

MATERIALS SCIENCE

Giant room-temperature terahertz photothermoelectric response mediated by hot carriers at the metal-semimetal interfaces

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The hot-carrier photothermoelectric (PTE) effect in two-dimensional materials can be used to develop room-temperature, fast, and sensitive detectors for microwave, terahertz, and far-infrared radiations. Here, we present a van der Waals semimetal PtSe₂ terahertz detector and find a giant terahertz PTE response mediated by hot carriers at the metal-PtSe₂ interfaces. The detector demonstrates an excellent zero-bias responsivity of 0.62 ampere per watt (A/W), a low-noise equivalent power of 19.6 picowatt per square root of hertz (pW/Hz^{1/2}), and a fast response time of 4.5 nanosecond (ns) at 0.1 terahertz (THz). The metal doping effect and asymmetric terahertz heating play vital roles in the hot-carrier PTE response at the metal-semimetal interfaces. Moreover, scanning photocurrent mapping shows that the short-wave zero-bias photoresponse is closely localized to the metal-semimetal interfaces, further revealing the metal doping effect. The bias-dependent photocurrent indicates that the short-wave photoresponse still originates from the PTE effect and the photovoltaic response is negligible. Our results provide important guidance for developing high performance semimetal detectors.

INTRODUCTION

Terahertz (THz) detectors have wide applications in many fields such as high data-rate communication, real-time imaging, spectroscopy, and sensing (1–4). The photon energy of the terahertz wave (0.42 meV@0.1 THz, 42 meV @10 THz) is very close to the room temperature ($T_0 = 300$ K) thermal energy ($k_B T_0 = 26$ meV, with k_B the Boltzmann constant), which makes uncooled photon-type detectors in this band extremely challenging. The thermal detectors are not limited by the bandgap and can achieve room temperature and wide spectral detection, which has advantages in the terahertz band. However, the slow response speed and low sensitivity of traditional thermal detectors limit their application range (5).

Recently, it has been found that the hot-carrier photothermoelectric (PTE) effect in two-dimensional (2D) materials can overcome these limitations (6–10). In this case, because of the free carrier absorption, the terahertz energy is transferred to the electron subsystem, and because of the weak electron–longitudinal optical (LO) phonon scattering in the low-energy range, the energy relaxation channel to the lattice subsystem is blocked. Further, because of the strong electron–electron scattering, the electron subsystem is thermalized to form a steady-state distribution with a higher electron temperature. If the electron subsystem is heated locally through free carrier absorption of terahertz radiation, a temperature gradient will be formed, and the hot carriers will diffuse from the hot region to the cold region, generating a fast PTE response (11). Song *et al.*

theoretically and Gabor *et al.* experimentally demonstrated the hot carrier–mediated intrinsic photoresponse in graphene homojunctions, revealing a sixfold photovoltage pattern due to the Seebeck coefficient variations, which distinguishes the PTE effect from the photovoltaic (PV) effect (12, 13). Cai *et al.* (6) first demonstrated an application of graphene hot-carrier PTE effect in terahertz detection. This device was accomplished by applying dissimilar metals to form contacts with graphene and achieved a sensitivity exceeding 1100 pW/Hz^{1/2} at 2.52 THz. Castilla *et al.* (7) presented a graphene terahertz PTE detector by using a split-gate/antenna structure with a narrow feed gap, which funnels the incident terahertz wave exactly to the small photoactive area of the detector, leading to a strongly enhanced terahertz absorption in graphene and an excellent sensitivity (80 pW/Hz^{1/2}). In addition, researchers have greatly improved the performance of graphene hot-carrier PTE detectors through various strategies to enhance the light-graphene interaction, such as graphene plasmonics (14, 15), optical resonant antennas (16), Fabry-Perot microcavities (17), and metasurfaces (18, 19).

Recently, PtSe₂, as a layered 2D transition metal dichalcogenide (TMDC) material, has become a hot research topic in optoelectronics because of its excellent properties including high carrier mobility, layer-dependent indirect bandgap (0 ~ 1.2 eV), broadband light absorption, strong thermoelectric effect, and excellent atmosphere stability (20–23). These properties make it comparable to other widely investigated 2D materials like graphene, MoS₂ and black phosphorus (24). Notably, as the layer number increases, PtSe₂ undergoes a transition from semiconductor (single layer and double layer) to semimetal ($\geq$ three layers) (23). As an electron-dominant thermoelectric material, semimetal PtSe₂ has a considerable Seebeck coefficient of about -73 μ V/K, and compared with graphene, it has a lower thermal conductivity, which is beneficial for obtaining a stronger thermoelectric response (25, 26). However, the hot-carrier PTE effect of semimetal PtSe₂ and its application in terahertz detection have not been investigated systemically.

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Here, we realize a room-temperature, zero-bias, fast, and sensitive semimetal PtSe₂-terahertz detector integrated with an asymmetric bow-tie antenna and systematically investigate the hot-carrier PTE effect of semimetal PtSe₂. At the resonant frequency (0.1 THz) of the antenna, the detector shows an excellent room-temperature and zero-bias detection performance of a high responsivity of 0.62 A/W, a low noise equivalent power (NEP) of 19.6 pW/Hz^{1/2}, and a fast response speed of 4.5 ns. On the basis of electrostatic and heat transfer simulations, we construct a hot-carrier PTE response model for the metal-PtSe₂-metal device. In this model, a simplified PTE response formula is attained by considering the metal-semimetal interface doping effect and the asymmetric hot-carrier temperature distribution induced by local terahertz heating. We find that the PTE effect mediated by hot carriers at the metal-semimetal interfaces plays a key role in terahertz response. When the maximum electron temperature position is overlapped with the depletion edge of the metal-semimetal interfaces, the PTE response reaches up to the largest value. Moreover, a bias voltage-dependent terahertz PTE response is observed, which is contributed to the direct dark-current-induced Peltier heat flow and the resultant shift of the maximum electron temperature position. The zero-bias scanning photocurrent imaging of the PtSe₂ device under 780-nm interband photoexcitation shows that photocurrents are limited to the metal-semimetal interfaces. The bias-dependent behavior of the photoresponse confirms that the photocurrent still originates from the PTE effect. The

weak PV response at the metal-semimetal interfaces can be attributed to the narrow depletion-region width, the unique band structure of electron-hole coexistence on the Fermi surface, and the short photo-generated carrier lifetime.

RESULTS

The PtSe₂ crystal has a typical 1T-type hexagonal crystal structure with a *P-3m1* space group, as shown in Fig. 1A. The monolayer PtSe₂ consists of three atomic layers stacked in the order Se-Pt-Se, and the layers are held together by vdWs force to form a layered crystal structure (20). Because of the weak interlayer vdWs force, PtSe₂, analogous to other TMDCs, can be mechanically exfoliated to PtSe₂ flakes with different layers. PtSe₂ has a layer-dependent band structure and bandgap (23, 27). The monolayer PtSe₂ is an indirect semiconductor with a bandgap ($E_g = 1.23$ eV). The bandgap of bilayer PtSe₂ is reduced to 0.23 eV. For the trilayer or more layers PtSe₂, the conduction and valence bands overlap indirectly and the material becomes a semimetal, with the coexistence of electrons and holes on the Fermi surface. As the layer number increases, the overlap of conduction band (CB) bottom and valence band (VB) top tends to 0.87 eV. Figure 1B shows the band structure of multilayer semimetal PtSe₂ (28). As shown in the inset of Fig. 1B, the intrinsic semimetal PtSe₂ Fermi surface consists of two types of pockets and one hole pocket, indicating that electrons and holes coexist at the Fermi surface,

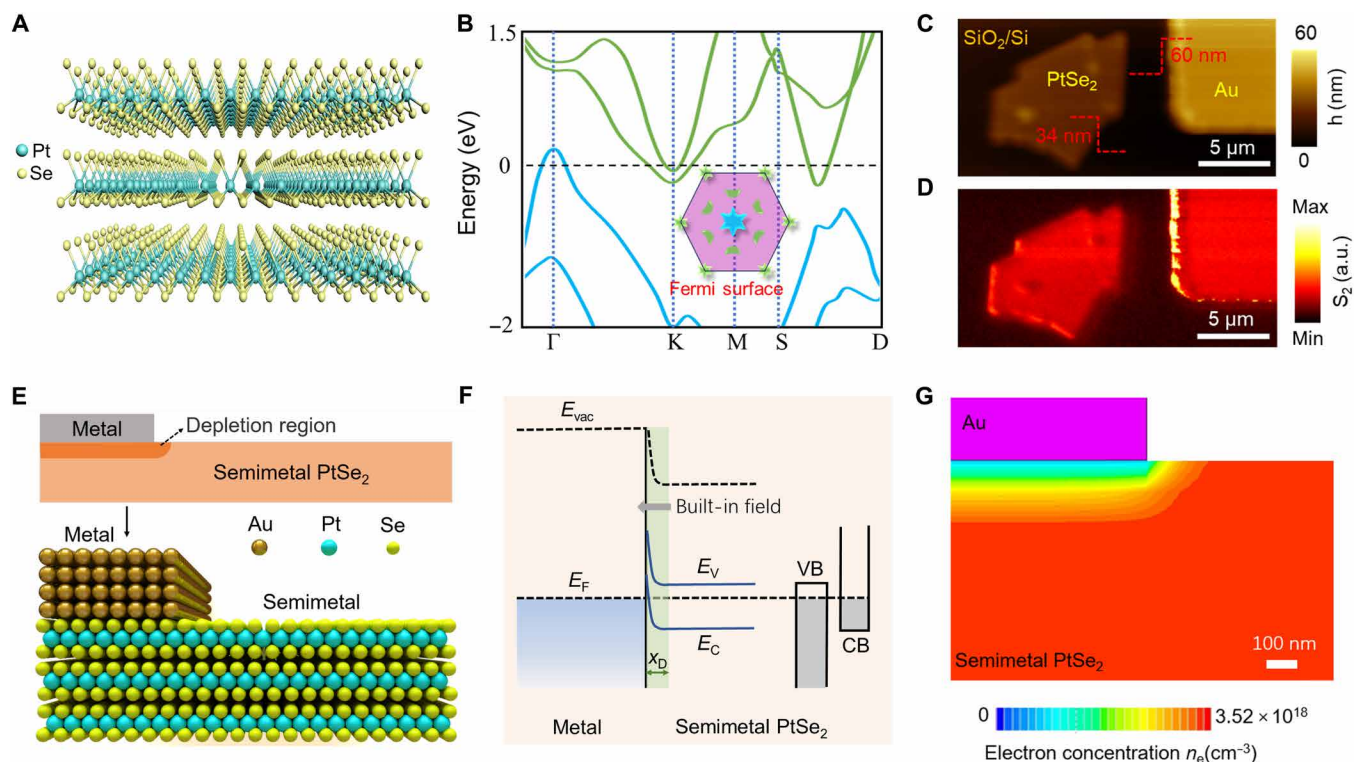


Fig. 1. The properties of semimetal PtSe₂ and metal-semimetal contact. (A) Schematic crystal structure of PtSe₂. (B) Band structure of semimetal PtSe₂. The inset is the top view of the Fermi surfaces in the first Brillouin zone. The green and blue pockets correspond to electron and hole pockets, respectively. (C) The AFM topography of the sample. *h*, hours. The thicknesses of the PtSe₂ flake and Au film are 34 and 60 nm, respectively. (D) The second-order (*S*₂) terahertz near-field scattering image of the sample. (E) Metal-semimetal contact diagram. (F) Band diagram of metal-semimetal contact in thermal equilibrium. E_F and E_{vac} represent the Fermi energy and the vacuum level, respectively. x_D corresponds to the width of the depletion region. (G) The simulation result of electron concentration distribution in the metal-semimetal contact region. a.u., arbitrary units.

and electrons are the major charge carriers. The shape of the hole pocket resembles an octahedron (colored blue), and there are two types of electron pockets (colored green): six octahedron electron pockets located around the K points and six crescent electron pockets located between the D and S points. The zero bandgap of PtSe₂ enables it to absorb broadband electromagnetic radiation. In the mid- and far-infrared and terahertz bands, light absorption is mainly achieved by intraband free-carrier absorption. In the visible and near-infrared bands, the intraband absorption efficiency is low because of the large wave vector compensation, and the light absorption is mainly due to the interband transition (29). Typical Raman signatures of PtSe₂ are observed in the corresponding Raman spectrum and Raman intensity mapping images (fig. S1), indicating the good quality of the mechanically exfoliated multilayer PtSe₂ flakes (30). The high electrical conductivity of semimetal PtSe₂ can be revealed by terahertz near-field optical imaging (31–33). We used a commercial terahertz scattering-type near-field optical microscope system (Neaspec GmbH) to perform terahertz near-field imaging of mechanically exfoliated semimetal PtSe₂ flakes (fig. S2A). Since Au has a strong terahertz near-field scattering signal, an Au thin-film electrode is used as a reference for near-field imaging (31). Figure 1 (C and D) shows the simultaneously collected atomic force microscope (AFM) topography and near-field images of the sample, respectively. As shown in Fig. 1D, PtSe₂ and Au show similar strong terahertz near-field signals, and there is an obvious contrast to the weak near-field signals of the SiO₂ substrate. The strong and homogeneous terahertz near-field scattering signals of the PtSe₂ flake reflect its semimetallic character. The second-order (S₂) time-domain near-field signals at different locations were Fourier transformed to obtain the frequency-domain amplitude spectra of PtSe₂, Au, and SiO₂ (fig. S2B). It can be seen that the near-field amplitude spectrum of PtSe₂ is close to that of Au in a wide frequency range, indicating high electron concentration and mobility in the PtSe₂ flake.

For semimetal PtSe₂ detectors, the simplest device configuration is the metal-PtSe₂-metal structure. Considering that the metal doping effect will influence the Seebeck coefficient of PtSe₂ in the contact regions, we first investigate the metal-PtSe₂ contacts (34–37). The schematic diagram of metal-semimetal contact is shown in Fig. 1E, where Au, as the electrode material, forms an electrical contact with PtSe₂. Because of the different work functions of Au and PtSe₂, there is a net charge transfer at the metal-semimetal interfaces and the generation of a built-in electric field, leading to the formation of a depletion region. Figure 1F shows the band diagram of metal-semimetal contact in thermal equilibrium. Since the work function of Au (5.1 eV) is higher than that of semimetal PtSe₂ (4.82 eV) (38), electrons are forced to transfer from the semimetal to the metal until the two Fermi energies of Au and PtSe₂ are aligned. At the metal-semimetal interface, the charge transfer leads to a built-in electric field that points from the semimetal channel to the metal electrode, and the semimetal CB edge E_c and VB edge E_v will bend upward. Because of the high electron concentration of PtSe₂ and the coexistence of electrons and holes at the Fermi surface, analogous to contact between the metal and heavily doped semiconductor, the metal-semimetal interface is expected to form a narrow depletion region. We use the Silvaco-ATLAS software (TCAD package) (39) to simulate the carrier concentration distribution and depletion region width of the Au-PtSe₂ contact. As shown in Fig. 1G, there is a thin depletion region (tens of nanometers) on the PtSe₂ side of the metal-semimetal interface at zero-bias voltage, which is thin enough for

direct tunneling of carrier across the barrier, resulting in the formation of good ohmic contacts. Because of the metal-semimetal interface doping effect, the decrease of electron concentration near the metal-semimetal interface regions leads to a reduced Seebeck coefficient compared with the undoped channel region (23), thus introducing a nonuniform distribution of the Seebeck coefficient along the PtSe₂ channel.

For 2D material detectors, the optically active area is typically on the nanoscale or microscale, much smaller than the wavelength of terahertz wave. The optical antenna can enhance terahertz absorption and achieve a local illumination through the terahertz field localization and enhancement. As shown in Fig. 2A, we design an asymmetric bow-tie antenna integrated semimetal PtSe₂ terahertz detector, in which the bow-tie antenna is an optical coupler to achieve asymmetric local terahertz heating. The structural parameters of the bow-tie antenna are presented in fig. S4A. The electric field enhancement factor is defined as the ratio of surface-averaged electric field amplitudes with and without the metal structures. As depicted in Fig. 2B, the feed gap of the designed bow-tie antenna shows excellent field enhancement performance with a peak field enhancement factor of 180 for the fundamental dipole resonance at 0.1 THz. The simulated surface electric field distribution (Fig. 2B) of the asymmetric bow-tie antenna at 0.1 THz exhibits a strong asymmetry. Figure 2C shows the optical micrograph of the fabricated PtSe₂ device. In this device, mechanically exfoliated PtSe₂ flakes are transferred to the channel region of the asymmetric bow-tie antenna structure, with the bow-tie antenna and the metal strip electrodes serving as the source (S) and drain (D), respectively. The AFM image of the fabricated PtSe₂ device is shown in fig. S4C. The measured thickness of the PtSe₂ flake is 72 nm. The electrical measurements (fig. S4D) show that the current-voltage (I - V) characteristic curve of the PtSe₂ device exhibits good linear characteristics and low resistance (123 ohm), indicating the formation of excellent ohmic contact between the Au electrodes and the PtSe₂ flake.

Since the terahertz field is localized and enhanced in the antenna feed gap, free carriers near the metal-semimetal interface are heated by the focused terahertz wave via the intraband free carrier absorption (Fig. 2D). Because of the low terahertz photon energy, the energy relaxation channel via electron-LO phonon scattering is blocked, and the terahertz energy is mainly transferred to the electron subsystem rather than the lattice subsystem (11). As shown in Fig. 2E, the electron subsystem of PtSe₂ is thermalized to form a steady-state distribution with a higher electron temperature (T_e) via the strong electron-electron scattering, and the lattice temperature (T_p) changes very little. Under continuous terahertz illumination, a steady-state distribution of electron temperature (T_e) gradient is established along the channel. When the terahertz illumination is turned off, the temperature gradient is destroyed through hot-carrier diffusion and the most excess energy is dissipated to the metal electrodes. Considering the imbalance between electron (hot-carrier effect) and lattice temperatures during terahertz heating, a two-temperature heat transfer model is used to analyze the hot-carrier temperature distribution of the PtSe₂ channel under asymmetric terahertz illumination (fig. S5). The 1D two-temperature heat transfer coupled equations are (40, 41)

$$\frac{\partial T_e}{\partial t} = \frac{\kappa_e}{C_e} \nabla^2 T_e - \frac{T_e - T_p}{\tau_{ep}} - \frac{T_e - T_0}{\tau} + \frac{Q}{C_e} \quad (1)$$

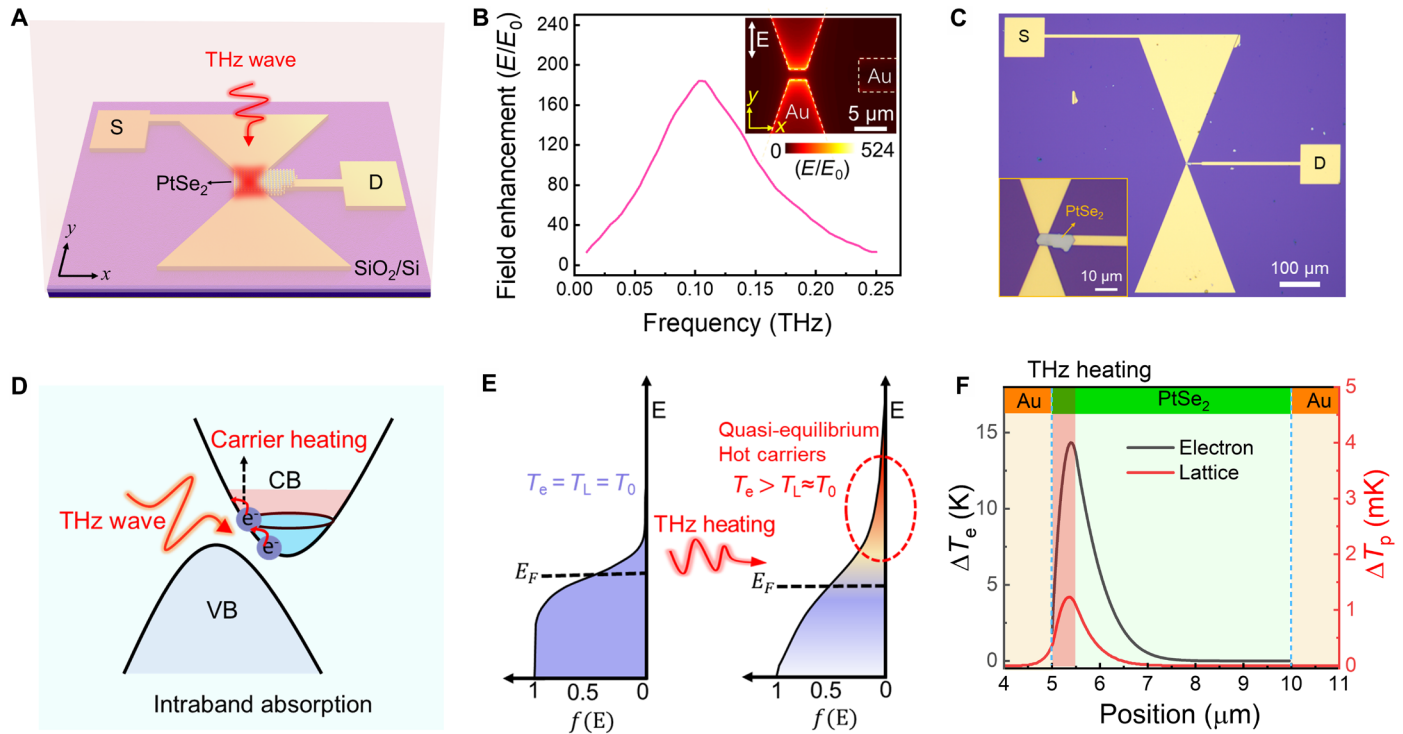


Fig. 2. The hot-carrier effect of semimetal PtSe₂ under terahertz illumination. (A) Schematic of the asymmetric bow-tie antenna integrated semimetal PtSe₂ terahertz detector. (B) The surface-averaged electric field intensity as a function of frequency in the feed gap of the bow-tie antenna. The terahertz wave is vertically incident to the device surface, and the polarization is along the antenna arm direction. The inset is the simulated electric field distribution of the asymmetric bow-tie antenna at 0.1 terahertz. (C) Optical micrograph of PtSe₂ device. (D) Schematic illustration of PtSe₂ carrier thermalization via intraband free carrier absorption under terahertz excitation. (E) The diagram of the hot-carrier effect of PtSe₂. Terahertz radiation-induced electron heating in PtSe₂ leads to a broader Fermi-Dirac distribution, in comparison with that without terahertz excitation. (F) Temperature distribution of electron and lattice between source and drain electrodes when the incident terahertz energy is localized within the 0.5-μm-wide region close to the Au electrode edge. ΔT_e and ΔT_p correspond to changes of electron and lattice temperatures, respectively.

$$\frac{\partial T_p}{\partial t} = \frac{\kappa_p}{C_p} \nabla^2 T_p + \frac{C_e}{C_p} \frac{T_e - T_p}{\tau_{ep}} - \frac{T_p - T_0}{\tau} \quad (2)$$

$$\frac{\partial T}{\partial t} = \frac{\kappa}{C} \nabla^2 T - \frac{T - T_0}{\tau} \quad (3)$$

where T_0 is the environment temperature (300 K). T , κ , τ , and C are temperature, thermal conductivity, heat relaxation time to environment, and heat capacity for Au electrodes, respectively. T_e (T_p), κ_e (κ_p), and C_e (C_p) are temperature, thermal conductivity, and heat capacity for electron (lattice) subsystem of PtSe₂. τ_{ep} is the heat relaxation time from the electron subsystem to the lattice subsystem. Q is input terahertz power density. The electron heat capacity C_e and electron thermal conductivity κ_e of PtSe₂ are calculated by the following formula (41, 42)

$$C_e = \frac{3}{2} n_e k_B \quad (4)$$

$$\kappa_e = \frac{1}{3} C_e v_F^2 \tau_F \quad (5)$$

where n_e , v_F , and τ_F are the electron concentration, electron Fermi velocity, and electron scattering time of PtSe₂, respectively. The

definitions and values of each parameter in the two-temperature heat transfer equations and the numerical results are shown in table S2. Figure 2F shows the temperature distributions of the electron and lattice along the PtSe₂ channel, obtained by numerically solving the two-temperature heat transfer equations. The numerical results show that the variation of electron temperature T_e is much greater than that of lattice temperature T_p , and the electron temperature exhibits a remarkable asymmetric distribution along the channel. Because of the rapid energy relaxation of hot carriers through the metal electrodes, a large electron temperature gradient ∇T_e exists near left the metal-semimetal interface. In particular, the large heat capacity and thermal conductivity of Au leads to a slight increase of electron temperature at the left metal-semimetal contact.

On the basis of the metal-semimetal interface doping effect and the two-temperature heat transfer process, we can quantitatively analyze the hot-carrier PTE response of the PtSe₂ device under asymmetric terahertz heating. As shown in Fig. 3A, we assume that the Seebeck coefficient in the doped region is S_1 and that in the undoped region is S_0 . The simplified distribution of the Seebeck coefficient in the channel corresponds to the green line in Fig. 3A. The purple line presents the hot-carrier temperature distribution under local terahertz heating at the metal-PtSe₂ interface, where T_{em} is the highest electron temperature in the channel, and T_{e1} and T_{e2} are the electron temperatures at the left and right doped region edges of the PtSe₂ device, respectively. Considering the 1D temperature and Seebeck coefficient as functions of x ($0 \leq x \leq L$, $x = 0$ at the source and

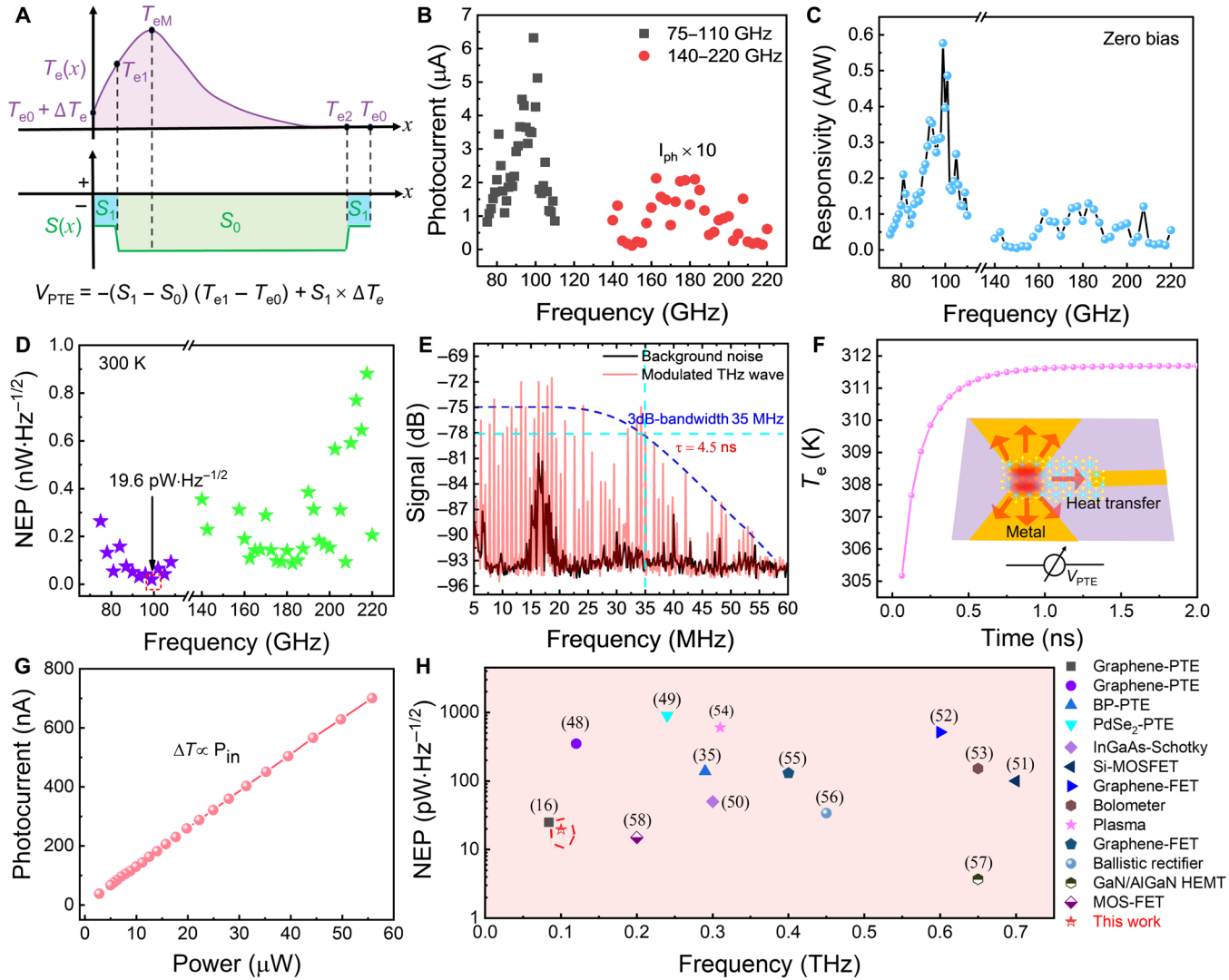


Fig. 3. The terahertz hot-carrier PTE response of PtSe₂ device at 300 K. (A) The hot-carrier PTE response model of PtSe₂ device under asymmetric terahertz heating. The purple line represents the simplified distribution of Seebeck coefficients and S_1 and S_0 correspond to the Seebeck coefficients for the doped and undoped regions, respectively. The purple line corresponds to the electron temperature distribution under asymmetric terahertz heating, where T_{eM} is the highest electron temperature in the channel, and T_{e1} and T_{e2} are the electron temperatures at the left and right doped region edges of the PtSe₂ device, respectively. ΔT_{e0} corresponds to the temperature change of Au electrode. (B) Zero-bias terahertz photocurrent spectrum of PtSe₂ device. The polarization of the terahertz wave is along the bow-tie antenna arm direction. (C) Photocurrent responsivity spectrum. (D) NEP spectrum. (E) Zero-bias photocurrents spectrum (red) of the PtSe₂ detector illuminated by the 100-GHz and continuum 100-MHz modulated terahertz radiation and the background noise spectrum (black). (F) Simulated intrinsic fast hot-carrier PTE response of PtSe₂. (G) Power-dependent photoresponse. (H) Performance comparison with the reported room temperature terahertz detectors. BP, black phosphorus; FET, field effect transistor; HEMT, high electron mobility transistor.

$x = L$ at the drain, where L is the length of the device channel), the PTE response can be calculated by integrating the local PTE voltage gradient generated by the temperature gradient $[\nabla T_e(x)]$ together with a spatially varying Seebeck coefficient $[S(x)]$ (43)

$$V_{PTE} = \int S(x) \nabla T_e(x) dx \quad (6)$$

As shown in Fig. 3A, ΔT_e is the electron temperature difference between the temperatures at the left and right end of the PtSe₂ channel, and $T_{e2} \approx T_{e0}$. A simplified expression of PTE response is

$$V_{PTE} = -(S_1 - S_0)(T_{e1} - T_{e0}) + S_1 \Delta T_e \quad (7)$$

According to Eq. 7, it can be seen that the metal doping effect and the position of the local terahertz heating have an important influence on the magnitude of the hot-carrier PTE response. If the metal doping effect is not considered, the integral PTE response would be $S_1 \Delta T_e$. Since ΔT_e is very small (Fig. 3A), the PTE response would be extremely weak. When the peak temperature position overlaps with the depletion edge of the Au-PtSe₂ interface, $T_{e1} = T_{eM}$, the PTE voltage reaches up to the largest value. Therefore, a strong hot-carrier PTE response can be achieved by introducing asymmetric terahertz heating of the device channel, especially by enhancing the terahertz electric field intensity near the metal-semimetal interface.

To verify the terahertz response performance of the PtSe₂ detector, we systematically characterized the detection parameters, including

photocurrent, responsivity, NEP , and response speed. As displayed in Fig. 3B, the PtSe₂ device exhibits an excellent zero-bias photocurrent response in the frequency range of 0.075 to 0.22 THz at room temperature. At the resonant frequency (0.1 THz) of the bow-tie antenna, the detector shows a peak photocurrent of up to 6.0 μ A. According to the PTE response model we constructed, the bow-tie antenna-induced asymmetric terahertz heating will drive the PTE current from the PtSe₂ channel toward the antenna, which is consistent with the measured photocurrent polarity. To evaluate the photoelectric conversion efficiency of the PtSe₂ terahertz PTE detector, it is necessary to quantify the responsivity that can be calculated as: $R_A = I_{ph} / (P_{in} \times S_A)$, where I_{ph} is the photocurrent, P_{in} is the terahertz power density, and S_A is the effective photoactive area of the detector. Here, we use the effective receiving area of the antenna as the effective photoactive area of the detector, that is, $S_A = 3\lambda^2 / 8\pi$ (44). Figure 3C shows the zero-bias spectral responsivity of the PtSe₂ detector calculated according to the measured photocurrent and the corresponding effective receiving power. At 0.1 THz, the peak responsivity of the device exceeds 0.62 A/W. The sensitivity (NEP) can be calculated as $NEP = i_n / R_A$, where i_n is the noise current. For the zero-bias PTE response, thermal noise is the only intrinsic noise source that can be estimated using the following formula: $i_n = \sqrt{4k_B T / R}$, where R is the resistance of the device. Figure S7B shows the measured noise spectra and the theoretical thermal noise value. Following the aforementioned analysis, the NEP of the PtSe₂ terahertz PTE detector can reach less than 19.6 pW/Hz^{1/2} at room temperature (Fig. 3D). To better evaluate the performance of the PtSe₂ device, error analyses are provided in note S6 and fig. S8, and a relative uncertainty of 3.6% for responsivity and NEP is derived. For a terahertz detector, response speed is important, especially in applications such as fast imaging and high-speed communication. Considering the intrinsic fast hot-carrier PTE response, we measured the terahertz response time of the PtSe₂ detector using a wideband current preamplifier (Femto DHPA-100, maximum bandwidth: 200 MHz) and a high-speed modulated terahertz source (45). The detailed process is described in the note S8 and fig. S9. Figure 3E presents the experimental zero-bias photocurrent spectrum with the illumination of the 100-GHz and continuum 100-MHz modulated terahertz radiation. The comb teeth in the range of 0 to 55 MHz can be resolved, and the amplitude fluctuations of comb teeth maybe originate from the interference and the air absorption. The 3-dB bandwidth of 35 MHz is derived from the response spectrum shown in Fig. 3E. The modulation frequency-dependent response of the detector is expressed as $R(f) = R_0 / \sqrt{1 + (2\pi f \tau_d)^2}$, where f is the modulation frequency and R_0 is the DC responsivity (46, 47). The relation between the 3-dB bandwidth f_{3dB} and the detector response time τ_d is expressed as $\tau_d = 1 / (2\pi f_{3dB})$ from which $\tau_d = 4.5$ ns is determined. The fast response also proves that PtSe₂ has a strong hot-carrier effect in the terahertz band. The simulated time evolution of the electron temperature under terahertz illumination in the two-temperature model identifies the intrinsic fast response of semimetal PtSe₂ device (Fig. 3F) due to the hot-carrier effect. Another important characteristic of the detector is the linear dynamic range. Figure 3G shows that the photocurrent has a good linear power dependence in a power range of 2.2 to 55 μ W at 0.02 THz. The reason for the linear photoresponse as a function of light power is the fact that the PtSe₂ PTE detector operates in the weak heating regime, the change in temperature of the electron system is approximately proportional to

the receiving light power. To facilitate better performance comparisons, we summarize the performance metrics of the reported room temperature terahertz detectors (Fig. 3H) (48–58).

Figure 4A shows the bias voltage-dependent terahertz response of the PtSe₂ device. When the polarities of dark and PTE currents are opposite, the photocurrent increases with increasing bias voltage. Conversely, a dark current in the same polarity as the PTE current will result in a suppression of the photoresponse. The zero-bias photoresponse and the bias polarity-dependent variations of photocurrent completely exclude the bolometric detection mechanism. Understanding the bias-dependent photocurrent is helpful for gaining deeper insight into the PTE effect near the Au-PtSe₂ interface. According to the temperature distribution evaluated by the two-temperature heat transfer model (Fig. 2F), we find that if the incident terahertz energy is localized into the 0.5- μ m-wide region close to the Au electrode edge (chosen as the origin of coordinate, $x = 0$ μ m), the peak temperature is located at about $x = 0.4$ μ m. The TCAD simulations show that the Au-PtSe₂ depletion width is insensitive to the bias voltage (fig. S3B). The maximum difference of the Au-PtSe₂ depletion width is 6 nm when the bias voltage changes from -0.4 to 0.4 V. Such a small change in depletion width cannot be responsible for the bias-dependent PTE response. Since Joule heating of the dark current heats the channel uniformly, it does not affect the distribution of the electron temperature gradient. Another reasonable explanation is that the Peltier heat flux carried by the dark current leads to the change of the temperature distribution along the PtSe₂ channel, which results in the bias-dependent photoresponse. According to the Peltier effect, the current-carried heat flux density J_Q can be expressed as (59)

$$J_Q = T \times S \times J \quad (8)$$

where J is the current density. Figure 4B shows the schematic diagram of electron temperature distribution changes caused by the Peltier effect at positive bias voltage. The Seebeck coefficient of PtSe₂ is negative, and the corresponding Peltier heat flux direction is the same as the PTE current direction. Because of the equal temperature of points a and b (the intersection points of the red dashed line and the temperature distribution curve shown in Fig. 4B), the net Peltier heat flux is zero for the regions of a and b. However, there is a net Peltier heat flux from the regions c and b to the region a and c, where c corresponds to the peak temperature position. Such a net Peltier heat flux will lead to the left shift of the peak temperature position. According to Eq. 7, the PTE response will be enhanced. On the basis of the same reasoning, the PTE response will be suppressed for the case of negative bias voltage. The bias voltage-dependent terahertz response also provides evidence that the metal doping effect plays a key role in the PTE response.

To gain deeper insights into the photoresponse mechanism, we use a scanning photocurrent microscope (60) to measure the spatially resolved photocurrents of the PtSe₂ device at zero-bias voltage under the 780-nm laser radiation. The zero-bias scanning photocurrent image (Fig. 4C) at room temperature shows that the photocurrent is mainly located at the metal-semimetal interfaces and has opposite polarities at the two metal-semimetal interfaces of the device. The zero-bias photoresponse at the metal-semimetal interfaces may originate from the PV effect or the PTE effect. For the PV effect, the electron-hole pairs generated through interband transition are separated by the built-in electric field (Fig. 4C) near the metal-semimetal interface to form a PV current from the semimetal to the metal electrode. The PTE effect is another possible detection mechanism. Because

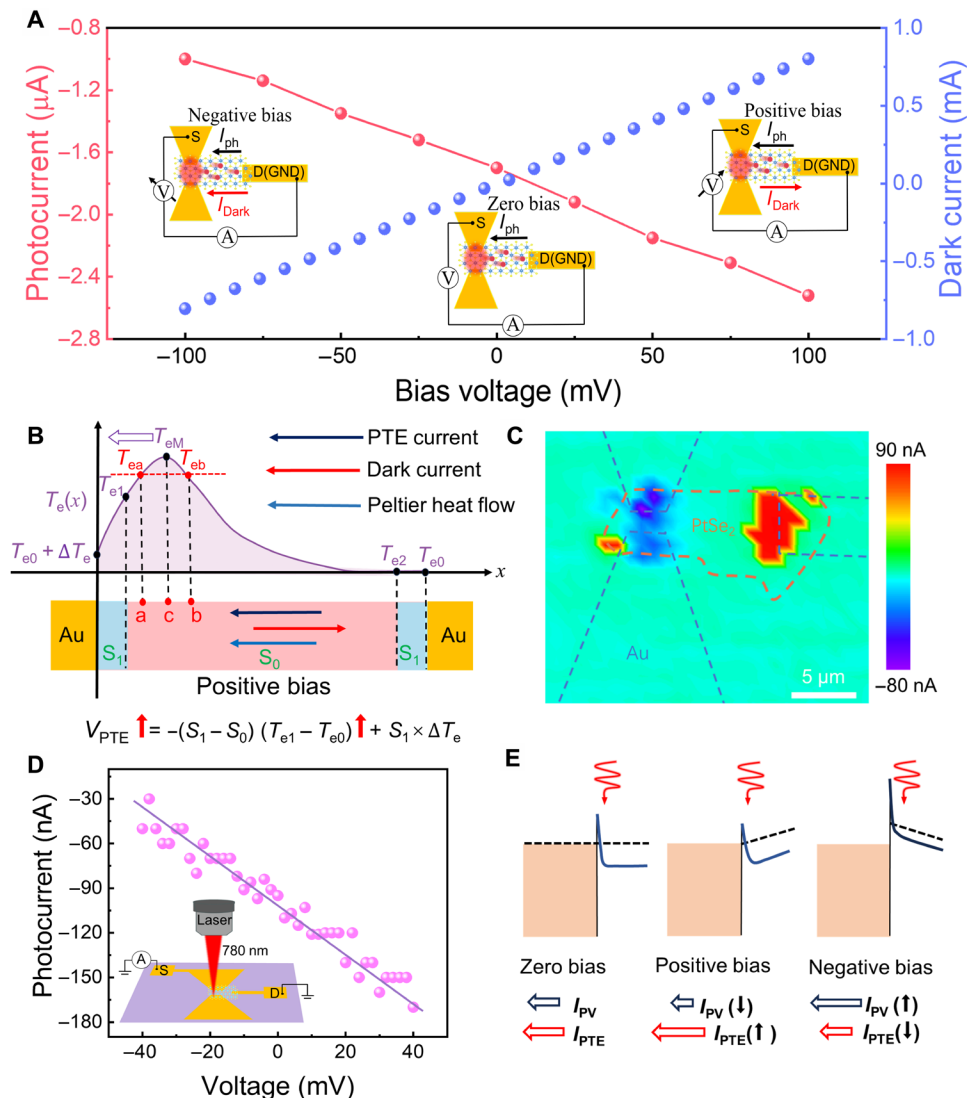


Fig. 4. The bias voltage-dependent terahertz response and scanning photocurrent imaging (780 nm) of the PtSe₂ device. (A) The bias voltage dependencies of the photocurrent (red) and dark current (blue). The illustrations correspond to different measurement configurations, and the black and red arrows indicate the direction of the photocurrent and the dark current, respectively. (B) Mechanisms of bias-dependent terahertz photoresponse. The purple line is the electron temperature distribution at zero bias under asymmetric terahertz heating, and electron temperatures at different locations in the channel are labeled. The two intersection points a and b of the red line and the temperature distribution curve represent two equal-temperature positions, and point c labels the position of maximum temperature. The black, red, and blue arrows represent PTE current, dark current, and Peltier heat flow, respectively. The purple arrow indicates the movement of the highest point of the electron temperature. (C) Scanning photocurrent image obtained at zero bias under a 780-nm focused laser with a power of about 35 μW . The dashed blue and orange lines correspond to the metal electrode and the multilayer PtSe₂ flake, respectively. Scale bar, 5 μm . (D) Bias voltage-dependent photocurrent of the metal-semimetal interface with the device being illuminated by a fixed laser spot located at the metal-semimetal contact edge near the bow-tie antenna. (E) Schematic diagram of bias-dependent photoresponse at the metal-semimetal interface. The direction of the built-in electric field is directed from the PtSe₂ channel toward the metal electrode. The black and red arrows indicate the directions of the PV currents and the PTE currents, respectively.

of the energy band overlap of semimetal PtSe₂, photoexcited non-equilibrium electrons and holes rapidly form a hot-carrier distribution through carrier-carrier scattering. On the basis of the proposed PTE response model of the PtSe₂ device, the carrier heating near the metal-semimetal interfaces will lead to a strong PTE response. According to the simplified expression of the PTE response (Eq. 7), the PTE response and the PV response near the metal-semimetal interfaces have the same polarity and need to be further distinguished.

Figure 4D shows the bias voltage-dependent photocurrent near the metal-semimetal interfaces. We find that when a positive bias

voltage is applied, the photoresponse is enhanced. On the contrary, a negative bias voltage results in the suppression of photoresponse. The bias-dependent photoresponse for the 780-nm laser illumination is consistent with that for the terahertz illumination (Fig. 4A). As shown in Fig. 4E, considering that the applied positive electric field leads to the weakening of the built-in electric field at the metal-semimetal interface, the corresponding PV response should be reduced, which is contradicted with the experimental results. In addition, when the positive bias voltage is applied, the polarity of the bolometric photocurrent is opposite to the measured photocurrent

polarity; thus, the bolometric effect should be excluded. Therefore, we can conclude that in the optical frequency band, the photoresponse near the metal-semimetal interfaces originates from the PTE effect. The negligible PV response at the metal-semimetal interfaces can be attributed to the following reasons: (i) the thin depletion region of the metal-semimetal interface, (ii) the unique band structure of electron-hole coexistence on the Fermi surface, and (iii) the short lifetime of photoexcited carriers. The fact that the photocurrent is very close to metal-semimetal contact further indicates the metal-doping effect on the PTE photoresponse. In addition, we measured the zero-bias PTE responses with visible to near-infrared local illuminations (six frequency points) at the Au-PtSe₂ interface, and the results are shown in fig. S11. The trend of responsivity with respect to wavelength shown in fig. S11C is in accordance with that of the reported absorption spectrum (61). The experimental results shown in fig. S11 clearly indicate the wideband PTE detection ability of PtSe₂. All the above results show that the hot-carrier PTE effect at the metal-semimetal interfaces dominates the wideband photocurrent generation of PtSe₂ device from visible to terahertz band.

Beyond its outstanding detection capabilities in the terahertz range, the PtSe₂ detector also demonstrates potential for terahertz communication applications. As depicted in Fig. 5A. We demonstrate a terahertz communication system, comprising binary data input, optical signal acquisition and conversion, and data output. The letters “USST” are first converted into a binary data stream (ASCII code) and encoded

onto a timing-modulated square wave signal from a function signal generator (AFG3022C), where the high level represents “1” and the low level represents “0,” with 0 used as a separator between each letter (Fig. 5B). The encoded square wave signal is used to modulate a microwave source (SMA100B), and the modulated microwave drives the frequency multiplier to generate 100-GHz radiation. Then, the 100-GHz modulated terahertz radiation carrying the encoded information is radiated into free space through a horn antenna. Our PtSe₂ detector served as the signal receiver, converting the terahertz signal into a photocurrent signal, which is then amplified by a current amplifier (SR570) and displayed on an oscilloscope. Last, the transmitted information was processed. As shown in Fig. 5C, the fidelity of the output signal curve compared to the input signal highlights the potential of the device in terahertz communication.

DISCUSSION

In conclusion, we systematically explore the hot-carrier PTE effect of semimetal PtSe₂ and its application in terahertz detection. On the basis of the metal-semimetal interface doping effect and the two-temperature heat transfer model, we construct a PTE response model and reveal a giant terahertz PTE response mediated by hot carriers at the metal-semimetal interfaces. In the terahertz band, the PtSe₂ PTE detector shows a high zero-bias responsivity of 0.62 A/W, a low NEP of 19.6 pW/Hz^{1/2}, and a fast response time of 4.5 ns. Because of

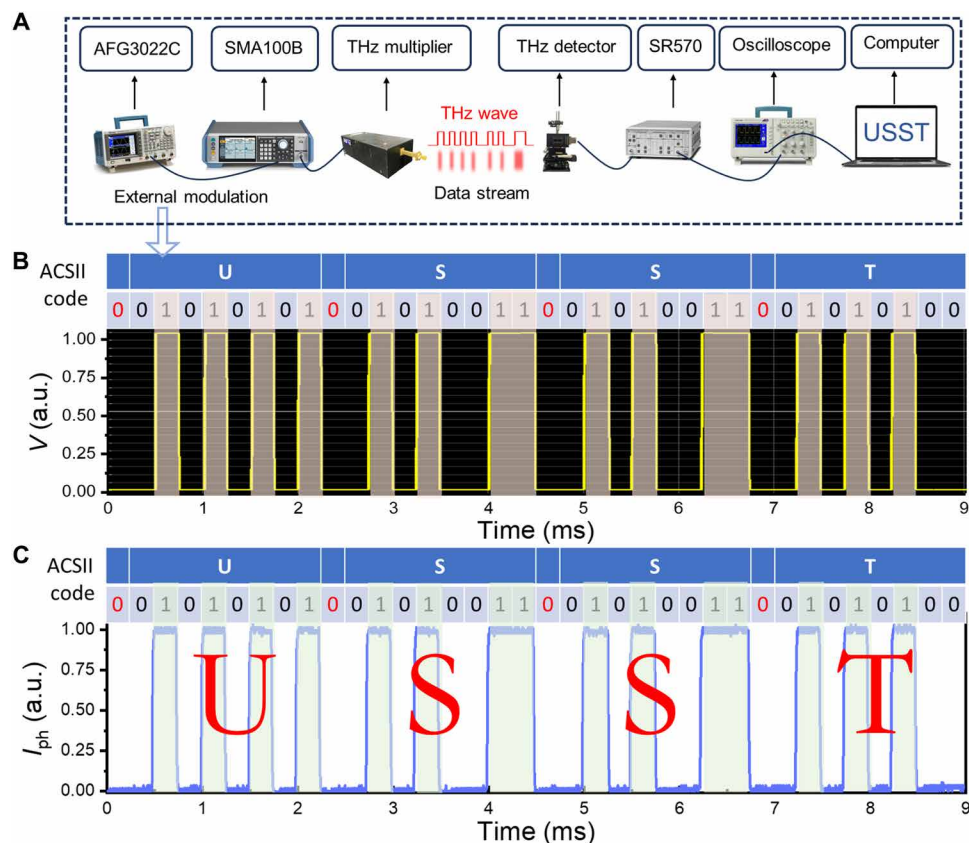


Fig. 5. Terahertz communication demonstration of the PtSe₂ detector at a carrier frequency of 100 GHz. (A) Schematic diagram of the terahertz communication setup at a carrier frequency of 100 GHz. (B) Normalized modulation voltage waveform with ASCII code information, where zero voltage represents “0” and the high level represents “1.” (C) Time-dependent photocurrent response of the PtSe₂ detector receiving the encoded information.

the difference between Au and PtSe₂ work functions, the doping effect at the metal-semimetal interface causes the change of the Seebeck coefficient, which is crucial to the PTE effect. Through TCAD simulation, we reveal the influence of metal doping effect and obtain the doping width. In the terahertz band, the electromagnetic energy is mainly transferred to the electron subsystem rather than the lattice subsystem due to weak electron–LO phonon interaction. By solving the two-temperature heat transfer equation under asymmetric terahertz heating, we reveal the intrinsic hot-carrier effect of PtSe₂. The large heat capacity and thermal conductivity of Au result in a large electron temperature gradient near the metal–PtSe₂ interface. Considering the electron temperature and Seebeck coefficient distributions in the PtSe₂ channel, we obtain the expression of the terahertz hot-carrier PTE response of the PtSe₂ detector. We find that the hot-carrier PTE effect at the metal-semimetal interfaces plays an important role in the terahertz response. The fast PTE response proves that PtSe₂ detector has a strong hot-carrier effect in the terahertz band. The two-temperature heat transfer simulations also identify that the intrinsic fast response of semimetal PtSe₂ device is due to the hot-carrier effect. In addition, the bias-dependent terahertz response reveals a hot-carrier PTE response associated with Peltier heat transport. Scanning photocurrent image shows that the zero-bias photocurrent of short-wave interband absorption is limited to the metal-semimetal interfaces. Because of the rapid spatial variation of carrier density at the metal–PtSe₂ interfaces, the presence of the built-in electric field and the change in the Seebeck coefficient lead to the coexistence of PV and PTE responses. In the visible spectral region, we theoretically proved that the PTE and PV responses near the metal-semimetal interface would show opposite bias-dependent behaviors. According to the bias-dependent behavior of the photoresponse, it is concluded that the zero-bias response at the metal-semimetal interfaces still originates from the PTE effect. Because of the narrow depletion-region width, the unique band structure of electron-hole coexistence on the Fermi surface, and the short photo-generated carrier lifetime, the PV response at the metal-semimetal interfaces is weak. In addition, our study provides a guideline for finding previously unknown material systems beyond graphene and PtSe₂. For an advanced material system, we believe that the following five requirements need to be considered: Good thermoelectric properties such as high Seebeck coefficient and low thermal conductivity, large light absorption coefficient, strong hot-carrier effect, broadband response, high environmental stability, and low cost of material growth. These findings promote the understanding of the hot-carrier PTE effect of semimetal PtSe₂, which is very valuable for further detector optimization and application.

MATERIALS AND METHODS

Device fabrication

High-quality PtSe₂ single crystals were grown by the flux zone method. Firstly, the planar metal asymmetric bow-tie coupling antenna structure was defined by electron beam lithography on a high-resistance intrinsic silicon substrate ($\rho > 20,000$ ohm-cm) with a thermally grown oxide layer SiO₂ (300 nm). Subsequently, a thermal evaporator was used to deposit Cr/Au (15/45 nm) film, after which a lift-off process was performed to obtain the desired electrodes. The PtSe₂ flakes were exfoliated from a bulk crystal using scotch tape and then transferred onto the prefabricated metal electrode channel

region based on the asymmetric bow-tie coupling antenna structure to form electrical contact by using a “PDMS transfer” technique.

THz photoresponse measurements

The *I*-*V* characteristic curve of the device was measured by a semiconductor parameter analyzer (Keithley, 4200A-SCS). For terahertz photoresponse measurements, the linearly polarized radiation at 75 to 110 and 140 to 220 GHz was generated from a 6× frequency multiplier and an 8× frequency multiplier driven by a microwave source (SMA, 100B). The radiation wave was modulated by a square waveform at the frequency of 1 kHz. The output power of the terahertz radiation was calibrated using a pyroelectric meter (Vector, H410). The power density at 75 to 110 and 140 to 220 GHz were 1 and 0.5 mW/cm², respectively. Photoresponse signal was recorded by a lock-in amplifier (Stanford, SR830) and an oscilloscope (Tektronix, MSO2024B) after a low-noise current preamplifier (Stanford, SR570). All measurements were carried out at room temperature and in the atmospheric environment.

Scanning photocurrent measurements

The photocurrent scanning imaging experiment of the PtSe₂ device is performed using a homemade scanning photocurrent imaging system. A fiber laser (Menlo, C-Fiber 780) is used as the light source to generate a 780-nm radiation, and a focused laser spot with a diameter of 2.5 μm is formed through a confocal optical system. The 2D step movement is achieved by an electrically controlled 2D translation stage, and the current values of the device under focused spot irradiation were recorded at each step point by the current source meter at the same time. The photocurrent of the device at each step point is defined as the difference between the current at laser irradiation and the dark current. After scanning the entire device area, the 2D photocurrent mapping was acquired by extracting the photocurrent value of each step point.

Supplementary Materials

This PDF file includes:

Supplementary notes S1 to S8
Figs. S1 to S11
Tables S1 and S2
References

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