Acoustic monitoring of microplasma formation and filamentation of tightly focused femtosecond laser pulses in silica glass

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Contact acoustic technique has been employed to perform spatially resolved in situ detection of microplasma formation and filamentation of tightly focused femtosecond laser pulses with supercritical pulse powers in bulk dielectrics, via corresponding acoustic emission. Investigation of acoustic generation mechanisms related to the plasma formation and filamentation effects reveals the critical character of the opaque microplasma and provides estimates of its gigapascal-level pressures and energy densities of a few kJ/cm³. The acoustic measurement enables real-time in situ monitoring and revealing of basic mechanisms of ionization and filamentation in bulk dielectrics.


Femtosecond lasers have been proved to be highly efficient in producing various nano- and microscale features for photonic and biomedical microfluidic devices. However, transient ionization, filamentation, and structural modification phenomena underlying fabrication of such features by tightly focused intense femtosecond laser pulses are not well understood yet, requiring high (submicrometer) spatial resolution in probing opaque dense laser-induced plasmas and subsequent phenomena at the nano- and microscales. Since standard time-resolved optical techniques, such as shadowgraphy, interferometry, scattering and transient lens techniques do not provide direct and informative insight into the essential parameters of transient laser plasmas, filaments, and structural modifications, other acquisition techniques insensitive to plasma screening are necessary. Recently various acoustic techniques have been demonstrated to enable sensitive and informative but low-frequency (>1 µs) measurements of breakdown thresholds and acoustic response of femtosecond laser plasmas, as well as the following transient features (shocks and bubbles) in water and air. Though such acoustic probing studies exhibited low temporal resolution and data acquisition far away from the (sub)micrometer-sized femtosecond laser focal region, their obvious advantage is the absolute insensitivity to dense plasma screening, enabling informative measurements in critical and supercritical plasmas. Importantly, near-field acoustic detection at high temporal resolution being of interest for such microscale basic studies is a technical challenge to be overcome by means of optical pump-probe techniques—interferometry, shadowgraphy, and transient lens imaging—at the expense of more laborious data acquisition and signal processing. However, the abovementioned important opportunities of acoustic detection in basic studies of femtosecond laser plasmas have not been realized so far.

In this letter, we report on spatially resolved in situ contact acoustic studies of micro-plasma formation and filamentation in bulk dielectrics produced by tightly focused femtosecond laser pulses with variable supercritical pulse powers. Our investigation of the related acoustic generation mechanisms and acoustic parameters (waveforms, amplitudes, and pulsewidths) shows the critical character of the microplasma and gives a measure of the plasma pressure and energy density, which are sufficient to provide formation of high-contrast ablative optical damage in the plasma region.

In these studies, we used a femtosecond laser nanomachining setup described elsewhere. Briefly, linearly polarized, prechirped 800 nm femtosecond laser pulses of 150 fs duration having variable microjoule energies were focused into a submicron spot [estimated waist radius \( w_0 = \lambda/2(n^2 - NA^2)/(\pi NA) \approx 0.45 \mu m \) and Rayleigh length \( l_0 = \lambda(n^2 - NA^2)/(\pi NA^2) \approx 0.75 \mu m \)] at an \( \approx 150 \mu m \) depth inside a synthetic silica glass VIOSIL-SQ plate \( n_0(800 \text{ nm}) \approx 1.5 \); Shin-Etsu Chemical Co.] using a microscope objective with numerical aperture NA=0.75 (Fig. 1). The precleaned 200 µm thick glass sample was taped to a 1 mm thick borosilicate microscope slide (Matsunami, Inc.) and placed onto a manually controlled, three-axis microstage of an optical microscope (Olympus X-70). The sample was irradiated through the slide by single laser pulses selected using a manual laser control option, and was moved between the laser pulses parallel to the focal plane within a small area of 0.5×0.5 mm². An acoustic transducer (sensitive area of 8×8 mm², bandwidth with a preamplifier of \( \Delta f \approx 30 \text{ MHz} \), sensitivity of \( \sim 10 \text{ V/atm} \); UC VINFIN) was attached on the top of the glass via a 0.5 mm vacuum grease layer providing their acoustic contact and was centered along the laser beam axis. Voltage transients from the transducer,

FIG. 1. (Color online) Experimental geometry for acoustic monitoring of femtosecond laser breakdown and filamentation in the slide/VIOSIL glass/ vacuum grease combination under focusing by a lens with NA=0.75.
representing acoustic signals generated after each single-shot laser irradiation of the glass, were recorded using a 50 Ω input of a digital storage oscilloscope Tektronix TDS-5104, which was triggered via another 50 Ω input by a fast photodetector UPD-200-SP (175 ps rise time; Alphalas, GmbH), fed by a weak split laser beam.

The acquired acoustic transients exhibit main asymmetric bipolar pulses with major compression (positive) and accompanying minor rarefaction (negative) phases at \( t=0.7 \, \mu s \) and broad compressive (positive) pedestals on a 0.3–1.2 \( \mu s \) timescale (Fig. 2), where the 0.6 \( \mu s \) delay time for the main pulse results not from the intermediate construction elements but presumably from the grease layer. At lower laser pulse energies of \( \approx 4 \, \mu J \), the pedestal is almost absent, while the compression phase has a bandwidth-limited half-maximum width of 20 ns and the minor rarefaction phase is hardly observable in the noise [Fig. 2(a)]. At slightly higher pulse energies of \( \approx 5 \, \mu J \), the pedestal appears at \( t=0.3–1.2 \, \mu s \) above the noise level and the main pulse becomes much stronger and broader considerably to the full width of 40 ns, covering the entire 200 \( \mu m \) thick silica sample [the longitudinal sonic speed in silica is \( C_{\text{sil,}\text{c}}=5 \, \text{km/s} \) (Ref. 23)], with its rarefaction tail also becoming more prominent. At the maximum laser pulse energies of \( \approx 6–7 \, \mu J \), dictated by the optical strength of the objective used, the compressive phase amplitude of the main pulse increases further, preserving its full width of 40 ns, and the accompanying weak rarefaction phase emerges within the noise [Fig. 2(b)]. Simultaneously, the broad compressive pedestal becomes very pronounced in the time interval of 0.4–1.2 \( \mu s \), including the intact downstream 0.5 mm thick grease layer \( [t \approx 0.3–0.6 \, \mu s, C_{\text{grease}}=1.4 \, \text{km/s}] \) for the grease (Ref. 23)] and the upstream 1 mm thick slide \( [t=0.8–1.0 \, \mu s, C_{\text{sil,}\text{c}}=5 \, \text{km/s}] \) for the borosilicate glass (Ref. 23), while the origin of the rest later part is not clear, being potentially related to acoustic reverberations in the sample/slide/transducer combination.

Under the conditions of acoustic propagation and detection used in our experiments, the differentiating far-field diffraction effect on the propagating pressure transient \(^{24}\) is completely compensated via spatiotemporal integration over the large aperture of the transducer.\(^ {25}\) Hence, the pipolar pulses having dominant compressive and minor rarefaction parts indicate that the stress initially induced in the focal volume is of compressive nature. The far-field diffraction occurs for the axial distance between the acoustic source and transducer of about 0.5 mm (presumably, the grease thickness) and for much shorter diffraction length \( L_D \approx \pi w_0^2/C_{\text{fg}} \approx 10^{-3} \) mm (Ref. 24) estimated accounting for significant broadening of femtosecond laser-generated ultrasonic pulses in bulk materials to pulsewidths \( \tau \approx 10^{-9} \) s (Ref. 26), enabling their quite accurate measurement by means of the implemented multimegahertz transducer. Then, the predominantly compressive character of the stress source may be explained either by generation of electron-ion (e-i) plasma \(^ {16}\) or intensive point defects \(^ {18}\) in the focal volume but not by electronic \(^ {27}\) and/or thermoelastic \(^ {28}\) stresses resulting in bipolar acoustic transients due to the mechanical momentum conservation.\(^ {28}\) Also, the pulse energy dependence of the main pulse (compressive pressure) amplitude in Fig. 3, when corrected to an experimentally measured preamplifier characteristic nonlinear at the minimum, near-threshold voltages (Fig. 3, inset), gives an additional evidence of the plasma origin of the source pressure. It demonstrates for the intensity-independent, almost constant constant lateral size of the transient plasma a slope of 0.59 ± 0.12 characteristic of pressure \( (P_{\text{pl}}) \) dependence on laser intensity \( I \) for critical e-i plasma \( \left( P_{\text{pl},\text{c}} \equiv N_{\text{c}} k_B T_e \approx 2 \times 10^{23} \text{ cm}^{-3} \right) \) for the plasma density \( N_c \) and particle thermal energy \( k_B T_e \approx 10^3 \), rather than that for subcritical with \( P_{\text{pl,subc}} \approx 3 \times 10^{21} \text{ cm}^{-3} \). The way the critical plasma in an electronically excited solid \( [N_c(800 \, \text{nm})=2 \times 10^{21} \text{ cm}^{-3}] \) can behave as a gaslike one even at a rather low ion/atom ratio of \( \approx 0.1 \) will be discussed in our forthcoming publication. Here, we just mention that the corresponding energy density for the critical plasma in VIOSIL glass, exceeding \( N_c(800 \, \text{nm}) \Delta_{\text{bg}}=3 \, \text{kJ/cm}^3 \) for its dielectric bandgap \( \Delta_{\text{bg}}=9 \, \text{eV} \) is sufficient for its high-contrast ablative damage,\(^ {31}\) which has also been observed as microcracks inside the glass by optical microscopy of the irradiated spots (not shown). Importantly, such energy densities correspond to gigapsascal-level source stresses much lower than the Young modulus of the VIOSIL glass,\(^ {29}\) and, thus, producing in the acoustic near-field region (close to the source/plasma) intense sonic waves, rather than shocks.

The pulse energy-dependent temporal (axial) shift of the main acoustic source, i.e., the plasma core, position within the VIOSIL glass sample upstream to its front surface (the buried glass/slide interface) in Fig. 2 shows a distinct self-focusing effect for the femtosecond laser pulses with the

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**Figure 2.** (Color online) Waveforms of acoustic signals from the slide/glass/grease combination at laser pulse energies of 4 (a) and 7 \( \mu J \) (b). The vertical dashed line shows the trigger (zero time) instant, while the large and small arrows delimit the main pulse and the pedestal [in (b)], respectively. The major pulse of the zero-time acoustic response results from the direct laser heating of the front electrode of the transducer.

**Figure 3.** (Color online) Experimental (white circles) pulse energy dependencies of main pulse compressive amplitude and linear fitting curve (straight line, slope \( S \)) for the latter. Inset: Characteristic of the preamplifier with different slopes \( S_{1,2} \) at low and medium amplitudes of input signals.
highly supercritical, multimegawatt (7–70 MW) pulse powers $P$ [the critical power at 800 nm for fused silica is $P_{cr}(800 \text{nm}) = 2$ MW (Ref. 32)], shortening the effective working distance of the objective. As a result, one can expect formation of a light filament and a subsequent plasma channel within the glass sample [see in Fig. 2 the corresponding broadening of the main acoustic pulses from 20 to 40 ns, the maximum possible for its 200 $\mu$m thickness] with its densest part, which represents the critical plasma, moving upstream versus the increasing laser pulse energy (power). However, the limited bandwidth $\Delta f = 30$ MHz of the acoustic detection system does not allow us to resolve temporally and spatially the acoustic response of the denser plasma region within the 40 ns main pulse width.

Finally, the quite flat leading part of the compressive pedestal in Fig. 2(b) at $t \approx 0.3–0.7 \mu$s can be interpreted via long-term, most probably, filamentary propagation of the multimegawatt laser pulses, leaking from the glass sample into the succeeding grease layer. Such propagation provides formation of a channel of electronically excited molecules and e-i plasma via significant nonlinear absorption of the grease and the resulting detected compressive ablative response comparable to that from the critical plasma in the glass. In contrast, the noisy trailing part of the compressive pedestal in Fig. 2(b) at $t = 0.7–1.2 \mu$s may represent damped acoustic oscillations in the slide/sample/transducer combination, rather than an acoustic response from the preceding slide excited by the supercritical but weakly focused laser pulses.

In conclusion, spatially resolved in situ acoustic studies of bulk dielectrics excited at microscale by tightly focused femtosecond laser pulses with variable supercritical pulse powers have revealed formation of microplasma and filamentation of the pulses. The effective focal point shifts upstream within the VIOSIL silica glass sample versus increasing supercritical power due to self-focusing, approaching its front surface and leaving behind a long plasma channel, while the densest part of the plasma in the focal volume has the critical density and provides ablative optical damage of the glass. These new acoustic experiments and their findings may be of interest for real-time in situ monitoring and basic understanding of microscale ionization, filamentation, and structural modification in bulk dielectrics.

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