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Ultrasound generated by a femtosecond and a picosecond laser pulse near the ablation threshold

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We have investigated high-frequency ultrasound generated by single laser pulses in thin ($50\text{ }\mu\text{m}$) aluminum foils as a function of the laser fluence. Laser-pulse durations of 80 fs and 270 ps were used to compare the ultrasound generated in two very different regimes: thermoelastic and ablation. The measured rear-surface displacement induced by the ultrasound pulse is similar after $50\text{-}\mu\text{m}$ propagation through the foils for the two laser-pulse durations in the fluence range of $0.1\text{--}0.7\text{ J/cm}^2$. For fluences greater than the ablation threshold (0.25 and 0.63 J/cm^2 for the 80-fs and 270-ps pulses, respectively), the ultrasound amplitude generated by the 270-ps laser pulse is increased significantly due to absorption of laser energy by the ablating plasma. This is not observed for the 80-fs laser pulse since ablation is produced well after the laser-pulse irradiation of the target. The measured surface displacement as a function of laser fluence is compared to the calculations of a one-dimensional fluid code for both laser-pulse durations. The model calculations agree in many ways with the experimental results, but some discrepancies are observed. © 2005 American Institute of Physics. [DOI: 10.1063/1.1999827]

I. INTRODUCTION

Because of its simplicity, low cost, and accuracy, high-frequency ultrasound find important industrial applications in the measurement of thickness of thin coatings. Although there are many ways to generate high-frequency ultrasound (acoustic microscopes, surface acoustic wave devices, thin piezoelectric film transducers), none of these technologies offer the unique combination of advantages of laser ultrasonics: noncontact with large standoff distances, ability to work on hot or moving parts, ability to work on parts with complex shapes, and ultrawide bandwidth.¹

Early models and experiments reviewed in Refs. 2–4 showed that there are two main regimes for laser generation of ultrasound in a solid: the thermoelastic and ablative regimes. The thermoelastic regime is characterized by surface heating due to laser absorption with an ensuing rapid local thermal expansion, which launches the ultrasound wave. The ablative regime occurs at fluences above some threshold (called the ablation threshold) for which partial ablation of the sample surface is achieved. The acoustic pulse is produced partly by thermal expansion and partly by momentum transfer from the ejected matter (a plasma) to the sample surface.

The thermoelastic and ablative regimes have been extensively studied both experimentally and theoretically by means of models involving various assumptions. As a result, in the thermoelastic regime, the relationship between the laser pulse and the acoustic pulse is now well understood. In that regime, the ultrasound amplitude is proportional to the

absorbed thermal energy, and the high-frequency limit depends on the light penetration depth and the Fourier transform of the light pulse.⁵ In the ablation regime, however, experimental and modeling efforts have been more recent.^{6–15} Although it is well known that the ablation mechanisms depend strongly on the laser-pulse duration (see, for example, Refs. 16 and 17), the investigations on ultrasound generation in the ablative regime essentially deal with long laser pulses (typically in the nanosecond range). For long laser pulses, the end of the pulse interacts with the matter ablated by the front of the pulse. This induces plasma heating, which increases the amplitude of the ultrasound launched inside the solid as a consequence of the additional pressure applied on the material's surface. On the contrary, sufficiently short laser pulses (typically in the subpicosecond range) do not have enough time to build up a plasma that interacts with the light pulse. Moreover, since less thermal energy is dissipated in the target, ablation is more efficient for short laser pulses at a given fluence. Therefore, one expects significant differences in the ultrasound generated by ablation with long and short laser pulses in the ablative regime.

The main goal of this paper is to investigate, by means of experiments and modeling, the influence of the ablation threshold on the ultrasound amplitude in two regimes of laser-pulse duration for which the ablation mechanisms are expected to be different. For that purpose we present a comparison of ultrasound amplitudes as a function of laser fluence generated with a (relatively) long laser pulse of 270 ps and with an ultrashort laser pulse of 80 fs in $50\text{-}\mu\text{m}$ -thick aluminum samples. It is worth pointing out that our previous investigations clearly indicate that all the features of ablation

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that characterize long pulses should hold for a 270-ps pulse since the long-pulse regimes start to be effective for pulse durations near 10 ps in the case of aluminum targets.^{16,17}

In our experiments the ablation thresholds were determined by measuring the broadband white light emission produced at the sample surface by the ablated matter. Ultrasound amplitudes were characterized by the surface displacements of the foil's rear side, measured by means of a high-frequency interferometry technique.^{18–20} This technique presents the considerable advantage over the more usual and higher-frequency pump-probe piezoreflectance detection technique^{21–26} that the time dependence of the rear surface displacement can be measured with a single laser shot. Oppositely, the pump-probe piezoreflectance technique requires as many laser shots as there are measured times. Multiple laser shots induce particular difficulties in ensuring the reproducibility of the measurements in the ablation regime. Moreover, estimations indicate that interferometry should be more sensitive for ultrasound measurements below about 10 GHz,²⁷ which is well beyond the limit of 800 MHz for the optimal response of the Fabry-Perot interferometer used in our experiments.²⁰

Measured ultrasound amplitudes were compared to the predictions of a one-dimensional fluid code to help determine the specific nature of ultrasound generation in these two regimes of laser-pulse duration. This code, used in previous work,^{17,28–30} provides a consistent description of the absorption of the laser energy in the target and in the plasma, of the ablation process, and of the generation of acoustic pulses in the solid.

This article is structured as follows: In Sec. II, we present the experimental configurations for the measurements of the ablation (plasma formation) threshold and of the surface displacement associated with the laser-generated ultrasound wave. The experimental measurements and their interpretation are presented in Sec. III. Section IV presents a comparison of these measurements with the calculations of surface displacement performed using a numerical model. Finally, a summary of the main results is presented in the Summary and Conclusion.

II. EXPERIMENTAL DETAILS

The ultrasound generation laser pulses were produced using the INRS-EMT 10-TW Ti:sapphire laser system at a wavelength of 800 nm. This laser system can produce pulses from 60 fs to 3 ps in duration at energies of up to 600 mJ as well as uncompressed pulses of 270 ps of up to 1 J of energy. Pulse durations of 80 fs and 270 ps with energies of up to 30 mJ were used for these experiments. One notes that the energy of the prepulse (i.e., a longer low-energy pulse occurring before the main short laser pulse) is more than 10^6 times smaller than the energy of the main pulse, and thus plays a negligible role in our experiments. Each measurement reported here was performed using a single laser pulse on a clean and fresh target surface because it was observed that successive pulses at the same location did not produce repetitive results. The energy of each pulse was measured with a microjoulemeter.

FIG. 1. Schematic of the experimental setup used for ablation threshold measurements.

The Al foil used for these measurements has 99.99% purity and was cleaned with methanol. The foil is approximately 50- μm thick and mounted on a 3-in. diameter ring such that the only mechanical constraints are at the edges. Samples from the same roll were used for the ablation threshold and surface displacement measurements. The dull side faced the ablation laser, while the more reflective side was used to increase the amount of light entering the surface displacement measurement system. This system was made of a continuous infrared laser and a 1-m confocal Fabry-Perot interferometer.¹⁸

A. Measurement of the ablation thresholds

As mentioned above, experiments performed with long (nanosecond) pulses indicate that there are two main regimes for ultrasound generation: the thermoelastic and ablation regimes. The transition between these two regimes occurs at the ablation threshold, which is usually defined in terms of fluence, i.e., the incident laser energy divided by the surface containing this energy.^{3,4} This threshold varies as a function of the laser-pulse duration.³¹ The ablation thresholds of Al samples used in the present ultrasound experiments were determined by measuring the broadband white light emitted from the sample surface, indicative of plasma generation and significant ejection of matter. The experimental layout for these measurements is shown in Fig. 1. The light emitted after laser impact from the sample surface was detected by a DET 110 Si PIN [ThorLabs] that has a spectral response of 320–1100 nm. A BG-39 blue filter (2-mm thick) was used to block the generation laser wavelength. This filter attenuates the 800-nm laser wavelength by a factor in excess of 10^4 and transmits with better than 80% efficiency in the 350–650-nm range. The detector was positioned at an approximate angle of 30° to the normal of the sample surface and 150 mm away from the generation spot. The intensity of light emission was acquired with an oscilloscope.

In these measurements, it is important to evaluate the fluence of laser pulses characterized by a Gaussian-like intensity profile in time and space. The value that is usually used is the average fluence, defined as the total laser energy divided by the area corresponding to the full width at half maximum (FWHM) diameter of the beam. This definition of fluence is acceptable when the plasma is produced over a

FIG. 2. Schematic of the experimental setup used for ultrasound generation and rear-surface displacement measurements.

relatively large fraction of the beam diameter, but is not very meaningful for determining the ablation threshold since, for increasing values of energy, plasma formation begins at the center (peak) of the pulse where the fluence is maximum. Thus, to determine the ablation threshold, it is more appropriate to use the fluence near the peak of the spatial distribution. The definition of fluence adopted here is 90% of the maximum fluence. To this value corresponds a small circular region around the center of the pulse, which will be referred to as the “flat-top” region. For a Gaussian profile, this definition of fluence corresponds to 66% of the average fluence. (That the average fluence is higher than that at the center results from the fact that all the laser energy is considered in the average fluence.) To evaluate this fluence, the size of the pulse was determined by fitting the measured beam spatial distribution to a Gaussian profile. The spatial energy profile of the laser beam was acquired with a Spiricon camera and LBA PC 32 software. The generation laser pulse used for these experiments was focused on the sample surface using a 2-m focal-length lens. The corresponding radius of the flat-top region used for defining the fluence is $152 \pm 7 \mu\text{m}$.

B. Generation and detection of ultrasound

As mentioned above, the irradiation of a sample surface by a laser beam induces a localized compression (positive stress) wave front that propagates into the sample. This compression wave is attenuated during propagation through the sample and reflected at the sample's free surface, returning as a rarefaction wave (negative longitudinal stress or tension).

Ultrasound generation and detection are performed on opposite sides of the foil to avoid interference in the surface displacement measurement between the detection laser pulse and the matter ablated by the generation laser pulse. The experimental layout of laser generation and detection of surface displacements is shown in Fig. 2. The ultrasound detection system comprises three main parts: a detection laser, a Fabry-Perot interferometer, and a photodiode detector. The detection laser is a LightWave Electronics Nd: yttrium aluminum garnet (YAG) laser, which produces 500 mW of continuous optical power at a wavelength of 1064 nm and is very frequency-stable. Surface displacements produced by the ultrasound wave can be detected through a phase modu-

FIG. 3. Measurements of plasma light-emission intensity as a function of laser fluence for an 80-fs (a) and a 270-ps (b) laser pulse on a 50- μm -thick Al foil target. The ablation threshold is indicated by the dashed lines.

lation of the detection laser beam induced by reflecting the beam on the sample surface.^{18,32} A half-wave plate and a beam-splitting cube are used to redirect the detection laser beam toward the Fabry-Perot interferometer after reflection on the sample surface. Phase modulation of the laser beam is converted to intensity modulation by the interferometer operating in a reflection mode. A quarter-wave plate and a beam-splitting cube then direct the detection laser beam exiting the interferometer to an 800-MHz bandwidth InGaAs photodiode detector with processing electronics. A 10-MHz high-pass filter was used to eliminate low frequencies. The signal is amplified by 26 dB using a 0.1–1300-MHz amplifier. The signal is acquired with a 1-GHz bandwidth digital oscilloscope. The detection laser beam was focused on the opposite surface of the sample with respect to the generation laser and aligned in the center of the flat-top generation zone. The focal spot radius of the detection beam was $130 \pm 25 \mu\text{m}$, hence about 85% that of the generating pulse radius (i.e., $152 \pm 7 \mu\text{m}$) as defined by the 90% rule discussed above.

III. EXPERIMENTAL RESULTS

In this section we describe (a) the broadband white light emission measurements used for determining the ablation threshold, and (b) measurements of the rear surface displacements characterizing the ultrasound amplitude, for both the 80-fs and 270-ps pulses.

A. Ablation threshold

The light-emission measurements as a function of the laser fluence are presented in Fig. 3 for the two pulse durations considered in this paper. One observes for both pulse durations a white light emission threshold occurring at 0.02 ± 0.01 and $0.04 \pm 0.02 \text{ J/cm}^2$ for laser pulse durations of 80 fs and 270 ps, respectively. Above this threshold, the white light emission steadily increases with fluence and eventually undergoes a strong break that we attribute to the formation of a plasma of ejected matter.³³ We define the ablation threshold by the fluence corresponding to this break. The physical explanation for the low-level light emission below the ablation threshold is not clear yet. Quantitative estimates support the assumption that this emission is due to black body radiation at the surface of the target. However, another possible explanation is that the emission would be

FIG. 4. (a) Typical measurement of rear sample surface displacement as a function of time. Ultrasound were generated by an 80-fs laser pulse with a fluence of $0.69 \pm 0.03 \text{ J/cm}^2$ on a $50\text{-}\mu\text{m}$ -thick Al foil. (b) Enlarged view of the first ultrasound echo. The value of displacement used in this article is illustrated by the two horizontal lines.

due to an intensity modulation in the generation laser beam (hot spots) producing narrow intense filaments, which would induce very localized ablation of the target.

The ablation thresholds of Al foils irradiated by an 80-fs and a 270-ps laser pulse are clearly 0.25 ± 0.03 and $0.63 \pm 0.03 \text{ J/cm}^2$, respectively. Using a 20X optical microscope, we have observed damage to the surface of the Al foil at fluences greater than 0.23 ± 0.03 and $0.53 \pm 0.03 \text{ J/cm}^2$ for the 80-fs and 270-ps pulses, respectively. The fact that the values found for the visible-damage thresholds are somewhat lower than the ablation threshold is consistent with the criterion used for the ablation threshold, since surface damage such as melting would naturally occur at a fluence lower than for ablation. However, these observations do not help in identifying the specific nature of the white light emission below the ablation threshold. More investigations would be required to draw definite conclusions.

B. Surface displacements

A typical signal corresponding to the rear surface displacement as a function of time in the ablative regime, in a

$50\text{-}\mu\text{m}$ -thick Al foil, is shown in Fig. 4. The compression wave front induces the first rear sample surface displacement [magnified in Fig. 4(b)], which is toward the detection laser. The amplitude of this first displacement peak (hereafter called “displacement amplitude”), measured at various fluences and laser durations, is used as a measurement of the ultrasound amplitude. This surface displacement peak is followed by a displacement away from the detection laser and back into the sample, which is associated with a tension wave following the compression wave front. The time delay between two consecutive echoes (compression peaks) corresponds to a round trip of the acoustic wave within the sample. The average long time scale displacement in Fig. 4(a) is associated with the global movement of the foil’s center of mass in reaction to the ejection of matter.²⁰

Measurements of the rear surface displacement amplitude as a function of laser fluence for the 80-fs and 270-ps laser pulses are presented in Figs. 5(a) and 5(b), respectively. One observes that the displacement amplitudes measured after traveling once through the foil are similar for the two pulse durations in the fluence range of $0.1\text{--}0.7 \text{ J/cm}^2$. However, the displacement amplitudes generated by a 270-ps laser pulse increase significantly in the fluence range of $0.7\text{--}1.5 \text{ J/cm}^2$. Measurements could not be performed with the 80-fs laser pulse in this fluence range because they were limited by the maximum energy available with this laser configuration.

As observed for the light-emission measurements (Fig. 3), there is also a break in the curves of the displacement amplitude measurements. This break occurs at the ablation threshold for the 270-ps pulse, but is approximately a factor of 2 greater than the ablation threshold for the 80-fs pulse. No surface displacements were detected at fluences for which no light emission was detected.

FIG. 5. Experimental results and simulations of rear sample surface displacements as a function of laser fluence for both an 80-fs and a 270-ps laser pulse on a $50\text{-}\mu\text{m}$ -thick Al foil. The slopes of the lines are represented by s , shown in units of m^3/J . Presented are the experimental measurements for 80 fs (a) and 270 ps (b), and the simulations for 80 fs, without and with 800-MHz low-pass filter (c), and 270 ps (d). The ablation threshold is indicated by the dashed lines.

The change in slope across the break is relatively small for the 80-fs pulse (3×10^{-13} – 9×10^{-13} m³/J) compared to the 270-ps pulse (3×10^{-13} – 43×10^{-13} m³/J). The small change in slope for the 80-fs pulse makes it difficult to precisely determine the break point. This might explain why this break point does not seem to coincide with the ablation threshold determined using the white light emission.

IV. SIMULATION RESULTS AND COMPARISON WITH EXPERIMENTS

To provide physical insight into the experimental results, we simulated the experiments using a one-dimensional fluid code that describes the generation and propagation of acoustic waves in bulk Al. A one-dimensional model is justified if the source of ultrasound is large enough so that essentially plane waves propagate in the thickness of the foil. Plane waves take place only in the near field of the acoustic source, as defined by the criterion $s \leq 0.1$, where $s = \lambda z / a^2$ is the Fresnel parameter. Here, λ is the acoustic wavelength, z is the propagation distance, and a is the radius of the source. In our experiments, $z = 50$ μm and $a = 152$ μm (the radius of the flat-top region). Then, since the velocity of sound in Al is $c_s = 6500$ m/s, the near-field criterion is verified for frequencies $c_s / \lambda > 141$ MHz, which is acceptable for ultrasound generated by short and ultrashort laser pulses (see Sec. I).

The code used for this project is adapted from previous work.^{17,28–30} It calculates the spatial profile of different hydrodynamic quantities such as density, temperature, pressure, and velocity as well as surface displacements of the target as a function of time by solving numerically the usual continuity equations for mass, momentum, and energy. The interaction of the laser pulse with the solid surface and with the ejected matter is described by solving the laser wave equation with the appropriate electrical permittivity, which depends on the local mass, density, and temperature of the medium. The equation of state for Al included in the code takes into account a single phase transformation from a dense state to a gaseous state. This seems to be justified because the latent heat of vaporization is much larger than the latent heat of fusion. The equation of state also takes into account the change in sound speed, resulting from plastic deformations of matter at high pressures.

For reasons of length and clarity, we show in this paper only the simulation results that can be directly compared with the experimental measurements. Details on the intermediate results of the simulations will be presented elsewhere.

The simulation results for the surface displacement amplitudes as a function of laser fluence in a 50- μm Al foil are presented in Figs. 5(c) and 5(d) for the 80-fs and 270-ps pulses, respectively. A comparison with Figs. 5(a) and 5(b), respectively, shows that the simulation and experimental results are qualitatively similar. However, significant quantitative differences are observed. In the following, we will discuss the physical interpretation of the general behavior observed in Fig. 5 as obtained from the simulations, as well as the possible reasons for the quantitative discrepancies between modeling and experiments.

A. Ablation thresholds

In the model calculations, the ablation threshold is unambiguously defined as the fluence required to produce a nonzero density of matter at some large distance away from the sample surface and at some large positive time.²⁸ These modeled thresholds are identified in Figs. 5(c) and 5(d).

For the 80-fs pulse, the calculated ablation threshold is 0.4 J/cm², while the threshold measured optically is near 0.25 J/cm² and the break point in the measured ultrasonic amplitude is 0.45 J/cm². We consider that this calculated ablation threshold agrees with experiment, given the uncertainties on the measurements and on the numerical model.

For the 270-ps pulse, the calculated ablation threshold is 4.5 J/cm², while the measured threshold is near 0.65 J/cm². Although the reasons for this large overestimation of the calculated ablation threshold are not yet clear, we suspect that this discrepancy could be due to the fact that some phase transformations are not taken into account in the model. Indeed, the numerical model predicts that, at the ablation threshold, the internal pressures are so high that the surface essentially explodes. However, it is possible that in the case of the longer 270-ps pulses, the surface would have time to melt and evaporate somewhat before the internal pressure builds up. This would create a gaseous Al atmosphere, which could greatly enhance the absorption of the later part of the light pulse and greatly lower the ablation threshold. (This effect would be similar to that occurring above the ablation threshold, as discussed below.) The numerical calculations could not predict this phenomenon because thermal evaporation is not included in the code. This phenomenon does not occur for the 80-fs laser pulse because the pulse ends before the surface atoms have time to move a significant distance as compared to the interatomic distance.

B. Displacement amplitudes

For the 270-ps laser pulse, there is a strong break in the surface displacement amplitude curve near the ablation threshold for both simulations [Fig. 5(d)] and experimental measurements [Fig. 5(b)] albeit at significantly different threshold values. The slope of both the simulated and experimental data is of the order of 10^{-13} m³/J for fluences below their respective ablation threshold, increasing to 50×10^{-13} m³/J (simulation) and 43×10^{-13} m³/J (measurements) for fluences above each ablation threshold. Therefore, except for the value of the ablation threshold (as discussed above), modeled and experimental displacement amplitudes agree well.

For the 80-fs laser pulse, and in contrast with the 270-ps laser pulse, the simulations predict that the ablation threshold has no effect on surface displacement amplitudes, whereas the measurements seem to show a modest break near 0.45 J/cm² [Figs. 5(b) and 5(d)]. Therefore, there is a marked difference between the ultrasound-generation mechanisms for the 80-fs and 270-ps pulses. However, there seems to be some qualitative disagreement between simulations and experiments for the 80-fs pulses.

The difference in calculated behavior between the surface displacement amplitudes for the 80-fs and 270-ps pulses

near their respective ablation threshold, i.e., the presence or absence of a break in displacement amplitude near the ablation threshold, can be explained as follows. In the 270-ps case, the time required to deposit the laser energy on the sample surface is sufficiently long to cause an interaction of the end of the laser pulse with the plasma produced at the beginning of the laser pulse. The laser energy is absorbed efficiently in the plasma, inducing a significant increase of temperature of the ejected matter. In turn, the higher plasma temperature increases the plasma pressure on the sample surface. As a result, the ultrasound waves generated at fluences above the ablation threshold have significantly greater amplitudes than those generated at fluences below the ablation threshold. This interpretation is in line with the normally accepted view for nanosecond laser-pulse generation.²⁻⁴ In contrast, the deposition of the laser energy of a femtosecond laser pulse on a sample surface occurs in a time interval so short that the atoms do not have time to move significantly. Therefore, the plasma ejected after the light pulse is not subsequently heated to higher pressures, and no increase in generation efficiency is predicted to occur at the ablation threshold. In partial agreement, both simulations and experimental results indicate that the ablation threshold has no or only a relatively small contribution to displacement amplitudes generated by femtosecond laser pulses. (It should be noted that both momentum and energy are naturally conserved in the model as a consequence of the continuity equations solved, and they have been checked to be conserved with high accuracy in the simulations.)

Still for the 80-fs pulse, the comparison between Figs. 5(a) and 5(c) shows that the model predicts an increase in ultrasound amplitude with a slope of $40 \times 10^{-13} \text{ m}^3/\text{J}$, while the measured value is probably $3 \times 10^{-13} \text{ m}^3/\text{J}$ but possibly up to $9 \times 10^{-13} \text{ m}^3/\text{J}$. Clearly, the model and experiment disagree quantitatively on surface displacement amplitude.

A partial explanation for these significant discrepancies is that the measurement of surface displacements is limited by the bandwidth of the InGaAs detector. As a result, the higher-frequency components of the measured signal are electronically attenuated. The frequency limit in the simulation results is much higher, and corresponds to the Nyquist criterion for a time step of 10 ps (50 GHz). Applying an 800-MHz low-pass filter (second-order Chebyshev) to the simulation results significantly decreased the amplitude of the surface displacement for the 80-fs pulse, as shown in Fig. 5(c). However, even with the filter, the calculated displacement amplitudes dependence on fluence is $33 \times 10^{-13} \text{ m}^3/\text{J}$, which is still substantially larger than the measured value for the 80-fs pulse. The same filter was found to have no effect on the displacement amplitudes for the 270-ps pulse. This is a natural consequence of the fact that the 270-ps laser pulse generates longer acoustic pulses than the 80-fs laser pulse and thus has lower frequency components.

The remaining discrepancy in surface displacement amplitudes cannot be attributed to any of the usual scattering or absorption mechanisms that cause ultrasonic attenuation for small-amplitude ultrasound because previous work consistently report signal attenuation in the range of 4.8–6 dB/mm near 530 MHz^{19,20,34} as compared to the ~ 400 dB/mm that

would be necessary to explain our results (i.e., roughly a factor of 10 for a propagation distance of 0.05 mm). Moreover, we do not think that the simulations can overestimate the amount of laser energy absorbed sufficiently to explain the discrepancy, since this quantity is in good general agreement with the available experimental data obtained for somewhat different laser parameters.³⁵

We believe that the observed attenuation is likely due to the high pressure generated in the solid by the 80-fs laser pulse. In the numerical model, the generated compressive stresses reach values in excess of 10 GPa near the ablation threshold. These stresses far exceed the static yield stress of Al, which is about 30 MPa or less for an annealed Al alloy of the 1000 series (more than 99% pure Al) and about 150 MPa or less for a heavily cold-deformed alloy of the same series (such as H18 treatment).³⁶ Although the dynamic yield stress is higher than the static one, it is unlikely to increase in the gigapascal range. As a result, both elastic and plastic deformation waves are generated, with most of the energy in the plastic wave.

It is known, from metallurgy, that plastic deformation results in the creation and motion of dislocations. Both of these processes convert mechanical energy into heat. Therefore, the attenuation of plastic waves could reduce the displacement amplitude substantially. We believe that the disagreement in the absolute displacement amplitude is due to the fact that the model did not take into account the appropriate wave-attenuation processes occurring in real solids. The only kinetic-energy absorption mechanism included in the code is the von Neumann artificial viscosity required for numerical stability. However, this contribution seems to be too small to describe the attenuation actually occurring in the aluminum samples used.

In the case of the 270-ps laser pulses, the calculated compressive stresses are only of the order of 1 GPa. Therefore, the propagating plastic deformation would be at least ten times smaller or perhaps absent altogether if the dynamic yield stress approaches 1 GPa. Correspondingly, the predicted and measured surface displacement amplitudes are found to agree.

V. SUMMARY AND CONCLUSION

The effects of laser-pulse duration and laser fluence on the amplitude of the rear sample surface displacements due to ultrasound waves propagating in thin (50 μm) Al foils were investigated experimentally. The two laser-pulse durations used in these experiments were 80 fs and 270 ps. The laser fluences investigated range from zero up to two or three times the ablation threshold for each pulse duration. A comparison with a one-dimensional fluid code was performed to provide physical insight into the ultrasound properties in these two regimes of laser-pulse duration.

Experimental results showed that displacement amplitudes of the rear surface as a function of laser fluence were similar when ultrasound were generated by 80-fs and 270-ps laser pulses in a common fluence range (0.1–0.7 J/cm²). Furthermore, it was discovered that the ablation threshold had a modest effect on ultrasound-generation efficiency when 80-fs laser pulses were used to generate ultrasound,

while the effect of the ablation threshold is far more significant for the 270-ps laser pulses. The significant influence of the ablation threshold on the surface displacement amplitudes for the 270-ps pulse is attributed to the heating of the ejected plasma by the incoming laser pulse, which increases the pressure at the target surface and induces additional momentum to the acoustic pulse. For the 80-fs pulse, this effect does not occur as ablation starts well after the interaction between the laser pulse and the target.

Comparison of the calculated and measured displacement amplitudes in the 270-ps case is not straightforward due to large discrepancies in the calculated and measured ablation thresholds. However, a significant break in the surface displacement amplitudes as a function of the laser fluence was predicted and experimentally observed at the ablation threshold for ultrasound generation. Moreover, the slopes of the surface displacement amplitudes associated with generation of ultrasound by a 270-ps laser pulse were very similar for both experiments and simulations below and above the ablation threshold.

For the 80-fs pulses, both experiments and simulations showed no (simulations) or relatively modest (experiments) break in the rear surface displacement amplitudes as a function of the laser fluence at the ablation threshold. The simulations predicted that very high pressures (a few gigapascal) generated at the sample surface by an 80-fs laser pulse would induce strong displacement amplitudes of the rear sample surface. Even when the limited bandwidth of the detection system was included in simulations, displacement amplitudes observed experimentally were much smaller than predicted by simulations. This seems to indicate a significant attenuation of the short high-pressure waves generated by ultrashort laser pulses. This strong attenuation was apparently not properly accounted for in the model.

Although several quantitative discrepancies have been observed between simulations and experiments, we believe that the code already encompasses several important features of ultrasound generation using short optical pulses. Improvements of the model would include realistic ultrasound absorption mechanisms, which depend on the ultrasound amplitude and frequency. Moreover, target evaporation below the ablation threshold should also be included in the code to investigate the role of early evaporated matter in triggering the ablation process for the 270-ps laser pulses.

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¹J.-P. Monchalín, in *Review of Progress in Quantitative Nondestructive Evaluation*, Green Bay, Wisconsin, 27 July-1 August, 2003, edited by D. O. Thompson and D. E. Chimenti (AIP, Melville, NY, 2004), Vol. 23A, pp. 3-31.

²D. A. Hutchins, Ultrasonics generated by pulsed lasers, in *Physical Acous-*

tics, edited by W. P. Mason and R. N. Thurston (Academic, San Diego, 1988), Vol. 18, pp. 21-123.

³D. A. Hutchins, *Can. J. Phys.* **64**, 1247 (1986).

⁴C. B. Scruby and L. E. Drain, *Laser Ultrasonics: Techniques and Applications* (Adam Hilger, Bristol, 1990).

⁵B. Betz and W. Arnold, *J. Phys. Colloq.* **44**, C6-61 (1983).

⁶J. A. Smith and K. L. Telschow, in *Review of Progress in Quantitative Nondestructive Evaluation*, Brunswick, Maine, 28 July-2 August, 1991, edited by D. O. Thompson and D. E. Chimenti (Plenum, New York, 1992), Vol. 11, pp. 625-630.

⁷R. Hrovatin and J. Mozina, *J. Appl. Phys.* **75**, 8207 (1994).

⁸S. Dixon, C. Edwards, and S. B. Palmer, *J. Phys. D* **29**, 3039 (1996).

⁹A. Hoffmann and W. Arnold, *Appl. Surf. Sci.* **96-98**, 71 (1996).

¹⁰R. J. Conant and S. E. Garwick, in *Review of Progress in Quantitative Nondestructive Evaluation*, Brunswick, Maine, 28 July-2 August, 1996, edited by D. O. Thompson and D. E. Chimenti (Plenum, New York, 1997), Vol. 16A, pp. 491-498.

¹¹T. W. Murray and J. W. Wagner, *J. Appl. Phys.* **85**, 2031 (1999).

¹²A. Hoffmann and W. Arnold, in *Review of Progress in Quantitative Nondestructive Evaluation*, Montreal, Canada 25-30 July, 1999, edited by D. O. Thompson and D. E. Chimenti (Plenum, New York, 2000), Vol. 19A, pp. 279-286.

¹³I. Apostol, R. Stoian, R. Dabu, A. Stratan, A. Sebanescu Oasa, and C. Angelescu, *SIOEL '99: Sixth Symposium on Optoelectronics*, Bucharest, Romania, 22-24 September, 1999 [Proc. SPIE 4068, 506 (2000)].

¹⁴R. J. Conant and S. E. Garwick, in *Review of Progress in Quantitative Nondestructive Evaluation*, edited by D. O. Thompson and D. E. Chimenti, (Plenum, New York, 1997), Vol. 16A, pp. 491-498.

¹⁵B. Mi and I. Charles Ume, *J. Nondestruct. Eval.* **21**, 23 (2002).

¹⁶B. Le Drogoff, F. Vidal, S. Laville, M. Chaker, T. W. Johnston, O. Barthélemy, J. Margot, and M. Sabsabi, *Appl. Opt.* **44**, 278 (2005).

¹⁷S. Laville, F. Vidal, T. W. Johnston, O. Barthélemy, M. Chaker, B. Le Drogoff, J. Margot, and M. Sabsabi, *Phys. Rev. E* **66**, 066415 (2002).

¹⁸J.-P. Monchalín, R. Héon, P. Bouchard, and C. Padiou, *Appl. Phys. Lett.* **55**, 1612 (1989).

¹⁹A. Moreau and M. Lord, in *Nondestructive Characterization of Materials*, Boulder, Colorado, 15-20 June, 1997, edited by R. E. Green, Jr. (Plenum, New York, 1998), Vol. VIII, pp. 27-32.

²⁰F. Blanchard *et al.*, in *Review of Progress in Quantitative Nondestructive Evaluation*, Bellingham, Washington, 14-19 July, 2002, edited by D. O. Thompson and D. E. Chimenti (AIP, Melville, NY, 2003), Vol. 22A, pp. 319-325.

²¹C. Thomsen, H. T. Grah, H. J. Maris, and J. Tauc, *Phys. Rev. B* **34**, 4129 (1986).

²²G. Eesley, B. Clemens, and C. A. Paddock, *Appl. Phys. Lett.* **50**, 717 (1987).

²³T. C. Zhu, H. J. Maris, and J. Tauc, *Phys. Rev. B* **44**, 4281 (1991).

²⁴G. Tas, R. J. Stoner, H. J. Maris, G. W. Rubloff, G. S. Oehrlein, and J. M. Halbout, *Appl. Phys. Lett.* **61**, 1787 (1992).

²⁵O. B. Wright and K. Kawashima, *Jpn. J. Appl. Phys., Part 1* **32**, 2452 (1993).

²⁶B. Bonello, B. Perrin, E. Romatet, and J. C. Jeannet, *Ultrasonics* **35**, 223 (1997).

²⁷A. Moreau, Comparative sensitivities of the piezoreflexive and interferometric ultrasound detection techniques, NRC-IMI Internal Report No. 2005-109923-G-CNRC.

²⁸F. Vidal, T. W. Johnston, S. Laville, O. Barthélemy, M. Chaker, B. Le Drogoff, J. Margot, and M. Sabsabi, *Phys. Rev. Lett.* **86**, 2573 (2001).

²⁹F. Vidal, S. Laville, T. W. Johnston, O. Barthélemy, M. Chaker, B. Le Drogoff, J. Margot, and M. Sabsabi, *Spectrochim. Acta, Part B* **56**, 973 (2001).

³⁰S. Laville, F. Vidal, T. W. Johnston, M. Chaker, B. Le Drogoff, O. Barthélemy, J. Margot, and M. Sabsabi, *Phys. Plasmas* **11**, 2182 (2004).

³¹P. P. Pronko, S. K. Dutta, D. Du, and R. K. Singh, *J. Appl. Phys.* **78**, 6233 (1995).

³²Q. Shan, A. S. Bradford, and R. J. Dewhurst, *Meas. Sci. Technol.* **9**, 24 (1998).

³³D. Du, X. Liu, and G. Mourou, *Appl. Phys. B* **63**, 617 (1996).

³⁴H. K. Wickramasinghe, R. C. Bray, V. Jipson, C. F. Quate, and J. R. Salcedo, *Appl. Phys. Lett.* **33**, 923 (1978).

³⁵H. M. Milchberg, R. R. Freeman, and S. C. Davey, *Phys. Rev. Lett.* **61**, 2364 (1988).

³⁶American Society for Metals, *Metals Handbook*, Desk ed. (American Society for Metals, Metals Park, OH 1985).