons of the irradiated material. In metals, the presence of conduction-band electrons enables the direct absorption of laser energy by electrons undergoing collisions with nuclei (i.e., by inverse bremsstrahlung). In semiconductors and dielectrics, electrons have to be excited across the bandgap before they can directly absorb photons.^{27,28} The electronic excitation in materials with a bandgap may strongly modify the interatomic bonding and can lead to ultrafast phase transitions, including

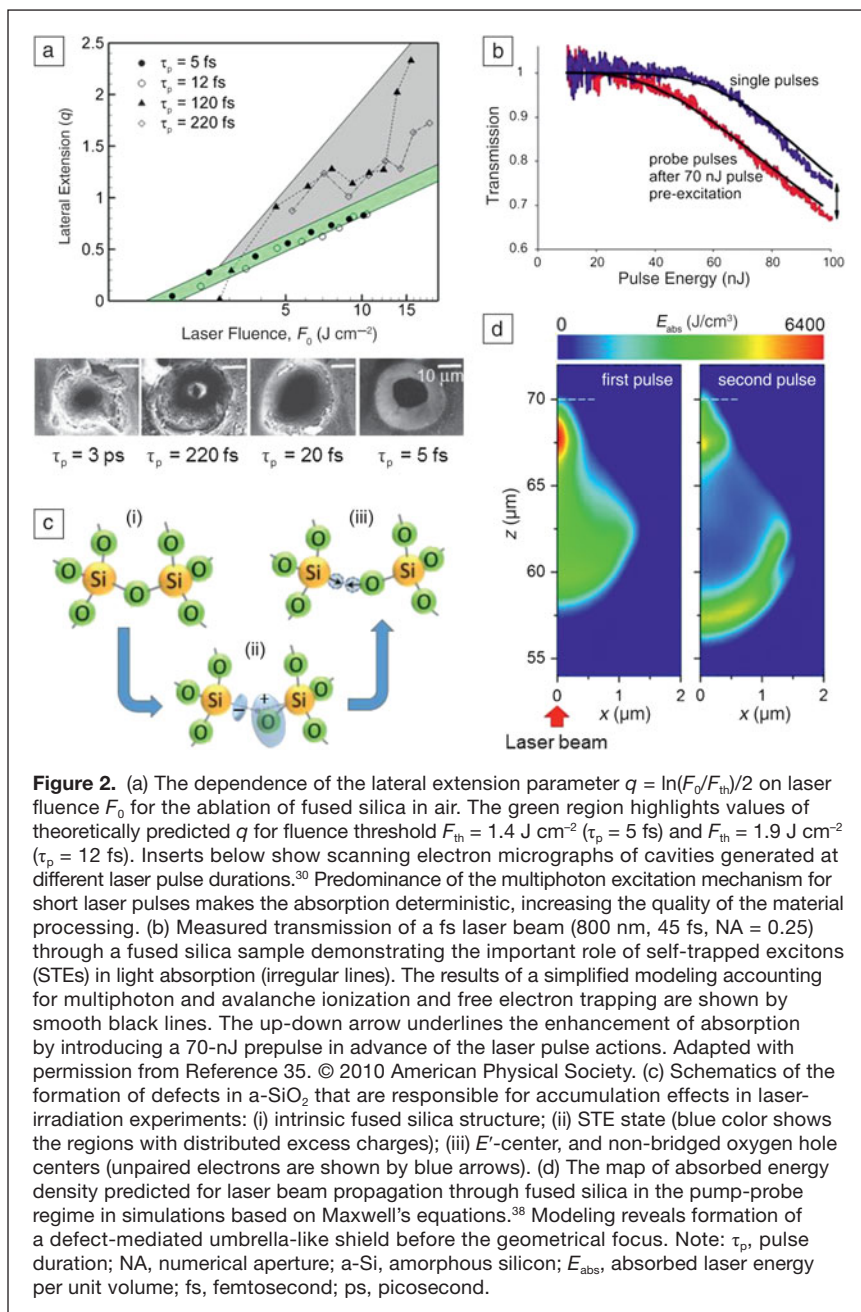
ultrafast melting⁵ and solid–solid phase transformations.⁶ Such phase transitions are referred to as “non-thermal,” as they are directly stimulated by the laser excitation, take place in the electronically excited state on the sub-picosecond (ps) time scale, and require no electron–phonon equilibration and lattice heating.

In wide bandgap dielectrics, the main mechanisms involved in the generation of free electrons by photons having lower energy than the bandgap are multiphoton and avalanche ionizations, which are highly nonlinear processes.²⁷ Multiphoton excitation (i.e., simultaneous absorption of several photons) is sensitive to the laser intensity as its probability exhibits a power law dependence on the laser intensity, which, in effect, makes the absorption process deterministic. It becomes effective for ultrashort laser pulses whose intensities are greatly increased due to the reduction of the pulse duration τ_p . At high laser intensities, the potential barrier for the escape of electrons from the atoms is distorted, and ionization by electron tunneling through the reduced barrier can dominate over multiphoton ionization.²⁹ Once the electrons are excited to the conduction band, they absorb laser energy

via inverse bremsstrahlung and can produce a sequence of electron–hole pairs through collisions with valence-band electrons. This process leads to avalanche multiplication of the density of conduction-band electrons.

The competition between these two mechanisms of free carrier generation is responsible for considerable improvement of machining quality with the use of ultrashort laser pulses, as illustrated in **Figure 2a**. Absorption of ultrashort laser pulses with durations of tens of femtoseconds (fs) and peak intensities above 10^{15} W/cm² is mostly governed by the photoionization mechanisms (multiphoton or tunneling), whereas for free electrons the laser pulse is too short to trigger the avalanche process. In this case, the dependence of the lateral extension parameter q , which relates the characteristic size of a generated structure divided by the illuminated area to the incident laser fluence, is logarithmic for a Gaussian beam profile. Therefore, laser-generated cavities can be fabricated with sub-10 nm lateral and vertical precision.³⁰ For $\tau_p > 100$ fs, the value of q strongly deviates from the logarithmic dependence, and the modified zone has a rugged splintered morphology, suggesting collateral damage outside the area where the laser fluence exceeds the damage threshold. This is likely to be conditioned by the “stochastic” avalanche process, which becomes efficient at longer τ_p .³¹

The defects that introduce intra-bandgap states also play an important role in laser excitation/absorption of wide-bandgap dielectric materials. Much work has been done on identifying the kinds of laser-generated defects in optical materials, especially in fused silica glass (bandgap of ~ 9 eV),^{32,33} and their roles in laser-induced ionization processes. The pump-probe



method, where an excitation pulse is followed by another incoming pulse with an adjustable time delay to probe the transient changes in reflection or transmission within the irradiated spot, is one of the most informative experimental techniques for such studies.^{34,35}

In particular, the characteristic lifetimes of free electrons³⁴ and self-trapped excitons (STEs)³⁵ ($\sim 150 \text{ fs}$ and $>30 \text{ ps}$, respectively) have been determined for fused silica by varying time delay between the pump and probe pulses. Figure 2b³⁵ demonstrates that absorption of energy from a laser beam tightly focused inside the bulk glass can be considerably enhanced by a prepulse, which couples to the sample only 3 ps in advance of the probe pulse and forms a population of STEs. The most

important defects produced in fused silica by laser irradiation are schematically presented in Figure 2c. Free electrons created by the pump pulse through ionization of intrinsic glass structure are swiftly trapped into STE states. At the ps time scale, the STEs recombine with $\sim 0.1\%$ of STEs evolving into permanent defects,³⁶ E'-centers (undercoordinated silicon atom), and non-bridged oxygen hole centers (oxygen atoms bonded to one silicon atom with one unpaired electron each) with the absorption peak of 5.8 eV.³³ Accumulation of permanent defects is one of the key factors that affects the quality of laser processing of wide-bandgap dielectrics.³⁷

An example of the strong effects of laser-generated defects on laser beam propagation is provided in Figure 2d, where the results of simulations based on Maxwell's equations supplemented by hydrodynamic-type equations for free electrons oscillating in the laser field³⁸ are shown for pump-probe irradiation conditions with laser energy in each pulse of $0.2 \mu\text{J}$, $\tau_p = 150 \text{ fs}$, and time separation between the pulses of $\sim 3 \text{ ps}$. The simulations, which take into account the formation of STEs and their re-excitation by the probe pulse, predict the formation of a defect-mediated umbrella-like shield and explain the enhancement of light absorption in the pump-probe experiments.³⁵

As demonstrated in a recent work on high-impact polystyrene³⁹ (see Figure 3), defect-mediated laser absorption can lead to the reduction of the threshold fluence for material modification with an increase in the laser beam radius, w , and the number of laser pulses applied to the same spot on the surface, N . The darkening of the modified surface in Figure 3a is due to ablation and phase separation, as illustrated by the scanning electron microscope image of the surface topography. The reduction of the threshold for large beam radii ($w \gg \Omega^{-0.5}$,

where Ω is the areal density of active low-density defects [LDDs], within the absorption volume), shown in Figure 3b, is ascribed to LDDs embedded in the matrix material and schematically illustrated in Figure 3c.³⁹ These defects act as donors of free electrons that can initiate an electron avalanche and, as discussed, increase laser absorption. At small beam radii ($w \ll \Omega^{-0.5}$), the laser radiation interacts mainly with the matrix material. Accumulation and incubation of LDDs upon repetitive laser irradiation significantly reduces the damage threshold (Figure 3b). This reduction is due to the generation and accumulation of optically active high-density defects (Figure 3c) with a separation distance that is smaller than the irradiation wavelength.

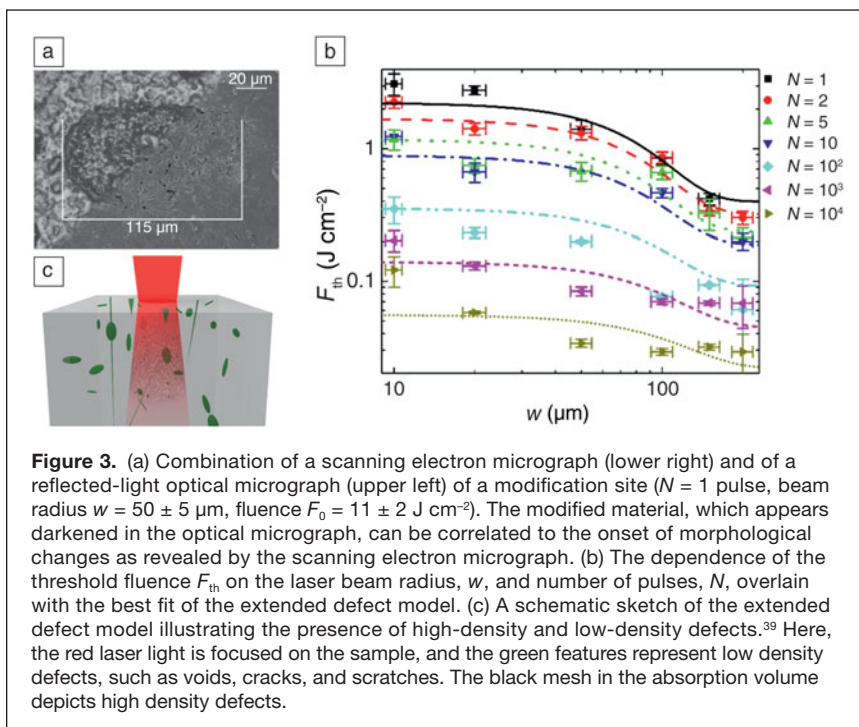


Figure 3. (a) Combination of a scanning electron micrograph (lower right) and of a reflected-light optical micrograph (upper left) of a modification site ($N = 1$ pulse, beam radius $w = 50 \pm 5 \mu\text{m}$, fluence $F_0 = 11 \pm 2 \text{ J cm}^{-2}$). The modified material, which appears darkened in the optical micrograph, can be correlated to the onset of morphological changes as revealed by the scanning electron micrograph. (b) The dependence of the threshold fluence F_{th} on the laser beam radius, w , and number of pulses, N , overlain with the best fit of the extended defect model. (c) A schematic sketch of the expanded defect model illustrating the presence of high-density and low-density defects.³⁹ Here, the red laser light is focused on the sample, and the green features represent low density defects, such as voids, cracks, and scratches. The black mesh in the absorption volume depicts high density defects.

Laser-induced structural and phase transformations

The initial non-thermal material response to the ultrafast laser excitation is quickly, within picoseconds, followed by the onset of thermal processes induced by the energy transfer from the excited electrons to atomic vibrations, commonly described as electron–phonon coupling. Rapid electron–phonon equilibration can produce ultrafast heating rates exceeding 10^{14} K/s, opening unique opportunities for investigating the kinetic limits of achievable superheating of solid phases. A combination of time-resolved optical, x-ray, and electron diffraction probing of short pulse laser melting^{40–42} with theoretical analysis⁸ and molecular dynamics (MD) simulations^{9,43,44} have provided important information on the time scales and mechanisms of the melting process occurring under conditions of strong superheating.

However, the short pulse laser irradiation may not only cause rapid heating and melting of localized surface or internal regions of the target, but also produce sharp temperature gradients leading to rapid quenching of the transiently melted material. In strongly absorbing materials such as metals or semiconductors, a combination of shallow depth of the laser energy deposition with high thermal conductivity of the irradiated material can lead to cooling rates approaching and even exceeding 10^{12} K/s.^{22,23} The cooling rates can be even higher, in excess of 10^{13} K/s,⁴⁵ when the laser energy deposition is confined not only within the thin surface layer of the irradiated target, but also in the lateral dimensions, creating conditions for even faster two- or three-dimensional heat transfer from the absorption region. The laterally localized laser energy deposition can be achieved by using tightly focused

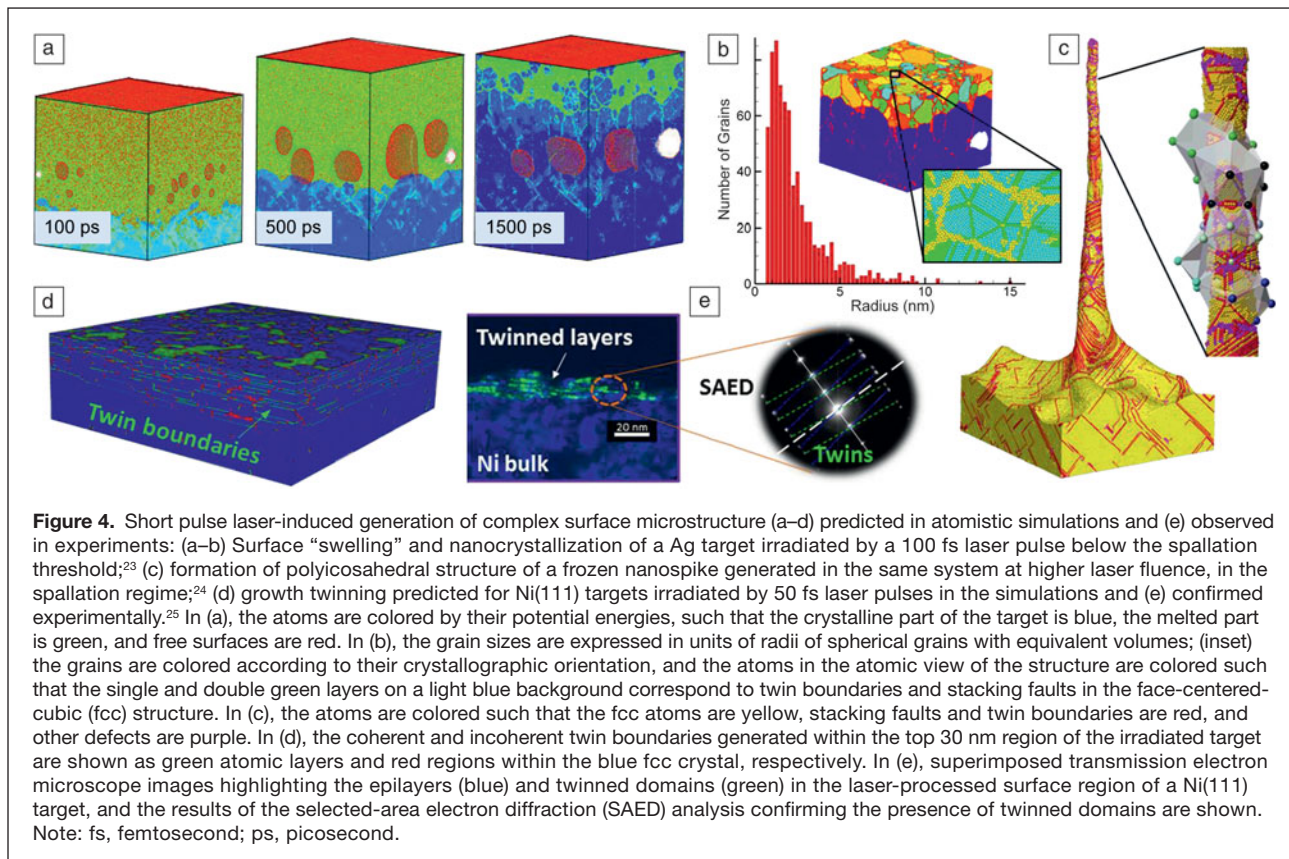
laser beams^{46,47} or taking advantage of the local field enhancement in the vicinity of a tip of a scanning probe microscope.^{48,49}

The ultrahigh rates of laser-induced cooling and the resulting resolidification, combined with the generation and relaxation of strong compressive stresses generated by the fast laser heating of the absorption region,^{17,18} create conditions for the formation of unusual metastable phases and unique material microstructures that are difficult if not impossible to produce by any other processing methods. In particular, metallic glass formation in systems not previously considered as glass formers has been demonstrated in ns- and ps-pulse laser quenching.^{22,26} Recent examples of exotic phases produced by lasers include a dense body-centered-cubic Al phase¹¹ and new tetragonal polymorphs of silicon¹² formed through ultrafast confined microexplosions generated by fs laser pulses tightly focused inside a sapphire ($\alpha\text{-Al}_2\text{O}_3$) crystal and at the interface between a transparent amorphous silicon dioxide layer (SiO_2) and an opaque crystalline Si substrate, respectively.

An unusual amorphous carbon phase featuring a large fraction of sp^3 -bonded carbon and a propensity for nano-/microdiamond nucleation has been generated by nanosecond laser treatment of amorphous carbon films.¹³

The laser-induced processes responsible for the generation of complex surface microstructures featuring ultrahigh densities of crystal defects are illustrated in **Figure 4**, where the results of large-scale MD simulations (Figure 4a–d) and experimental transmission electron microscopy characterization of the surface modified by fs laser irradiation (Figure 4e) are shown. The snapshots presented in Figure 4a illustrate how a complex interplay of fast laser melting, rapid cooling, resolidification, and the dynamic relaxation of laser-induced stresses in the surface region of a Ag target irradiated by a 100 fs laser pulse result in the formation of a subsurface porous region covered by a nanocrystalline surface layer.²³ The nanocrystalline layer with ultrafine grains (see distribution in Figure 4b) is produced by massive nucleation of crystallites triggered by a deep undercooling of the melted surface region down to $\leq 0.7T_m$ (T_m is the melting temperature) and features random crystallographic orientation of the nanograins (Figure 4b).

The enlarged atomic-scale view of the surface microstructure shown for one of the nanograins in Figure 4b reveals a high density of stacking faults, twins, and nanoscale-twinned structural elements with fivefold symmetry. The formation of similar pentagonal structural elements has also been observed in frozen nanospikes generated in a MD simulation performed at a higher laser fluence, in the spallation regime discussed in the next section.²⁴ Structural analysis of the Ag nanospike generated in the MD simulation (Figure 4c), reveals a remarkable variability of the structural motifs coexisting in the nanospike,



including a region of continuous network of pentagonal twinned structural elements arranged into a polyicosahedral structure.²⁴

The computational prediction of the formation of a thin nanocrystalline layer with high density of twins suggests that short pulse laser processing is a viable alternative to the severe plastic deformation⁵⁰ or pulsed electrodeposition⁵¹ techniques that are currently employed to yield maximum structural refinement and high strength of metallic materials. Moreover, the localization of the nanocrystalline structure within a thin surface layer of the target and the prominent presence of twin boundaries and pentagonal structural elements are two factors that may also increase the ductility of the laser-modified layer,^{52–54} a property that is generally incompatible with high strength.

Another example of unusual laser-generated defect configurations is the recent observation of nanoscale lamellar structures of interlayering twinned domains and epilayers in the surface regions of Ni(111) targets transiently melted and resolidified following exposure to fs laser pulses (Figure 4e).²⁵ The results have been explained by atomistic- and continuum-level simulations performed for the same system (Figure 4d). The simulations predict the onset of growth twinning at the resolidification front when the temperature drops below a certain critical level ($0.87T_m$ for the simulated Ni). The results of this study indicate that growth twinning can be used as a sensitive measure of the level of undercooling achieved in the transient laser-induced melting and resolidification of face-centered-cubic

metal targets/grains with $\{111\}$ surface orientation. In the absence of growth twinning, the structural signatures of the transient melting and resolidification (e.g., generation of a high vacancy concentration),⁵⁵ may be too weak to be detected by *ex situ* experimental characterization methods. In addition, the formation of a thin layer with high density of growth twins drastically increases the strength of the surface region.⁵⁶

Laser ablation: From photomechanical spallation to phase and Coulomb explosions

The process of laser-induced removal of material from an irradiated target, commonly called laser ablation, is at the core of many practical laser-enabled applications, including high-precision cutting and drilling, surface nanostructuring, pulsed laser deposition of films and coatings, and generation of nanoparticles with narrow size distributions and well-controlled compositions. Several mechanisms driving the collective ejection of material from the irradiated target are briefly outlined in the next section.

As mentioned, the fast rate of energy deposition by short laser pulses may lead to the generation of compressive stresses. The laser-generated stresses are particularly high in the regime of stress confinement,^{17,18} when the time of the laser heating (defined by the laser pulse duration, τ_p , or the time of the electron–phonon equilibration, τ_{e-ph} , whichever is longer) is shorter than the time required for the mechanical relaxation

(expansion) of the heated volume, in other words, $\max\{\tau_p, \tau_{e-ph}\} \leq L_p/C_s$, where C_s is the speed of sound in the target material and L_p is the effective depth of the laser energy deposition (optical penetration depth or, in the case of metals, the depth of diffusive/ballistic energy transport during the time of the electron–phonon equilibration).¹⁸

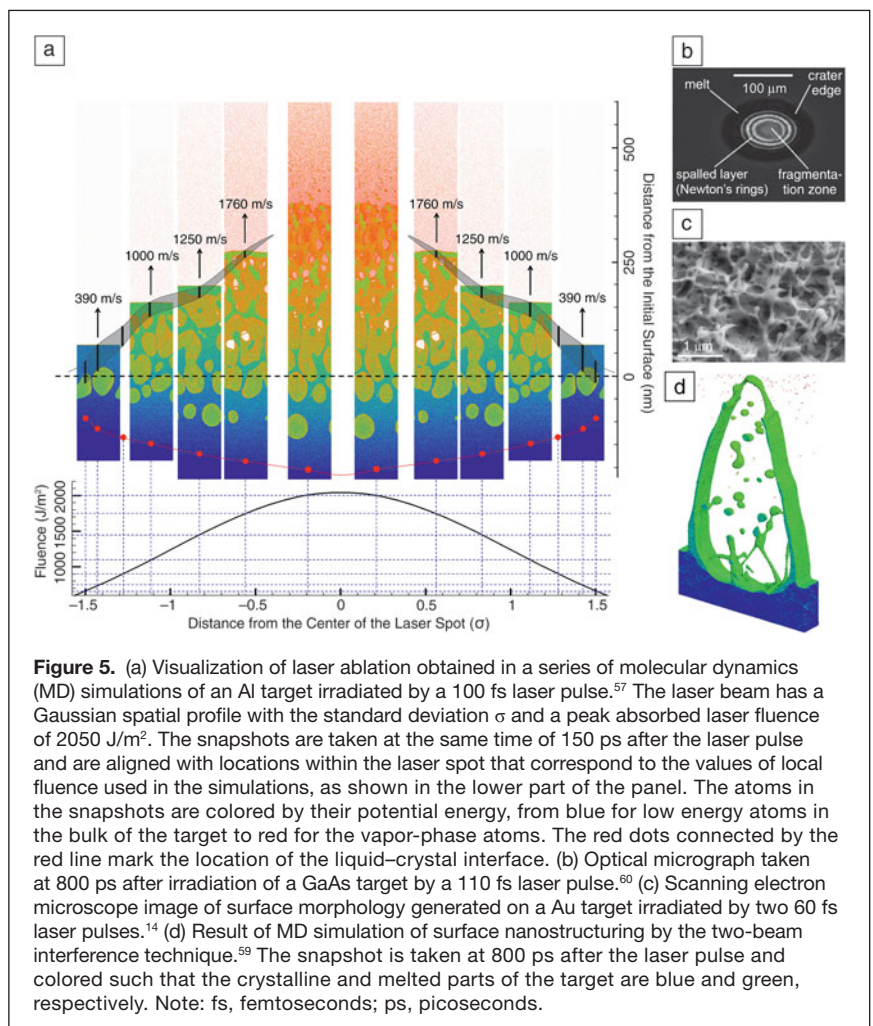
The interaction of the laser-induced compressive stresses with the free surface of the irradiated sample can result in the generation of a tensile wave sufficiently intense to cause the formation subsurface voids (Figure 4a). The growth and percolation of the voids may lead to the separation and ejection of a top layer from the target. Analogous to the term “spallation,” commonly used to describe the dynamic fracture that results from the reflection of a shock wave from the back surface of a sample, material ejection driven by the relaxation of laser-induced stresses is commonly referred to as photomechanical spallation (admittedly, “cavitation” would be a more appropriate term when the photomechanical processes take place in the melted part of the target).

Further increase in the laser fluence above the spallation threshold results in the separation and ejection of thinner layers and/or multiple droplets from the target, and, at a sufficiently high laser fluence, induces a transition to a different ablation regime commonly referred to as “phase explosion” or “explosive boiling.” In this regime, the melted surface region of the irradiated target is overheated above the limit of thermodynamic stability of the liquid phase,^{19–21} leading to a rapid decomposition of the overheated melted material into a mixture of vapor and liquid droplets. The transition from photomechanical spallation to phase explosion is signified by a sharp (threshold-like) increase in the amount of the vapor-phase atoms in the ejected plume,^{57,58} which reflects the difference in the physical mechanisms responsible for the material ejection in these two regimes. An explosive release of vapor, rather than the relaxation of photomechanical stresses, provides the main driving force for the collective ejection of the overheated surface region of the target in the regime of phase explosion. In the case of ablation by longer laser pulses, in the absence of stress confinement, spallation is not activated and explosive boiling is the mechanism responsible for the direct transition from the regime of surface melting and evaporation to the phase explosion.^{21,58}

Due to the spatial variation of laser intensity within laser beams that often have Gaussian profiles, photomechanical spallation and phase explosion may coexist and jointly contribute to the material ejection induced by the same laser pulse. This is illustrated by **Figure 5a**, where a “mosaic approach” combining the results

of simulations performed at different local laser fluences is used for representation of the laser-induced processes at the scale of the whole laser spot.⁵⁷ Two distinct sections can be easily identified within the laser spot: the central area, where the overheated top part of the surface region undergoes an explosive decomposition into vapor and liquid droplets, and the periphery of the spot, where spallation of well-defined liquid layers connected to the bulk of the target by transient foamy structures of interconnected liquid regions is observed. Although the liquid layers observed in individual simulations are flattened by the application of periodic boundary conditions, the integral picture of ablation by the Gaussian laser beam is likely to involve the peel off of a continuous liquid layer with its thickness decreasing toward the center of the laser spot, as shown schematically in Figure 5a by gray stripes and directly observed in Figure 5d, where a snapshot from a simulation of Au surface nanostructuring by periodically modulated energy deposition realized in two-beam interference experiments is shown.⁵⁹

Both spallation and phase explosion provide channels for effective transformation of the energy deposited by the laser irradiation into material disintegration and vaporization,



thus creating conditions for rapid cooling and freezing of the transient liquid structures generated in laser spallation or ablation.^{24,57} Indeed, the temperature of the liquid–crystal interface, shown in Figure 5a by the red line connecting red dots, drops down to the melting temperature of Al by ~600 ps, and the interface starts to move up, toward the surface. One can expect that the solidification process will freeze (either by epitaxial regrowth of the substrate or by rapid homogeneous nucleation of the solid phase under strong undercooling conditions) some of the evolving liquid structures, leading to the formation of a complex surface nanostructure,^{14–16} such as the one shown in Figure 5c.

The occurrence of both spallation and phase explosion processes within the same laser spot can also be related to the results of pump-probe experiments,^{10,60,61} where the observation of optical interference patterns (Newton's rings) can be explained by the spallation of a thin liquid layer from the irradiated target and the absence of interference fringes in the central part of the laser spot (Figure 5b) can be attributed to the transition to the phase explosion regime.

Another possible ablation mechanism intimately connected to the conditions of electronic nonequilibrium, discussed at the beginning of this article, is the electrostatic disintegration of a superficial layer of irradiated material caused by effective photo- and thermionic emission of electrons and associated positive charging of the material surface, the so-called Coulomb explosion (CE).⁷ This mechanism of ablation, which manifests itself as highly energetic ion emission, has been widely debated for different materials.^{62–65} The main feature of CE is the momentum scaling of differently charged ions and ions of different species in time-of-flight signals⁶² that suggests their emission and acceleration by the electric field.

The occurrence of CE is defined by the competition between the laser-induced electron photoemission leaving behind a positively charged surface layer of the material and the neutralization of the positive charge by the free electron current from the material bulk. It was proven theoretically that at fluences slightly above the ablation threshold, CE can be realized in wide-bandgap dielectrics while, in semiconductors and metals, high mobilities of charge carriers prevent accumulation of the positive charge up to the level sufficient for disintegration of a surface layer by electrostatic repulsive forces.⁷ When CE does occur, the exploded material leaves behind a smooth undamaged surface⁶² that makes this ablation mode highly attractive for nanoscale applications (e.g., ultrafine material polishing). There are indications of the possibility of CE in semiconductors at laser fluences above the plasma formation level.⁶³ However, at such conditions, the ions originating from the CE constitute a small fraction of the ablation products and are masked by the thermal ablation ions, including those accelerated by the plume double layer (two oppositely charged parallel layers created in the front of expanding plasmas due to faster electrons running ahead of slower ions).^{63,66} In thin metal wires exposed to ultraintense laser pulses, a substantial fraction of electrons is carried away

from the wire by the laser field, resulting in electrostatic disintegration of the ionic lattice left behind.⁶⁷

Concluding remarks

Rapid expansion of practical applications of ultrashort pulse lasers into new areas where precise nanoscale control of material modification is required can only be sustained with an improved fundamental understanding of laser–materials interactions. The ability to achieve high spatial resolution in material modification and microstructure engineering usually relies on fast and highly localized energy deposition that, unavoidably, creates conditions of strong electronic, thermodynamic, and mechanical nonequilibrium.

Theoretical and computational description of the materials behavior far from equilibrium is challenging and requires the revision of existing (or the development of new) theoretical models and computational approaches. In particular, the intricate connections between the ultrafast material response to laser excitation, generation of crystal defects, phase transformations, and mass transfer occurring under highly nonequilibrium conditions can only be accounted for by combining the research expertise and theoretical approaches from different disciplines. Close interdisciplinary collaborations and innovative design of new multiscale approaches incorporating dissimilar physical concepts and operating at different time and length scales are needed for disruptive progress in the theoretical and computational description of laser–materials interactions.

On the experimental side, the development of advanced imaging techniques capable of providing time-resolved information on the ultrafast structural and phase transformations is crucial for gaining fundamental understanding of laser-induced processes. Not unlike the theoretical efforts, the imaging/probing of different laser-induced processes requires a combination of optical pump-probe techniques with variable delay and temporal pulse-shaping, advanced diffractive imaging of transient atomic dynamics and ultrafast structural transformations, as well as the imaging/probing of ablation plume dynamics capable of tracking the spatial distribution and physical characteristics of different plume components.

Overall, investigations of ultrafast laser–materials interactions are expanding the limits of the fundamental understanding of material behavior under extreme conditions and providing opportunities for the generation of new material phases and microstructures.

Acknowledgments

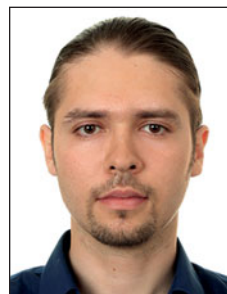
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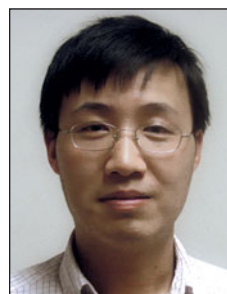
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