Terahertz sensing based on the nonlinear electrodynamics of the two-dimensional correlated topological semimetal $TaIrTe_4$

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Abstract

The development of terahertz (THz) sensing technologies is limited by the lack of sensitive, broadband, and fast terahertz detectors. Thermal bolometers are bulky and slow, whereas electronic terahertz detectors (such as Schottky diodes) are fast, but their sensitivity degrades quickly outside a narrow frequency window. Here, we show that a two-dimensional correlated topological semimetal, tantalum iridium telluride ($TaIrTe_4$), has a large room temperature nonlinear Hall effect, and that the interaction between this effect and terahertz nonlinear electrodynamics can be used as a mechanism for terahertz sensing. Our photodetectors exhibit a high sensitivity (noise equivalent power of around 1 pW $Hz^{-1/2}$) and a large zero-

bias responsivity (around 0.3 A W^{-1}) a broadband spectral range (0.1–10 THz) at room temperature with intrinsic ultrafast response time (around ps). The zero-bias responsivity and noise equivalent power performance can be further improved (to 18 A W^{-1} and 0.05 pW $Hz^{-1/2}$, respectively) by introducing gate-tunable electron correlations.

Main

Terahertz (THz) technology is critical to quantum information technology and biomedical sensing because its frequencies (0.1–10 THz) resonate with low-energy collective excitations in quantum materials and molecular vibrations in biological matter. The ultrahigh bandwidth of the THz band could also of be use in high-speed wireless communication[1–4]. However, the widespread adoption of THz technologies has been hindered by the lack of sensitive, broadband, and fast THz detectors[5–7]. Current primary THz detectors can be classified into two categories[5,8]: thermal detectors that use THz-induced heating (such as bolometers and Golay cells), and electronic detectors that use THz-driven unidirectional electron flow across junction barriers to achieve nonlinear frequency rectification (such as Schottky diodes). Thermal-type THz detectors are bulky and slow due to thermal heating and transport, with response times on the order of milliseconds. In contrast, electronic rectifiers are compact and can detect nanosecond signal modulation at room temperature, but their responsivity decreases with frequency (less than 100 V W^{-1} or 0.1 A W^{-1} beyond 1 THz)[9,10] , and they have narrow operating bandwidth (~ 0.1 THz) due to intrinsic junction capacitance and resonant tunnelling at the metal-insulator interfaces[5]. It is therefore critical to establish a strong THz light–matter interaction mechanism and develop new material platforms to simultaneously boost responsivity, sensitivity (or noise-equivalent power; NEP), spectral range, and response speed[11].

Recently, topological effects in non-centrosymmetric quantum semimetals, such as Weyl semimetals, have been found to cause prominent nonlinear properties[12,13]. For example, it has been shown that the nonzero quantum geometric property[14–17] — often referred to as Berry curvature $\Omega_n(k)$ — that occurs in the layered topological semimetal tungsten ditelluride (WTe2) can lead to a novel nonlinear Hall effect even at room temperature, where longitudinal a.c. driving currents can be rectified into a large transverse d.c. current without invoking any diode or junction region. The underlying mechanism is related to the asymmetric distribution of Berry curvature in momentum space. When an electric field is applied, electrons in the material redistribute in momentum space, leading to an imbalance in the Berry curvature experienced by the electrons. This imbalance gives electrons an anomalous transverse velocity (proportional to the product of the Berry curvature imbalance and the applied electric field), thus leading to a d.c. rectification current[18,19]. Due to their divergent Berry curvature and asymmetric scattering, the second

order nonlinear susceptibility χ_{abc} of topological semimetals are several orders of magnitude higher (~ 10^{-6}-10^{-1} m V^{-1})[20,21] than that in typical nonlinear optical materials such as $LiNbO_3$ (~ 10^{-10}-10^{-9} m V^{-1}) [22,23], providing highly efficient frequency conversion. These findings at ultra-low frequency (ULF; < 3 kHz) highlight the critical role of quantum geometrical properties and corresponding band structure engineering in enabling new rectification mechanisms. If this junction-free mechanism can be extended to the THz regime[24], it could bypass junction-induced performance limitations in existing electronic THz detectors. Gapless and conducting semimetals also intrinsically support broadband and fast photodetection.

In this Article, we report a zero-bias and junction-free THz rectification driven by an intrinsic large Berry curvature dipole (BCD) in few-layer tantalum iridium telluride (TaIrTe4), a non-centrosymmetric 2D correlated topological semimetal. We first examine the THz electrodynamics in atomically thin TaIrTe4 via nonlinear Hall transport measurements and demonstrate a large nonlinear Hall effect. This is then used to develop a THz rectification method that differs from previous bulk centrosymmetric semimetals[25,26], We show that a THz photodetector using this mechanism can offer a fast response (~ ps), high sensitivity (NEP ~ 1 pW $Hz^{-1/2}$) and a large room-temperature responsivity (R~ 0.3 A W^{-1}). We also find that strong electron correlation in the 2D limit leads to substantial band renormalization and can further boost responsivity (R ~ 18 A W^{-1}) and sensitivity (NEP ~ 0.05 pW $Hz^{-1/2}$). The electron correlation and THz rectification performance can be modulated by in situ electrostatic gating due to the weak dielectric screening at the few-layer limit.

Large room-temperature nonlinear Hall effect in atomically thin $TaIrTe_4$

$TaIrTe_4$ crystallizes in an orthorhombic structure that belongs to the space group $Pmn2_1$. Each monolayer of $TaIrTe_4$ comprises a layer of Ta (or Ir) atoms sandwiched between two layers of Te atoms in a distorted octahedral coordination. Because $TaIrTe_4$ is formed by stacking these monolayers with alternating layers rotated by 180 degrees, inversion symmetry is broken. Bulk $TaIrTe_4$ is recognized as a type-II Weyl semimetal with nontrivial Berry curvature monopoles[27]. Thinning down to few-layer limit (Fig. 1a), its space group changes to Pm due to the lack of the screw-axis and glide-plane symmetries at the surfaces. This symmetry change is critical for enabling the nontrivial Hall effect via in-plane driving electric field, which also has been observed in isostructural WTe_2[21,28,29]. In our study, we obtained ultrathin $TaIrTe_4$ flakes by mechanical exfoliation and dry transfer to target SiO_2/Si substrates with thin h-BN capping (see Methods and Supplementary Section 1 and 2). To confirm the change from $Pmn2_1$ to Pm, we have conducted polarization-resolved SHG and observe a two-fold pattern with minor lobes. This fits well with the nonlinear susceptibility of the Pm space group (Fig. 1b, see Supplementary Section 3). Based on the Boltzmann equation and the anomalous motion equation of Bloch electrons[18,19], the nonlinear Hall

rectification current density can be expressed as $J_a(\omega = 0) = \left(\frac{1}{1+\omega^2\tau^2}\right)\chi_{abc}E_b(\omega)E_c(-\omega)$. Here χ_{abc} is nonlinear conductivity tensor, $E_b(\omega), E_c(-\omega)$ are alternating electric field at angular frequency ω (E_b equals to $E_c$for single AC input) and τ is the average scattering time. It is proportional to the product of nonlinear conductivity tensor χ_{abc} at DC limit and a frequency dependent factor $\tau/(1+\omega^2\tau^2)$. Furthermore, χ_{abc} is directly related to Berry curvature $\Omega_n(k)$ in the following expression: $\chi_{abc} = e^3\tau \int_k (\partial_b f_0)\,\Omega_n(k)$ $= -e^3\tau \int_k (f_0)\,\partial_b\Omega_n(k)$, f_0 is the equilibrium electron distribution. To understand the subsequent THz rectification results, it is essential to first examine the nonlinear conductivity at DC limit. Accordingly, we have conducted nonlinear Hall transport measurements in a few-layer sample with Hall bar device geometry at room temperature (Fig. 1c inset), which can probe the nonlinear conductivity around the Fermi level. In particular, we use an AC driving current (I^{ω}) at 500 Hz applied along the *a* axis and measure the generated NHE voltage ($V^{2\omega}$) along the *b* axis. The $V^{2\omega}$ shows quadratic dependence of input I_{AC} (Fig. 1c), which is a hallmark of the NHE effect. Moreover, the NHE generation efficiency $\eta_{ab} = \frac{V_{ab}^{2\omega}}{(V_{aa}^{\omega})^2}$ and nonlinear Hall conductivity χ_{baa} are found to be significant, where a and b represent directions along which voltages are applied and measured. For the measured bilayer $TaIrTe_4$, its χ_{baa}can reach to ~ 6.6×10^{-2} μm V^{-1} Ω^{-1} at room temperature (See Supplementary Section 4). This value is more than three orders of magnitude larger than that in previous transport study on thick $TaIrTe_4$ flakes (> 20 nm)[30]. On one hand, the lower symmetry at ultrathin limit enables intrinsic Berry curvature contribution which are not present in bulk limit. On the other hand, our DFT and model calculation suggests strongly hybridized Te-p and Ta or Ir-d orbitals near the Fermi level in $TaIrTe_4$, leading to a nontrivial Berry curvature dipole and corresponding large nonlinear Hall effect (Fig. 1d, and Supplementary section 5).

High-performance Room-temperature THz sensing mediated by nonlinear Hall effect

Building upon the large NHE in few-layer $TaIrTe_4$, we now interrogate the interplay of NHE with THz electrodynamics as a new mechanism for THz sensing (Fig. 2a). To implement THz sensing, we designed and fabricated atomically thin $TaIrTe_4$ THz photodetectors in contact with a pair of Cr/Au pre-patterned electrodes and a bow-tie antenna (Fig. 2b inset). Specifically, the antenna is positioned along the *a* axis of $TaIrTe_4$, while two electrode contacts are aligned along the *b* axis for collecting the corresponding rectified current via NHE. To benchmark the responsivity, the ratio between photocurrent and incident THz irradiation power, we use an CW IMPATT diode with calibrated power as the THz excitation sources (0.1 THz). The incident THz light is focused onto the photodetector for THz photocurrent measurements (see Methods and Supplementary Section 6). Firstly, the incident power-dependent photovoltage measurement shows linear (or quadratic) relationship between the photovoltage and the incident power (or electric field)

at room temperature (Fig. 2b), consistent with the power law of the NHE rectification mechanism. In Fig. 2c, we show the responsivity of multiple $TaIrTe_4$ samples with different thicknesses for the same incident power (See Supplementary Section 7 for the calculation procedure). As a comparison, we also measured the THz photocurrent response of a graphene detector based on the same device geometry. Compared with the response of all $TaIrTe_4$ devices, the reference graphene sample shows significantly lower photovoltage. This comparison under the same device geometry and the THz photocurrent measurement setup suggests that the strong response in $TaIrTe_4$ should come from the NHE, which is not possible in the centrosymmetric graphene device. Moreover, a responsivity enhancement has been observed from thick layers down to bilayer (Fig. 2c). This may result from both the crystal symmetry and electronic band mixing difference at Fermi level between bulk and ultrathin samples. Note that the THz responsivity of bilayer $TaIrTe_4$ has achieved 0.3 A W^{-1} without any bias or preamplifier amplification at room temperature. The value is significantly higher than that in previously studied bulk $NbIrTe_4$ and bulk 1T-$CoTe_2$ without bias[25,26]. In bulk $NbIrTe_4$, the screw-axis and glide-plane symmetries are preserved to eliminate in-plane photocurrent current generation only by normal incident THz wave with in-plane driving electrical field. Thus, an additional voltage bias must be applied to induce asymmetry and promote carrier diffusion, however this also generates large dark current even without light irradiation, which significant undermine the detection sensitivity. While for 1T-$CoTe_2$, it is mediated by extrinsic disorder contribution and its responsivity includes additional gain from an external preamplifier. In contrast, the rectification in ultrathin $TaIrTe_4$ is primarily attributed to the intrinsic quantum geometrical property as verified by the scaling law analysis of nonlinear Hall response (See Supplementary Section 8), resulting in a large responsivity at room temperature. Such a large responsivity also leads to high sensitivity of our THz photodetector, giving a noise equivalent power (NEP) down to pW $Hz^{-1/2}$ (Fig. 2d and Supplementary Section 9 for calculation procedure). This value is comparable to the top performance of room-temperature THz photodetectors such as GaN high-electron-mobility transistor (HEMT, 0.6 pW $Hz^{-1/2}$), and orders of magnitude better than commercialized bolometers (~ 30 -200 pW $Hz^{-1/2}$), Golay cells (~ 100 pW $Hz^{-1/2}$) and Schottky diodes (10 – 30 pW $Hz^{-1/2}$)[5,8]. Such high responsivity and low noise equivalent power via nonlinear Hall mechanism in the atomically thin topological semimetals can enable practical applications such as high-performance room temperature THz imaging (See Supplementary section 10 for more information).

To further investigate the NHE mechanism, we examine its anisotropic response with incident THz polarization. In Fig. 2e, we show the photovoltage response at different angles of THz polarization, from the transverse (*b* axis) to the longitudinal (*a* axis). We consistently observe a maximum photovoltage when the incident polarization angle is aligned with the *a* axis. Additionally, the photovoltage measured along the longitudinal direction (*a* axis) is significantly lower than that along the transverse direction (*b* axis). This photocurrent generation anisotropy is well aligned with the expected nonlinear Hall response in a Pm

space group crystal. Building upon the confirmation of nonlinear Hall rectification mechanism, we further investigate its potential for THz rectification at higher incident frequency (Fig. 2f and Supplementary Section 11). Surprisingly, the responsivity remains above 0.2 A W^{-1} for 3 to 10 THz with a less than 25% drop compared to the responsivity at 0.1 THz. This behavior overcomes the grand frequency cutoff challenge in conventional electronic THz rectifiers such as Schottky diodes[9,10], which typically only have narrow operating bandwidth ~ 0.1 THz. Such a broadband response is suspected to result from the junction-free device nature and relatively short carrier lifetime of semimetals at room temperature ($\omega\tau < 1$ and prefactor $\frac{1}{1+\omega^2\tau^2} \sim 1$). Indeed, based on Hall measurements the carrier lifetime at room temperature is only tens of femtoseconds (See Supplementary Section 12).

Besides responsivity, NEP and response bandwidth, response time is another important device metric for THz photodetectors. For the fundamental nonlinear Hall dynamics and device bandwidth characteristics, both the intrinsic and extrinsic response times need to be considered. In few-layer $TaIrTe_4$, the intrinsic response time represents the fundamental speed limit governed by the nonlinear rectification mechanism for photocurrent generation, while the extrinsic response time is longer and accounts for an additional bandwidth limit from photodetector circuits. In our photodetectors, we determine the intrinsic time response by ultrafast autocorrelation measurements[31]. In this configuration (Fig. 2g inset), picosecond THz pulses are generated by optical rectification of femtosecond NIR pulses in BNA organic crystals[32] (See Supplementary Section 13). Each THz pulse is split up into two pulses and subsequently recombined collinearly at the device under test. The photovoltage is measured as a function of the two-pulse delay time. Given the nonlinear rectification process, a maximum enhancement of photocurrent signal is observed at zero delay (Fig. 2g). As the time delay increases, the less overlap of the two THz pulse envelopes results in a smaller interference of NHE current or voltage rectification. Finally, the measured photovoltage reaches to a constant level, where the two input THz pulses are completely separated in the time domain. By fitting the autocorrelated photocurrent data with input THz pulse convolution, we estimate an intrinsic response time on the order of ps, suggesting its great potential for high-speed applications. We also characterized the extrinsic response time of our prototype THz sensor (See Supplementary Section 14), giving a rise time of $4.2 \pm 0.4\ \mu s$. Further device optimization can be implemented on the ohmic contacts, the dielectric environment and the device geometry to further reduce resistance and capacitance for achieving the intrinsically fast speed benefiting from its unique junction-free rectification feature[33].

Enhanced THz rectification by an emergent correlated charge ordering

In addition to the comprehensive THz rectification characterization at room temperature, we observed a strong temperature dependence of the THz photovoltage in ultrathin $TaIrTe_4$ (Fig. 3a). When

cooling from 300 K to 65 K, the responsivity exhibits only minor changes. However, upon cooling below 65 K, the responsivity dramatically increases and continues to increase down to the lowest temperatures of our setup (4 K), reaching a maximum of 13.7 A W^{-1}. Concomitantly, the absolute photovoltage is also substantially enhanced as compared to that at room temperature. Moreover, we find a nearly π phase shift in photocurrent simultaneous to the dramatic amplitude change (Fig. 3b). Given the same incident THz power and frequency, the nonlinear Hall formular suggests an abrupt change in both amplitude and sign for χ_{baa}. Furthermore, the detection sensitivity of the device is also boosted (Fig. 3c) with the NEP of the bilayer $TaIrTe_4$ reaching 5×10^{-14} W $Hz^{-1/2}$ at 4 K, better than that of quantum well photodetectors and Si bolometers at similar temperatures[5,8].

To understand the origin of such a dramatic property change, we have conducted the temperature-dependent resistance measurement on another bilayer $TaIrTe_4$ device (Fig. 3d). Here, *R* represents the four-probe resistance along the crystalline a axis. Upon cooling from room temperature, the resistance decreases and can be fit by $R \sim T$, which is consistent with reduced phonon scattering in metals or semimetals[34]. Below 100 K, the resistance dependence changes and can be fit by $R \sim T^2$. This scaling law change suggests that the sample starts to exhibit Fermi-liquid behavior due to the nontrivial electron-electron interactions[35,36]. Further cooling down, a resistance anomaly has been observed around the transition temperature T_C = 66 K. This signature is more evident in *dR/dT* plot (Fig. 3d inset). Such a resistance anomaly has been widely recognized as a hallmark for phase transitions mediated by strong electron correlation in quantum semimetal and metals[37–39]. And the transition temperature is typically defined at the anomaly peak or dip positions in *dR/dT* plot. Indeed, such a strong electron correlation has also been observed in centrosymmetric monolayer $TaIrTe_4$ via quantum transport[40]. Their electronic susceptibility calculations suggest a phase instability in conduction band at a nesting vector of Q ($0.068 \cdot \frac{2\pi}{a}$, $0 \cdot \frac{2\pi}{b}$), connecting two neighboring van Hove singularities. This instability may lead to an electron density modulation along the one-dimensional Ta chain (*a*-axis), with a relatively large period of about 15 atomic unit cells. Future experimental study such as scanning tunneling microscopy would enable the direct visualization of the proposed charge density wave pattern.

Nevertheless, this emergent electron correlation could result in nontrivial band renormalization with flatten band and abundant band inversions around the Fermi level. This leads to a more drastic change of Berry curvature in momentum space with greatly enhanced $\partial_k \Omega_n(k)$, which is proportional to the strength of nonlinear Hall effect. Besides, the emergence of less dispersive bands with more density of states available around Fermi level can also promote the THz rectification response. Indeed, our calculations based on Hartree Fock method point out the introduction of electron correlation can modify the bands around Fermi level and results in Berry curvature dipole change (Supplementary Section 15). In

particular, we find that Berry curvature dipole D_{ac} can be enhanced when the electron correlation occurs. Also, the sign of the Berry curvature distribution can be flipped, consistent with the observed 180-degree phase change of the THz photocurrent after the transition.

Beyond the transport anomaly observation, such an electron correlation scenario as the emerging new state is further evinced by optical SHG and polarization-resolved THz photocurrent measurements. For the former (Fig. 3e), we observe an associated transition in the SHG intensity, which we suspect is from an increase in the electronic density of state due to band renormalization and folding. In addition, we observe a monotonic enhancement of the THz photocurrent anisotropy when the topological semimetal transitions into this new correlated state (Fig. 3f). For example, when the incident linear THz wave polarization is rotated, the *b*-axis photocurrent of ultrathin $TaIrTe_4$ shows an intensity anisotropy ratio of 14:1 (*a* axis: *b* axis incidence wave) in this new state at 4 K, about a factor of 4 larger than the room temperature value. Similar enhancement of anisotropy arises from the phase transition has also been observed in the temperature-dependent optical linear dichroism (see Supplementary Section 16).

Electrostatic gate control of the correlated charge ordering and the nonlinear THz electrodynamics

Finally, we examine how in-situ electrostatic gating can influence the correlated charge ordering and corresponding THz electrodynamics. Using a SiO_2/Si back gate in a bilayer $TaIrT_4$ device, we measured the THz photocurrent response as a function of gate bias and temperature. The gate-temperature responsivity mapping shows nontrivial modulation of the phase diagram and the further enhancement of the THz responsivity (Fig. 4a, b and Supplementary Section 17). Two dome-like regions with enhanced THz photoresponsivity are observed, one at positive and the other at negative gate biases. For example, the responsivity reaches ~ 18 A W^{-1} at 4 K for a gate bias at 30 V and -100 V, respectively. A gate modulation of nonlinear Hall strength, which varies over a factor of 4, is also observed at 4 K (Fig. 4c), with its maximum value about 1.5 times at room temperature (Fig. 4d and Supplementary Section 17). Another interesting observation is the gate-controllable phase switching between the electron correlated state and the noninteracting semimetal state, resulting in on-demand modulation of THz rectification strength and photocurrent flow direction. In particular, the 2D phase mapping (Fig. 4b) clearly outlines the gate-controllable phase boundaries between a noninteracting semimetal state and the correlated electronic phase with a tunable transition temperature from 40 to 80 K. As shown in Fig. 4e, this gate controllable switching is especially visible near the pristine transition temperature, where V^{DC} completely switches sign. Thus, we demonstrate that both the nonlinear Hall strength and THz rectification current direction can be controlled on-demand (See Supplementary Section 17 for further comparison). The observed gate-dependent THz electrodynamics can be understood in the following manner: the intrinsic BCD influences on nonlinear electrodynamics are highly susceptible to the position of the Fermi level and the applied electric field. A

recent theoretical analysis highlights the Fermi level dependence for intrinsic Berry curvature, side-jump and skew scattering contributions in nonlinear Hall rectification[41]. Our DFT calculations also indicate the electric field, imparted by gate, can modify the Berry curvature and corresponding nonlinear Hall strength (Supplementary Section 18), showing qualitative agreement with our experimental data. Finally, the electron correlation can also be influenced by the gate bias leading to the observed gate-controlled phase transition near the intrinsic transition temperature (See Supplementary Section 17 for more details). Taken together, the band nesting and resulting flat band effects on the nonlinear Hall rectification process can be substantially modified and enhanced by applying electrostatic control, which is a unique tuning knob for 2D layered topological semimetals.

Conclusions

We have investigated the interplay between quantum geometrical properties, gate-tunable electron correlation and THz electrodynamics in atomically-thin layers of a topological semimetal, TaIrTe4. Combined with first-principles calculations, we found the electron correlation in 2D TaIrTe4 induces substantial band renormalization with abundant band crossings and inversions around the Fermi level, which leads to drastic changes in the Berry curvature and enhanced nonlinear Hall effect. This nonlinear Hall effect can be used for THz rectification, allowing electrical THz photodetectors to be created with a room temperature responsivity of 0.3 A W^{-1}, low NEP of ~ 1 pW $Hz^{-1/2}$, fast intrinsic speed (~ps), and broadband THz response (0.1–10 THz). Moreover, we found that the responsivity can be increased by almost a factor of 50 (to 18 A W^{-1}) and NEP of 0.05 pW $Hz^{-1/2}$ when the topological semimetal transitions to a correlated electronic phase. The responsivity and sensitivity of few-layer TaIrTe4 detectors show several advantages compared to current electronic-type THz detectors based on conventional technology or 2D materials (Fig. 4f). Our work advances the understanding of THz nonlinear electrodynamics in 2D correlated topological semimetals and could be used to develop high-performance THz sensing technology[47].

Methods

Single crystal synthesis

Single crystals of $TaIrTe_4$ were synthesized via a flux method using excess tellurium. Ta powder (99.98%), Ir powder (99.99%) and Te lumps (99.9999%) were loaded in a 1:1:20 ratio (Ta:Ir:Te) into an alumina Canfield crucible set and sealed in a quartz ampule under vacuum (5×10^{-6} Torr). The regents were then

heated to 1100 ℃ for over 24 hours and dwelled for 5 days, before cooling to 600 ℃ at 1 ℃ /hr. The ampule was then quickly cooled to 525 ℃ and centrifuged to remove excess tellurium. To remove any residual Te on the surface, the resulting single crystals were sealed in another evacuated ampule and annealed at 425 ℃ for 2 days with a ~200 ℃ temperature gradient.

THz sensing device fabrication

Multilayer hexagonal boron nitride (hBN) and few-layer $TaIrTe_4$ flakes were mechanically exfoliated and picked up using a dry stacking method. Then the entire hBN/$TaIrTe_4$ stack was transferred onto a 280 nm SiO_2/Si substrate with pre-patterned metal contacts and bow-tie antenna. All device fabrication processes are conducted within a nitrogen-filled glove box with O_2 and H_2O level less than 0.01 ppm. The thickness of the $TaIrTe_4$ flakes was confirmed by optical contrast and atomic force microscopy.

THz photocurrent characterization

0.1 THz CW wave is generated by an IMPATT diodes (Terasense Group Inc) with electrical trigger modulation. The modulated THz wave is routed by a set of gold parabolic mirrors, then focused onto the sample. The sample is mounted on a copper sample holder inside an optical cryostat (Cryo Industries of America, Inc) with temperature range from 4 to 500 K. The cryostat is mounted on an XYZ stage for signal optimization. After the THz signal is transformed to DC voltage by the few-layer $TaIrTe_4$ rectifiers, the voltage signal is then collected and analyzed by a preamplifier (Stanford Research, SR570) and a lock-in amplifier (SSI-Instrument, OE1022D). The responsivity calculation has divided the preamplifier gain to obtain the intrinsic responsivity of the device.

Ultrafast autocorrelation photocurrent measurements

We used a mode-locked Ti:Sapphire laser (Astrella, Coherent) capable of delivering 35 fs optical pulses (λ_{center} = 800 nm) at a repetition rate of 1 kHz to generate coherent THz pulses for the autocorrelation measurements. In particular, coherent THz pulses were induced through an optical rectification process via optical pumping of a pair of BNA crystals. The pair of THz pulses are subsequently recombined collinearly onto the device under test. The photovoltage is measured as a function of the two-pulse delay time. Given the nonlinear rectification process, a maximum enhancement of photocurrent signal is expected at zero delay. As the time delay increases, the less overlap of the THz pulses envelopes will result in smaller current rectification profiles interference until it reaches to a constant level, where the rise edges and fall edges of rectification currents by the two pulses are not affecting with each other. By fitting the autocorrelated photocurrent data, one can estimate an intrinsic response time determined by the distinct THz rectification

dynamics.

Nonlinear optical spectroscopy

The measurements were conducted using an optical second harmonic detection setup coupled with a cryogenic system (OptiCool, Quantum Design) capable of operating from 1.7 K to 300 K. The excitation light at 1040 nm was generated by a tunable femtosecond laser (Discover NX, Coherent Inc.). This excitation laser was linearly polarized with polarization rotating using a half waveplate and focused on the sample through a 50x NIR objective, achieving a spot size around 3 μm. The second harmonic generation (SHG) signal was detected in a backscattering configuration, transmitted through a polarizer aligned parallel to the crystal *b* axis and collected using photon-counting PMT modules. For the SHG polarization pattern study, a half-wave plate was used to control the polarization of the incident light, while a polarizer was employed to analyze the emitted SHG light from the sample.

First Principles Calculation

We use the Vienna ab initio simulation package (VASP) [48]to determine the most stable structure and analyze the electronic properties of $TaIrTe_4$. The projector-augmented-wave (PAW) pseudopotentials are utilized to describe the valence electron configurations for Ta, Ir, and Te., specifically $5p^6\ 6s^2\ 5d^4$, $6s^1\ 5d^8$, and $5s^2\ 5p^4$, respectively. The Perdew-Burke-Ernzerhof (PBE) functional is employed to address electronic interactions. The plane waves' energy cutoff is set at 500 eV, and the Gaussian smearing method's width is chosen to be 0.05 eV. A Gamma-centered k-mesh of 16×8×1 is used for Brillouin Zone (BZ) integrations, with a spacing resolution of 0.02 2π/Å. To achieve the ground state, the convergence criteria for lattice optimizations are set at 10^{-6} eV and 0.1 meV/Å for total energy and ionic forces, respectively. The optB88-vdW correlation functional is applied to account for the van der Waals (vdW) interactions. To investigate the transportation feature, we transformed the eigenstates of DFT calculations into a set of Maximally localized Wannier functions. [49]

As for the NLHE, the intrinsic part of the NLH conductivity $\chi_{\alpha\beta\beta}$ can be described by the BCD [50], which can be calculated as follows:

$\chi_{\alpha\beta\beta} = -\epsilon_{\alpha\beta\gamma}\frac{e^3\tau}{2\hbar^2(1+i\omega\tau)}D_{\beta\gamma}$,

$D_{\beta\gamma} = \int_k f_n^0(\boldsymbol{k})\frac{\partial\Omega_\gamma^n}{\partial k_\beta}$,

Where $D_{\beta\gamma}$ is the BCD, $f_n^0(\boldsymbol{k})$ is the equilibrium Fermi-Dirac distribution, τ is the relaxation time, $\epsilon_{\alpha\beta\gamma}$ is the third rank Levi-Civita symbol, $\alpha, \beta = a, b$ and $\gamma = c$ in 2D. The Berry curvature Ω_c in the 2D system can be calculated by [51],

$\Omega_c(\boldsymbol{k}) = -2Im \sum_{m \neq n} \frac{<n|\partial_{k_a} H|m><m|\partial_{k_b}|n>}{(\epsilon_n - \epsilon_m)^2}$,

Where ϵ_n and $|n\rangle$ are eigenvalues and eigenvectors, respectively.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Code availability

The codes used for the calculation are available from the corresponding authors on reasonable requests.

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Author Contributions

J.X. and T.X. conceived the research and designed the experiments. Y.W. and J.X. supervised the project. Y.H. synthesized the bulk high-quality $TaIrTe_4$ crystals under the guidance of D.R.; T.T. and K.W. provided the high-quality h-BN bulk crystals. Y.G. designed the THz sensing device with H.J. and T.X., under the guidance of D.V.W., Y.W. and J.X.; H. J., C.F., T.X. and Y.M. fabricated the devices under the guidance of Y.W. and J.X.; T.X. performed the THz photocurrent and SHG measurements and analyzed the data with J.X.; J.R. conducted the AFM measurements under the guidance of J.X.; J. Li. , L. P. and Y. Z. performed first-principles and Hartree-Fock calculations. All authors discussed the results and jointly wrote

the paper.

Competing interests

J.X. Y.W. and D.V.W. have submitted a patent application ('Terahertz radiation detectors based on thin films of non- centrosymmetric layered topological semimetals'; US no. 18/448,648) that covers a specific aspect of the manuscript. The other authors declare no competing interests.

Figure Legends/Captions

Figure. 1: Large nonlinear Hall effect in few-layer $TaIrTe_4$ at room temperature. a, Top and side views of the crystal structure of few-layer $TaIrTe_4$. The lack of the screw-axis and glide-plane symmetries at few-layer limit allows an in-plane polar axis along the mirror line for nontrivial in-plane nonlinear Hall effect. **b,** Polarization-resolved second harmonic generation (SHG) of a four-layer $TaIrTe_4$, whose pattern fits well to Pm space group. Here only the incident light polarization is rotating. 0 degree refers to the polarization aligned with crystal *b* axis, and it is also the fixed SHG detection polarization direction. **c,** The nonlinear hall transport measurement of a bilayer $TaIrTe_4$ at 300K. Second-harmonic transverse voltage $V^{2\omega}$ in response to an applied a.c. I^{ω} along crystalline *a* axis, shows quadratic power law and large second-order nonlinear conductivity χ_{baa} about 0.1 μm V^{-1} Ω^{-1}, which is about three orders of magnitude than that in bulk $TaIrTe_4$ [30]. The inset is the schematics of a Hall bar device and measurement configuration. **d,** Calculated local Berry curvature dipole distribution $D_{ac} = D_{xz} = \partial_{k_x}\Omega_z(k)$ in the two-dimensional Brillion zone at $E=E_F + 0.02$ eV for bilayer $TaIrTe_4$. Here k_x is along the *a* axis in the calculations. The Berry curvature dipole unit here is Å.

Figure. 2: Room-temperature THz rectification in few-layer $TaIrTe_4$ topological semimetals.
a, Schematics for a new THz sensing mechanism in 2D topological semimetal $TaIrTe_4$. In particular, large nonlinear Hall effect efficiently rectifies THz wave to DC voltage mediated by the diverging Berry curvature in layered topological semimetals. Photovoltage is maximized along the *b*-axis with incident THz electric field along the *a*-axis. The resulting THz photodetector may enable sensitive, broadband, and ultrafast THz wave detection. **b,** Incident THz fluence dependent photovoltage. It shows linear (or quadratic) relationship between the photovoltage and the incident power (or electric field). Error bars of black circles (mean) represent standard deviations result from $n = 1{,}000$ random samplings at each data point. The inset shows the typical THz sensor based on few-layer $TaIrTe_4$ (cyan flake) covered by thin h-BN (sapphire blue flake), the scale bar is 10 μm. **c,** Photovoltage response comparison of few-layer $TaIrTe_4$ with various thickness and reference graphene sample. **d,** Noise equivalent power (NEP) characterization of a bilayer $TaIrTe_4$ THz sensing device shows superior sensitivity at room temperature. **e,** THz polarization dependence of photovoltage measured along *a* and *b* axis. The legends in the figure (a and b) represent along which crystalline direction the photocurrent is measured. The horizontal axis represents the incident THz wave polarization direction,

the 0 degree refers to the polarization aligned parallel to the crystalline *b* axis. The signal variation is consistent with underlying nonlinear Hall rectification mechanism with certain Pm space group symmetry constraint. **f,** Broadband THz response characterization of a bilayer $TaIrTe_4$ device. It shows large responsivity over the entire THz regime from 0.1 to 10 THz before suffering significant cut-off. Broadband blackbody radiation source and THz filters are used (see Supplementary Section 11 for more details). Error bars represent standard deviations result from $n = 2{,}000$ random samplings at each data point. **g,** Intrinsic response time characterization by ultrafast autocorrelation measurements, suggesting an ultrafast rectification response down to picosecond level. The inset shows the pulsed autocorrelation measurement schematics.

Figure. 3: Enhanced THz electrodynamics by correlated charge ordering in few-layer $TaIrTe_4$.

a, Temperature dependent THz responsivity of a bilayer $TaIrTe_4$ device. A dramatic enhancement of THz rectification is observed below a critical transition temperature ($T_c \sim 66$ K). **b,** Temperature dependent THz photovoltage phase of the same device. Below the T_c, a synchronized π phase shift is observed, suggesting a sign change in intrinsic Berry curvature dipole and corresponding nonlinear Hall rectification. **c,** NEP characterization after the transition shows greatly enhanced THz sensitivity performance down to 50 fW $Hz^{-1/2}$. **d,** Temperature dependent resistance in a bilayer $TaIrTe_4$ device. Here *R* represents the four-probe resistance along the *a* axis. Upon cooling below 100 K, the sample starts to show Fermi liquid behavior ($R \sim T^2$) associated with electron-electron interaction. A resistance anomaly is observed around the transition temperature ~ 66 K, which is more evident in the *dR/dT* plot (Fig. 3d inset). **e,** Temperature dependent second harmonic generation (SHG) of a few-layer $TaIrTe_4$. SHG intensity increases is found below the transition temperature. The incident light polarization is fixed along crystalline axis *b*, with no polarizer inserted for detection. Error bars of black rectangle represent standard deviations result from random samplings with gate number of 100 collected by PMT. **f,** Incident THz polarization dependent photovoltage at varying temperature. The angle in the horizontal axis represents the incident THz wave polarization direction with 0 degree defined as the polarization parallel to the crystalline *b* axis. The photovoltages are normalized by the values measured at 0 degree. Also, all photovoltages here are collected along the crystalline *b* axis. Error bars of circles represent standard deviations result from $n = 1{,}000$ random samplings.

Figure. 4: Electrostatic gate control of electron correlation and THz rectification

a, Mapping of THz rectification responsivity as a function of gate bias and temperature. **b,** Corresponding mapping of THz rectification voltage phase. **c, d,** Single gate modulation of THz photovoltage at 4K and 300K, respectively. **e,** Gate control of phase transitions and THz photovoltage flip at 66 K. **f,** THz sensing performance comparison among electronic-type detectors based on different 2D layered materials and conventional THz rectifiers with zero source-drain bias to avoid large dark current. BP: black phosphorous[42], QW: quantum well[43], SD: Schottky diode[10]. The data points of Bi_2Se_3, $PdTe_2$ and graphene are adapted from the literature[44–46].

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