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# A candidate material $\text{EuSn}_2\text{As}_2$ -based terahertz direct detection and imaging

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Exploring the photoelectric performance of emerging materials represented by graphene, black phosphorus and transition metal dichalcogenides is attracting enormous research interest for a wide range of electronic and photonic applications. The realization of low-power consuming photodetectors with high sensitivity and fast photoresponse in the terahertz band remains one of the profound challenges in optoelectronics. In this study, a material- $\text{EuSn}_2\text{As}_2$  has been successfully implemented to realize highly sensitive terahertz photodetectors. The non-equilibrium dynamics in a two-dimensional plane allow an optionally switching between different styles of direct photon-conversions: the analogous photoconductive and photovoltaic modes spontaneously supported by the intrinsic electronic system. The prototype devices exhibited excellent sensitivity of 0.2–1.6 A/W (0.3–2.4 kV/W) from 0.02 to 0.30 THz at room temperature, corresponding to a noise-equivalent power  $<30\text{pW/Hz}^{0.5}$  and a fast response time  $<16\ \mu\text{s}$ . The versatile switching behaviour and performance of the  $\text{EuSn}_2\text{As}_2$  flakes-based terahertz detectors were validated via rigorous full-dimension and imaging experiments. These results open the feasibility avenues for low-energy photoelectric applications of  $\text{EuSn}_2\text{As}_2$  material.

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## INTRODUCTION

Terahertz (THz) radiation (0.1–10 THz), which connects electronics to optics, encompasses a wide band of the electromagnetic spectrum from microwaves to infrared light, and its mixed property enables broad applications, including wireless communication, spectroscopy, sensing, and imaging<sup>1</sup>. The emerging materials such as graphene, topological insulators, and three-dimensional Dirac systems, with low-energy electrostatics is characterized by mass-less Dirac fermions, exhibit an intrinsically THz response<sup>2,3</sup>. However, THz detection based on layered two-dimensional (2D) materials like graphene<sup>4</sup>, and black phosphorus<sup>5</sup> exhibits several drawbacks imposed by a slow response speed, high manufacturing costs, and the necessity of cryogenic cooling because of the strong background thermal noise at room temperature. Photodetection makes use of topological properties in system supporting gapped bulk excitation, opening feasibility of convenient platform driven by the global properties of quantum wavefunction to beyond traditional landmarks of band-structure engineering<sup>6</sup>. The photocurrent generation in topological insulators, mainly based on  $\text{Bi}_2\text{Se}_3$ ,  $\text{Bi}_2\text{Te}_3$  and  $\text{Sb}_2\text{Te}_3$  materials, has been studied under different configurations such as overdamped plasma waves<sup>7</sup>, and asymmetric scattering of topological surface states<sup>8</sup>, with an aim to achieve a detectivity, comparable to or even exceeding that of graphene<sup>9</sup>. Topological semi-metallic materials such as  $\text{Cd}_3\text{As}_2$ ,  $\text{MoTe}_2$  and  $\text{TaIrTe}_4$  have been recently introduced into the field of photoelectric detection<sup>10</sup>. Especially, the Berry-curvature-enhanced giant photoreponse of these semimetals enables their highly sensitivity photodetection in the technologically significant mid-infrared and THz regions. Thus, the practical applications of THz detection

can be realized by exploring emerging materials with attractive properties, especially a fast and improved response to THz radiation.

The existing photodetectors generally detect high-energy photons above the bandgap energy via photoconductive or photovoltaic effect. However, detection via thermal effects such as bolometric, photothermoelectric (PTE) and pyroelectric effects can occur theoretically for any specific wavelength without the bandgap limitation. Among these photodetectors, the PTE-based detectors are considered as viable candidates for THz detection because of their simple geometry, zero-bias operation, and low power consumption. Wang et al. demonstrated<sup>11</sup> the PTE-based detection capability of the  $\text{EuBiSe}_3$  single crystal, which exhibited a room-temperature self-powered photovoltage responsivity up to 0.59 V/W at THz frequencies, and a noise-equivalent power below 1 nW/Hz lower than reported photothermoelectric detectors by at least an order of magnitude. Recently, quantum materials<sup>12</sup>  $\text{EuSn}_2\text{As}_2$ ,  $\text{MnBi}_{2n}\text{Te}_{3n+1}$  and  $\text{Co}_3\text{Sn}_2\text{S}_2$  have garnered significant attention because of their phenomena such as the quantum anomalous Hall effect (even at room temperature), chiral Majorana fermions and axion electrodynamics<sup>13</sup>. However, the photodetection capabilities of these materials have been rarely investigated, possibility because of the difficulty in manipulating their inherent properties, leading to a general consensus on their inapplicability in terahertz electronics.

In this communication, we reported the candidate material  $\text{EuSn}_2\text{As}_2$ —as an ideal platform to study 2D plane THz photoresponse that relates to the intrinsic electronic system.  $\text{EuSn}_2\text{As}_2$  possesses an unconventional bulk band structure, in which metal states contribute significantly below the Fermi energy level while

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the nonmetal states contribute significantly above the Fermi energy level<sup>12</sup>. The low-energy-photons absorption exhibited by  $\text{EuSn}_2\text{As}_2$  is related to the low heat capacity associated with the mass-less Dirac electrons. The temperature of non-equilibrium carriers due to the THz illumination can be a strong modification of the electronic chemical potential, although some deviation may occur due to thermal excitation of electrons from the surface and bulk because of the presence of massive bulk carriers. The definite feature of an  $\text{EuSn}_2\text{As}_2$ -based photodetector is its strong photoresponse up to THz frequencies, followed by a flexible switching of operation with the involvement of analogous photoconductive and photovoltaic modes under the electromagnetic driven force. Based on the experimental results presented,  $\text{EuSn}_2\text{As}_2$  material can be heralded as a promising candidate for realizing a highly sensitive and fast low-energy photodetection, promoting cutting-edge research and technological advancements in the field of THz optoelectronics.

## RESULTS

### Materials characterization

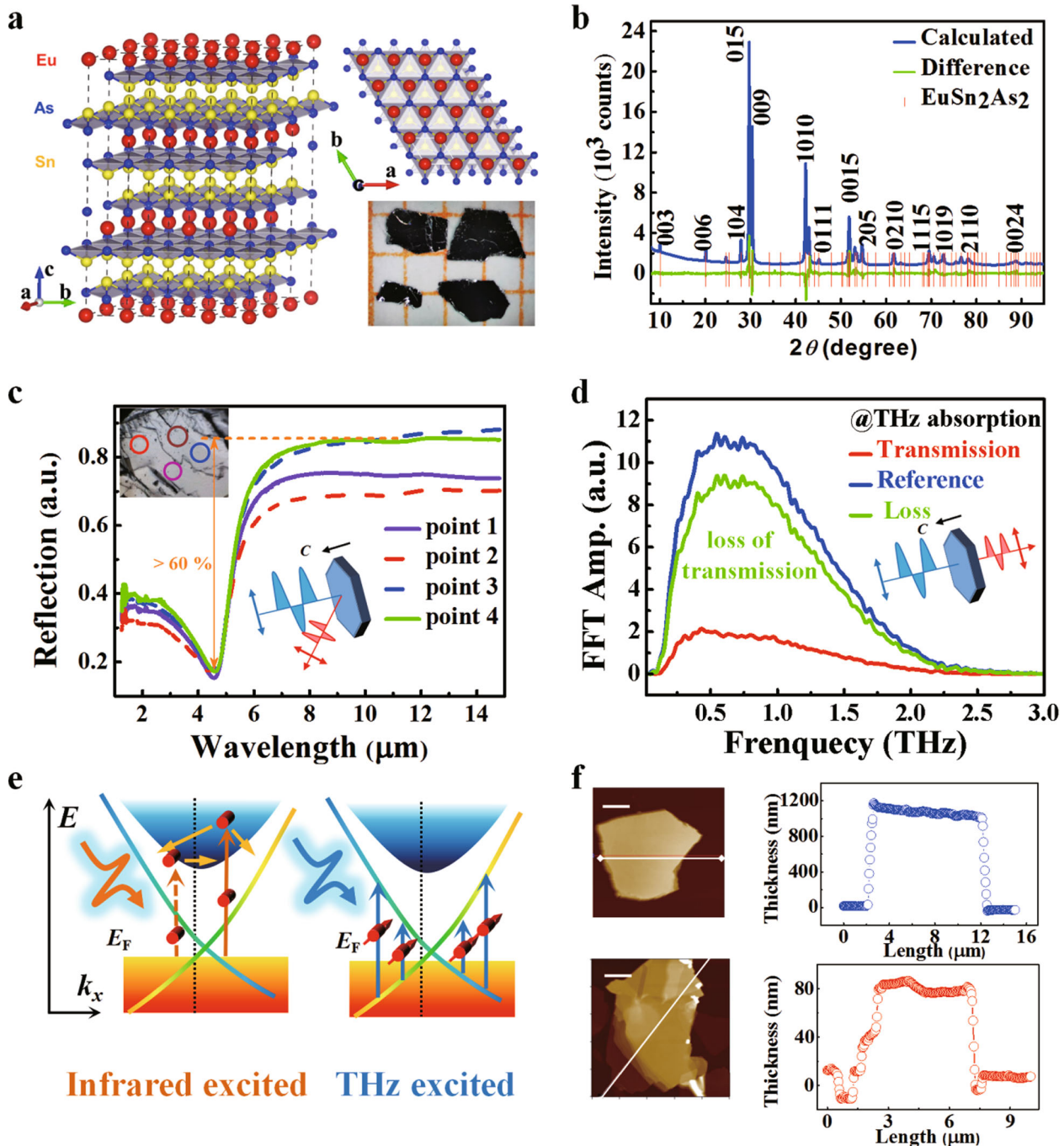
Single-crystal  $\text{EuSn}_2\text{As}_2$  were synthesized by melting stoichiometric amounts of Eu: Sn: As (1.1: 20: 2) in an alumina crucible enclosed in a quartz tube jacket under 50 mTorr pressures and at 850 °C for 12 h (See the “Methods” section for details).  $\text{EuSn}_2\text{As}_2$  flakes, exfoliated from the bulk part, are integrated with multiple antennas through a combination of lithography, electron-beam evaporation and lift-off processes (See Supplementary Methods), to enhance the photodetection sensitivity up to THz frequencies. The photosensitive  $\text{EuSn}_2\text{As}_2$  crystal structure consists of hexagonal Eu, and As layers surrounding an Sn bi-layer as shown in Fig. 1a<sup>14</sup>. Similar to that observed in  $\text{EuSn}_2\text{P}_2$  material<sup>15</sup>, the results from single crystal X-ray diffraction patterns reveal  $\text{EuSn}_2\text{As}_2$  crystallizes in a rhombohedral lattice with the space group  $\bar{R}3m$  analogous to a well-known  $\text{Bi}_2\text{Te}_3$  material. To elucidate the photodetection behaviour, the spectroscopy in relevant with low-energy or ground excited-states was performed to identify the capability of optical transition. The infrared reflection spectrum as shown in Fig. 1c demonstrates that  $\text{EuSn}_2\text{As}_2$  material exhibits a strong absorption of up to 60% at  $\sim 5 \mu\text{m}$  of characteristic wavelength (photon energy: 240 meV). High-energy photons (energy > 240 meV) can excite electron-hole pairs and induce relaxation over the bandgap, whereas the middle energy photons (energy < 240 meV) can be strongly reflected and vanished because of the metallic state. The optical transition to the bulk states increases the absorption of photons with energy  $\sim 240$  meV (wavelength  $\sim 5 \mu\text{m}$ ) because of the scattering between the bulk and the metallic states. Further, the transmission spectrum of the  $\text{EuSn}_2\text{As}_2$  single crystal in the frequency range of 0.02–3.0 THz was obtained by THz spectroscopic measurement techniques, displaying a huge transmission loss or a broadband absorption in this particular frequency regime as depicted in Fig. 1d (See Supplementary Fig. 4a, b). The spectroscopic results indicate that the excitation processes<sup>16</sup> shown in Fig. 1e induce the broadband absorption in  $\text{EuSn}_2\text{As}_2$  material. Photons with energy higher than the bulk bandgap value can excite the electrons in the valence band to the corresponding conduction band as illustrated on the left side of Fig. 1e. For THz photons with energies within the absorption band of the metallic state of  $\text{EuSn}_2\text{As}_2$ , the observed enhanced absorption might be ascribed to the gapless metallic state-induced intra-Dirac-band excitation dynamics of the Dirac electrons (free-carrier absorption) as shown on the right side of Fig. 1e<sup>17</sup>. The visible/infrared absorption possibly originates from the bulk state, whereas the metallic state is responsible for the absorption in the microwave/THz band. To devise our detectors, the topographic atomic force microscopy (AFM) scan of derived  $\text{EuSn}_2\text{As}_2$  flakes, shown in Fig. 1f, substantiates the layered feature

with two thicknesses of  $\sim 1 \mu\text{m}$  and 100 nm, indicating that the exfoliation of large-areas atomically-thin layered  $\text{EuSn}_2\text{As}_2$  is a challenging task. To ensure an ideal trade-off between the detector fabrication and the coupling performance, flakes with thicknesses in the range of 80–100 nm, which is roughly coincident with the out-of-plane thickness of the coupling antenna, are preferred.

### Device design and characterizations

Figure 2a schematically illustrates the structure of the multiple-antenna-coupled  $\text{EuSn}_2\text{As}_2$  material detectors for THz band detection at room temperature. The photosensitive area located at the intersection of the pair of crossed bow-tie antennas (similar to the four-leaf clover shape), which has a total length ( $2R$ ) of  $\sim 400 \mu\text{m}$  and a flare angle of  $45^\circ$ , in resonance with 0.375 THz radiation covering the measured frequency range. The channel length ( $L$  of antenna gap) is  $8 \mu\text{m}$  ( $2r = 8 \mu\text{m}$ ), forming a photosensitive area of  $S_a = \pi r^2 = 50.24 \mu\text{m}^2$  (see Supplementary Fig. 3a for the detector image). Owing to the strong dipole-oscillating charges in the multiple-antenna configurations, shown in Fig. 2b, the deep-subwavelength gap ( $L = 2r = 8 \mu\text{m} \approx \lambda/3000$ ) between the antenna pairs behaves as a gap-capacitor charged by the THz-induced currents. The oscillating electric field, under different polarizations ( $0^\circ$  and  $45^\circ$ ) of the incident 0.1 THz and 0.3 THz radiation, displays an enhancement of several orders of magnitude (an enhancement of power  $|E/E_0|^2 = 200\text{--}300$  times) at the gap to facilitate electromagnetic absorption by the  $\text{EuSn}_2\text{As}_2$  flakes<sup>18</sup> (see Supplementary Figs. 1 and 2 for the local field distribution at 0.02–0.6 THz). Based on the FDTD simulation analysis, it could be inferred that a net photocurrent can flow to different directions in the antenna-coupled detectors owing to the changes of the localized field distributions in a 2D plane ( $x$ – $y$  plane) of the material at different electromagnetic polarizations<sup>19</sup>. The standard behaviours of the different detection modes can be understood using the simplified model shown in Fig. 2c under low-resistance states circumstances (see Supplementary Fig. 3 for electrical characteristics). A probable explanation for the low-energy-photon absorption of  $\text{EuSn}_2\text{As}_2$  materials, similar to graphene, is related to its low heat capacity associated with the mass-less Dirac electrons. The temperature of the electron bath increases significantly upon an intense THz excitation, thus resulting in a response dominated by incoherent thermal effects due to a less effective interaction between the electrons and the lattice. The increase in the electronic temperature under the THz illumination can strongly modify the electronic chemical potential<sup>17</sup>, indicating that the excited non-equilibrium carrier's temperature ( $\Delta T_e$ ) is much higher than the lattice temperature ( $\Delta T_l$ ) under THz radiation. Consequently, a 2D potential gradient can form in the 2D photosensitive area because of the interplay between the radiation-induced heating and heat-sinking of the antenna pads. This potential gradient can trap and de-trap the non-equilibrium carriers under an applied bias voltage, resulting in a directional photocurrent  $I_{\text{ph}} \sim (E_{\text{THZ}}^2(t) + E_{\text{dc}}^2(t))$  with a high photoconductive gain (The photocurrent direction is the same as that of the applied bias voltage). Moreover, a non-directional photocurrent  $I_{\text{ph}} \sim E_{\text{THZ}}^2$  exists even in the absence of applied voltage (The direction of this non-directional photocurrent is different from that of the applied bias voltage), and is steered by the localized field, non-equilibrium carriers' distributions or the Bloch electrons scattering process described later<sup>20</sup>.

To clarify the above-mentioned phenomenon, the current characteristics of the devices fabricated in this study, were recorded under the “ON/OFF” modulated radiation illumination as shown in Fig. 2d, e. The photocurrent ( $I_{\text{ON}}/I_{\text{OFF}}$ ) is deemed to be more than  $1 \mu\text{A}$  at room temperature, can even increase up to  $6 \mu\text{A}$  in the absence of voltage (See Supplementary Fig. 5), revealing a broadband response in THz band irrespective of the

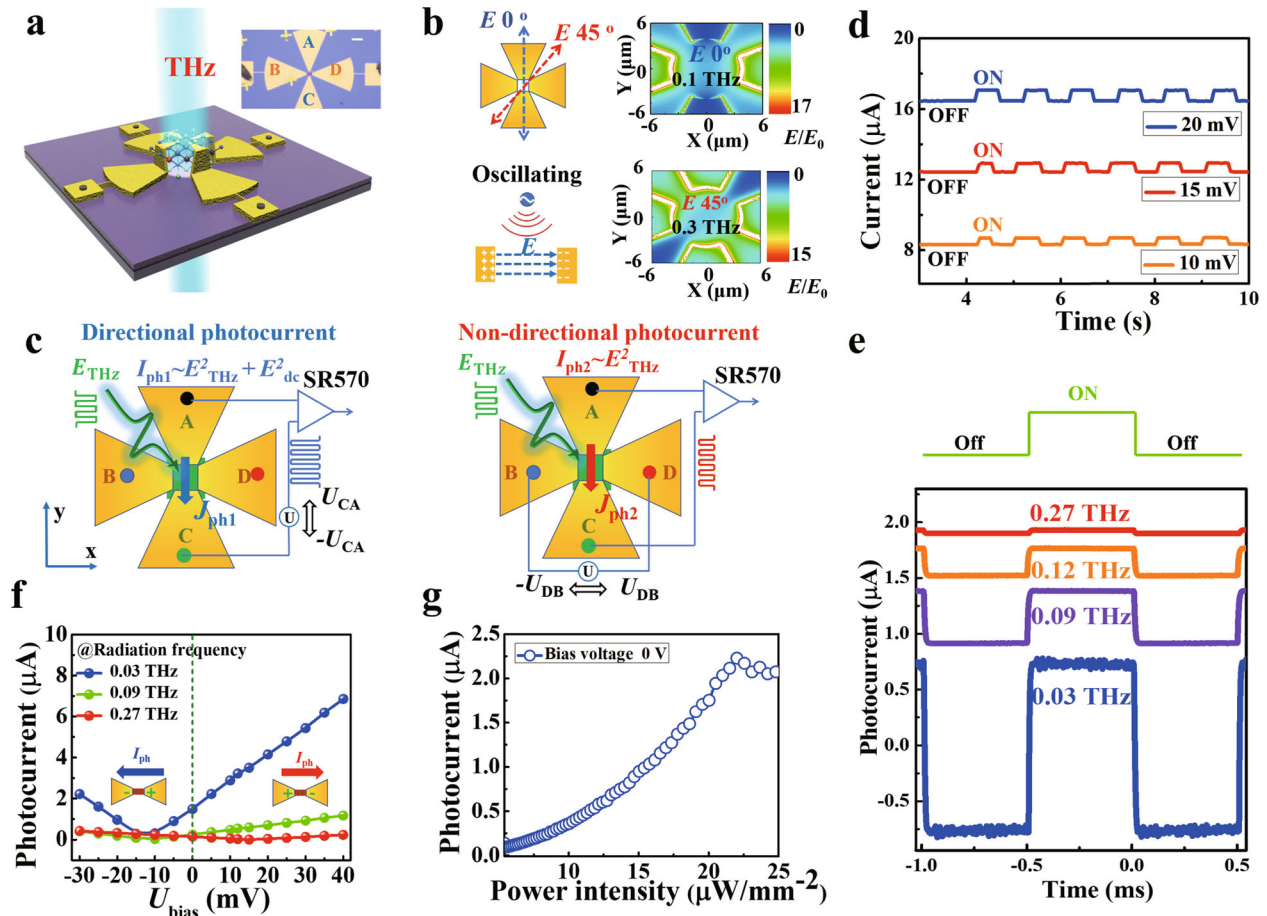


**Fig. 1 Basic characterization of the single-crystal  $\text{EuSn}_2\text{As}_2$ .** **a** Crystalline structures of  $\text{EuSn}_2\text{As}_2$ , in which the atoms are arranged in puckered honeycomb layers bounded together by van der Waals' forces, and an optical image at the corner with the grid length of 1 mm. **b** X-ray diffraction spectrum of the  $\text{EuSn}_2\text{As}_2$  flakes with the corresponding Miller indices in parentheses. **c** Infrared reflection spectra at different sampling points. **d** The THz frequency-domain transmission spectra of with/without the samples. **e** Schematic illustration of the broadband absorption mechanisms between the bulk state and the gapless metallic state; and the blue and orange arrows indicate the low- and high-energy photons excitations, and the red solid circle indicates an electron with a spin, respectively. **f** The AFM image of  $\text{EuSn}_2\text{As}_2$  flakes for device patterning indicates the thickness of the sample, and the scale bars are 4  $\mu\text{m}$  (upper) and 2  $\mu\text{m}$  (lower).

different coupling efficiency of the frequency-dependent antenna (See Supplementary Fig. 2 for the local field distribution). Notably, the photocurrent ( $I_{\text{ph-CA}}$  or  $I_{\text{ph-DB}}$ ) increases linearly with the increasing bias voltage ( $U_{\text{CA}}$  or  $U_{\text{DB}}$ ) at 0.03 THz, 0.09 THz and 0.27 THz as shown in Fig. 2f in accordance with the photoconduction routes (See Supplementary Fig. 5b in detail). In addition, the zero-bias photocurrent component ( $I_{\text{ph-CA}}$  or  $I_{\text{ph-DB}}$ ), shown in Fig. 2g, survives during these measurements, and demonstrates a sub-

linear dependency of the zero-bias photocurrent on the power intensity, validating the effectiveness of the analogous photovoltaic mode. The experimental results shown in Figs. 1 and 2 suggest that the intra-Dirac-band excitation enable the detection of low-energy photons below the bandgap energy (photocurrent  $I_{\text{ph}} > 1 \mu\text{A}$ , even up to 6  $\mu\text{A}$  at RT as shown in Supplementary Fig. 5), owing to the stable non-equilibrium dynamics of the electronic collective system. In contrast, for a weak photocurrent





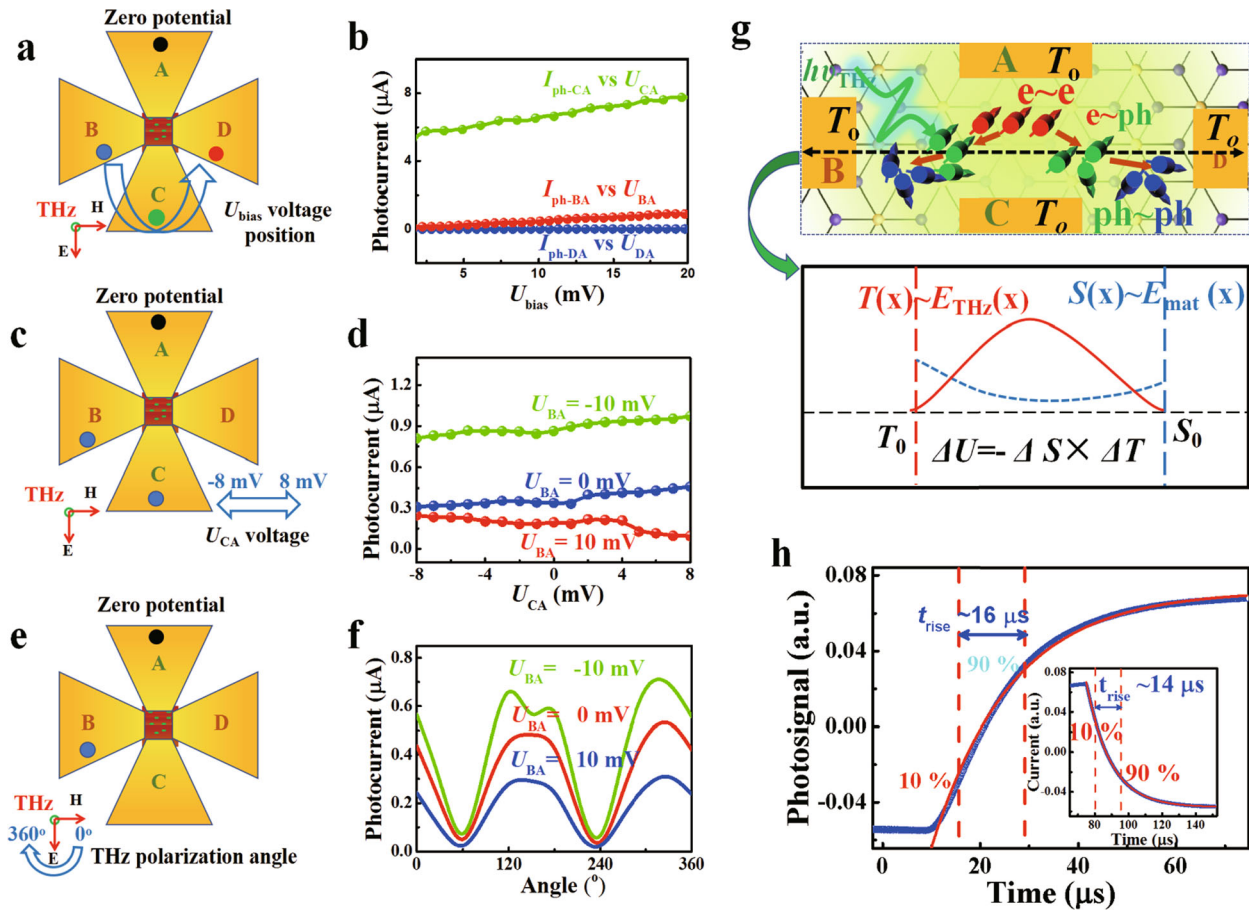
**Fig. 2** The  $\text{EuSn}_2\text{As}_2$  flakes-based THz detector and its photoresponse features. **a** Schematic illustration of the detector exploiting  $\text{EuSn}_2\text{As}_2$  flakes integrated with multiple bow-tie antennas; inset: an optical micrograph image of the detector with the scale bar of  $50 \mu\text{m}$ . **b** The FDTD simulations of the field distributions in the antenna gap at 0.1 and 0.3 THz radiation with incident polarization angles of  $0^\circ$  and  $45^\circ$ . Right side, the antenna-coupled incident electric field ratio  $E/E_0$  onto the active area. **c** THz detection with two representative photocurrents (directional and non-directional photocurrents), where  $E_{\text{THz}}$  is the THz field,  $E_{\text{dc}}$  is the applied DC electric field and  $J_{\text{ph}}$  ( $J_{\text{ph1}}$  and  $J_{\text{ph2}}$ ) is the photocurrent density. **d**, **e** Temporal photocurrent  $I_{\text{ph}}$  at bias voltage range of 10–30 mV and under 0.02–0.27 THz radiation; “ON” and “OFF” stand for illumination and no illumination, respectively. **f** Photocurrent ( $I_{\text{ph-CA}}$  or  $I_{\text{ph-DB}}$ ) as a function of the bias voltage  $U_{\text{bias}}$  ( $U_{\text{CA}}$  or  $U_{\text{DB}}$ ) under 0.03 THz, 0.09 THz and 0.27 THz radiation. **g** Photocurrent ( $I_{\text{ph}}$ ) as a function of the power density ( $P_{\text{density}}$ ) at a zero-bias voltage.

(photocurrent  $I_{\text{ph}} < 2 \text{ nA}$  at RT under a laser output power of 30 mW as indicated in Supplementary Fig. 3b) generated by visible and infrared photons, the intra-Dirac-band excitation is eliminated owing to the fast relaxation of the carriers before they reach the electrodes after the visible/infrared photoexcitation within a timescale of few femtoseconds to picoseconds<sup>21</sup>.

### Photoresponse controlling effects and non-equilibrium dynamics

Based on the above-mentioned analyses, it can be expected to occur that a net photocurrent flows in different directions and can be changed by tuning the applied voltage in the designed device. Its dominant dynamics depend on the following factors<sup>22</sup>: (i) the symmetry (or asymmetry) of the antenna and metal-materials contact; and (ii) the relative orientation between the electric field (or THz polarization) and the crystallographic axis of the material. Based on this, the photoelectric conversion process was vigorously evaluated via different designated strategies. “Strategy A” was designated to extrude the toggled phenomena stemming from the anisotropy of the crystallographic orientation as shown in Fig. 3a. Under the same THz polarization, the different linear behaviour of the photocurrent ( $I_{\text{ph-BA}}$ ,  $I_{\text{ph-CA}}$ ,  $I_{\text{ph-DA}}$ ) with the sweeping voltage ( $U_{\text{BA}}$ ,  $U_{\text{CA}}$ ,  $U_{\text{DA}}$ ) (because of the anisotropy of the orientation resistivity<sup>23</sup> as depicted in Supplementary Fig. 3a) is well preserved

as shown in Fig. 3b, which conforms to the phenomenological explanation of photoconduction features. Based on this analysis, it can be concluded that directional photocurrent can be severely affected by combined effects of the non-equilibrium carriers’ distribution and the orientation resistivity at different electrostatic contact points (electrode B, C, D). “Strategy B” was designed to examine the effect of boundary potential on the photoconduction phenomenon as shown in Fig. 3c. Under the same THz polarization and different bias voltage  $U_{\text{BA}}$  (10 mV, 0, and  $-10$  mV), the sub-linear behaviour of the photocurrent ( $I_{\text{ph-BA}}$ ) with the sweeping voltage ( $U_{\text{CA}}$ ) from  $-8$  mV to 8 mV, is evidently compensated by the photocurrent spilled over from the two bias voltages ( $U_{\text{BA}}$  and  $U_{\text{CA}}$ ) as shown in Fig. 3d. Notably, the photoconduction is possibly affected by the following phenomena: (i) the boundary potential-induced changes in the THz field distribution or non-equilibrium carriers distribution (See Supplementary Fig. 3c for the peak response variation at different bias voltages), and (ii) the existence of two photosensitive areas ( $S_{\text{B-A}}$  and  $S_{\text{C-A}}$ ) that produce compensated photocurrents. “Strategy C” was designed to certify the non-equilibrium carriers’ distribution induced by different THz polarizations as shown in Fig. 3e. Under the same bias voltage ( $U_{\text{BA}}$ ) and without any boundary voltage, the THz-light polarization angle ( $\theta$ ) dependence of the photocurrent  $I_{\text{ph-BA}}$  can be well fitted by a simple sine function ( $I_{\text{ph-BA}} \sim$

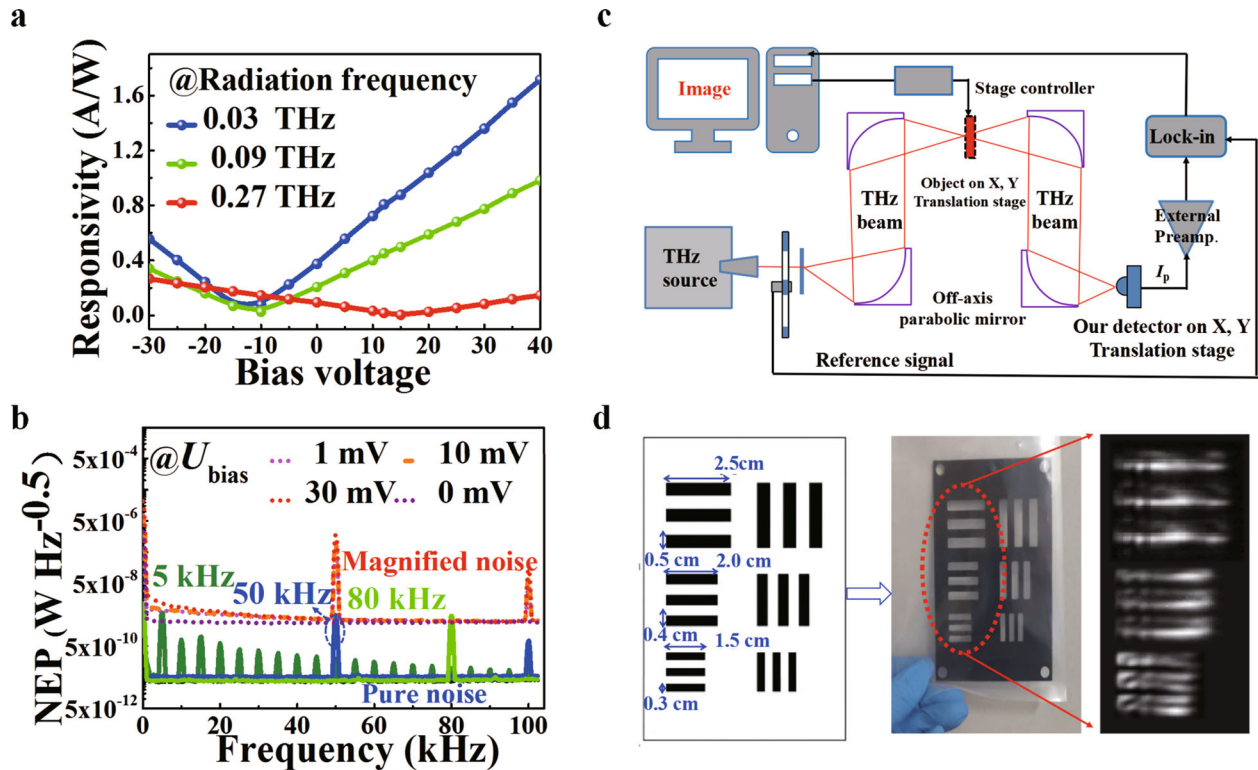


**Fig. 3 Multi-dimensional controlling effects under 0.03 THz radiation.** **a, b** Schematic and the photocurrents ( $I_{\text{ph-BA}}$ ,  $I_{\text{ph-CA}}$ ,  $I_{\text{ph-DA}}$ ) were measured under a sweeping voltage,  $U_{\text{bias}}$  ( $U_{\text{BA}}$ ,  $U_{\text{CA}}$ ,  $U_{\text{DA}}$ ), from 0 mV to 20 mV at different electrode positions. **c, d** Schematic and the photocurrent ( $I_{\text{ph-BA}}$ ) was measured under a sweeping voltage,  $U_{\text{CA}}$ , from  $-8$  mV to  $8$  mV at the same material orientation. **e, f** Schematic and the photocurrent ( $I_{\text{ph-BA}}$ ) measured under a sweeping THz polarization angle ( $0^\circ$  to  $360^\circ$ ) at a certain voltage  $U_{\text{BA}}$  ( $-10$  mV,  $0$  mV and  $10$  mV). **g** Proposed non-equilibrium carriers' transport under electromagnetic radiation:  $T_0$  is the reference temperature. Only the intraband free carrier (Drude) absorption can be generated at such a low-photon energy.  $T_{\text{materials}} > T_{\text{sink}} \sim T_0$ , where  $T_{\text{materials}}$  is the temperature of the flakes, and  $T_{\text{sink}}$  is the temperature of the metal sink. **h** Time-resolved photoresponse showing the deduced response time,  $\tau_{\text{rise/fall}}$  near  $16 \mu\text{s}$  and  $14 \mu\text{s}$ , with the fitting function:  $\exp(\pm t/\tau)$ .

$E_{\text{THz}}^2 \times \sin\theta$ ) as presented in Fig. 3f, revealing that the PTE effect depends on the square of the electric field. Thus, the polarization-dependent photocurrents ( $I_{\text{ph-CA}}$  and  $I_{\text{ph-BD}}$ ) are not caused by the accidental symmetry breaking, and show a  $\pi/2$  phase-shift<sup>24</sup>.

Based on the aforementioned design, analysis and experimental results, the operational principle of the fabricated detectors can be reasonably explained by the activated non-equilibrium dynamics of the intrinsic electronic system, and this tentative assignment comes with a few caveats. Owing to the free-carrier absorption predominantly induced by the intrinsic electronic system<sup>17</sup>, the non-equilibrium carrier's temperature ( $\Delta T_e$ ), is higher than the lattice temperature ( $\Delta T_l$ ) and is not uniformly distributed owing to the higher thermal conductance of the metal contacts and the non-uniform polarized THz coupling when  $\text{EuSn}_2\text{As}_2$  is impinged by the THz photons as shown in Fig. 3g. Therefore, a temperature gradient of  $\nabla T_e(x, y)$  along 2D photosensitive area can be formed. In addition, stemming from the intermixing of the different nanoscale junctions, the  $\text{EuSn}_2\text{As}_2$  layer thickness, and the metal- $\text{EuSn}_2\text{As}_2$  contact, the Seebeck coefficient,  $S(x, y)$  ( $S = -\pi^2 k_B^2 T d\sigma/dE_F/3e\sigma$  given by the Mott relation)<sup>25</sup> is not completely symmetric across the photosensitive region (See Supplementary Figs. 7 and 8 for asymmetric photoresponse caused by the non-uniform contact and impedance). Consequently, the diffusion of non-equilibrium carriers creates a potential gradient  $\nabla U(x, y) = -\nabla S(x, y) \times \nabla T_e(x, y)$ , and the total signal is the integral of  $\nabla U(x, y)$  over the 2D photosensitive region.

Based on this, the graphene- $\text{EuSn}_2\text{As}_2$  flakes-metal heterojunction device exhibits a large asymmetry, and a large photocurrent is expected to achieve a higher detection performance<sup>26</sup>. Apart from the explanation of non-equilibrium carriers-induced non-directional photocurrent as shown in Fig. 2c mentioned above, a rectification detection mechanism attributed to skew scattering due to the emergence of inherent chirality of the itinerant electrons in a time-reversal invariant but inversion-breaking system can also prevail<sup>21</sup>. Naively, the intriguing anisotropic properties may<sup>15</sup> influence the electron's trajectory under the condition of external excitation (low temperature or other factors), and this anisotropy combined with the chiral Bloch electrons induce skew scattering under both the crystal and electromagnetic fields<sup>23</sup>. The anisotropic scattering of the chiral Bloch electrons and pertinent second-order nonlinear response of  $\text{EuSn}_2\text{As}_2$ -based THz detectors possibly induce a non-directional photocurrent (similar to the scattering photocurrent produced by the Hall effect) in response to the THz field. Although the conclusive mechanisms for scattering origins remain elusive, these mechanisms are possibly responsible for the low-energy high-sensitivity photoresponse inherent to  $\text{EuSn}_2\text{As}_2$  material. To better verify the existence of non-equilibrium carriers' processes in terms of response speed, the time-resolved photocurrent response was evaluated, which showed the response time  $\tau_{\text{rise/fall}}$  near  $16 \mu\text{s}$  and  $14 \mu\text{s}$  with a fitting function of  $\exp(t/\tau)$  under  $0.03$  THz radiation (See Supplementary Figs. 6 and 7a for fast response and high signal-to-



**Fig. 4 Performance evaluation of  $\text{EuSn}_2\text{As}_2$ -based THz detectors.** **a** Current responsivity as a function of bias voltage; input power  $P_{\text{in}} \sim P_{\text{density}} \times S_{\text{antenna}}$ ; here,  $S_{\text{antenna}} = \pi R^2 \sim 0.13 \text{ mm}^2$  is the whole area of the antenna, and  $P_{\text{density}}$  is the power density reaching the detector. **b** NEP of the detector under the without/with modulated radiation (5 kHz, 50 kHz, and 80 kHz) and bias voltages (0 mV, 1 mV, 10 mV and 30 mV). The pure noise of a single device is indicated using the solid line ( $U_{\text{bias}} = 0 \text{ mV}$ ), and the magnified noise (>100 times) of single device and test system is indicated using the dotted line. **c** Schematic of transmission-mode THz imaging for macroscopic objects. **d** Image of a scale plate scanned at 0.27 THz in the single-pixel transmission mode.

noise ratio)<sup>27–33</sup>. Moreover, the fast photoresponse can be attributed to the absence of slow trapping-related carrier recombination owing to an intrinsic electronic system involving low-energy photons. Moreover, the static electric field accelerates the photo-excited carriers towards the electrodes quickly, thereby facilitating an efficient charge carriers' collection<sup>8</sup>.

### Detection performance and imaging application

Further, the performance of the detector in reference to the active structure-area rather than the  $\text{EuSn}_2\text{As}_2$ -material's area (See Supplementary Fig. 9 for high sensitivity by all-around calculated responsivity), was analyzed as well. The current responsivity  $R_I = 0.2\text{--}1.6 \text{ A/W}$  (cal.  $R_v = 0.3\text{--}2.4 \text{ kV/W}$ ) is observed to grow linearly with the bias voltage, even when the frequency is extended to 0.27 THz as shown in Fig. 4a, benefiting from the broadband absorption characteristics of the  $\text{EuSn}_2\text{As}_2$  flakes, irrespective of the different coupling efficiency of the frequency-dependent antenna. A low noise-equivalent power (NEP) value of  $30 \text{ pW/Hz}^{0.5}$  (pure noise) and  $2 \text{ nW/Hz}^{0.5}$  (magnified noise about >100 times), without/with the bias voltage, is obtained via the noise spectral density measurements performed using a dynamic signal analyzer (SR785) as shown in Fig. 4b. The photoresponse of the fabricated detectors was found to be favourable even over a modulated frequency of 80 kHz, and is evidently faster than the currently available room-temperature thermal detectors with millisecond response times, substantiating the capability of the high signal-to-noise ratio (SNR) observed for the designed detectors (See Supplementary Figs. 6, 7a for fast response and high signal-to-noise). Finally, to further evaluate the application performance of a single-pixel detector at room temperature under an ambient environment, elaborate imaging experiments were devised as

shown in Fig. 4c (See Supplementary Fig. 9d for transmission experiment results). For a 2D raster scanning imaging under 0.27 THz radiation with a 1 kHz repetition rate, the detector and the imaging object were placed at the THz beam focus, and  $200 \times 200$  points of the object were acquired through a combination of pre-amplification and lock-in techniques. As evident from Fig. 4d, a resolution of the scale plate better than 0.3 cm is obtained, which is close to the diffraction limit of the optical system. More than 1 h was required to acquire the image using a non-optimized setup via mechanical scanning of the half-scale plate. The clearly resolved feature of the target objects indicates the feasibility of these detectors in applications such as quality controlling. The comparative survey of the emerging THz photodetectors built from different materials is shown in Table 1 from three calculated responsivities, and the real photoresponse performance of our device is appealing to engineer applications for large-area and fast imaging. Finally, major improvements can be envisioned by engineering various device architectures with improved efficiencies, such as reducing the losses due to coupling with THz radiation.

### DISCUSSION

In summary, we have explored and experimentally validated terahertz detectors based on multiple antennas-coupled  $\text{EuSn}_2\text{As}_2$  flakes, showing both high sensitivity and a fast response for multiple working modes. The controllable non-equilibrium carriers' transport in the intrinsic electronic system allows a highly sensitive direct THz detection with photoconductive and photovoltaic modes contributed from the non-equilibrium dynamics or self-driven scattering origin, rendering these nanodevices suitable for real-time and low-power consumption imaging at room



**Table 1.** Comparison of the latest reported emerging THz photodetectors.

Frequency [THz]	Responsivity [V/W]	NEP [W Hz <sup>-1/2</sup> ]	Mechanism	Normalization area ( $A_{\text{norm}}$ )	Refs.
0.3–1	0.1, 10, 32, 764, 4 k	$1100 \times 10^{-12}$ , $3 \times 10^{-12}$	Plasma waves, PTE Ballistic & Tunnelling rectification	$\lambda^2/4$ , $\lambda^2/4\pi$	Graphene <sup>4, 27–30</sup>
0.15–0.298	0.15	$4 \times 10^{-8}$ $1 \times 10^{-9}$	Plasma waves, PTE	$\lambda^2/4$	BP <sup>31</sup>
0.293	0.21	$1.36 \times 10^{-8}$	PTE	$\lambda^2/4$	Bi <sub>2</sub> Te <sub>2.2</sub> Se <sub>0.8</sub> <sup>7</sup>
0.12	475 A W <sup>-1</sup>	$3.6 \times 10^{-13}$	Photogalvanic & Photon-drag effect	material area	Bi <sub>2</sub> Se <sub>3</sub> <sup>8</sup>
0.17	27 k	$2 \times 10^{-13}$	EIW	material area (300 $\mu\text{m}^2$ )	Bi <sub>2</sub> O <sub>2</sub> Se <sup>32</sup>
0.12	1.4 k	$10^{-11}$	PTE	material area	PtTe <sub>2</sub> /graphene <sup>33</sup>
0.02~0.30	0.2–1.6 A/W (0.3–2.4k) 6–20 A/W (9–30k)	$30 \times 10^{-12}$	PTE, Anisotropic scattering	device area (0.13 mm <sup>2</sup> ), $\lambda^2/4$ , material area (50.24 $\mu\text{m}^2$ )	This work (EuSn <sub>2</sub> As <sub>2</sub> )

The normalization area  $A_{\text{norm}}$  refers to the area to which the incident power was normalized:  $P_{\text{norm}} = P_{\text{in}} \times A_{\text{norm}}/A_{\text{focus}}$ . In our work, we use three normalization area  $A_{\text{norm}}$  to quantitatively analyze the responsivity, whereas the incident power was not normalized to any area, and the incident power was account for losses occurring in the unfocusing system.

temperature. The design and evaluation strategies proposed in this work pave the way to activate, probe and exploit the activity non-equilibrium dynamics of EuSn<sub>2</sub>As<sub>2</sub>-material for devising THz devices with tailorable performance through a combination of optimization methods, such as alternative structures integrated with metamaterials, and experimental techniques, such as polarization- and time-resolved angle-resolved photoemission spectroscopy<sup>21,34</sup>. These results are crucial to bring to fruition in the field of THz optoelectronics to enable large-area, fast imaging applications as well as high-speed wireless communication.

## METHODS

### Single-crystal growth and device fabrication

The EuSn<sub>2</sub>As<sub>2</sub> crystals were synthesized via melting Eu, Sn and As in a stoichiometric ratio of Eu: Sn: As (1.1: 20: 2) in an alumina crucible enclosed inside a quartz tube jacket, under 50 mTorr pressures, at temperature 850 °C for 12 h. Afterwards, the tube was cooled to 600 °C at a rate of 3 °C/h and the excess Sn was removed via centrifugation after reaching the temperature of 600 °C, and shiny hexagonal single crystals were obtained. Next, the multi-layered EuSn<sub>2</sub>As<sub>2</sub> (~80 nm) was exfoliated using a blue Nitto tape onto a 300-nm thick SiO<sub>2</sub> substrate over a high resistance Si. The standard ultraviolet lithography technique was used to pattern the electrodes, followed by electron-beam evaporation of Cr/Au metal. The characteristics of the EuSn<sub>2</sub>As<sub>2</sub> flakes were assessed via a combination of X-ray diffraction, optical microscopy, and AFM imaging.

### THz detection and imaging

For THz detection, the device was uniformly illuminated with a chopped electronic source, and the open-circuit photovoltaic signal or the short-circuit photocurrent signal was detected after processing the initial signal generated by the device using a preamplifier, lock-in amplifier and high-speed sampling oscilloscope. The evolution of the photocurrent curves of the devices was also measured under the “ON/OFF” modulated radiation. The radiation frequency was tuned up to 0.12 THz (WR 9.0 Tripler ~ 7.5  $\mu\text{W mm}^{-2}$ ) and 0.24–0.29 THz (WR 2.8 Tripler ~ 10  $\mu\text{W mm}^{-2}$ ) using a multiplier (Virginia Diodes Inc.) connected to a microwave source emitting radiation frequency in the range of 0.02–0.04 THz (Agilent E8257D ~ 25  $\mu\text{W mm}^{-2}$ ). The power flux intensity was calibrated by a TK100 power meter. For imaging application, as shown in Fig. 4c, a 0.27 THz beam was focused on the detector using two pairs of off-axis parabolic mirrors, and the images were acquired via raster scanning of the object at the beam focus, consisting of 200 × 200 points with 100 ms integration time at every point.

## DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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## AUTHOR CONTRIBUTIONS

L.W. and X.S.C. conceived the project. Y.L. provided the photosensitive material. C.L., L.Z., and Z.C. designed the experiments. C.L., Y.L., Z.C., S.Z., and C.S. conducted all the experiments. C.L., G.L., and L.Z. undertook theoretical calculations. C.L. and L.W. participated in writing the manuscript. X.Y., Z.X., W.L., Y.L., and W.Z. analyzed the data. All authors contributed to the organization of the manuscript.

## COMPETING INTERESTS

The authors declare no competing interests.

## ADDITIONAL INFORMATION

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