Intense terahertz emission from atomic cluster plasma produced by intense femtosecond laser pulses

Fazel Jahangiri,[1,a)] Masaki Hashida,[1] Takeshi Nagashima,[2] Shigeki Tokita,[1] Masanori Hangyo,[2] and Shuji Sakabe[1]
[1]*Advanced Research Center for Beam Science, ICR, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan and Department of Physics, GSS, Kyoto University, Kitashirakawa, Sakyo, Kyoto 606-7501, Japan*
[2]*Institute of Laser Engineering, Osaka University, 2-6 Yamadaoka, Suita, Osaka 565-0871, Japan*

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Terahertz (THz) emission from argon cluster plasma, generated by intense femtosecond laser pulses in the energy range of 10–70 mJ, has been investigated. THz polarization, energy dependence, and angular distribution were measured to provide an initial discussion on the mechanisms of THz emission. THz pulses of much higher energy were generated from argon clusters than from argon gas, which indicates that plasma produced from atomic clusters holds considerable promise as an intense THz source. © *2011 American Institute of Physics*. [doi:10.1063/1.3672814]

Terahertz (THz) waves are widely used in many applications such as remote sensing, spectroscopic analysis of organic and inorganic compounds, biomedical diagnostics, and threat detection. To expand the range of possible applications, efficient high-energy sources are needed. Intense THz pulses have been generated using free-electron lasers[1] and employing large nonlinear crystals.[2] Meanwhile, THz radiation from plasmas produced by femtosecond laser pulses has also attracted attention,[3] and THz emissions from gas plasmas, which are convenient and replenishable THz sources, have been extensively studied.[4–6] Different schemes such as electric field biasing of the plasma[4] and frequency-mixing methods[5,6] have been employed to enhance THz pulse energy. Using a frequency-mixing scheme, THz pulses with energy up to 570 nJ at frequencies below 5.5 THz have been generated.[6] Furthermore, Rodriguez and Dakovski[7] showed that, at laser energies higher than 4 mJ, stronger THz radiation is generated from argon gas than from air, neon, krypton, and xenon.

Our initial study indicated that atomic clusters with laser absorption much higher than that of gases are suitable for generating strong THz radiation.[8] To investigate the feasibility of atomic clusters as sources of intense THz radiation, in the present study, we precisely measured the angular distribution of THz waves. Such measurements are important for considering the generation mechanism and estimating the total energy of THz pulses. In a comparison of the THz pulse energy generated under irradiation of femtosecond laser pulses (10–70 mJ) from argon clusters with that from argon gas, an enhancement of two orders of magnitude has been observed.

Femtosecond laser pulses from a chirped-pulse amplified Ti:sapphire laser[9] with energy of 10-70 mJ, duration of 130 fs, and center wavelength of 800 nm were focused by a spherical lens with a focal length of 200 mm, onto argon clusters in a spot with diameter of 17 μm. The experimental setup is schematically shown in Fig. 1. Argon clusters were generated by injecting argon gas[10] with pressure up to 8 MPa into a vacuum chamber with diameter and wall thickness of 100 mm and 5 mm, respectively. Suitable for measuring angular distribution, the chamber was made of fused silica glass with refractive index of 1.95 and transparency of 90% at 0.5 THz.[11] High-energy ions generated by Coulomb explosion of argon clusters, detected in a separate experiment using time-of-flight measurements, were used to confirm cluster production. THz radiation from the argon clusters was collected and collimated by a polyethylene lens (focal length, 150 mm) and directly imaged on the input window of a helium-cooled InSb bolometer by a parabolic mirror (focal length, 119.2 mm). Wire grids polarizers (extinction ratio, $\sim 10^{-5}$ at 0.5 THz) were used to measure the horizontally and vertically polarized components. Multiple layers of polystyrene foam and a thin black polyethylene filter are installed behind the lens and in front of the bolometer, respectively, to exclude the laser pulses and unwanted lights emitted or scattered from the plasma.

The intensity interferogram and spectrum of THz waves were measured with a Martin–Puplett interferometer and the bolometer (spectral responsivity below 2 THz). A typical interferogram of a THz pulse and its power spectrum (backing pressure, 7 MPa; laser energy, 30 mJ) is shown in Fig. 2. The maximum observed frequency was $\sim$1.0 THz with a peak at $\sim$0.5 THz. The spectrum of THz waves did not strongly depend on laser pulse energy, and a nearly identical

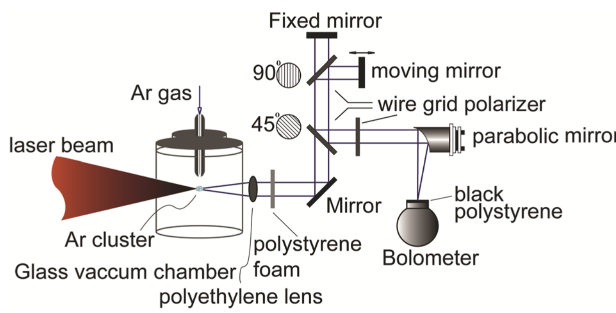

FIG. 1. (Color online) Schematic of experimental setup.

a)Electronic mail: fjahangiri@laser.kuicr.kyoto-u.ac.jp.

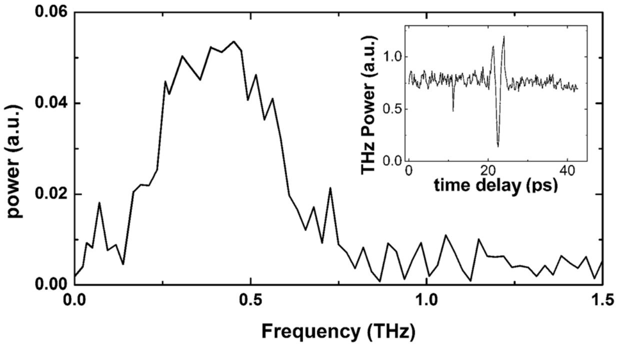

FIG. 2. Typical power spectrum and interferogram (inset) of THz pulse emitted from argon clusters (laser energy, 30 mJ; backing pressure, 7 MPa).

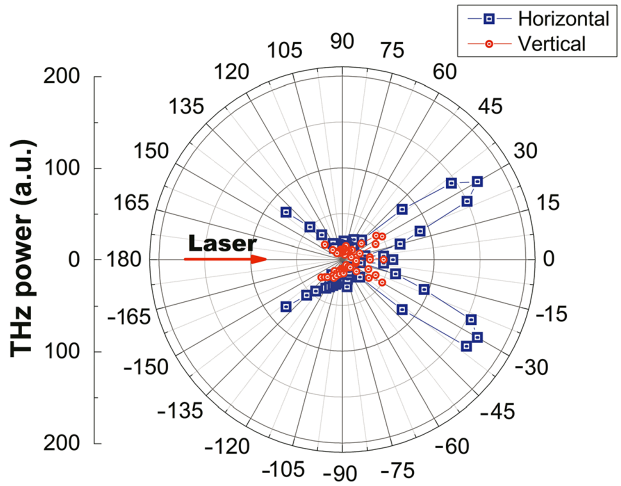

FIG. 3. (Color online) Angular distribution of THz radiation for laser energy of 30 mJ and backing pressure of 7 MPa.

spectrum was observed in the forward and backward directions. The angular distribution of THz waves was measured with resolution of 4° by rotating the detection setup about the center of the glass chamber. Figure 3 shows a typical angular distribution measured at 7 MPa backing pressure. Each data point was plotted by averaging of 150 pulses, and the standard deviation was 5%. The highest THz power was detected at ±30° and ±140° with respect to the laser propagation direction. In a symmetric pattern, the backward peaks are expected to be at ±150°, but the setup configuration limited our measurement to an angle of ±140°. Nonetheless, the peaks observed at ±140° appeared consistent with the expected symmetric pattern. The direction of maximum THz emission was found to depend on plasma length, which was estimated from images of fluorescent light scattered from the plasma, and was controlled by the F-number of the focusing lens. The angle of the maximum THz peak changes in proportion to $(\lambda/L)^{1/2}$, where λ and L are the wavelength and plasma length, respectively (Fig. 4(a)). In addition to the directions of strongest THz emission, considerable THz radiation was also observed in the forward direction (0°). Through the experiments, this emission was found not to be caused by asymmetry in the plasma's shape. The measurements show that the THz waves were radially polarized. However, the THz radiation observed in the forward direction (0°) was elliptically polarized.

We compared the THz radiation from argon clusters with that from argon gas at the same atomic density of $10^{17}\,\mathrm{cm}^{-3}$, for different laser energies. THz radiation from argon gas was generated and detected by using the setup shown in Fig. 1, after filling the chamber with argon gas. At laser energy of 70 mJ, an average signal of 0.2 KV/sr (4 V through an ∼10 mm aperture at 70 mm from the plasma) and 22.5 KV/sr (90 V through an ∼10 mm aperture at 150 mm from the plasma) was detected from the plasma of argon gas and argon cluster, respectively. Slabs of glass with known absorption coefficients[11] were used to attenuate the signal. Therefore, THz energy per unit solid angle observed from argon clusters was about two orders of magnitude higher than that from argon gas. Since the total solid angles, in which these average signals could be detected from argon gas and argon clusters, were ∼0.2 sr and ∼1.1 sr, respectively, a roughly 600-fold enhancement in total THz pulse energy was found. Fig. 5 shows the energy dependence of total THz radiation from argon clusters as compared with that from argon gas, at the same atomic density. The strong THz radiation originates from the local density of the cluster target, which is much higher than that of argon gas and results in high laser absorption. From the bolometer parameters (amplification factor, 100; optical responsivity, 1.6 V/mW), the total THz pulse energy was roughly estimated as ∼1 nJ/pulse for argon gas, which is consistent with previous reports.[3,12]

The THz polarization properties and the dependence of emission divergence on plasma length observed in this experiment are consistent with the expectations for radiation generated by ponderomotive charge separation[3] and transition-Cherenkov radiation (TCR).[12] However, those mechanisms can explain only forward radiation; and backward radiation which has been observed here (Fig. 3) has not

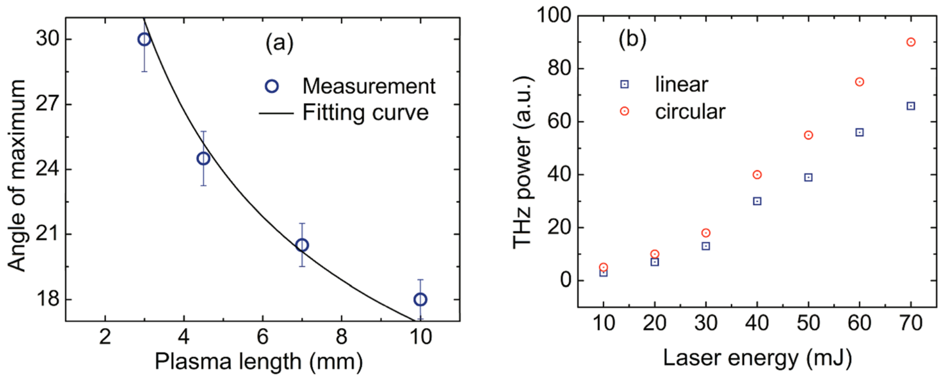

FIG. 4. (Color online) (a) Angle of maximum THz emission versus plasma length. (b) Enhancement of THz wave power in the forward direction by circularly polarized laser light compared with linearly polarized laser light.

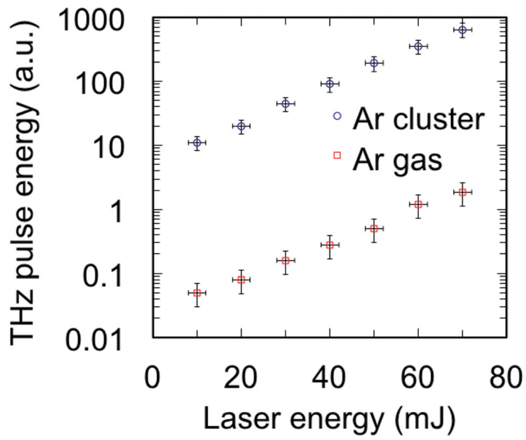

FIG. 5. (Color online) Energy dependence of THz pulse radiation from argon clusters and argon gas, at same atomic density, produced by linearly polarized laser pulses with energy from 10 to 70 mJ.

been theoretically predicted or experimentally observed for those mechanisms.[3,12] The observed four-lobed angular distribution is reminiscent of the radiation pattern expected from time-varying electric quadrupoles,[13,14] which are considered here to explain the whole angular distribution. Ponderomotive force originating from the gradient of the electric field acts on electrons near the laser pulse, expelling them from the laser-produced plasma in a direction parallel or perpendicular to the laser propagation direction. Then, the positively charged core and surrounding electrons are distributed so that the charges of $-e$, $+2e$, and $-e$, where e (>0) is the electron charge, line up in a line segment parallel or perpendicular to the direction of laser pulse propagation, in the region where the laser pulse exists. This series of charges can be considered as a quadrupole. As the laser pulse passes through the plasma, each quadrupole in the plasma oscillates with a period of the order of the femtosecond laser pulse duration and radiation in the THz range is generated. The power of radiation from axial electric quadrupoles is distributed in proportion to $\sin^2(\phi)\cos^2(\phi)$, where ϕ is the angle of the radiation direction with respect to laser propagation direction. The consistency of quadrupole radiation in the forward direction with the TCR mechanism has been recently studied.[14] Regardless of the laser pulse polarization, radially polarized radiation is expected from this mechanism, which is consistent with the polarization properties observed in the present experiment. Furthermore, since the quadrupoles are induced by ponderomotive forces within the plasma, a square dependence on laser energy is expected for THz radiation;[3,15] in fact, such dependence was observed in the present experiment. Therefore, the polarization properties, angular distribution, and energy dependence observed here can be understood on the basis of electric quadrupole radiation. The elliptical polarization of the strong forward (0°) radiation suggests a contribution from four-wave (FW) mixing in the presence of spontaneous second harmonic (SH) light.[16] To test this hypothesis experimentally, we relied on the fact that THz power linearly depends on SH intensity in FW mixing.[5,6] As shown in Fig. 4(b), the energy enhancement observed for circular polarization compared with linear polarization is 1.5-fold, which is the same as the enhancement of SH light[17] for a circularly polarized laser.

In conclusion, the polarization and angular distribution of THz radiation form argon clusters irradiated with intense single-color femtosecond laser pulses were measured, and an enhancement of two orders of magnitude was observed for THz radiation from argon clusters as compared with that from argon gas. The observed properties suggest the laser-induced electric quadrupoles with a contribution from the FW mixing process as the mechanisms of THz generation. Since the enhancement with atomic clusters was obtained via single-color laser excitation, atomic clusters in a two-color excitation scheme have promise as a more intense THz source.

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