Coherent terahertz wave emission from laser-induced plasma in isotropic transparent solid dielectrics

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In this Letter, we employ fused silica and two types of optical glass as examples to investigate the coherent terahertz (THz) wave emission from laser-ionized isotropic transparent dielectrics. Based on the laser energy and incident angle dependences, we ascribe the THz emission to the ponderomotive force-induced dipole oscillation. Additionally, our investigation on the dependence of THz amplitude on the laser pulse duration confirms the dominant role of avalanche ionization in solid dielectrics. The THz emission can be utilized to indirectly monitor the ultrafast dynamics of carrier generation and motion during the laser ionization process of solid dielectrics.

Keywords: terahertz wave emission; laser-induced plasma; laser-solid interaction; ponderomotive force; fused silica.
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1. Introduction
Terahertz (THz) emissions from single-color, laser-ionized gaseous[1,2] and liquid[3–5] materials have been extensively investigated, with their origin commonly ascribed to the ponderomotive force-induced dipole emission[6]. Furthermore, THz wave emissions from laser ablated solid-state metallic materials have also been observed under the irradiation of both nonrelativistic[7–9] and relativistic[10–12] ultrafast laser pulses. The interaction of laser pulses with dielectric materials is quite different from that of the metallic materials[13,14]. The free carriers generation during the interaction of laser pulses with solid dielectrics mainly arises from two ionization processes: multiphoton ionization (MPI) and avalanche ionization[13]. When a transparent solid dielectric is ionized by intense ultrafast laser pulses, the generated electrons and ions in the material are ejected from the material surface, leading to plasma formation both inside and outside of the material surface and, consequently, the ablation of material. Driven by ponderomotive force, electrons in the plasma gain a net velocity and move directionally. The moving electrons and the quasi-static ions cause charge separation and form a transient dipole moment, emitting THz waves. Therefore, the emitted THz wave can reflect the charge dynamics during the laser ionization process. Eventually, a crater is created on the material surface through the laser ablation process, which is the basis of laser processing techniques. The study of the THz generation through laser interaction with solid dielectrics contributes to a comprehensive understanding of their laser ionization and ablation processes. However, systematic investigation on THz emission from laser-ionized transparent dielectric solid material is relatively rare.

As a typical isotropic dielectric solid material, fused silica (FS) has high transmittances to both visible light and ultraviolet[15], making it widely used throughout the optics field. Various experimental approaches have been utilized to investigate the laser ablation process of FS material[16–19] since it is crucial for applications that employ FS as the optical material; whereas THz emission from the laser-induced ionization process in transparent isotropic solid materials has never been demonstrated. In this Letter, we experimentally investigate the THz emission from FS plate (as well as other isotropic glass plates, such as BK7 and K9 glasses) irradiated by nonrelativistic sub-picosecond laser pulses. The typical waveform and spectrum of the THz wave emitted from laser-ionized FS plate are demonstrated in comparison with those from air plasma and BK7 and K9 glass plates. The dependences of the peak THz amplitude on the laser incident angle, laser pulse chirp, and laser pulse energy are systematically investigated. Our work not only demonstrates the THz emission process from laser-ionized FS plates but also provides an alternative method for investigating the ultrafast laser ablation process of FS material.

2. Experiments
The experimental setup is shown in Fig. 1(a). In our experiment, laser pulses are delivered by a commercial femtosecond
the charge separation in the plasma forms a transient dipole that
stimulates the optical field, giving rise to THz emission. A plasma is formed both at the surface and inside of the FS plate, as shown in Fig. 1(c). Stimulated by the optical field, laser irradiation exceeding 1000 seconds, the damage trace forms through the superposition of a series of individual damage traces on the FS plate. The laser irradiation time of the upper and bottom traces is about 100 seconds and 1000 seconds, respectively. (c) Schematic diagram of the THz emission from the plasma formed by the laser ablation process of the fused silica plate.

3. Results and Discussion

The temporal waveform of the THz radiation emitted from the laser-ionized FS plate, under irradiation of laser pulses with a pulse energy of 800 μJ, a pulse duration of ~500 fs, and an incident angle of ~50 degrees is shown by the black curve in Fig. 2(a). When the FS plate is removed, only a very small THz signal emitted from the air plasma can be detected since the THz wave generation from the air plasma prefers a shorter laser pulse. As a comparison, we measure the THz waveform generated from the air plasma when the laser pulse duration is tuned to be about 90 fs (close to the Fourier transform limited pulse condition) as shown by the red curve in Fig. 2(a). The peak amplitude of the THz wave emitted from the FS plate irradiated by 500-fs laser pulses is about ten times of that from air plasma induced by 90-fs laser pulses. When 90-fs laser pulses are used to irradiate the FS plate, the peak amplitude of the THz wave emitted from the FS plate is about 3.5 times of that from the air plasma as shown by the black dashed line in Fig. 2(a).

For comparison, we also investigate the THz emission from the laser ionization process of other common isotropic dielectric solid materials that have high optical transmittance, such as BK7 and K9 glasses. Both BK7 and K9 are optical borosilicate glass, which are widely used in commercial optics. BK7 glass is composed of 70.0% SiO2, 11.5% B2O3, 9.5% Na2O, 7.5% K2O, and 1.0% BaO (in mass fraction of each component) [20], while K9 glass is composed of 69.13% SiO2, 10.75% B2O3, 10.40% Na2O, and 6.29% K2O [21]. The blue and green curves in Fig. 2(a) show the THz waveforms emitted from laser-ionized BK7 and K9 glass plates under the same experimental conditions.
used for the FS plate, respectively. The amplitudes of the THz waves emitted from these glass plates are even smaller than that from air plasma since the absorption in these glass materials greatly damps the strength of the transmitted THz waves. Since the frequency dependent absorption coefficients of the BK7 and K9 glasses increase rapidly with the THz frequency in the range of 0.1–1.0 THz and exceed 80 cm\(^{-1}\) at 1 THz\(^{-1}\), only low-frequency components of a THz pulse can pass through the BK7 and K9 glass plates with a 2-mm thickness, which is verified by the THz Fourier transform spectra shown by the blue and green curves in Fig. 2(b).

On the contrary, the absorption coefficient of the FS is no more than 10 cm\(^{-1}\) within the frequency range of 0.1–2.0 THz\(^{-1}\), although it also increases with THz frequency. Therefore, THz frequency components within the range of 0.1–2.0 THz can efficiently transmit the FS plate, which is consistent with the experimental result shown by the black curve in Fig. 2(b). However, the bandwidths of all the THz spectra emitted from laser-ionized dielectric solid plates are narrower than that from the air plasma induced by laser pulses with a 90-fs pulse duration, which can be attributed to the frequency dependent absorptions and the much longer pump laser pulse duration. It is noteworthy that when 90-fs laser pulses are used to irradiate the FS plate, the spectral bandwidth of the THz wave is broadened compared to that when the 500-fs laser pulses are used, as shown by the black dashed line in Fig. 2(b).

Similar to the THz generation process in single-color laser-ionized gases and liquids, we suppose that the ponderomotive force-induced dipole emission contributes mostly to the THz wave detected from the laser-ionized FS plate in our experiment. In this case, the THz wave is emitted in the lateral directions with respect to the moment of the ponderomotive force-induced dipole. The orientation of the ponderomotive force-induced dipole moment changes with the laser incident angle, leading to the variation in the efficiency of coupling the THz wave out of the FS plate and collecting the THz wave by PM2 with a 2-inch clear aperture. Consequently, the detected peak THz amplitude is a function of the laser incident angle, as shown in Fig. 3(a). When the laser beam is normally incident on the FS plate, corresponding to the incident angle of 0 degrees, only a very small THz signal can be detected in the laser propagation direction since the dipole moment is aligned with the laser axis in this case. When the laser incident angle deviates from 0 degrees, the peak THz amplitude increases and reaches its maxima at an incident angle between 50 degrees and 55 degrees. When the absolute value of the incident angle exceeds 55 degrees, the peak THz amplitude decreases with the laser incident angle. It is worth noting that the dependence of the peak THz amplitude on the laser incident angle is essentially an odd function, as shown in Fig. 3(a). The strengths of the THz wave are nearly identical while the polarities of the THz amplitude are reversed for laser incident angles with opposite signs. As shown in Fig. 3(b), the THz waveforms obtained at laser incident angles of 45 degrees and −45 degrees are completely inverted in comparison to each other. These experimental results are consistent with the ponderomotive-induced dipole model since the opposite laser incident angles lead to mirrored orientations of the dipole moment. Therefore, based on the ponderomotive force-induced dipole model, we can calculate the on-axis THz amplitude as a function of the laser incident angle using the following equation:

\[
E_{\text{THz}}(\theta) \propto \frac{1 - R(\theta)}{\cos(\theta)} \cdot \sin[\beta(\theta) - \gamma(\theta)]t_{\text{THz}}(\theta) \cdot \exp[-\alpha L(\theta)/2].
\] (1)

The first term on the right hand of Eq. (1) represents the laser intensity at the focus spot on the FS surface, where \(R(\theta)\) is the reflectivity of light. The second term represents the electric field distribution of a point dipole source induced by the laser field, where \(\beta(\theta)\) is the orientation of the dipole moment, \(\gamma(\theta)\) is the emission angle of the THz wave from the dipole with respect to the laser axis, and \(t_{\text{THz}}(\theta)\) is the transmittance of the THz electric field when the THz wave is coupling out of the FS plate at the rear FS-air interface. The last term represents the absorption loss of THz wave in FS plate, where \(\alpha\) is the absorption coefficient, and \(L(\theta)\) is the propagation length of the THz wave in the FS plate. As shown by the black curve in Fig. 3(a), the calculation result is consistent with the experimental results, further validating the ponderomotive force-induced dipole model.

The THz generation process begins with the generation of free carriers via different ionization mechanisms that depend on material properties and laser parameters, such as the ionization potential of the material, the peak laser intensity, and the laser pulse duration. Owing to the characteristics of the isotropic dielectric solid material, the ionization process of the FS is quite different from that of gaseous, liquid, and metallic materials. For laser pulses with a sub-picosecond pulse duration, MPI and, subsequently, avalanche ionization dominate the ionization process of the laser irradiated FS. MPI generates free electrons through multiphoton absorption and provides seed electrons for the avalanche ionization, acting as an initial step of the ionization process. Then, these seed electrons are further heated by the laser field, leading to the avalanche ionization through electron impact. The avalanche ionization takes place within the laser pulse duration. Therefore, a longer laser pulse duration enables a longer avalanche process, leading to a higher electron density.

![Fig. 3](https://example.com/figure3.png)

**Fig. 3.** (a) Dependence of the peak THz amplitude emitted from the laser-ionized fused silica plate on the laser incident angle. (b) THz waveforms measured at incident angles of 45 degrees and −45 degrees, respectively.
in the generated plasma. Driven by the ponderomotive force, the generated electrons form a transient dipole and consequently emit THz radiation. However, the ponderomotive force is directly proportional to the gradient of the laser intensity. In this case, when the laser pulse energy is fixed, the ponderomotive force decreases with the laser pulse duration. Therefore, the trade-off between the electron density and the ponderomotive force determines the optimal laser pulse duration for the THz generation from the laser-ionized FS plate, which can be verified by the experimental results shown in Fig. 4(a). The dependences of the peak THz amplitude on the laser pulse duration measured at laser pulse energies of 200, 400, 800, and 1600 μJ are shown in Fig. 4(a). The tunable pulse duration is achieved by pre-chirping the laser pulses. The laser chirp is presented in terms of the group delay dispersion (GDD) imparted to the laser pulses in the top label of Fig. 4(a). The left panel shows the experimental results obtained when the laser pulse is positively chirped and vice versa. For all pump laser pulses with positive or negative chirp and different pulse energies, the peak THz amplitude first increases and then decreases with the pulse duration. However, the optimal laser pulse duration for the THz emission tends to increase with laser pulse energy. When the laser pulses are negatively chirped, the optimal laser pulse durations are approximately 385, 460, and 496 fs for laser pulse energies of 400, 800, and 1600 μJ, respectively. Such laser pulse durations limit the electron density generated from the avalanche ionization process. By using a longer laser pulse, the electron density in the plasma can be even higher, but the THz emission may be weaker since the ponderomotive force depends on the gradient of the laser intensity. A much stronger THz emission can be expected if the solid target is ionized by a pre-pulse with a long pulse duration, and the THz wave is generated by a subsequent pulse with a short pulse duration \[26\].

Moreover, the dependences of the peak THz amplitude on the laser pulse energy measured at 496-fs (negative chirp), 200-fs (positive chirp), and 92-fs (close to the Fourier transform limited pulse condition) laser pulse durations are shown in Fig. 4(b). The experimental results measured at the laser pulse durations of 496 fs and 200 fs can be fitted linearly, whereas the result measured at 92 fs shows a saturation trend. When the laser pulse duration is fixed, the THz electric field emitted from ponderomotive force-induced dipole is directly proportional to the laser pulse energy. Thus, the fitting results shown by the red and blue dashed lines in Fig. 4(b) confirm the linear correlation between the peak THz amplitude and the laser pulse energy, providing further evidence that the THz wave emitted from the laser-ionized FS plate originates from the dipole emission induced by the ponderomotive force.

However, the saturation trend of the pump pulse energy dependence measured at the pulse duration of 92 fs may be associated with the intensity clamping effect. The peak power of the 92-fs laser pulse is relatively higher than that of the chirped pulses at the same laser pulse energy. The self-defocusing in the FS is more significant for laser pulses with higher peak powers, leading to the intensity clamping effect and, consequently, the THz emission saturation trend at high pulse energies, as evidenced by laser pulse spectrum broadening in our previous work when liquid lines were used as the THz source \[4\]. The use of chirped laser pulses with longer pulse durations can mitigate the THz emission saturation and obtain a linear trend of the pump pulse energy dependence.

### 4. Conclusion

In conclusion, we experimentally investigate the THz emission from the laser-ionized FS plate. The amplitude of the THz wave emitted from the laser-ionized FS plate is approximately ten times of that from air plasma. The THz emission from other isotropic dielectric solid materials, such as BK7 and K9 glasses, has also been investigated. However, the amplitudes of the THz waves emitted from these glass materials are much smaller than that from the FS plate owing to their relatively high absorption coefficients in the THz frequency range. The THz emission from the laser-ionized FS plate is attributed to the ponderomotive force-induced dipole oscillation, which is verified by the dependences of the peak THz amplitude on the laser incident angle and laser pulse energy. The dependence of the peak THz amplitude on the laser incident angle shows that the optimal laser incident angle for the THz emission falls between 50 degrees and 55 degrees, and the polarities of THz amplitude are reverse for laser incident angles with opposite signs. Moreover, the linear relationship between the peak THz amplitude and the laser pulse energy further verifies the ponderomotive force induced dipole model. Additionally, our investigation on the dependence of the peak THz amplitude on the laser pulse duration reveals the ionization process of the laser-irradiated FS plate. The optimal pulse duration for the THz emission is on the sub-picosecond scale, which is determined by the trade-off between the avalanche ionization and the ponderomotive force. By means of the double-pump technique, a much stronger THz emission from a laser-ionized isotropic dielectric solid material can be anticipated. Our work not only demonstrates the physical mechanism of the THz emission from the laser.
ionization process of the FS material but also may have contributions to laser processing applications by offering a potential method to study the laser ablation process of solid dielectrics.

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