Damage in a Thin Metal Film by High-Power Terahertz Radiation

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We report on the experimental observation of high-power terahertz-radiation-induced damage in a thin aluminum film with a thickness less than a terahertz skin depth. Damage in a thin metal film produced by a single terahertz pulse is observed for the first time. The damage mechanism induced by a single terahertz pulse could be attributed to thermal expansion of the film causing debonding of the film from the substrate, film cracking, and ablation. The damage pattern induced by multiple terahertz pulses at fluences below the damage threshold is quite different from that observed in single-pulse experiments. The observed damage pattern resembles an array of microcracks elongated perpendicular to the in-plane field direction. A mechanism related to microcracks' generation and based on a new phenomenon of electrostriction in thin metal films is proposed.

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Introduction.—The ongoing progress in terahertz technology has resulted in the generation of intense coherent sub-picosecond pulses of terahertz radiation. Increasing attention has been drawn towards the use of so-called intense terahertz transients as a powerful tool in basic science and applied research, including leading-edge optoelectronic technologies [1–3]. The highest electric fields (up to 100 MV/cm) in the 0.1–10 THz range have been effectively generated using optical rectification of terawatt near-infrared femtosecond laser pulses in organic crystals [4,5]. At present, such laser-based sources of high-field terahertz pulses could pave the way to a new research area of strong-field-induced effects in matter causing changes of crystalline lattice, ultrafast phase transitions, modification of a surface layer, damage, formation of through holes, and craters in various materials [6].

The first experimental studies of laser-matter interaction carried out with femtosecond laser pulses in the optical and x-ray regions of the spectrum were aimed at observational studying of surface-layer modifications and damages [7,8]. As a result, the first theories related to mechanisms of radiation absorption and damage formation were developed [9].

Recently, there have been very few studies related to similar phenomena induced by ultrafast terahertz transients. Irreversible damage in a thin film of vanadium dioxide has been observed at terahertz electric fields of 4 MV/cm [10]. At a fluence of terahertz radiation above 89 mJ/cm^2, there has been experimentally observed modification and damage of thin metal films [11,12]. However, the damage mechanisms in these works have not been analyzed.

We present the experimental data on damage behavior in aluminum films with a thickness less than a terahertz skin-depth layer after irradiation with single and multiple THz pulses at fluences up to 300 mJ/cm^2. Similar experiments with bulk metal require very high radiation fluences, since the reflection coefficient in the 1–2 THz range is about 0.99 [13]. Owing to electron heat conductivity during a picosecond pulse, one should consider the heating of a surface layer that is much thicker than the skin-depth layer. Therefore, thin aluminum films of 20 nm in thickness (a skin-depth layer at 1–2 THz is about 30–60 nm [13,14]) were used in the experiments.

In this study, for the first time, we managed to induce damage (a kind of a through hole) to a thin aluminum film by a single terahertz pulse and measure its fluence threshold. The damage pattern induced by multiple terahertz pulses at fluences below the single-pulse damage threshold is quite different from that observed in single-pulse experiments. The experimental data were qualitatively analyzed, suggesting the damage mechanisms caused by single and multiple terahertz pulse irradiation.

Experimental setup.—The experimental layout, the terahertz electric field waveform, and its spectrum are shown in Fig. 1. Linearly polarized terahertz radiation was tightly focused on the sample surface at normal incidence.

Terahertz pulses were generated by optical rectification in a $400-\mu$m-thick organic nonlinear crystal [4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate (DSTMS)] [5] of 8 mm in diameter pumped by 100 fs laser pulses at a central

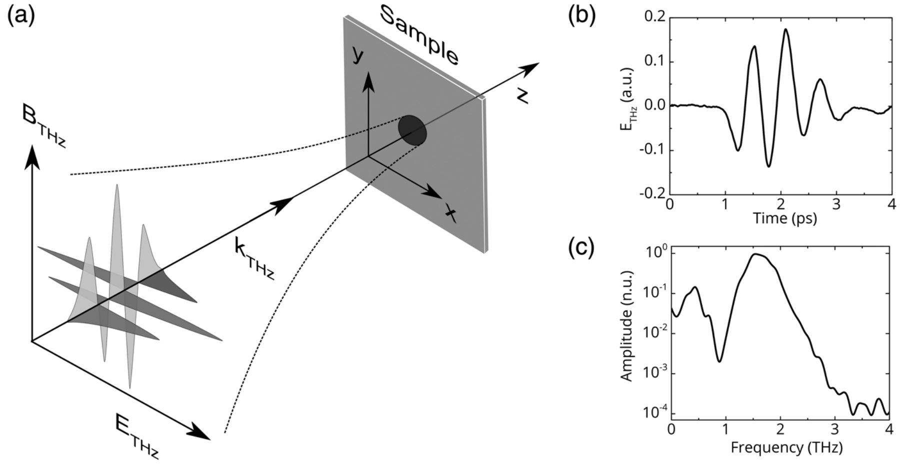

FIG. 1. (a) Experimental layout. (b) The terahertz electric field waveform in the time domain and (c) its Fourier-transformed spectrum. The electric field waveform and spectrum are obtained in free-space electro-optical sampling (EOS) carried out in a gallium phosphide crystal (GaP).

wavelength of 1240 nm delivered by an amplified Cr: forsterite laser [4]. A terahertz low-pass filter (LPF 8.8-47, Tydex) was used for cutting wavelengths below 34 μm. The attenuation coefficient of the terahertz low-pass filter at a wavelength of 1240 nm is no less than 10^5. A telescope 6∶1, consisting of two off-axis parabolic mirrors with reflected focus lengths of 25.4 and 152.4 mm, was used to compensate terahertz beam divergence. The terahertz beam was focused to a diffraction-limited spot size using an off-axis parabolic mirror with a focus length of 50.8 mm and a diameter of 50.8 mm. Terahertz pulse energy was measured by means of a calibrated optoacoustic detector (Golay cell, GC-1D Tydex). A polarized attenuator placed into the beam of a pump laser was used to adjust the energy of terahertz pulses. The terahertz electric field waveform and spectrum are obtained in electro-optical sampling. A nonlinear bilayer 2.1-mm-thick GaP crystal with a 2-mm-thick GaP (100) and a 0.1-mm-thick GaP (110) was used as an electro-optical crystal.

The optical rectification process provides a terahertz output of 76 μJ at a pump fluence of 20 mJ/cm^2. The terahertz electric field strength of 15 MV/cm has been estimated from the terahertz pulse energy, duration, and beam radius, assuming a Gaussian pulse shape. The terahertz electric field strength was derived from the equation

$$E_{\mathrm{THz}}\left[\frac{\mathrm{V}}{\mathrm{cm}}\right] = 27.45\sqrt{I_{\mathrm{THz}}\left[\frac{\mathrm{W}}{\mathrm{cm}^2}\right]}, \tag{1}$$

where I_{THz} is the terahertz pulse intensity:

$$I_{\mathrm{THz}} = \frac{4\sqrt{\ln 2}}{\pi\sqrt{\pi}}\frac{W_{\mathrm{THz}}}{w^2\tau_{\mathrm{FWHM}}}, \tag{2}$$

where W_{THz} is the terahertz pulse energy, w is the terahertz beam waist at a level of $1/e^2$, and τ_{FWHM} is the full-width at half-maximum terahertz pulse duration.

The experiments were carried out in an enclosed housing with relative air humidity less than 2%. The sample was an aluminum film deposited on a polished glass substrate 180 μm thick (a coverslip) using magnetron sputtering at a rate of 1 nm/s in an argon atmosphere (purity of 99.999%) at a pressure of 5×10^{-2} bar. The thickness of the magnetron-sputtered film was 20 nm. The study of surface morphology using atomic force microscope and scanning electron microscope (SEM) revealed a surface roughness (R_a) of 4.5 nm.

Results.—Single-shot damage of aluminum film:For the first time, the experiment on single-terahertz-pulse-induced damage in a thin aluminum film was a success. SEM images of film damages at different fluences F of a terahertz pulse are presented in Fig. 2.

SEM images of aluminum film damages induced by single terahertz pulses at fluences slightly below the damage threshold are shown in Figs. 2(a)–2(c). The SEM images clearly show delamination and cracking of the film along with its surface modification. At higher fluences, the film is completely removed at the center, forming a through hole, as shown in Figs. 2(d)–2(f). However, at the edges of the removed film, there is a distinctive feature presented by a rim of 1–2 μm in width that looks as if the film edges were wrapped. Around the rim, there is a modified surface wherein surface roughness is greater compared to the unirradiated surface.

The single-pulse damage threshold of the film was measured using a standard technique [15] where the squared radius of damaged regions (produced through holes) is plotted versus the logarithm of the energy of incident THz pulses (Fig. 3).

The experimental data are well approximated with a linear function indicating a Gaussian profile of the terahertz

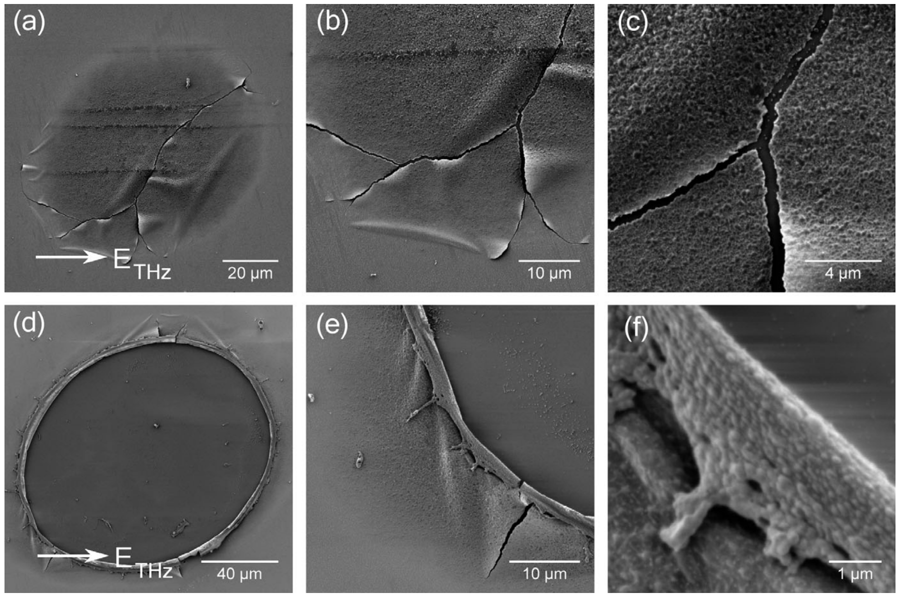

FIG. 2. SEM images of through holes and their edges in aluminum film produced by single terahertz pulses: (a)–(c) $F = 140$ mJ/cm^2; (d)–(f) $F = 300$ mJ/cm^2. The arrows indicate the direction of the terahertz electric field.

beam and high homogeneity of the used metal film. The measured radius at a level of $1/e$ was of $r_0 \approx 90$ μm, which gives a single-pulse damage threshold of the incident fluence of $F_a \approx 150 \pm 10$ mJ/cm^2.

Multiple-shot damage of aluminum film:SEM images of damages induced by multiple terahertz pulses at different fluences and numbers of pulses are shown in Fig. 4. The results were obtained at a pulse repetition rate of 10 Hz.

The observed damages occur at a fluence over 10 times less compared to that of the single-pulse damage threshold F_a. The damages have the form of elongated surface

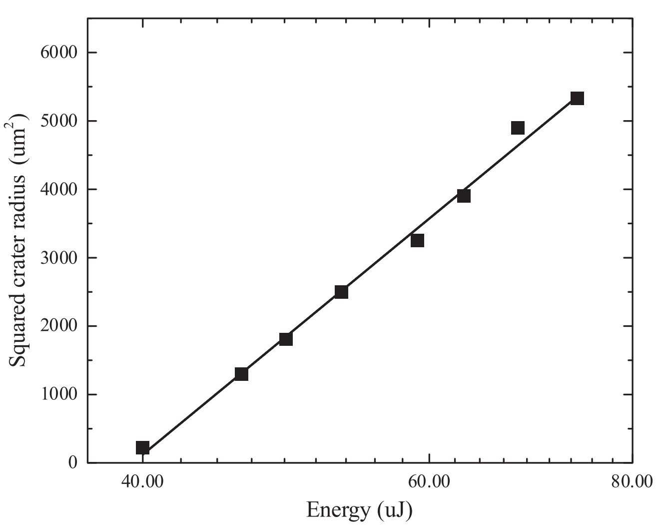

FIG. 3. Determination of the damage threshold in aluminum film irradiated with a single terahertz pulse; experimental data are shown with points approximated with a linear function (black solid line).

discontinuities (channels) that are perpendicular to the electric field vector of the terahertz pulse. The length of channels increases with a number of pulses; however, the width of channels decreases with increasing distance from the center to the periphery. The maximum width of the channels is about 10 μm at the center and decreases up to several hundreds of nanometers at the periphery.

For comparison, similar studies of damage were carried out on the same aluminum thin film using subpicosecond laser pulses. The source of radiation was a Ti:S laser that generates pulses of 400 fs duration at a wavelength of 800 nm. The *s*-polarized laser beam was focused on the sample's surface at an angle of incidence of 45° in a spot of $r_{0x} \approx 70$ μm and $r_{0y} \approx 40$ μm at the $1/e$ level with Gaussian spatial distribution. The experimentally measured damage threshold of the aluminum film in this case was $F_a^L = 50 \pm 5$ mJ/cm^2.

Single-pulse laser irradiation with a fluence above the threshold F_a^L resulted in a damage pattern of the aluminum film [Fig. 5(a)] similar to that observed under the action of a single terahertz pulse with $F > F_a$ [Fig. 2(d)].

At the same time, the damage pattern after multiple laser and terahertz exposures is distinctly different. Figure 5(b) shows the morphology of the surface exposed to the irradiation of *s*-polarized laser pulses ($N = 100$) with a fluence of $F \approx F_a^L$. In contrast to the irradiation with terahertz pulses, any damage pattern in the form of channels or cracks oriented relative to the polarization of laser radiation was not observed under similar experimental conditions.

Discussion.—We suppose the observed single-terahertz-pulse-driven damages (through holes) in thin metal film to be formed as a result of absorption of the terahertz pulse

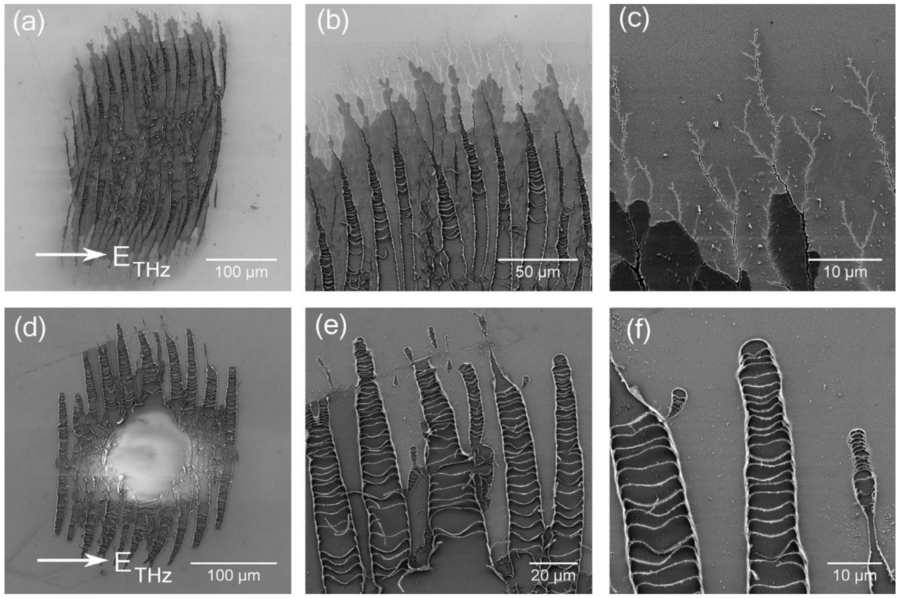

FIG. 4. SEM images of aluminum film damages induced by multiple terahertz pulses at different fluences: (a)–(c) $F = 80$ mJ/cm^2, $N = 600$; (d)–(f) $F = 240$ mJ/cm^2, $N = 60$. Here F is the fluence at the center of the focal spot, and N is the number of pulses. The arrows indicate the direction of the terahertz electric field.

energy by conduction electrons with the following generation of hot electrons that transfer energy to the lattice and heat it. Depending on the fluence, the lattice heating after the terahertz pulse could result in film damage due to thermal expansion or melting and ablation processes.

After study of the SEM images at threshold fluences [Figs. 2(a)–2(c)], we suppose thermal expansion of the heated film to be the cause of the observed damages. Due to differences in acoustical impedances at film boundaries, the film center of mass gains momentum in the direction from the substrate. Besides this, a tensile stress wave results in adhesive debonding [16]. Delamination of the film occurs when exceeding the adhesion strength. With increasing fluence of the terahertz pulse, the film expands into free space and breaks down with the formation of a roll-like rim at the edges of the through hole.

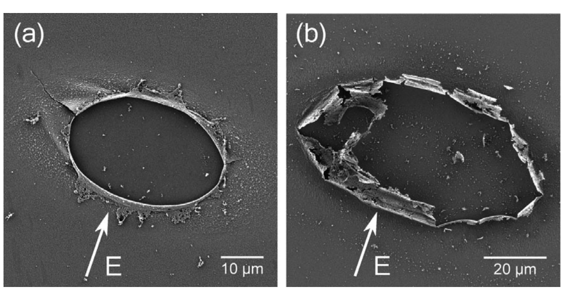

FIG. 5. SEM images of damage in aluminum film produced by (a) single and (b) multiple laser pulses: (a) $F = 60$ mJ/cm^2, $N = 1$; (b) $F = 50$ mJ/cm^2, $N = 100$. The vector of the electric field of the incident laser beam E is oriented along a small axis of the focal spot (white arrow). N is the number of pulses.

The internal pressure in the film heated to its melting point could be roughly estimated to be ∼1 GPa using the following relation: $P \approx \Gamma c \rho (T_m - T_0)$, where $\Gamma = 2$ is the Grüneisen parameter for aluminum; $c = 880$ J/(kg K) and $\rho = 2700$ kg/m^3 are the heat capacity and aluminum density, correspondingly; $T_m = 930$ K is the melting temperature of aluminum; and $T_0 = 300$ K is the initial temperature. The adhesion strength for a metal film measured by a tearing-off method is about ∼100 MPa [17]. Hence, it follows that at temperatures below the melting point, delamination and cracking of the film could occur.

The damage behavior of aluminum film irradiated with multiple terahertz pulses at fluences below the single-pulse damage threshold is rather challenging to explain. We suppose that the terahertz pulse at the center of the focused spot (a region with the peak electric field) generates a microcrack as a result of induced stress. It might be supposed that for the subsequent terahertz pulses there is a local field enhancement in the gap of the induced microcrack. This results in a microcrack growth perpendicular to the electric field vector and an increase in width in the across-track direction. The microcrack growth occurs along with film ablation in microregions similar to the mechanism of the single-pulse damage.

The mechanism of microcrack generation seems to be the most intriguing. The spatial period of damages induced by multiple terahertz pulses can be related neither to a radiation wavelength nor to a sound wavelength at the radiation frequency. That is why we think the periodicity of damages is not the result of resonance effects but can be attributed to the critical stress of damage under an ultrafast dynamical load.

We suppose the microcrack generation to be based on the electrostriction phenomenon that has not been previously observed in thin metal films. During two-cycle terahertz pulse irradiation, the applied electric field should completely penetrate through the film and causes an electric current. The current density is given by Ohm's law $\vec{j} = \sigma\vec{E}$ with a dc conductivity σ. The applicability of Ohm's law with static conductivity requires the fulfillment of the inequality $\omega\tau_c \ll 1$, where ω and τ_c are the frequency of the electric field oscillations and the time of electron-lattice collisions, respectively. In our case, this condition is easily satisfied, since $\omega \sim 10^{12}$ s^{-1} and $\tau_c \sim 10^{-14}$ s.

An estimate of charge density derived from a continuity equation $\partial\rho/\partial t + div\vec{j} = 0$ gives $\rho \sim (\sigma E\tau/r_0)$, where $E = |\vec{E}|$, and τ is a time comparable to the terahertz pulse duration. The force per unit volume acting on the charge by electric field $\vec{E}$ could be determined as $\vec{F} \sim (\sigma\tau/r_0)E\vec{E}$. A stress that is parallel to the electric-field vector of the terahertz pulse arises according to $\Sigma \sim (\Lambda/r_0)\sigma\tau E^2$, where Λ is an order of magnitude of distance between adjacent linear damages of the film that are perpendicular to the field. Assuming $\Lambda/r_0 \sim 10^{-2}$, $\tau \sim 10^{-12}$ s, $E \sim 10^4$ CGSE ($\sim$3 MV/cm), and taking into account that the aluminum conductivity is $\sigma \sim 10^{17}$ s^{-1}, we get an estimate $\Sigma \sim 10^4$ MPa that is bigger than the breaking stress for aluminum [18,19].

Conclusion.—In conclusion, the complete destruction of a thin metal film irradiated with a single 700 fs terahertz pulse has been observed for the first time and has resulted in ablation and through holes. The measured damage (ablation) threshold of the film was 150 mJ/cm^2 at the center of the focal spot. Delamination and film cracking was observed at fluences near the damage threshold. The study shows that the mechanism of aluminum film damage produced by a single terahertz pulse could be attributed to the thermal expansion of the film resulting in exceeding the adhesion strength with the substrate surface, cracking and ablation. The damage behavior induced by multiple terahertz pulses at fluences below the damage threshold is quite different from that observed in the single-pulse experiments. The damages are presented by a group of cracks elongated perpendicular to the in-plane field direction. A mechanism related to the generation of such microcracks and based on a new phenomenon of electrostriction in thin metal films is proposed. For the development of a terahertz-driven destruction model, future experimental and theoretical studies need to be carried out.

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