**Data associated with the manuscript titled “Towards terawatt-scale spectrally-tunable terahertz pulses via relativistic laser-foil interactions”**

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This dataset includes experimental and numerical data from the manuscript 'Towards terawatt-scale spectrally-tunable terahertz pulses via relativistic laser-foil interactions'. The experimental data are obtained in the experiment titled “Intense terahertz radiation from picosecond laser-produced plasmas” (App No.: 16110035, PI: David Neely), performed at Vulcan TAW. The data are taken to demonstrate efficient production of spectrally-tunable terawatt (TW)-level THz pulses from high-intensity picosecond laser irradiation on a metal foil and identify the underlying physics. The data acquisition and analysis methods can also be found in the manuscript.

**Keywords:** Terahertz (THz) radiation, Laser-plasma interaction, Tunable THz source

**Data description**

1. Data for Figure 1(c): Calculated THz radiation (total, TR, SR and BR) spectra at given parameters as a function of the electron bunch duration.

The THz radiation spectra is calculated numerically by substituting reasonable parameters into the analytically derived formula describing the transition radiation (TR), sheath radiation (SR) and bremsstrahlung-like radiation (BR). A premise is adopted that the electron bunch current remains constant (corresponding to a constant laser intensity and laser absorption efficiency in the experiment).

1. Data for Figure 2: Experimental results and theoretical fitting of THz spectra at given (a) laser pulse durations and (b) target sizes.

Circle points represent the experimental data of the discretized spectra of the THz radiation at 75°, which are characterized with a set of calibrated bandpass filters. The THz radiation is split into multiple beams by high-resistivity silicon beam splitters. In each beam, narrowband THz band-pass filters with different central frequencies *ω*i were inserted. The filtered THz beams were detected with cross-calibrated THz detectors (response range 0.1−250 THz). The whole THz detection system was housed in a grounded metal shielding box to avoid the unwanted interference by the EMP. The THz energy emitted from the target, *S*0, within the transmission bandwidth of the bandpass filters, Δ*ω*i, was retrieved from the signal recorded by the detector, *S*det, via the following formula

*S*det(*ω*i)=*S*0(*ω*i)⋅*T*col(*ω*i)⋅*T*bs(*ω*i)⋅*T*filter(*ω*i)⋅*R*det(*ω*i)

where *T*col, *T*bs and *T*filter represent the transmittance (at the frequency *ω*i) of the THz collection optics, silicon beam splitters and the THz filter, respectively, *R*det is the calibrated detector responsivity. The THz spectral intensity at *ω*i was evaluated eventually as *I*(*ω*i) = *S*0(*ω*i)/(Δ*ω*i⋅ΔΩ).

Curves are fits using the analytical models. In the fitting, the electron bunch duration is taken to be the same as the laser pulse, and the experimentally measured electron temperature, angular distribution and proton energy are used as the input.

3. Data for Figure 3:

(a) Retrieved bunch charge of the escaping electrons as a function of the experimentally measured values.

(b) Retrieved sheath electron density as a function of the target size and laser pulse duration.

(c) Retrieved SR peak spectral intensity and sheath charge, and their dependence on the measured proton charge.

(d) Total SR energy and proton beam energy as a function of the retrieved electron sheath energy.

The retrieved parameters are obtained from the modeling fitting of the experimental spectra. The experimental values for electrons and protons are obtained with the following methods. An electron spectrometer placed in the laser forward direction is used to measure the energy spectrum of the escaping electrons. A wraparound array of optical fibers is used to sample electrons at different directions. After calibration with wraparound image plate stacks, the electron charge is obtained by integrating the signal from each fiber loop, with consideration of the correction factor caused by the bend loss in the fiber. Protons emitted in the rear target normal are detected with a Thomson parabola spectrometer. The proton charge was obtained by integrating the measured proton energy spectrum, and the total proton beam energy was evaluated by considering the proton beam’s spatial distribution measured with stacked dosimetry films (RCFs).

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