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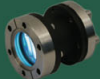



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Enhancing the energy of terahertz radiation from plasma produced by intense femtosecond laser pulses

Fazel Jahangiri,^{1,2,a)} Masaki Hashida,^{1,3,a)} Shigeki Tokita,^{1,3} Takeshi Nagashima,^{3,4} Masanori Hangyo,^{3,4} and Shuji Sakabe^{1,3}

¹Advanced Research Center for Beam Science, ICR, Kyoto University, Kyoto, Japan

²Laser and Plasma Research Institute, Shahid Beheshti University, Tehran, Iran

³Department of Physics, GSS, Kyoto University, Kyoto, Japan

⁴Institute of Laser Engineering, Osaka University, Osaka, Japan

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Terahertz (THz) radiation from atomic clusters illuminated by intense femtosecond laser pulses is investigated. By studying the angular distribution, polarization properties and energy dependence of THz waves, we aim to obtain a proper understanding of the mechanism of THz generation. The properties of THz waves measured in this study differ from those predicted by previously proposed mechanisms. To interpret these properties qualitatively, we propose that the radiation is generated by time-varying quadrupoles, which are produced by the ponderomotive force of the laser pulse.

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The application of THz radiation is expanding in remote sensing, imaging, spectroscopic analyses, and other promising areas. As a promising source for THz radiation, plasma induced by femtosecond laser pulses has been extensively studied.^{1–7} Comparison of different gas and solid targets has revealed that solid targets are more efficient than gas targets,^{8–12} owing to their much higher laser absorption.¹³ However, considerable THz reflection at the vacuum-plasma interface and generation of a large amount of debris are significant problems associated with THz radiation produced from solid targets. We have recently proposed atomic clusters as an alternative target for the production of THz radiation.^{9,14} These combine advantages of both gas and solid targets: laser absorption is higher than for gases, resulting in stronger THz emission, and there is much less debris generation than for solid targets. We have reported that the plasma of argon clusters as well-isolated solid targets can generate THz pulses with energies two orders of magnitude higher than argon gas, distributed in a scissors-like shape in the horizontal plane.¹⁴ Although, this improvement can be directly ascribed to higher laser absorption in atomic clusters,^{15,16} a deep understanding of the physical mechanism is still desirable in order to explain the overall characteristics of THz radiation. Precise measurements of THz power in both horizontal and vertical planes around the plasma together with polarization analyses are presented here to inform the discussion about the physical mechanism of THz generation. The measurements reveal a conical angular distribution, in both forward and backward directions, with radial polarization and also a significant emission in the forward direction with elliptical polarization. The total THz power has a square dependence on laser pulse energy. These results are not consistent with previously proposed mechanisms, including electron oscillations driven by the laser wake field,^{1,2} nonlinear frequency mixing processes in air plasmas^{4–8} and transition radiation from relativistic electrons induced by the laser

wake field passing through the plasma-vacuum boundary.^{11,12} Based on the observed properties, a qualitative mechanism is proposed and discussed.

Argon clusters were produced by injection of high pressure argon gas (<8 MPa) into a high-vacuum chamber, through a conical nozzle. High-energy pulses (<80 mJ) from a Ti:sapphire laser¹⁷ with a center wavelength of 800 nm and a duration of 130 fs were focused onto the argon cluster beam by a plano-convex lens with a focal length of 200 mm. The gas injection was synchronized with the laser pulses for maximum laser energy absorption (details can be found in Ref. 18). The vacuum chamber was made of fused silica with a transparency of ~80% for THz waves at frequencies of <3 THz.¹⁴ The experiment used the same setup as described in Ref. 14. The mean size of the clusters, which depends on the backing pressure, was determined by the Rayleigh scattering method¹⁹ to be ~10⁴ atoms at 7 MPa. This corresponds to a mean cluster diameter of ~10 nm.²⁰ Considering the atomic density of argon gas flowing at backing pressure of 7 MPa that is estimated to be ~10¹⁷ cm⁻³ (Ref. 21) and also considering the mean size of the clusters (~10⁴ atoms), the average density would be equivalent to 10 cluster/μm³ and the mean distance between adjacent clusters is estimated to be of the order of ~1 μm. The laser spot diameter was ~17 μm. A schematic diagram of our experimental setup is shown in Fig. 1(a). The laser pulses were linearly polarized in the horizontal direction, parallel to the optical table. Argon clusters were irradiated by the laser pulses, and the high-energy ions generated by Coulomb explosion of the clusters were detected by a time-of-flight measurement.^{18,19} The energy distribution of the generated ions was used to confirm the production of the clusters. The THz waves produced from the interaction of laser pulses with the cluster beam were collimated by a polyethylene lens with focal length of 150 mm, which was placed outside the vacuum chamber. After passing through a wire grid polarizer, the THz waves were focused onto a liquid-He-cooled InSb bolometer by an off-axis parabolic mirror. A time interferogram of THz pulses was obtained by using a

^{a)}Authors to whom correspondence should be addressed. Electronic addresses: f_jahangiri@sbu.ac.ir and hashida@laser.kuicr.kyoto-u.ac.jp

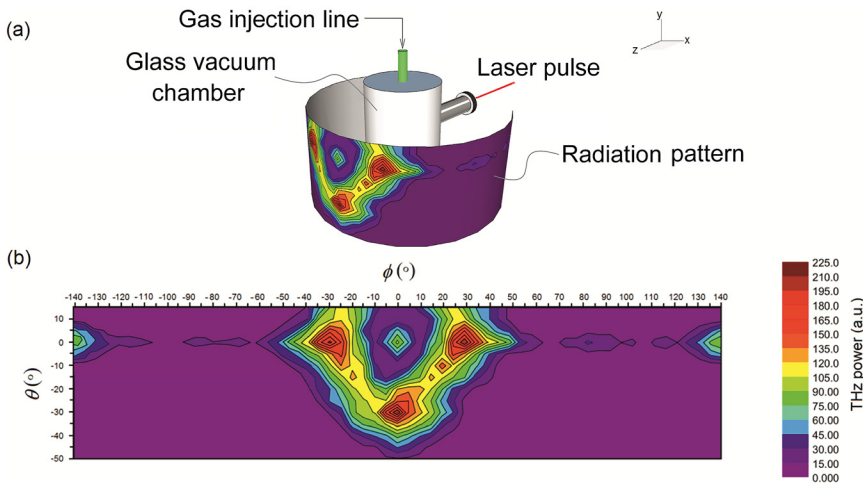


FIG. 1. (a) Schematic of experimental setup with (b) the 2D angular distribution of THz waves. The horizontal and vertical measurement angles were limited by the laser focusing setup and the gas injection line, respectively.

Martin-Puplett interferometer and the bolometer. The details of these measurements have been presented in Ref. 14. The spectrum of THz pulses was found by performing an inverse Fourier transform; the spectrum does not strongly depend on laser energy or backing pressure.¹⁴ A thick plate of polystyrene foam and a black polyethylene sheet were placed in front of the bolometer to block unwanted infrared and visible light emitted or scattered by the clusters.

The angular distribution of THz waves measured in two dimensions at a backing pressure of 7 MPa and a laser energy of 30 mJ shows that THz waves had a conical distribution in both the forward and backward directions (Fig. 1(b)). By moving the bolometer along a circular path around the chamber, THz power was measured in the horizontal (ϕ) and vertical (θ) directions at step sizes of $\Delta\phi = 5^\circ$ and $\Delta\theta = 5^\circ$. The reference angle was chosen such that the laser propagation direction was $(\phi, \theta) = (0, 0)$. This figure also shows that the radiation cone includes significant peaks, detected in planes containing the angles $\phi = 0$ and $\theta = 0$. Furthermore, considerable radiation was observed in the direction of laser propagation.

Using a wire grid polarizer, the horizontal and vertical components of the THz waves were separately measured at different angles. Comparing the horizontal and vertical components gives the polarization direction (see Fig. 2). The observed polarization pattern was reminiscent of that expected

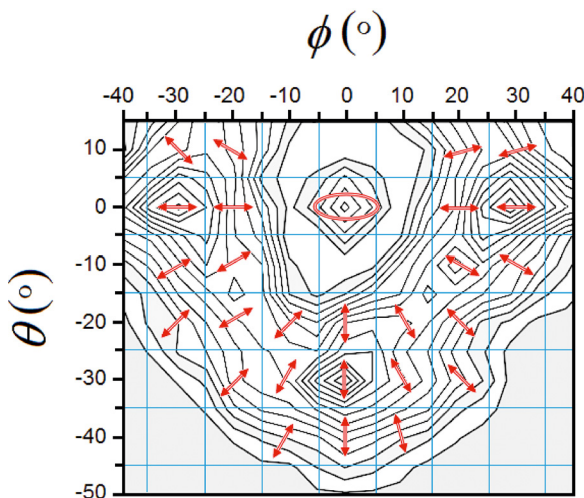


FIG. 2. Radial and elliptical polarization of THz waves observed on the radiation cone and on the laser propagation axis, respectively.

for a radial polarization. By inserting a half-waveplate in front of the focusing lens, we confirmed that the polarization of the THz waves was independent of laser polarization. Our measurements show that the considerable radiation observed in the direction of laser propagation cannot be a conical beam. The polarizations of THz waves were measured before and after passing through a quarter-waveplate designed for a frequency of 0.5 THz (WPCQD47-OW600L/4, Tydex Co.). Bandpass filters (BPF0.5-47, Tydex Co.), which were inserted in the path of the THz waves, passed the frequencies specified by the quarter-waveplates. The polarization of the THz waves after passing through the quarter-waveplate was almost linear, which shows that the initial THz polarization in this direction was elliptical with a measured ellipticity of ~ 0.5 (see Fig. 3). The elliptical polarization of THz waves generated along the laser axis is consistent with a four-wave mixing process.¹⁴

The measured properties of the THz waves differed from those expected from previously proposed mechanisms.^{1,2,11,12,22,23} The nonlinear current associated with laser intensity inside the plasma^{1,2} predicts a polarization dependent on laser polarization. However, we observed that the polarization state of the THz wave distributed through the cones was independent of laser polarization. This suggests that the contribution from the nonlinear current was negligible.

It has also been proposed that transition radiation generated by laser-wakefield-accelerated electron bunches passing through a plasma-vacuum boundary can result in THz radiation.^{11,12} According to the theory,²⁴ when the plasma is assumed to be a perfect conductor, the angular energy distribution of radiation from a single electron traversing the plasma-vacuum boundary is given by

$$W(\omega_{THz}, \Theta) \propto \frac{\beta^2 \sin^2 \Theta}{(1 - \beta^2 \cos^2 \Theta)^2}, \quad (1)$$

where Θ is measured with respect to the laser pulse propagation direction and $\beta = v/c$ (v : electron velocity; c : speed of light in a vacuum). For laser intensities of 10^{17} W/cm², such as those used in the present work, the ponderomotive energy $K = e^2 E^2 / (m \omega^2)$ is ~ 6 keV,²⁵ where e is the electron charge, E is the laser electric field, m is the electron mass, and ω is laser frequency. This corresponds to an electron velocity of $\beta \approx 0.15$. For these parameters, it is expected from Eq. (1)

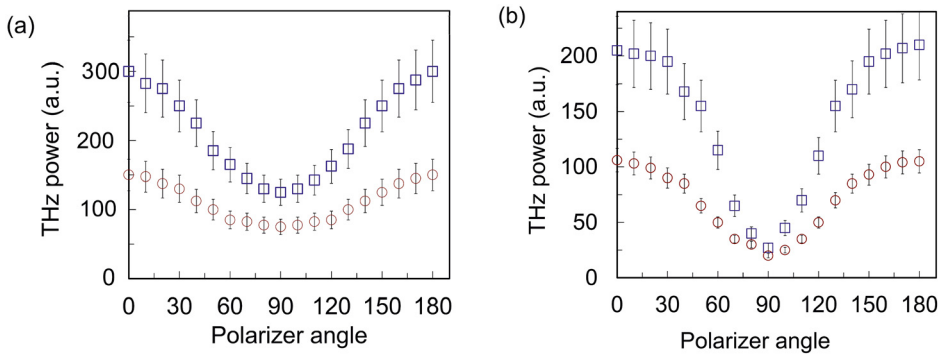


FIG. 3. Polarization analyses of THz radiation along the laser propagation axis (a) before and (b) after passing through a quarter-waveplate, at laser energies of 10 mJ (circle) and 40 mJ (square).

that the angular distribution of THz wave power caused by the transition radiation should exhibit a maximum in the direction of $\Theta \approx 90^\circ$, which is inconsistent with the measured angular distribution. Therefore, we conclude that here transition radiation can be ruled out as a dominant contributor to the production of THz radiation.

There is also the possibility that the THz radiation was Cherenkov radiation caused by a high-energy electron beam.^{22,23} The real part n_{THz} of the refractive index of the plasma at 0.5 THz is estimated to be ~ 40 , under the Drude model with an electron scattering time of ~ 0.01 ps. This value comes from the fact that, for an assumed electron density of $\sim 10^{18} \text{ cm}^{-3}$, the electrons would be scattered by the clusters within a scattering time of $\tau = d/(c\beta)$, where d is the mean distance between the clusters. Then, the Cherenkov radiation would be emitted in a direction of $\cos^{-1}(1/\beta n_{\text{THz}}) \approx 80^\circ$, if the boundary effect is ignored. If the THz radiation in direction of 30° (half-cone angle) were Cherenkov radiation, the value of β would be ~ 0.029 . This value is too small for the laser intensity we used; therefore, Cherenkov radiation cannot be a major factor. Moreover, this model cannot explain the backward THz radiation observed from the argon cluster plasma.

To interpret the angular distribution and polarization of THz waves, we propose a model based on radiation from time-varying electric quadrupoles^{26,27} generated by electrons moving in response to the ponderomotive force of the laser pulses. The electrons in the vicinity of the laser pulse are expelled to the outside of the laser-produced plasma by the ponderomotive force originating from the radial gradient of the electric field. This motion of the electrons produces a core of positive charge surrounded by symmetrically distributed electrons. Since we did not apply an external field, the plasma was axially symmetric with zero transverse dipole moment.²⁸ Such plasma volumes can be theoretically considered as a simple radiating quadrupole source.²⁶ An axial electric quadrupole is formed by the distribution of charges on a line with two ions ($+2q$) at the midpoint and two electrons ($-q$) located symmetrically around it, as shown schematically in Fig. 4. The axial quadrupoles prefer to align parallel to the directions of laser propagation, laser polarization, and gas injection. As the laser pulse passes through the plasma, the quadrupoles at each point in the plasma oscillate with a period of ~ 130 fs, which is the duration of the laser pulse. Therefore, waves radiated from the quadrupoles have a spectrum in the THz frequency region. Nonresonant THz radiation and scattering caused by the electrons may result in a broadening of the spectrum around the central wavelength. According to our

measurements, the wavelength of THz radiation is $\geq 300 \mu\text{m}$, much smaller than the plasma dimensions, which was estimated from images of fluorescent light scattered from the plasma.¹⁴ Therefore, the total radiation from plasma is over the sum of the radiation coming from each small region containing the electric axial quadrupoles. According to electromagnetic field theory, the angular distribution of radiated power from an axial quadrupole is a four-lobed pattern^{26,29}

$$\frac{dP}{d\Omega} \propto \sin^2\theta \cos^2\theta, \quad (2)$$

where θ is the angle between the observation point and the axis of the quadrupole. Therefore, THz radiation from quadrupoles oscillating in a defined direction is conically distributed around that axis in the forward and backward directions.

The local density of argon clusters is much higher than the density of the argon gas just after exiting the nozzle of injection system.^{14,15} Since the ponderomotive energy is proportional to the plasma density, it will be much higher inside the argon cluster plasma than in a gas plasma, resulting in much stronger quadrupole radiation. The laser absorption (A_{cluster}) of an argon cluster has been measured to be ~ 30 -fold that of argon gas, at the same atomic density and same laser energy. It has been found that THz radiation and laser absorption of argon clusters have a square ($I_{\text{THz}} \propto P^2$) and linear dependence ($A_{\text{cluster}} \propto P$) on backing pressure (P), respectively; resulting in $I_{\text{THz}} \propto (A_{\text{cluster}})^2$. Therefore, a 900-fold enhancement might be expected in THz power when a 30-fold increase is observed in laser absorption. The slightly smaller experimentally observed enhancement (600-fold)¹⁴ might be attributed to the saturation of THz power at high backing pressures.⁹ Furthermore, since the quadrupoles are induced by ponderomotive forces within the plasma, a square dependence on laser energy is expected for THz radiation.^{14,27} This is consistent with our observations,

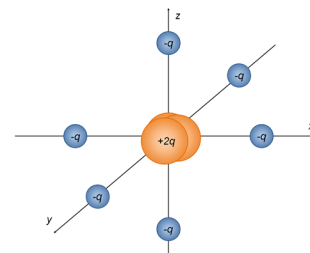


FIG. 4. Configuration of electric quadrupoles in the plasma, parallel and perpendicular to the laser propagation direction (z). The perpendicular directions are the directions of the laser polarization (x) and gas jet injection (y).

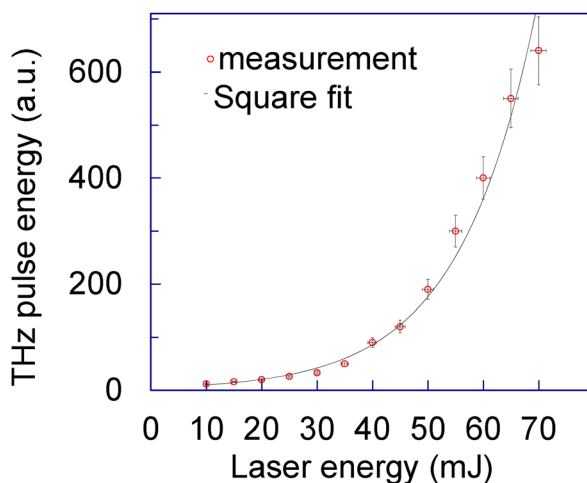


FIG. 5. The energy of the THz radiation has a square dependence on laser energy.

as shown in Fig. 5. It is worth pointing out that, due to technical limitations, the laser energy was kept less than 80 mJ, so we did not observe any saturation at high laser energies. The non-saturated THz power at high laser energies reveals the potential for the extraction of higher THz energies from argon cluster plasma.

In addition to the conical distributions, four significant peaks were also observed in the directions of $(\theta, \varphi) = (0^\circ, 30^\circ)$ and $(-30^\circ, 0^\circ)$, as shown in Fig. 1(b). It seems that quadrupole radiation can also explain the appearance of these peaks. As discussed above, the quadrupoles are generated parallel and transverse to the laser propagation direction. Just as the parallel quadrupoles generate THz waves in forward and backward cones aligned along the direction of laser propagation (z direction), the transverse quadrupoles can generate THz radiation in cones with axes in the x and y directions. In agreement with previous reports,²³ we have recently shown¹⁴ that the opening angle of the radiation cones is expected to be proportional to $(\lambda/d_i)^{0.5}$, where λ is the wavelength of radiation and d_i is the length of the plasma along the radiation direction ($i = x, y, \text{ or } z$). Assuming that the plasma is cylindrical along z direction with length of d and radius of R , plasma lengths are $d_x = R$, $d_y = R$, and $d_z = d$. Experimental measurements of plasma dimensions show that the radius of the plasma, R , is around a quarter of its length, d (i.e., $d_x = d_y = 1/4 d_z$). Thus the opening angle of the radiation cones in the x and y directions should be ~ 2 -fold larger than that observed in the z direction (30°), that is, $\sim 60^\circ$. Therefore, the radiation cones with axes along the x and y directions are, respectively, superposed at angles of $(\theta, \varphi) = (0^\circ, 30^\circ)$ and $(-30^\circ, 0^\circ)$ on the radiation cones along the z direction, exactly where the four observed peaks appear.

In summary, the polarization properties, angular distribution, and laser energy dependence of THz waves generated from argon cluster plasma have been studied and compared with the properties expected if they were produced by previously proposed mechanisms. Differences between the expected and observed properties allow us to reject all previously proposed models for the generation of THz radiation. We propose that the THz radiation is generated by electric quadrupoles. This model can explain the polarization

properties, energy dependence, and backing pressure dependence of THz waves.

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