Looking at Femtosecond Laser-Induced Black Metals at Different Polarizations

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Recently, we demonstrated a femtosecond laser blackening technique that makes reflective metals highly absorptive, creating the so-called "black metals."[1,2] The essence of this metal blackening technique is to apply intense femtosecond laser pulses on a metal surface and create a variety of unique surface structures at nano- and micro-scales that dramatically enhance light coupling into the metal.[1,2]

Here, we address if the enhanced absorption has a polarization effect. To answer this question, we performed experiments using an amplified femtosecond Ti:sapphire laser system that generates about 66-fs pulses with the central wavelength of 800 nm.

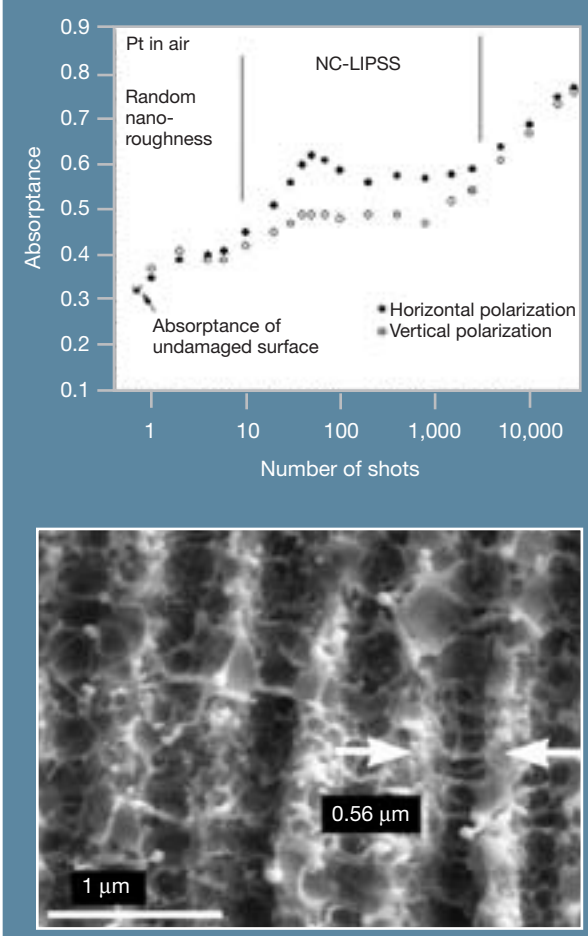
We used several metals, including platinum, tungsten and titanium. To blacken the metals, we polarized the laser beam horizontally and focused it onto a sample at normal incidence. Absorptance of the blackened spot was measured using a calorimetric technique.[1] After blackening the sample with a number of pulses, we reduced the incident laser fluence to a level much below the damage threshold and used this low fluence to measure absorptance change of the irradiated spot. For the probing beam, both horizontally and vertically polarized light were used to measure the absorptance change following laser blackening.[1]

The top portion of the figure shows the absorptance change of platinum as a function of the number of blackening pulses at a moderate laser fluence of 0.17 J/cm^2. Only partial blackness was obtained at this moderate laser fluence. However, there was an interesting section under these experimental conditions, where absorptance showed a clear polarization dependence when the number of incident pulses was within 20-1,500.[3]

To understand what causes this polarization effect, we studied the irradiated spots using scanning electron microscopy. We found that the polarization effect was caused by a unique type of nanostructure-covered laser-induced periodic surface structure (NC-LIPSS).[3] A typical image of NC-LIPSSs following 200-pulse treatment is shown in the bottom part of the figure.

Our study shows that absorptance is higher when light polarization is perpendicular to the grooves of NC-LIPSSs. This is due to the fact that the grating-like NC-LIPSSs effectively couple the incident light into a propagating surface electromagnetic wave in the form of surface plasmons when the light polarization is perpendicular to the NC-LIPSS grooves, providing an additional energy absorption channel. Furthermore, at this partially blackened region, the metals exhibit various colors at different viewing angles due to the grating effect.

The polarization effect demonstrated here suggests a controllable way to modify the optical properties of metals with femtosecond laser processing. The treated metals should find important applications in photonics, plasmonics, thermal radiation sources, thermophotovoltaics, surface-plasmon-polariton-based sensors and bio-optical devices.



(Top) Absorptance of platinum measured at different light polarizations as a function of the number of laser shots at a moderate fluence of 0.17 J/cm^2 at normal incidence. (Bottom) Image of NC-LIPSSs formed at the fluence of 0.17 J/cm^2 following 200 shots.

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References

1. A.Y. Vorobyev and C. Guo. Phys. Rev. B **72**, 195422 (2005).
2. P. Daukantas. Opt. Photon. News **18**(2), 8 (2007).
3. A.Y. Vorobyev and C. Guo. Appl. Phys. A (Rapid Comm.) **86**, 321-4 (2007).

Turning Aluminum Liquid in Picoseconds

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The interactions between the electrons and the lattice in solids define many basic properties of these materials. Exciting the electrons in a solid with intense ultrashort laser pulses is one way to induce non-equilibrium processes, observe the resulting dynamics and obtain direct information on electron-lattice interactions. An ultrashort laser pulse can rapidly heat electrons to temperatures on the order of 10^3 K, while leaving the lattice near room temperature.

This transient two-temperature system tends to reach quasi-equilibrium within a few picoseconds via electron-phonon interactions, as well as electron diffusion out of the excited region. For sufficiently intense laser pulses, structural changes such as melting and ablation take place. Thermal melting, which results from a rise of the lattice temperature above the material's melting point, can be readily observed with picosecond laser pulses.

After the development of femtosecond lasers, a non-thermal melting mechanism was reported in semiconductors. This mechanism occurs when the laser-induced electronic excitation rearranges the positions of the ions in a liquid-like disordered configuration before the lattice reaches the melting temperature. Non-thermal melting usually occurs within hundreds of femtoseconds after photoexcitation, in contrast to the picosecond time scale of thermal melting.

The first metal for which non-thermal melting was observed was aluminum. It was reported that the material's dielectric constant at 800 nm reaches the value for liquid aluminum 500 fs after excitation of solid aluminum with a femtosecond laser pulse, a time scale that is significantly shorter than the picosecond time scale for lattice thermalization.[1]

This conclusion was later called into dispute, after researchers conducted electron-diffraction experiments of optically excited, thin aluminum films and found a 3.5-ps time scale for the solid-to-liquid phase transition, indicating that it is a thermal process.[2]

We recently measured the reflectivity of aluminum during the laser-induced solid-to-liquid phase transition over a broad wavelength range, from 350 to 730 nm, with a time resolution of 65 fs.[3] The figure shows the response of the reflectivity at 590 nm for four laser intensities above the melting intensity I_m.

At negative times—that is, before the arrival of the excitation pulse—the reflectivity remained constant at its equilibrium value. At positive times, after the arrival of the excitation pulse, the reflectivity started to drop as the solid-to-liquid phase transition began. At all excitation intensities, it took 1.5-2 ps for the transition to be complete. We observed the duration for this transition over the entire range of wavelengths over which reflectivity was measured.

Our results unambiguously settle the argument: The phase transition in optically excited aluminum is thermal, and is mediated by heat transferred from the excited electronic population to the lattice through electron-phonon interactions. Only in semiconductors, where covalent bonding causes strong electron-lattice correlations, is it possible to observe non-thermal melting.

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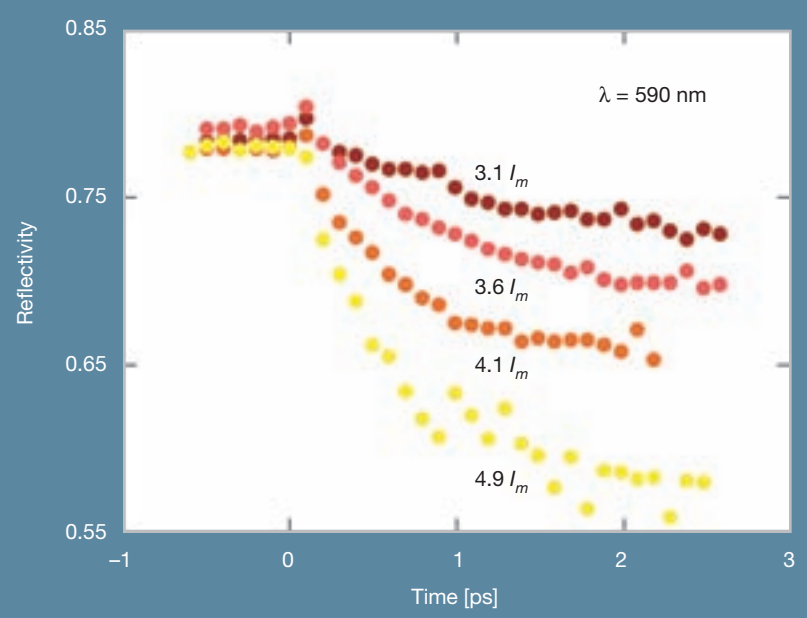
References

1. C. Guo et al. "Structural phase transition of aluminum induced by electronic excitation," Phys. Rev. Lett. **84**, 4493 (2000).
2. B.J. Siwick et al. "An atomic-level view of melting using femtosecond electron diffraction," Science **302**, 1382 (2003).
3. M. Kandyla et al. "Femtosecond dynamics of the laser-induced solid-to-liquid phase transition in aluminum," Phys. Rev. B **75**, 214107 (2007).



Time-resolved reflectivity dynamics of aluminum following single-pulse photoexcitation at 590 nm for different absorbed intensities of the excitation pulse.

Ablation of Sub-100-nm Features with a Tabletop Soft X-ray Laser

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Tightly focused laser beams can be used to locally ablate materials in order to analyze their composition and to pattern small devices. Ablation with femtosecond, mid-infrared and ultraviolet lasers has achieved a spatial resolution of 200 nm.[1,2] Soft X-ray light can extend these capabilities into the sub-100-nm range.

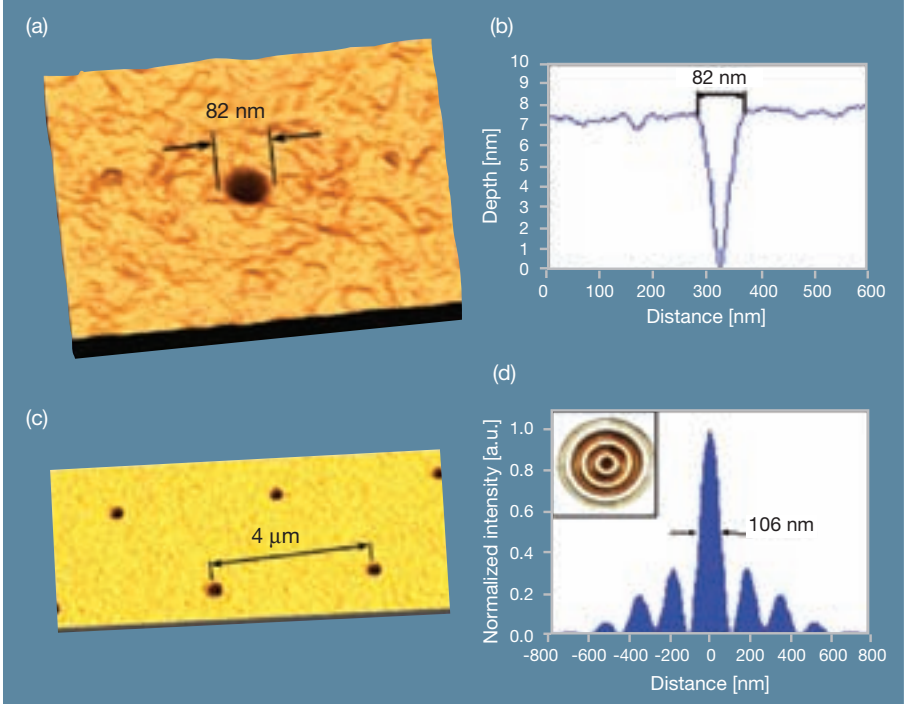
We recently demonstrated sub-100-nm laser ablation using the focused output from a compact soft X-ray laser.[3] We used a free-standing Fresnel zone plate (FZP)[5] to focus the collimated laser output from a 46.9 nm capillary discharge tabletop soft X-ray laser[4] onto a thin layer of poly-methyl methacrylate (PMMA) spun on top of a silicon wafer.

The laser produced roughly 0.1 mJ pulses of 1.2 ns duration, and was operated at a repetition rate of 1-2 Hz. The 10 percent efficiency FZP had a 0.5-mm diameter, an outermost zone width of 200 nm, and a numerical aperture of 0.12. When used in first-order diffraction, its Rayleigh-like spatial resolution at 46.9 nm wavelength is about 240 nm. The fluence onto the sample was controlled by introducing argon gas into the vacuum chamber to attenuate the laser beam.

We demonstrated the single shot ablation of 240-nm-diameter holes in PMMA by placing the sample in the first-order focus of the FZP and attenuating the laser beam 36×. The ablated holes showed very clean and smooth walls. Shot-to-shot reproducibility was excellent.

Smaller holes were obtained by placing the sample about 7.5 µm away from the third-order focus of the FZP. In accordance with simulations conducted using the Rayleigh-Sommerfeld diffraction integral, the intensity distribution at this location has a very narrow intensity peak, about 100 nm FWHM, which is surrounded by concentric rings of decreasing intensity. Attenuation of the intensity of the rings to values below the ablation threshold resulted in the ablation of 82-nm-diameter craters.

The high spatial resolution and small absorption depth of the ablation tool make it attractive for nano-patterning applications. This ablation technique will also enable development of surface nanoprobes based on techniques such as laser-induced breakdown spectroscopy.



(a) AFM image of the 82-nm-diameter craters obtained by placing the sample near the third-order focus of the Fresnel zone plate (FZP) and attenuating the laser intensity to a level such that only the central peak can ablate PMMA. (b) Cross-section of an 82-nm-diameter hole ablated in PMMA. (c) Atomic force microscope (AFM) image of a regular array of holes ablated in PMMA with the focused output from the 46.9 nm laser. (d) Calculated (blue) and measured (inset) laser intensity distribution at 7 µm from the FZP.

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References

1. P.P. Pronko et al. Opt.Comm. **14**(1-2), 106-10 (1995).
2. K. Chen et al. Appl. Phys. A **65**(4-5), 517-18 (1997).
3. G. Vaschenko et al. Opt. Lett. **31**(24), 3615-7 (2006).
4. D.R. Benware et al. Phys. Rev. Lett. **81**(26), 5804-7 (1998).
5. E.H. Anderson. IEEE J. Quantum Electron. **42**(1-2), 27-35 (2006).

Ultrafast Laser Microwelding

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In high-tech industries, welding on a micrometer scale has become vitally important. Laser microwelding is highly flexible, precise and fast and offers non-contact processing. However, in conventional laser microwelding, the welded substrates must be opaque, or one of the surfaces must be coated with an opaque material, because the beam penetrates the transparent substrate and is absorbed at the surface of the opaque material.

Previously, we have performed the ultashort laser microwelding technique for transparent materials without any intermediate layers by using a femtosecond laser system in a low-repetition-rate regime.[1,2,3] Now, we demonstrate the technique based on a localized heat accumulation effect in a high-repetition-rate regime.[4] The laser pulses act as a point source of heat at the focal volume within the bulk material when the time interval between successive pulses is much shorter than the time scale for the diffusion of heat out of the focal volume.

The accumulated energy around the focal volume makes it possible to achieve very high temperatures and melt the material around the focal volume.[5] The melted material, called a liquid pool, is created at the interface and fills the original gap between the materials. The liquid pool is then resolidified, thus joining the two materials. When the gap between samples is present, the samples could not be joined because of the ablation at the surface of each sample.

We performed the ultrashort laser microwelding of non-alkali alumino silicate glass substrates based on a localized heat accumulation effect using an amplified femtosecond Er-fiber laser (500 kHz, 947 fs).[4] The experiment was performed at room temperature. We stacked two samples and pressed them together. When the laser pulses were focused on the interface between the two samples, the filamentary propagation of fs laser pulses bridged the two substrates along the laser propagation axis. The focal region was translated two-dimensionally (perpendicular to the optical axis).

We formed the 3 × 3 array of joint volumes (100 µm × 100 µm × 25 µm) by irradiating fs laser pulses with a pulse energy of 0.8 µJ and a translation rate of 200 µm/s. We also welded a non-alkali glass substrate and a silicon substrate. The wavelength of the laser system was 1,558 nm where silicon and borolicate glass are transparent.

This result is an important step toward the welding of semiconductor materials. In addition, ultrafast laser microwelding is a versatile tool for joining dissimilar materials. It opens up possibilities in the production of electronic, electromechanical and medical devices. ◭

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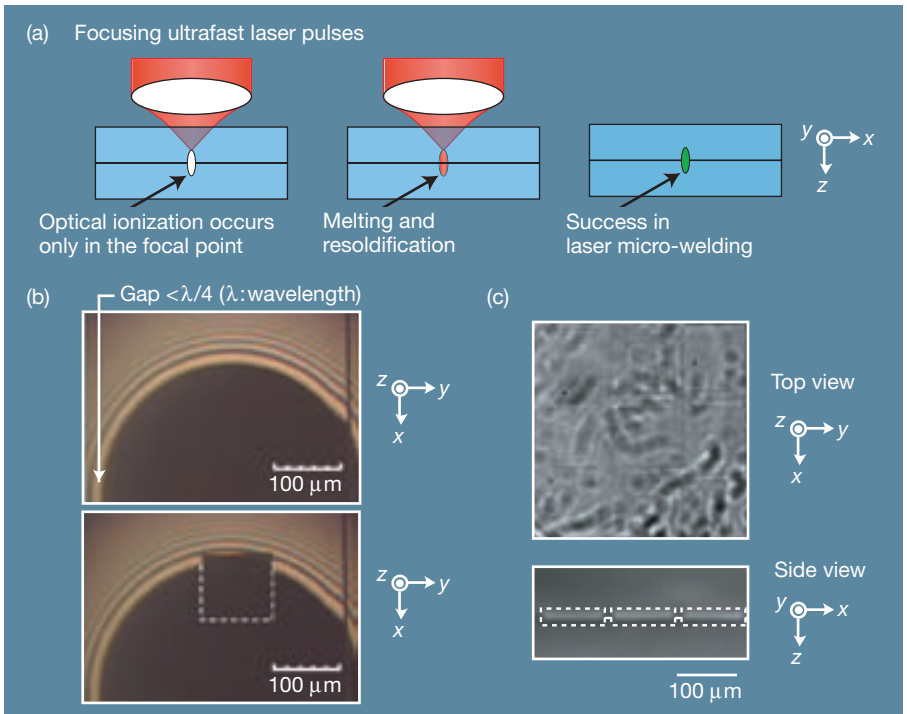
References

1. T. Tamaki et al. Jpn. J. Appl. Phys. Part 2, **44**, L687-9 (2005).
2. W. Watanabe et al. Appl. Phys. Lett. **89**, 021106 (2006).
3. W. Watanabe et al. Appl. Phys. B, **87**, 85-9 (2007).
4. T. Tamaki et al. Opt. Express **14**, 10460-8 (2006).
5. K. Itoh et al. MRS Bulletin **31**, 620-5 (2006).



(a) When an ultrashort laser pulse is focused in the interface of two substrates, optical ionization occurs only around the focal volume. Nonlinear absorption results in the creation of an electron-ion plasma. Subsequently, localized melting and quenching occur at the interface. Resolidification results in the samples being joined. (b) White-light fringes of welded samples, silica-glass substrate and lens, before (top) and after (bottom) irradiation by fs laser pulses. The incident pulse energy was 1.0 µJ and the translation speed of the filament was 5 µm/s. By irradiation of the pulses, a bright area was changed to a black area because a one-quarter-wavelength gap was almost filled. In other words, the microwelding was successful. (c) Optical images of the welding volumes produced at the scan speed of 20 µm/s. The dashed line shows the welding volumes.