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Multimillijoule coherent terahertz bursts from picosecond laser-irradiated metal foils

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High-intensity, picosecond laser pulse irradiating a metal foil. A transient state in materials (2) attribute the THz radiation produced at the rear side of thin solid targets to the target normal sheath acceleration of ions (33). On the other hand, transition radiation, induced by energetic electrons crossing the target–vacuum interface, also contributes (34), which has been proved indirectly by employing different target configurations (29).

Recently, laser-produced plasmas have attracted considerable interest as a damage-free medium for achieving compact intense THz sources (20, 21). Laser-driven gas-density plasmas usually deliver a few microjoules THz energy (22–25), which unfortunately saturates with increasing pump laser energy (26). Laser–solid interactions, by contrast, show advantages in the THz output energy, reaching hundreds of microjoules (27–32). The THz radiation generated usually has an ultrabroadband spectrum up to 30–100 THz, and the underlying THz generation mechanisms have not been fully understood. Gopal et al. (27) attribute the THz radiation produced at the rear side of thin solid targets to the target normal sheath acceleration of ions (33). On the other hand, transition radiation, induced by energetic electrons crossing the target–vacuum interface, also contributes (34), which has been proved indirectly by employing different target configurations (29).

In this report, utilizing an ultraintense, picosecond (ps) laser pulse to irradiate a metal foil, we demonstrate the efficient generation of low-frequency (<3 THz) coherent THz radiation with pulse energies exceeding multimillijoules, surpassing other THz sources (11–32). The direct manipulation of target-rear sheath fields with a controllable prepulse provides direct evidence that the THz radiation mainly originates from the coherent transition radiation induced by energetic electrons transiting the target.
Results

THz Source. The experimental setup (see Materials and Methods for a detailed description) is shown in Fig. 1. A high-intensity picosecond laser pulse was focused onto a copper foil. Intense THz radiation, along with energetic ions and electrons, was emitted from the target-rear surface. To simultaneously characterize the accelerated ions (primarily protons) and escaping electrons in the same laser shots, THz lens systems with limited acceptance angles were used to collect the THz radiation in different directions.

Intense THz radiation was observed. At a laser energy of ~60 J, the THz energy in 0.12 steradian (sr) at 75° was measured to be ~2.3 mJ at frequencies below 20 THz. Spectral measurements, either with a set of low-pass filters or narrowband pass filters, showed that the THz radiation was low-frequency (<3 THz) dominated (Fig. 2A). THz energy measurements at different directions showed that the THz radiation became weak near the rear target normal direction (Fig. 2B). By varying the laser energy, a power-law correlation of the THz energy to the electron charge was observed (Fig. 2C).

THz Generation Mechanism. To identify the roles of sheath fields, ions, and electrons in the THz generation, we modified the sheath fields directly by controlling the preplasma at the target rear. An additional laser beam was introduced and focused to the preplasma behind the main target front surface. The preplasma scale length was adjusted by varying the relative timing between the prepulse and main pulse. With increasing preplasma scale length, the maximum proton energy decreases significantly (Fig. 3A), resulting from the decrease in the sheath-field strength (35). By contrast, both the THz radiation and the electron charge measured increase significantly (Fig. 3A and B). The distinctly different dependence of the THz radiation and sheath fields on preplasma scale length excludes the possibility that the sheath fields and ions contribute significantly to the measured THz radiation.

The coherent transition radiation (CTR) model (36, 37) can readily explain the THz radiation. The CTR energy, \( W_{\text{CTR}} \), generated by an electron bunch with a Boltzmann energy distribution, scales with the bunch charge \( Q \) and the electron temperature \( T_e \) approximately as \( W_{\text{CTR}} \propto Q^2 T_e^{0.22} \) (Fig. 2C, Inset). With increasing laser energy, both \( Q \) and \( T_e \) increase. According to the experimentally measured \( T_e \propto Q^{2.3} \), it is anticipated that \( W_{\text{CTR}} \propto Q^2 \), which explains well the observed power-law fit of \( 2.3 \pm 0.7 \) (Fig. 2C). When introducing the target-rear preplasma, \( T_e \) does not vary much because the target-front electron acceleration is not affected. Hydrodynamic simulations with the MULTI–fs code (38) show that, the preplasma density remains comparably high for the low-frequency THz radiation in the <1-ns evolution time scale (39), and the density scale length at the plasma–vacuum interface remains much less than the THz wavelength (Fig. 3B). In this case, modeling the target-rear surface as a sharp metallic boundary is a good approximation (22, 37, 40), and the conventional CTR scenario is still applicable. This is also suggested by the quadratic dependency shown in Fig. 3C. Based on the measured angular distribution and energy spectra of electrons (Fig. 1 A and B, Insets), one can calculate the radiation spectrum and angular distribution from the CTR theory (Materials and Methods). The calculated values agree well with the measurements (Fig. 2 A and B).

One may wonder whether those low-energy electrons, which are dragged back to the target by the strong sheath field, are capable of forming secondary transition radiation and thus contributing to the total THz radiation. Our experimental results (Fig. 3) answer this question: the electrons confined at the vicinity of the target contribute little to the THz transition radiation. A simplified analytic model is proposed to understand this (Materials and Methods). As shown in Fig. 4A, the radiation spectrum, \( I(\omega) \), emitted by a single electron in the sheath, depends mainly on the lifetime of electrons in the sheath, \( \tau_e \), which is determined by the electron kinetic energy and sheath-field strength. The coherent radiation intensity, \( I(\omega) \), generated by an electron bunch, is approximately given as \( I(\omega) \propto \omega^2 I(\omega) \), where \( F(\omega) \) is the electron bunch form factor. For the picosecond laser bunch in our picosecond laser experiment, \( F(\omega) \) gets rather weak at the frequency region over 1 THz, overlapping...
little with $L_s(\omega)$ especially at high sheath fields (Fig. 4B), and hence those electrons confined in the sheath cannot efficiently generate coherent radiation below 1 THz, but possibly contribute to the weak high-frequency radiation observed in the experiment.

Qualitatively, the adverse role of sheath fields in the CTR can be understood by analogy to the formation-zone effect (41), where the backward radiation generated by electrons reentering the target interferes destructively with the forward radiation generated by electrons exiting the target surface, thus suppressing the total forward THz radiation. When the target-rear pre-pulse is on, the preplasma reduces the sheath-field strength (35), fewer electrons are dragged back to the target, resulting in weaker destructive interference, and meanwhile more electrons escape from the target (39), which more efficiently produces stronger THz radiation (Fig. 3).

**High-Field THz Pump Experiment.** A fraction of the THz radiation was focused onto high-resistivity silicon (Si) wafers. If the THz field is adequately high, the interband luminescence will emerge (42) despite the band gap (1.12 eV) of Si exceeding the central THz photon energy by a factor of $\sim 1,000$. A scientific-grade camera was used to observe the luminescence emitted from the Si samples. Bright near-infrared luminescence was observed within the THz focal spot. The Si wafer with a lower resistivity exhibits brighter luminescence. The luminescence intensity first increases exponentially with the THz energy, and then saturates when the THz energy on the Si sample is over $\sim 100 \mu J$ (Fig. 5) with an estimated field strength of $\sim 0.08$ GV/m at a spot size of $\sim 3$ mm.

The presence of observable luminescence from high-purity Si implies a substantial multiplication in the number of interband electron–hole (e-h) pairs. We attribute the generation of massive e-h carriers mainly to the process of impact ionization (43), where an energetic conduction-band electron collides with a valence-band electron, creating two conduction electrons and a hole. Since the electric field required to ionize the phosphorus donor in Si is only $\sim 180$ kV/cm (44), the impurity donors can be ionized fully on the rising edge of THz pulses, and subsequently are accelerated by the THz field. If an electron gains a kinetic energy exceeding the band gap, impact ionization will occur efficiently (45). After each impact ionization event, the carrier number is doubled, and the original electron and the newly born e-h pair can regain energy from the THz field in a cascade way. Such an avalanche-like ionization behavior will lead to the carrier density increasing exponentially over time and with THz pulse energy. This fully explains the experimentally observed nonlinear behavior of luminescence intensity with the THz energy. On the other hand, the generated high-density carriers enhance the carrier-phonon scattering and the Coulomb scattering among carriers, which will in turn reduce the energy of carriers and suppress impact ionization (43). This may account for the saturation of luminescence observed at high THz energies.

Another mechanism that possibly contributes to the carrier generation at high THz fields is Zener tunneling (42). According

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**Fig. 3.** Identification of THz generation mechanism. (A) Dependence of spectrally integrated THz energy (blue circles) and the maximum proton energy (magenta squares) on the timing of the prepulse at the target rear. (B) Measured electron charge (blue circles) and simulated density scale length at the plasma–vacuum interface (magenta squares) as a function of the timing of prepulse. The error bar of plasma scale length is caused by the different laser intensity used in simulations. (C) Dependence of THz energy on the electron charge and a power-law function fit (red curve).

**Fig. 4.** Radiation of electrons confined in the sheath. The electron kinetic energy is indicated. (A) Calculated normalized radiation spectra generated by a single electron. (Inset) Schematic illustrating the scenario where an electron crosses the target surface, and returns back under the action of the sheath field. The transient electric dipole consisting of the electron and its image charge emits electromagnetic radiation. (B) Central frequency as a function of the sheath-field strength, $E_s$. The cyan area sketches the coherent frequency region for a picosecond electron bunch.
to the interband tunneling model developed by Kane (46), the density of carriers generated via field-induced tunneling is estimated to be only \( 8 \times 10^{20} \text{ cm}^{-3} \) under the bias of 0.08-GV/m THz field lasting 1.5 ps, much less than the impurity concentration in samples. Hence Zener tunneling contributes little to the carrier generation in our experiment.

**Discussion**

Given the good agreement of THz measurements with the CTR model, one can evaluate the total THz pulse energy by extrapolating experimental measurements with model calculations (*Materials and Methods*). At a pump laser energy of \( \sim 60 \text{ J} \) in the case without target-rear preplasmas, the total energy of THz pulses emitted from the target rear is determined to be \( \sim 55 \text{ mJ} (\pm 20\%) \) within the frequency range up to 3 THz. This corresponds to a laser-THz energy conversion efficiency of \( \sim 0.1\% \) and a peak power of \( \sim 36 \text{ GW} \) (Fig. 6) for the pulse duration of \( \sim 1.5 \text{ ps} \) (see Fig. 24 for the retrieved quasi-half-cycle THz waveform). The parameters evaluated above can be even higher since the THz energy can be further boosted by the target-rear preplasma. For the case in which the THz energy is increased by a factor of \( \sim 4 \) (Fig. 3A), the THz electric field, \( E_{\text{THz}} \), at the target-rear surface is estimated to be \( \sim 4 \text{ GV/m} \) (*Materials and Methods*). Although a higher THz peak field of 8.3 GV/m has been reported at a multi-THz central frequency and thus in a much smaller focal spot (17), a comparably low central frequency of \( \sim 0.3 \text{ THz} \) here leads to a much higher ponderomotive potential \( U_p \sim 200 \text{ keV} \). The normalized vector potential, as a critical parameter to characterize the electromagnetic field strength, is estimated as \( a_0 = eE_{\text{THz}}/m_e c \omega_0 \sim 1.2 \), where \( \omega_0 \) is the central angular frequency, \( e \) and \( m_e \) are the charge and mass of the electron, respectively, and \( c \) is the speed of light. This already arrives at the realm of relativistic optics, which was not accessible on a half-cycle time scale previously.

Given that the THz radiation is emitted in a rather large divergence angle (Fig. 2B), one needs to use collection optics with large acceptance angles to deliver more available THz energy for practical applications. For example, if an ellipsoidal mirror with an acceptance solid angle of \( \sim 4 \text{ sr} \) was applied (27, 32), up to \( \sim 70\% \) of the total THz energy would be collected.

In addition to a high THz energy, our experimental results also present approaches to tuning laser-driven CTR-based THz sources. On the one hand, transferring the target-rear preplasma scheme to femtosecond laser systems, one may efficiently obtain millijoule-level THz pulses by adopting hundreds-of-millijoule tabletop femtosecond lasers, which can be operated in a high repetition rate (typically 10 Hz) and be available in university-scale laboratories. On the other hand, by comparing the broadband THz spectra (up to \( \sim 30 \text{ THz} \)) obtained previously in femtosecond laser-driven cases (29–32) with that in our current picosecond laser experiment, it is inferred that the THz spectra can be tunable by varying the pump laser pulse duration.

In conclusion, we have experimentally demonstrated the efficient generation of low-frequency THz pulses with an ultrahigh energy of \( \sim 50 \text{ mJ} \), exceeding other state-of-the-art THz sources by nearly one order of magnitude. A further THz energy boost is obtained by introducing preplasmas to enhance the number of electrons escaping the sheath field generated at the target-rear surface. The application potential of the THz source is illustrated with a preliminary THz pump experiment, where a strong-field THz pulse induces the extraordinary multiplication of carriers in semiconductors. Multimillijoule THz sources reported here could enable the study of relativistic optics in the THz regime. Together with intrinsically synchronized energetic particles and photons generated concomitantly in laser–plasma interactions, more opportunities in the extreme THz science (47) will be opened up via multifunctional pump–probe experiments.

**Materials and Methods**

**Laser System.** The experiment was carried out at the Rutherford Appleton Laboratory using the Vulcan laser (48) operating in a dual-laser beam (B7 and B8) configuration. The beam B7 (~1.5-ps pulse duration, ~1,053-nm central wavelength) as the main pump pulse, was focused by an f/3 off-axis parabolic mirror onto the 100-μm-thick copper (Cu) foil target at an incidence angle of 30° and an ~5-μm focal spot size (full width at half maximum, FWHM). For the maximum laser energy of ~60 J on target, the peak laser intensity was \( \sim 5 \times 10^{19} \text{ W/cm}^2 \) (\( \phi_e \sim 6 \)). The beam B8 (5 ± 2 J, ~10-ps pulse duration) as the prepulse, was focused by an f/15 OAP mirror onto the...
target-rear surface at an incidence angle of 75° and an enlarged focal spot size of ~0.5 mm x 1.9 mm, corresponding to a laser intensity in the range of ~3–6 x 10¹⁷ W/cm². The fairly large spot size of 88 produced a quasi-one-dimensional evolution of the preplasma, and ensured full overlap of the preplasmas with the B7-accelerated electron bunch. Neither THz radiation nor particle acceleration was detected when only B8 was applied on the target. Before the arrival of B7 at the target, the target-front surface was not perturbed by the target-rear preplasma. Evidences of this came from the fact that the target-front electron and optical diagnostic signals did not vary with the prepluse.

**THz Characterization.** The THz radiation at 75°, 45°, −20°, and −40° (Fig. 1D, Inset) with respect to the rear target normal was collected with similar lens configurations, and then relayed into THz energy and spectrum detection systems. To remove radiation at other wavelengths and avoid saturation of the THz detectors, THz filters and high-resistivity Si wafers were placed in the THz path. Owing to the high energies of the THz pulses produced in the experiment, a filter-based THz spectrometer was applied to measure the discretized THz spectrum in a single shot. In the THz spectrometer, the THz beam was split into eight beamlets. Low-pass or narrowband band-pass THz filters with varying cutoff or central frequencies were inserted in different beamlet paths, and the filtered THz radiation was measured by cross-calibrated pyroelectric detectors. The THz spectral intensity is retrieved as \( I_{\text{THz}} = \frac{e^2}{4\pi\varepsilon_0} \left( \frac{\sin^2 \theta}{G_w(\omega_s, \theta)} \right)^2 \), where \( G_w(\omega_s, \theta) = \int d\varepsilon_0 e^{i\varepsilon_0^2} P_c^2 \left( 1 + \Delta^2 \right) \cos^2 \left( \frac{\theta}{\sqrt{1 + p^2 - \frac{1}{1 + p^2}} \cos \theta} \right) \theta \) is the angle between the electron direction and the radiation wave vector.

**Calculation of CTR.** For an electron bunch crossing an ideal conductor–vacuum interface, the CTR energy, \( W_{\text{CTR}} \), emitted per unit angular frequency \( \Delta\omega \) and unit solid angle \( \Omega \) can be expressed as \( W_{\text{CTR}} = Q \int d^2 p \left| p \right|^2 \left( \frac{E}{\theta} \right)^2 \), where \( Q \) is the electron bunch charge, \( g(\theta) \) is the momentum distribution function, which can be decomposed into two parts, the energy spectrum and the angular distribution. Since the transverse formation of the electron and its image charge, the resulting radiation spectral intensity is retrieved as \( I_{\text{THz}} = \frac{e^2}{4\pi\varepsilon_0} \left( \frac{\sin^2 \theta}{G_w(\omega_s, \theta)} \right)^2 \), where \( G_w(\omega_s, \theta) = \int d\varepsilon_0 e^{i\varepsilon_0^2} P_c^2 \left( 1 + \Delta^2 \right) \cos^2 \left( \frac{\theta}{\sqrt{1 + p^2 - \frac{1}{1 + p^2}} \cos \theta} \right) \theta \) is the angle between the electron direction and the radiation wave vector.

**Data Availability.** Data associated with research published in this paper can be accessed at https://edata.stfc.ac.uk/handle/edata/747.

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