Abstract

Cryogenic Near-field Imaging Technology at Terahertz Frequency

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This dissertation reports on data acquisition method and the application of world’s first cryogenic apertureless near-field microscope designed for terahertz frequencies. The dissertation briefly summarizes the commonly used data acquisition methods and the existing challenges in applying near-field technology using broadband terahertz sources. We devised, implemented, and validated a novel measurement technique to resolve the challenges. The novel method improves the traditional method by providing the information of the carrier-envelop-phase of the terahertz pulse.

The physical properties of WTe₂ microcrystals depend sensitively on the layer number. By applying both the traditional and the novel techniques, we systematically explored the layer-dependent electromagnetic response of mono-layer and few-layer tungsten ditelluride (WTe₂) microcrystals. On tri-layer WTe₂, we discovered the plasmonic response and imaged the real-space pattern of the terahertz plasmon using the novel measurement technique. On bi-layer WTe₂, our measurements support that the band alignment is semi-metallic instead of semi-conducting.

Near-field technology at terahertz frequency is sensitive to the Drude behavior of condensed matters. We imaged the electromagnetic response of the transition of cadmium osmate (Cd₂Os₂O₇) crystals from a high temperature metal to a low temperature magnetic insulator. The result is consistent with the temperature dependence in the direct-current conductivity.
In the end, the dissertation discusses the theory and simulation of imaging hydrodynamic flow of materials with viscous electron systems via nano-photocurrent technique. In anisotropic material, nano-photocurrent measures the geometrical properties of the Shockley-Ramo auxiliary field or flux. As a result, the nano-photocurrent is a good candidate to detect the boundary layer and vortex flow pattern of a viscous electron system.
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Acknowledgments

First and foremost, I would like to express my gratitude to my advisor, professor Dimitri N. Basov for guiding me through the whole Ph. D. study. Dimitri’s dedication in the scientific research, insight and optimism towards scientific discoveries, patience in the weekly discussion is highly motivating. I’m very fortunate and grateful to have worked in his group.

My exploration in near-field technology would not be possible without the collaboration with my colleagues, Dr. Harry T. Stinson, and Rocco A. Vitalone. From Ted, I acquired first-hand knowledge in designing and constructing the near-field microscope. Working with Rocco, I accumulated valuable experience in operating high-field THz and nonlinear optics. These knowledge and experience enabled me to construct the instrument in San Diego with Ted, move and reconstruct it in New York, upgrade the THz measurement technique and successfully apply the THz near-field technique on various material systems.

I thank all the members in Dimitri’s group for their help and support. From my first day in the group, I always had extremely detailed discussion with Dr. Yiming Shao. These discussions are crucial in my scientific research. My collaboration with him is always fun and fruitful. He casted an important role in my research in TMD materials and nano-photocurrent technology. Dr. (now Prof) Alex Mcleod provided me with deeper theoretical understanding about the near-field technology, which triggered my innovation in upgrading the THz near-field measurement. I also had close collaborations with Dr. Lin Xiong, Yinan Dong, Dr. Shuai Zhang, Suheng Xu, Frank L. Ruta and Matthew Fu. I always enjoyed discussion with Dr. (now Prof) Siyuan Dai, Dr. Aaron Sternbach and Dr. Brian Kim. Without you, my Ph. D. journey would not be so rich and colorful.
I thank Professors Mengkun Liu, Michael Fogler and Andrew Millis for providing important comments in my work on THz near-field microscopy. My thanks also go to Dr. Zaiyao Fei, Elliott Runburg, Professors David Cobden and Xiaodong Xu for providing excellent WTe$_2$ devices.

Last but not the least, I must express my gratitude to my parents. Their understanding and patience supported me to finish my Ph. D. study oversees.
Chapter 1: Introduction

In this dissertation, we mainly introduce the novel method of measurement and application of world’s first cryogenic scattering-type Scanning Near-field Optical Microscope (s-SNOM) [1] for use at terahertz (THz) frequencies. Near-field technology overcomes the diffraction limit by utilizing the lightning-rod effect on the apex of a metal tip. The finite size (10 ~ 100 nm) of the apex couples to electromagnetic field near the sample surface at a finite momentum. The antenna-like metal lip out-couples the near-field to the far-field and the signal is collected by the same parabolic mirror which focuses the incoming illumination. The technique has been successfully applied in visible and IR frequencies. At THz frequencies, near-field technology breaks the diffraction limit by a factor of 1000 ~ 10000. However, due to the long wavelength and the semi-classic nature, collecting near-field signal at THz frequencies faces unique challenges. We innovated a detection method to overcome the difficulties. Utilizing the THz SNOM, we researched the electrodynamic properties of few-layer WTe₂ microcrystals and Cd₂Os₂O₇ single crystals.

1.1 Challenges

The major challenge of applying the near-field at THz frequencies stems from the semi-classic nature of THz wave. The metal tip used in THz SNOM are chosen to resonate with the illuminating electromagnetic wave as an antenna. Specifically, the length of the shaft \( L \) is a quarter of the wavelength \( \lambda \). when illuminated with a broadband light, resonance peaks and valleys can be observed in the frequency-domain. To extract the intrinsic material property, a common method is to normalize the signal collected on the sample to another well-studied material (e.g., Au, SiO₂, and sapphire). However, taking the tip-sample interaction into consideration, the resonance of the tip depends on the optical property of the sample as well [2].
As a result, the normalized spectrum contains artifact due to the different resonance features when the tip is scanning on varied materials. To extract the material property, accurate electromagnetic simulations with both the tip and different samples are required and hence, understanding material property via nano-spectroscopy is not straightforward.

Figure 1.1 Time-Domain and Frequency Domain near-field spectra of 10 nm thick BSCCO on SiO$_2$.

In addition to the antenna effect induced artifacts, the metal cantilever contributes to a second pulse in the time-domain spectrum and interference artifacts in the frequency-domain spectrum. Because the specific requirements of the metal tip used in THz SNOM, choices are extremely limited. The widely used tips are produced by etching a metal wire consist of pure platinum, tungsten, or PtIr alloy. The tip shaft ($\approx 80$ μm) and cantilever ($\approx 320$ μm) are shaped by curving the metal wire. After the tip couples to the THz illumination, the THz signal does not only directly scatter to the far-field, but also travels along the cantilever and is reflected by the other end [3]. As a result, the long metal cantilever generates an echo pulse arriving at the detector at $\approx 2.3$ ps later than the first pulse (Figure 1.1). The field strength of the second pulse is $\frac{1}{4} \sim \frac{1}{2}$ of the main pulse. The two pulses contribute to interference artifacts in the frequency-domain spectra corresponding to the resonance of the whole wire tip (Figure 1.1). Like the tip shaft antenna effect, such artifacts induce difficulties in spectra normalization between materials.
In addition to the artifacts and the difficulty in interpreting the spectrum, combining time-domain spectroscopy with real-space imaging (hyper-spectroscopy) is highly time-consuming and resource-hungry. In section 2.9.3, we present a 10 μm linecut with a resolution of 200 nm, which summarizes 2-hour data. The long data acquisition time is extremely inconvenient to identify real-space features. Besides, the THz near-field signal sensitively depends on the sample surface quality and tip apex morphology. At the base temperature, constant scanning is possible to induce contamination from other regions of the sample or icing. The tip apex is also likely to degrade due to accumulation of particles, icing and intense lightning-rod effect between the tip and sample. Therefore, large area hyper-spectroscopy is not pragmatic in most cases.

1.2 White-light THz imaging

Broadband THz near-field signal is detected via two major approaches. First, like far-field THz experiments, we can park the tip on the sample and acquire the time-domain spectrum. As stated in section 1.1, THz near-field spectroscopy with metal wire tips is contaminated with artifacts due to the antenna effect. Hyper-spectroscopy is not pragmatic for large area scanning. In the second method, instead of scanning the detection time-delay and park the tip, we scan the tip on the sample and park the detection time-delay at the main peak (Figure 1.1). We focus on the change of the \( \vec{E} \) field at different location on the sample surface. At the main peak, all frequency components are nearly in-phase, thus, the collected signal can be approximated by the integration of amplitude in the whole frequency domain. Hence, we call the signal white-light. White-light signal is efficient in acquiring real-space feature without knowing the information of the whole spectrum.

The deficiency of white-light signal resides in the fixed detection time-delay. The change of the \( \vec{E} \) field is contributed by two major degrees of freedom. Firstly, the overall amplitude of the
signal varies when scanning different location on the sample. Secondly, the amplitude is constant, while the pulse is delayed in time. The two degrees of freedom correspond to the change of the overall amplitude and the carrier-envelope phase (CEP). White-light imaging mixes the contribution from both degrees of freedom whose impact cannot be decomposed.

1.3 Phase-resolved white-light imaging

![Figure 1.2](image)

Figure 1.2: Resolving carrier-envelope phase (CEP) via zero-point signal. (a) Assuming the spectra of two materials are different by a non-zero CEP, the main-peak of the red curve is shifted to later time-delay relative to the black curve due to the CEP. We monitor the signal at the detection time-delay of the main peak ($R$) and the zero-point ($I$) of the black curve. (b), (c) The near-field signal collected at the main peak (R) and the zero-point (I). (d) The approximated CEP of $I/R$.

The straightforward way to upgrade the traditional white-light signal is to add an additional channel. Along with the main peak signal ($R$ in Figure 1.2), we monitor the signal at the zero-point ($I$ in Figure 1.2) near the main peak. When the overall amplitude varies and the CEP is constant, $R$ changes and $I$ is constant. When the signal acquires a positive CEP, both $R$ and $I$ change. The CEP can be approximated by $\phi \approx \tan^{-1} \lambda I/R$ and the white-light amplitude is approximated by $A \approx \sqrt{\lambda^2 I^2 + R^2}$. The factor $\lambda$ is a correction parameter according to the pulse
shape. In Chapter 2, we provide a more applicable method where we modulate the detection
time-delay and populate the signal $I$ in the first side-band of the demodulated near-field signal.

Phase-resolved WL imaging provides better insight into the material property. The phase-
channel reveals critical imaging proof of ultra-long wavelength low-quality factor surface
plasmon and potentially other propagating mode (Chapter 2). The complex-valued signal enables
determination of the optical constant (conductivity) of the investigated sample.
Chapter 2: Phase-resolved Terahertz imaging technique

2.1 Introduction

The scattering-type near-field microscopy (s-SNOM) is a powerful technique for exploring the nano-scale electrodynamics beyond the diffraction limit [4]. A conducting tip is used to enhance the electric field at the apex and scatters the incident light to the far-field, resulting in a spatial resolution that only depends on the size of the tip apex (typically 30~200 nm) instead of the wavelength (typically 1~500 um) [5, 6]. The technique has exhibited unique capabilities in studying nano and mesoscale phenomena, including polaritonic effects in low-dimensional systems [7, 8, 9, 10, 11, 12, 13], phase transitions in strongly correlated materials [14, 15, 16, 17, 1], electrodynamics of photonic crystals [12, 18, 19] and free carrier distribution in nano-devices [20, 21].

At infrared and visible frequencies, the s-SNOM technique employs two major kinds of detection strategies, pseudo-heterodyne detection for continuous-wave (CW) illumination and Fourier-transformed infrared spectroscopy for broadband pulses. Both strategies modulate the tip scattered signal by oscillating the tip vertically with respect to the sample surface. The near-field (NF) signal is subsequently demodulated at the integer harmonics of the tip oscillation frequency. For an imaging experiment with monochromatic lasers, pseudo-heterodyne interferometry is utilized to acquire both the amplitude and the phase of the artifact-free demodulated signal [22, 23, 24]. For spectroscopic studies using coherent broadband illumination, a Michelson interferometer is included in the light path to generate an interferogram, which is subsequently Fourier-transformed to determine the frequency-dependent NF contrast [25, 26]. Combining Fourier-transform infrared spectroscopy with real-space scanning produces hyper-spectral images that provide both spectrally and spatially dependent
information. At Terahertz (THz) frequencies, the recent integration of quantum cascade lasers (QCL) [27, 28], THz transceivers [29], gas lasers [30, 31], and backward-wave oscillators [32] provide possible options for CW imaging. However, the available frequencies are still much more limited compared to QCLs in the mid-IR.

For nanoscale implementation of the time-domain spectroscopy (TDS), high repetition rate broadband pulses generated with photoconductive antennas (PCA) are widely utilized [1, 13, 21, 33, 34, 35, 36]. Recently, high-power broadband THz pulses generated using nonlinear optics, such as optical rectification [37] and tilted-pulse-front schemes have also been applied in the THz NF microscopy [38]. The THz signal is detected via two major approaches, white-light (WL) imaging and TDS. The white-light imaging resolves the E field at the main peak $\hat{E}$ of the time-domain THz pulse, corresponding to a spectrally integrated NF contrast [1, 21, 13, 34, 33, 35]. Since the materials have a complex optical response, $\hat{E}$ is associated with both the amplitude and the phase of the scattered radiation. While WL imaging provides integrated nanoscale THz contrast, it is impossible to recover the full complex NF signal using this approach. The phase response, in particular, is typically inaccessible, limiting its spectral sensitivity. The TDS method [1, 33, 34, 39, 40, 41] yields the THz near-field amplitude and phase spectra via the Fourier transformation of the time-domain signal. However, TDS is not suitable for fast imaging especially since hyper-spectroscopy (a combination of WL imaging and TDS) demands an exceedingly long data acquisition time. To resolve those technical issues, we introduce here a novel phase-resolved detection method for pulsed illumination to rapidly access both the averaged amplitude and the phase information in broadband THz nano-imaging.

As a testbed for our new approach, we investigate mono- and few-layer tungsten ditelluride (WTe$_2$) microcrystals, which have attracted much attention due to the rich physical phenomena
observed in this van der Waals layered material. For example, when patterned into narrow ribbons, WTe₂ microcrystals show hydrodynamic current flow below 20 K [42]. When thinned down, the transport properties of WTe₂ reveal a crossover from 3D to a 2D electronic system. Spin-orbit coupling (SOC) gradually drives the conduction and valence band away from the Fermi energy, leading to reduced sizes of electron and hole pockets in momentum space [43]. Additionally, an anisotropic plasma frequency [44] gives rise to the in-plane hyperbolic plasmonic behavior [45]. Few-layer WTe₂ further shows quantum nonlinear Hall effect [46] [47], photocurrent [48], and photo-galvanic effects [49] due to the low symmetry of the material. Bilayer WTe₂ supports switchable out-of-plane ferroelectricity [50], whereas monolayer WTe₂ is argued to be a 2D topological insulator [51, 52] and excitonic insulator [53, 54].

2.2 Detection of the cavity-envelop phase of terahertz pulses

![Figure 2.1: Side-Band detection of THz NF signal via time-delay modulation. a Schematic diagram of the experimental configuration in analogy to ref. [38]. Additionally, the beam path of the gate pulses is modulated at frequency $M$. b Schematic time-domain THz signals of a substrate and a sample. For simplicity, we assume the materials share the same frequency dependence of the amplitude but include a finite CEP shift for the sample. Due to the CEP shift, the main peak of the sample and the substrate occurs at different gate delays. The shaded area indicates the range of the gate beam path modulation. The inset depicts the amplitude spectrum of the THz pulses.](image)
In a previous study [38], we overcame the diffraction limit and used the traditional WL imaging technique to study the nanoscale optical response of WTe$_2$. We concluded that the temperature-dependent NF signal of 2L -12L WTe$_2$ is dominated by the surface plasmon (SP). In this report, we apply the phase-resolved NF imaging technique to acquire well-defined maps of the amplitude and phase of the scattered light. The knowledge of the full complex-valued NF signal provides solid evidence for terahertz SP on WTe$_2$. The data reveals the negative temperature dependence of the wavelength $\lambda_P$ and the quality factor $Q$ of the SP. We also found an increase of the plasma frequency and a decrease of the scattering rate at low temperatures. The complex-valued NF signals also facilitate the extraction of the complex permittivity of WTe$_2$ by comparing real-space maps with patterns predicted by simulations.

The schematics of the proposed phase-resolved nano-THz experiments are displayed in Figure 2.1. In this experiment, THz radiation is generated through optical rectification of optical pulses at 1030 nm (17 W at 750 kHz repetition rate) with a tilted phase front onto a LiNbO$_3$ single crystal. The generated THz beam is filtered to be a Gaussian beam with a center frequency of 1 THz and a full width at the half maximum of 200~300 GHz. The detection is achieved through EO sampling of the tip-scattered THz pulse in ZnTe crystal gated by 20 fs pulses at 800 nm. The THz beam is focused onto an 80 $\mu$m long PtIr wire tip from Rocky Mountain Nanotechnology. The tip oscillates and modulates the scattered NF signal at $\Omega \sim 70$ kHz. The detected signal is demodulated at the integer multiples of the tip oscillation frequency. Instead of detection at a fixed time delay (as in conventional WL imaging), we periodically modulate the time delay of the gate pulses at a frequency $M \ll \Omega$. Consequently, the detected amplitudes occur at carrier-band frequencies ($n\Omega$) and side-band frequencies ($n\Omega \pm mM$). Here, the carrier-bands encode the in-phase part of the signal, and the first side-bands encode the out-of-phase part. By
combining signals from different channels, the method provides a reliable estimation of the CEP. In 2.9.1, we provide a detailed description of the method.

### 2.3 Terahertz near-field signal of representative materials

![Figure 2.2: THz near-field amplitude and phase for representative materials. The simulated NF amplitude and phase of 0.3 nm thin crystal with band insulator (a, b), Dirac materials at $\mu = 0$ (c, d), $\mu = 5 \text{ meV}$ (e, f) and $\mu = 25 \text{ meV}$ (g, h). The optical constants (conductivities) are listed in Table. 1. (i, j), Horizontal linecuts ($Y = 0$) of amplitude and phase are extracted for a range of chemical potential ($\mu = 0 \sim 37.5 \text{ meV}$). (k, l) NF signal map of an infinitely large metallic sheet sample with arbitrary 2-dimensional conductivity $\sigma_{2d} = \sigma \cdot d$ at 1 THz. The dots mark the results of Dirac materials simulated at different chemical potentials.

To demonstrate the sensitivity of the THz NF amplitude and phase on permittivity, we simulated signals at a representative frequency of 1 THz of a gapped insulator and Dirac semimetal at different doping levels (Figure 2.2). The parameters of the simulation are summarized in Table. 1. The sample is assumed to be a 0.3 nm thick $10 \mu m \times 10 \mu m$ square stacked on top of SiO$_2$/Si substrate. The simulation is based on the real-space NF modeling documented in Ref. [38]. Since
the simulation assumes $T = 0 \text{ K}$, the impact of thermal doping is not considered. In this simulation, we assume the Dirac semimetal has a relatively low scattering rate $\gamma = 10 \text{ cm}^{-1}$ to highlight the doping dependence of the NF signal. For the gapped insulator with $\varepsilon = 80$ (Figure 2.2a and b), the contrast in the amplitude is induced by the contrast of $\varepsilon_1$ ($\varepsilon_2 \sim 0$) between the sample and the substrate. The phase is identical to the substrate due to the lack of absorption. The neutral Dirac (Figure 2.2c and d) material shows a negligible contrast with respect to the substrate. Because of the absence of free charges, the optical response is solely contributed by the inter-band transition. However, the quantum conductance is not strong enough to generate observable near-field contrast in either amplitude or phase channels.

**Table 2.1:** The permittivity and conductivity of the simulated materials, the unscreened (screened) plasma wavelength $\lambda_p$ ($\lambda_p^*$) and $Q$-factor of the screened SPs in Fig. 1. The permittivity is calculated based on Graphene band structure at $T = 0 \text{ K}$ and $\gamma = 10 \text{ cm}^{-1}$.

<table>
<thead>
<tr>
<th>Band</th>
<th>Insulator</th>
<th>Dirac Semimetal</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\varepsilon$</td>
<td>$\mu = 0$</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>80</td>
<td>$1 + 3680i$</td>
</tr>
<tr>
<td>$\sigma_{1,2,d}(S)$</td>
<td>0</td>
<td>$1 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\lambda_p$ ($\lambda_p^*$), $Q$</td>
<td>$-$</td>
<td>$-$</td>
</tr>
</tbody>
</table>

For doped Dirac materials (Figure 2.2c to h), the surface electrodynamics is dominated by the SPs. At low chemical potential, the plasma frequency is low and comparable to the probing THz radiation. The weak oscillator strength gives rise to a low amplitude signal (Figure 2.2e). However, because the probing frequency is close to the SP resonance, a strong phase response is observed (Figure 2.2f). At higher chemical potential, the wavelength of the SP and the NF amplitude are increased (Figure 2.2g). The up-shift of the plasma resonance frequency reduces the overall phase response at the simulation frequency 1 THz (Figure 2.2h). Because of the long wavelength of the SP screened by the substrate and the low quality-factor, the clarity of the SP fringe pattern is suppressed. To study the chemical potential dependence (Figure 2.2i and j), we
extracted horizontal linecuts along the sample. With increasing chemical potential, the plasma fringes with growing wavelength on opposite edges gradually merge and weaken in both channels. As a result, the most significant feature of the SP at high chemical potential is the local enhancement in phase images near the sample edge. Considering the entire doping dependence (vertical axes of Figure 2.2j), the spatial modulation in scattering response is more apparent in the phase than that in the amplitude channel. In Figure 2.2k and l, we map out the near-field amplitude and phase for infinitely large sheet samples with arbitrary 2-dimensional conductivities $\sigma_{2d} = \sigma \cdot d$. By introducing the phase channel, we can estimate the conductivity or permittivity of the investigated sample based on Figure 2.2k and l.

2.4 Few layer WTe$_2$ microcrystal as a testbed

![Figure 2.3](image)

**Figure 2.3:** The phase-resolved THz imaging of a multi-terraced WTe$_2$ micro-crystal. (a, b) Temperature-dependent normalized amplitude ($S_2/S_1$) and phase images ($\phi_2 - \phi_1$) of WTe$_2$ as a function of temperature. The images are further normalized on the substrate. The boundaries of all terraces are demarcated with dashed lines in the image recorded at 33 K. Solid lines in the images taken at 295 K reveal the trajectory of the linecuts analyzed in panels (c) and (d). (c) The amplitude and (d) phase linecuts extracted from (a, b). The linecuts are shifted along the vertical axis for clarity. The modulations near the sample edges (marked with black arrows and highlighted with blue shading) extend into the 12L region and the
width of these peaks depends strongly on the temperature. (e, f) Temperature-dependent unnormalized amplitude ($S_2$) and phase ($\phi_2$) averaged over all pixels in each terrace.

We now demonstrate the utility of the phase-resolved WL imaging using multi-terraced semimetallic WTe$_2$ micro-crystals as a case study. The configuration of our homebuilt THz-SNOM and THz generation and detection are schematically displayed in Figure 2.1a [38]. In this experiment, we focus on the demodulation at the 2nd harmonic frequency of tip oscillation. The exfoliated micro-crystals of WTe$_2$ have a variety of terraces. The boundaries of all regions are demarcated, and the layer numbers are labeled in Figure 2.3a. The entire microcrystal is encapsulated between the top (6 nm) and bottom (20 nm) hexagonal boron nitride (hBN). The substrate of the structure is p-doped silicon with a 285 nm SiO$_2$ cover layer.

The NF amplitude and phase of WTe$_2$ are acquired from 33 K to 295 K (Figure 2.3a and b). In near-field imaging, dividing the higher-order signal by the lower-order signal is proven to generate more generic near-field information [55], thus the images and linecuts are presented as $S_2/S_1$ and $\phi_2 - \phi_1$. However, we observe that the ratio between signals of different orders depends strongly on the geometry of the near-field tip, which may change when scanning different images. As a result, tracking temperature dependence of the signal level is difficult. Therefore, we use $S_2$ normalized on the substrate in the same image to extract the averaged signal in Figure 2.3e and f. With decreasing temperature (Figure 2.3e), the amplitude recorded in 3L ~ 12L regions increases, consistent with our earlier nano-THz experiment using conventional data acquisition [38]. The 2L region exhibits a slightly enhanced amplitude at low temperatures. However, in contrast to the previous study where the signal on 2L is nearly temperature independent. The phase values for all regions are also higher at lower temperatures. As for the layer dependence, thicker WTe$_2$ regions show higher NF amplitude. The phase shows the opposite layer dependence.
The inconsistency between the 2L results here and the previous study [38] reveals the importance of acquiring the full complex-valued NF signal. Firstly, by incorporating the phase-resolved technique, we can separate the amplitude and phase degrees of freedom. Since 2L WTe$_2$ has a large phase compared to the substrate, the signal of 2L is delayed compared to that of the substrate. The WL time delay when scanning the 2L is not on the THz main peak. If we consider the main peak signal as the WL amplitude, the acquired signal on 2L will be significantly lower than the real WL amplitude due to the large phase contrast. Therefore, this phenomenon becomes more apparent at lower temperatures where the phase difference increases in magnitude.

As discussed in Ref. [38], the NF signal of few-layer WTe$_2$ is dominated by SP, which prompts the real-space scattering signal in both amplitude and phase channels. We extracted linecuts at identical locations in all images in Figure 2.3a and b and summarized the results in Figure 2.3c and d. The result exhibits strong spatial dependence in both channels. Enhanced $S_2$ (Figure 2.3c) in the middle of the 12L region is observed at high temperatures but not in the lowest temperature, consistent with the gradual shift of the maximal $S_2$ away from the linecut region. In the phase channel (Figure 2.3d), peaks (indicated by arrows) are observed at all temperatures and on both 12L/substrate and 3L/substrate edges. To clearly illustrate the temperature dependence of the extent of the edge peak, phase linecuts in Figure 2.3d are shifted vertically and the peaks are highlighted with blue shades. The peak becomes more prominent and extends deeper into the sample at lower temperatures. As discussed later, the strong and systematic temperature and thickness dependence of the edge peak in the phase profile is an important indicator for the underlying optical properties of the microcrystals.
2.5 Near-field Simulation

Figure 2.4: Real space modeling and tuning of SP patterns in WTe2. (a) The amplitude and (b) phase images. Simulations adequately reproduce key features of the $T = 295$ K experimental data in Fig. 3 using permittivity values listed in Table 2. The values are based on Ref. [38]. (c) and (d), the simulated amplitude and phase are calculated at $T = 33$ K. (e, f) The simulated amplitude and phase linecuts of 12L WTe2 at the location indicated in (a)–(d), identical to those extracted from experiments. (g, h) We mark the estimated conductivities of 3L ~ 12L WTe2 on the near-field amplitude and phase map generated by the lightning-rod model [56]. The dashed line highlights conductivities corresponding to $Q = 1$.

To understand the imaging results, we simulated the real-space SP patterns at 295 K (Figure 2.4a and b) and 33 K (Figure 2.4c and d). The simulation assumes the NF signal of a 2D material is proportional to the z-direction (along the tip) polarization of a dipole at a certain height near the sample surface. The concept of the method has been successfully applied to model previous NF measurements at both THz [38] and mid-IR [57] [58] [59] frequencies. The permittivity (conductivity) values of all regions in Figure 2.4a–d are summarized in Table 2.2. The permittivity values used in the simulation are based on the conclusion and model in Ref. [38] that the 2 L ~ 12 L WTe2 can be described by a semimetal with $\Delta = 10$~20 meV. The real-space simulation focuses on the SP pattern and the averaged signal is less important than that
produced by the lightning-rod model presented in Figure 2.4g and h. Nevertheless, the simulated signal shows the same layer number and temperature dependence as in the experiment.

Table 2.2: The permittivity of each region in the simulation in Fig. 4a and 4b. The permittivity value is estimated based on Ref. [38]. For 3L~12L, the wavelength is proportional to the layer number \( N \). We note that the difference in the NF signal of 3L~12L is solely caused by the difference of sample thickness.

<table>
<thead>
<tr>
<th>Layer number</th>
<th>Permittivity ( \varepsilon )</th>
<th>Conductivity ( \sigma_{1,2d} ) (( \mu S ))</th>
<th>( \lambda_p(\lambda_p^P), Q )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2L (295 K)</td>
<td>(-17 + 344i)</td>
<td>26.8</td>
<td>80.0 (27.6) nm, 0.05</td>
</tr>
<tr>
<td>2L (33 K)</td>
<td>(-454 + 454i)</td>
<td>35.6</td>
<td>2.00 (0.690) ( \mu )m, 1</td>
</tr>
<tr>
<td>3L~12L (295 K)</td>
<td>(-29 + 586i)</td>
<td>(22.8 \times N)</td>
<td>66.7 (23.0) nm ( \times N), 0.05</td>
</tr>
<tr>
<td>3L~12L (33 K)</td>
<td>(-757 + 757i)</td>
<td>(29.5 \times N)</td>
<td>1.67 (0.575) ( \mu )m ( \times N), 1</td>
</tr>
</tbody>
</table>

To highlight the key features in the modeling, we extracted linecuts in the simulation image at a similar location to those in the experimental images on 12 L. These linecuts are presented in Figure 2.4e and f. In the 295 K simulation result, we observe a narrow edge peak in the phase and unsaturated signal in the amplitude, consistent with the 295 K experiment result. In the result for 33 K, since the wavelength is comparable to the sample size, the SP fringe is still obscure. Consequently, the dramatic increase of wavelength doesn’t induce a change in the length scale of the real-space pattern proportional to the wavelength. Instead, the amplitude is boosted and flattened, and the edge peak in the phase extends deeper into the 12L region, which is consistent with the experiment result at 33 K.

Besides the real-space pattern, we mark the conductivity (permittivity) for 3L ~ 12L WTe\(_2\) in the NF signal map in Figure 2.4g and h together with the increasing of wavelength and quality factor. Because we assume 3L ~ 12L WTe\(_2\) shares the same optical constant, the 2d conductivities of different regions are along a diagonal line corresponding to a specific quality factor at each temperature. The maps clearly show that the dominating factor of increasing amplitude and phase on WTe\(_2\) from 295 K to 33 K is the dramatic increase in the quality factor.
The fast lengthening of the wavelength at 33 K boosts the $\sigma_{1,2d}$, consistent with the transport measurement [51].

Figure 2.5: Tuning of the THz plasma fringe pattern. In (a) and (b), we maintain the wavelength $\lambda_p = 10 \mu m$ and tune the Q-factor. The Q-factor dependence of the amplitude and phase linecuts are shown in false-color images. We highlight three curves with $Q = 0.4, 1, and 1.6$. The plasmon fringe is indicated by black arrows. In (c) and (d), we maintain the Q-factor $Q = 0.5$ and tune the wavelength $\lambda_p$. We highlight three curves with $\lambda_p = 8, 16, and 24 \mu m$. (e, f) The SP pattern depends critically on the phase-shift $\theta$ in the process of SP reflection from the sample edge. The $\theta$ dependence of SP pattern is plotted. In the inset, we illustrate the phase shift acquired by the reflected SP wave.

To exhibit the $\lambda_p$ and $Q$ dependence of the SP pattern, we tuned both parameters in a wide range in the simulation. By increasing the Q-factor from 0.2 to 2 (Figure 2.5a and b) and maintaining $\lambda_p = 10 \mu m$, the amplitude fringe (indicated by black arrows in Figure 2.5a) becomes visible. In the phase channel (Figure 2.5b), the overall value increases with increasing Q-factor and the phase fringe (indicated by black arrows) becomes visible as well. In Figure 2.5c and d, we demonstrate the $\lambda_p$ dependence of the SP pattern by fixing the Q-factor to 0.5. The wavelength dependence of the overall value is analogous to the result shown in Figure 2.2i and j.
The longer $\lambda_p$ diminishes the visibility of the plasmon fringe in both the amplitude and phase channels due to the limited sample size.

### 2.6 Phase-shift of Plasmon reflection at the sample boundary

The SP pattern is determined by the phase-shift $\theta$ acquired by the reflected wave from the sample edge (Inset of Figure 2.5e). In Figure 2.5e and f, we demonstrate modeling with $\theta = 0 \sim \pi$. To emphasize the features near the edge, the model calculates SP with screened wavelength $\lambda_p^* = 8 \, \mu m$ and $Q = 0.25$ at 1 THz near the left end of a one-dimensional ribbon ($L = 10 \, \mu m$). Because of the screening of the p-doped Si, the dispersion of the SP is linear, therefore the wave packet is not distorted in the propagation. In the amplitude channel (Figure 2.5e), the signal on the edge gradually decreases with increasing $\theta$. When $\theta = \pi$, the wave reflection is analogous to mechanical waves propagating on a string with a fixed end, leading to a low response near the edge. In the phase channel (Figure 2.5f), the peak near the edge becomes a valley at a high phase shift. Compared to higher frequencies, THz range SPs are more easily screened by the metallic gating material beneath the sample due to their long wavelengths. Therefore, the screened plasmon acquires a reflection phase-shift $\theta = 0$ [60]. In the current sample configuration, the Si substrate is highly p-doped and shows moderate metallicity ($\rho = 0.01 \sim 0.02 \, \Omega \cdot cm$). Accordingly, the reflection phase-shift of SP is tuned to 0 in the modeling in Figure 2.4 and Figure 2.5a ~ d, which matches reasonably well with the experimental data.

### 2.7 Delay time dependent surface plasmon pattern

Because the illumination in the experiment is pulsed instead of CW, we must consider the launching and detection of the SP pattern in the time domain. As shown in the inset of Figure 2.5e, the signal is the interference between the tip launched plasmon and the edge reflected
plasmon. The edge reflected plasmon is launched by the tip at an earlier time point. For WTe$_2$, the speed of the propagating SP is estimated to be on the order of $v \sim 10 \mu$m/ps [38]. Therefore, the reflected wave takes a non-negligible time (~picoseconds) to travel back to the tip. In the near-CW scenario, SPs are launched by the tip at a constant power and the detected signal is also power instead of E field. Therefore, detection at any point in time is identical. In this experiment, we detect near the main peak $E$ of the pulse. The reflected wave is launched at an earlier time $t$ when the THz excitation field is relatively lower. This phenomenon induces an artificial damping factor to the observed SP fringe. We therefore predict SP fringes patterns can be more easily observed at a larger time delay than the main peak.

2.8 Conclusion

In conclusion, we devised, implemented, and validated a side-band detection method capable of extracting the averaged amplitude and phase of broadband THz signals on few-layer WTe$_2$. With a small time-delay oscillation, the in-phase and out-of-phase frequency components of the signal encoded in alternating CB and SBs can be easily detected through lock-in amplification. Applying this method for nano-THz investigation of a multi-terraced semimetallic WTe$_2$ microcrystal, we demonstrated the NF amplitude and phase pattern of SP confined in a relatively small sample geometry. We numerically demonstrated the SP pattern dependence on a series of parameters (wavelength, quality factor, sample thickness, and reflection phase-shift). By comparing these simulations with the experiment, we extracted the permittivity of few-layer WTe$_2$. The advantage of the side-band detection method over the traditional white-light imaging is that the amplitude and the phase degrees of freedom can be independently extracted. The full complex-valued signal provided by the method is the key to study the damped plasmonic and polaritonic behaviors at THz
frequencies, which applies to metallic as well as dielectric systems. Compared to hyper-
spectral imaging, this represents a practical method to efficiently achieve averaged phase
information in the challenging THz frequency range with less beam time and resource
requirements.

2.9 Supplementary

2.9.1 Side-band detection of Gaussian beam

Figure 2.6: Side-Band detection of THz near-field signal employing the time-delay
modulation. a The schematic diagram of the experiment configuration. b Time-domain
spectra of two gaussian pulses with $\phi_0 = 0, \beta = 0, \alpha = 0$ (black, reference) and $\phi_0 = \pi/3, \beta = 0.1, \alpha = 0.01$ (red, sample). Two curves share the same $\omega_0 = 2\pi \cdot 1 \ THz$, $\Delta \omega = 2\pi \cdot 0.35 \ THz$. The grey area indicates the range of the time-delay modulation ($\gamma = 0.1 \ ps$). c Frequency-
domain spectrum of signal produced by the time-delay modulation shown in b. The carrier-
band value of $\phi_0 = 0$ is rescaled by a factor of 0.3.

we assume the signal pulse $S(t)$ can be described by a gaussian wave packet, with a
center frequency $\omega_0$ ($2\pi \cdot 1 \ THz$) and the full width half maximum FWHM = $2\pi \cdot 0.82 \ THz$
($\Delta \omega = 2\pi \cdot 0.35 \ THz$). For THz signal whose phase $\phi$ does not have strong frequency-
dependence, we assume $\phi \approx \phi_0 - \beta \frac{\omega_0 (\omega - \omega_0)}{\Delta \omega^2} + \alpha \frac{(\omega - \omega_0)^2}{2\Delta \omega^2}$ ($|\alpha| \ll 1$), where $\alpha$ and $\beta$ are unitless,
\[ S(t) \propto E(t) = \Re \{ \sqrt{2\pi \Delta \omega^2} e^{-\frac{\omega_0^2 \beta^2}{2 \Delta \omega^2}} e^{-i(\omega_0 t - \phi_0 + \frac{\omega_0^2 \beta^2}{2 \Delta \omega^2})} e^{-\frac{1}{2} \Delta \omega^2 (t - t^*)^2} \} \]

where \( t^* = \frac{\beta \omega_0}{\Delta \omega^2} = \phi'(\omega_0) \). In the following discussion, the term \( \frac{\omega_0^2 \beta^2}{2 \Delta \omega^2} \) in the phase is neglected. In practical experiments, the investigated surface consists of a reference material and a sample material. In the current example, we assume the phases of the reference and the sample spectrum are \((\phi_0, \beta, \alpha) = (0,0,0)\) and \((\phi_0, \beta, \alpha) = (\frac{\pi}{3}, 0.1, 0.01)\). The non-zero carrier-envelop phase \( \phi_0 \) and parameter \( \beta \) shifts the main peak of the signal pulse away from the original detection time point at \( t = 0 \) ps (Figure 2.6b). As a result, a pure sample-induced phase shift of the THz pulse causes observable contrast in the WL signal, which is difficult to differentiate from the contrast due to the change of the overall amplitude. By incorporating an oscillation of the time-delay at frequency \( M (M \ll \Omega) \),

\[ t = t_0 + \tau \cos Mt, \]

where \( t_0 = 0 \) ps, \( \tau = 0.1 \) ps, part of the signal is distributed from the carrier-bands (CB) at frequencies \( n\Omega (n \in Z^+) \) to side-bands (SB) \( n\Omega \pm mM (m \in Z^+) \) (Figure 2.6c). The range of the oscillation is highlighted with a grey box in Figure 2.6b. When \( t_0 = 0 \) ps (Figure 2.6c), the values of \( n\Omega + M \) and \( n\Omega - M \) side-bands are zero for the reference spectrum with \( \phi_0 = 0 \) and finite value for the sample spectrum with \( \phi_0 = \pi/3 \).
Figure 2.7: Phase calculation produced by the time-delay modulation. Values of a, the carrier-band and b, the 1st side-band and the first four contributing terms are compared side-by-side. Terms with small values are rescaled by 10 or 100 times. The rescale factor is displayed on the bar plot. c The \( t_0 \)-dependent spectrum of the carrier-band and the rescaled 1st side-band of the reference spectrum. The dashed line at \( t = 0 \) ps indicates where the values are collected to calculate the phase. d The same with c for sample spectrum. e \( \phi_{calc} \) is calculated at different delay \( t_0 \). When \( t_0 \) is near the interval \([0, 1.3] \) ps, the calculation result is close to \( \frac{\pi}{3} \). f The accuracy and the linearity of the model phase \( \phi_{calc} \) is calculated at different \( \gamma \) value. The linearity is represented by the residue of the linear regression to \( \phi_{calc}(\phi_0) \) with \( \phi_0 \in [0, 2\pi] \). The vertical dash line marks the limit of \( \gamma \Delta \omega / 2\sqrt{2} \) value below which the proposed method is valid. The horizontal dash lines marks zero level.

With Eq. 2.1 and Eq. 2.2, the time-dependent signal can be expanded into

\[
S(t) \propto \Re \left\{ e^{-i \Delta \omega t^2 (t^2 + 2(t_0 - t)^2)} e^{-i (\omega_0 t_0 - \phi_0 + \omega_0^2 t^2 + \omega_0^2 (t - t_0)^2)} \left( \sum_{m=1}^{3} (-1)^{m_1 + m_2 + m_3} J_{m_1} I_{m_2} I_{m_3} e^{i (m_1 + 2m_2 + m_3) M t} \right) \right\},
\]

Eq. 2.3

where \( J_{m_1}, I_{m_2} \) and \( I_{m_3} \) are Bessel function and modified Bessel functions of the first kind with arguments \( \omega_0 t, \Delta \omega^2 t^2 / 4 \) and \( \Delta \omega^2 (t_0 - t^*) \) respectively. For a certain side-band \( n \Omega + mM \), all terms with \( m_1 + 2m_2 + m_3 = m \) in the sum of Eq. 2.3 contribute to the detected signal. The
detailed frequency-domain analysis for \( \phi = \pi/3 \) spectra is shown in Figure 2.7a and b. The values of the CB \( S_{n\Omega} \), the 1st SB \( S_{n\Omega+M} \) and their first four contributing terms are compared side-by-side. Here, \( \Omega \) is the tip tapping frequency. When the conditions

\[
\begin{align*}
I_{m_2} & \sim \Delta \omega^2 \tau^2 / 8 \ll 1 \\
I_{m_3} & \sim \frac{\Delta \omega^2 \tau}{2} |t_0 - t^*| \ll 1
\end{align*}
\]

are satisfied, the contribution from terms with non-zero \( m_2 \) and \( m_3 \) are negligible. Consequently, the CB and SBs encodes the in-phase and the out-of-phase signal alternatively. Eq. 2.3 approximately reduces to

\[
\begin{bmatrix} S_{n\Omega} \\ S_{n\Omega \pm M} \end{bmatrix} \propto \begin{bmatrix} I_0 I_0 & 2I_1 I_0 I_{-1} \\ -I_0 I_1 & I_1 I_0 I_0 \end{bmatrix} \begin{bmatrix} \cos \theta \\ \sin \theta \end{bmatrix} \approx \begin{bmatrix} I_0 I_0 & 0 \\ 0 & I_1 I_0 I_0 \end{bmatrix} \begin{bmatrix} \cos \theta \\ \sin \theta \end{bmatrix}
\]

Eq. 2.5

where \( \theta = -\omega_0 t_0 + \phi_0 \). According to Eq. 2.5, the in-phase and out-of-phase signal can be independently extracted from the CB and the \( \pm 1 \) SBs. To correctly calculate the phase, we introduce the parameter \( \lambda = \frac{I_0(\omega_0 \tau)}{I_1(\omega_0 \tau)} \) to cancel the pre-factor. In Figure 2.7c and d, \( t_0 \)-dependent spectra of CB and rescaled 1st SB of the reference and sample are displayed. The calculated amplitude and phase can be expressed as

\[
\begin{align*}
S_n^{\text{calc}} &= \left( S_{n\Omega}^2 + \left( \lambda \frac{S_{n\Omega+M} + S_{n\Omega-M}}{2} \right)^2 \right)^{0.5} \\
\phi_n^{\text{calc}}(\phi_0) &= -\tan^{-1} \lambda \frac{S_{n\Omega+M} + S_{n\Omega-M}}{2S_{n\Omega}}
\end{align*}
\]

Eq. 2.6

Now, we discuss the accuracy and the linearity of \( \phi^{\text{calc}}(\phi_0) \). In Figure 2.7d, \( \phi^{\text{calc}} \) is calculated by applying the prescribed protocol at varying the time-delay \( t_0 \). In the current example, Eq. 2.4 requires \( \tau \ll \frac{2\sqrt{2}}{\Delta \omega} = 1.29 \) ps and \( |t_0 - t^*| \ll \frac{2}{\Delta \omega^2 \tau} = 4.14 \) ps. In our simulation of the protocol we use, \( t^*_R = 0.13 \) ps, \( t^*_S = 0 \) ps and \( \tau = 0.1 \). Therefore, we confirm the calculation is accurate for \( t_0 \) near the interval \([0,0.13] \) ps (Figure 2.7e). In practical nano-THz
experiments, to maximize the amplitude of the measured fields, $t_0$ is usually chosen to be the
time-delay of the WL peak of the reference spectrum. Due to the factor of $e^{-\frac{1}{2} \Delta \omega^2 (t-t^*)^2}$ in Eq.
2.1, the white light peak is located at $t_0 \approx t_R^* + \frac{\phi_0 - \omega_0 t_R^*}{\omega_0}$. Here, $\phi_0 - \omega_0 t_R^*$ is wrapped in the
interval $[-\frac{\pi}{2}, \frac{\pi}{2}]$. When $\omega_0 = 2\pi \cdot 1$ THz, $t_0 \in t_R^* + \left(-\frac{1}{4}, \frac{1}{4}\right)$ ps. Hence, Eq. 2.4 becomes,

$$\begin{align*}
\tau &\ll \frac{2\sqrt{2}}{\Delta \omega} |t^*_S - t^*_R| \\
|t^*_S - t^*_R| &\ll \frac{2}{\Delta \omega^2 \tau}
\end{align*}$$

Eq. 2.7

Besides the accuracy, the linearity of the mapping of $\phi_0 \rightarrow \phi_{\text{calc}}$ is an important indicator of the
validity of the method. The source of the non-linearity arises from the contribution of terms with
non-zero $m_2$ and $m_3$ in Eq. 2.3. The linearity is characterized by the residual of the linear-
regression on $\phi_{\text{calc}}(\phi_0)$. In Figure 2.7e, the $\tau$ dependent accuracy and the linearity of the
mapping $\phi_{\text{calc}}(\phi_0)$ share similar behavior. With our residual analysis, we find the side-band
detection method is sufficiently valid for $\frac{\tau \Delta \omega}{2\sqrt{2}} \leq 0.25$ (viz. $\tau \leq 0.32$ ps). By including more
terms in Eq. 2.5, it is possible to extend the validity of our protocol to higher values of $\tau$, which
increases the value of SBs.

2.9.2 Real-space THz near-field imaging of graphene
Figure 2.8: THz near-field amplitude and phase for graphene. The result is presented in same order and format with Figure 2.2 in the main text. Here, we increased the scattering rate from $\gamma = 10 \text{ cm}^{-1}$ to $30 \text{ cm}^{-1}$. With increased scattering rate, even if the plasmon pattern is smeared, the trend of the signal is identical to the result with lower scattering rate.

At THz frequencies, the scattering rate of graphene is higher than 1 THz at doping level below 50 meV (>77 K) [61]. Higher scattering rate smear the pattern of surface plasmon. In Figure 2.8, we recalculate the result presented in Figure 2.2 in the main text with increased scattering rate $\gamma = 30 \text{ cm}^{-1}$. The results show similar feature to those with lower scattering rate in Figure 2.2. The overall amplitude increases monotonically, and the phase is roughly maximized at a frequency slightly higher than the probe frequency (1 THz). Due to the increase of the scattering rate, the quality factor Q of the surface plasmon is reduced to 1/3 of the original value. As a result, the real-space plasmon pattern presented in Figure 2.8c to h is damped in a shorter distance to the sample edge. For reference, the plasmon wavelength and the Q-factor of different scattering rate is listed and compared in Table 2.3.

<table>
<thead>
<tr>
<th>$\gamma$</th>
<th>Dirac Semimetal</th>
<th>$\mu = 5 \text{ meV}$</th>
<th>$\mu = 25 \text{ meV}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10 \text{ cm}^{-1}$</td>
<td>$\epsilon (1 \text{ THz})$</td>
<td>$-4200 + 1930i$</td>
<td>$-25800 + 7950i$</td>
</tr>
<tr>
<td>$\lambda_P (\lambda^*_P), Q$</td>
<td>3.96 (1.90) μm, 2.18</td>
<td>24.3 (11.7) μm, 3.25</td>
<td></td>
</tr>
<tr>
<td>$30 \text{ cm}^{-1}$</td>
<td>$\epsilon (1 \text{ THz})$</td>
<td>$-2270 + 3810i$</td>
<td>$-15450 + 14400i$</td>
</tr>
<tr>
<td>$\lambda_P (\lambda^*_P), Q$</td>
<td>2.14 (1.02) μm, 0.60</td>
<td>14.6 (7.00) μm, 1.07</td>
<td></td>
</tr>
</tbody>
</table>
Figure 2.9: Thickness dependence of THz near-field signal. We assume the material has a Dirac band structure with $E_F = 12.5 \text{ meV}$ and $\gamma = 10 \text{ cm}^{-1}$. In the model, we only change the thickness of the sample.

In Figure 2.9, we maintain the dielectric property of the model material and we tune the thickness of the sample. We expect to two major impacts on the near-field signal. Firstly, the wavelength of the surface plasmon is changed due to its straightforward proportionality with the sample thickness. Secondly, the bulk near-field response of the material changes with the thickness. The first part is analyzed in Figure 2.5 of the main text. The second part is summarized in Figure 2.9. With increasing thickness, the near-field amplitude of the thin metal increases and the phase decreases.
2.9.3 Spectrum and hyper-spectroscopy

We acquired time-domain spectra across the 12L WTe₂ and performed Fourier-transformation. In Figure 2.10, we plot the linecuts at frequencies 0.5 THz to 1.1 THz at 295 K and 33 K. In the amplitude at room temperature (Figure 2.10a), the signal is higher in the interior of 12 L. The spatial dependence is nearly absent in the lower temperature result (Figure 2.10b). The frequency dependence of the amplitude is small. In the phase channel (Figure 2.10c and d), the edge peak is present at both temperatures. The low temperature peaks are wider and more prominent than high temperature ones, in agreement with the WL imaging. More interestingly, we observed a strong negative dependence of the width of edge peak with the frequency at low temperature. According to Figure 2.5d in the main text, the surface plasmon (SP) wavelength is longer at lower frequencies compared to higher frequencies.
Chapter 3: Terahertz imaging and nano-spectroscopy of layer-dependent properties of few-layer WTe$_2$ microcrystals

3.1 Introduction

The physical properties of the enigmatic material tungsten ditelluride (WTe$_2$) depend critically on the number of layers. Bulk WTe$_2$ is postulated to be a type-II Weyl semimetal [62] [63] with Fermi-arc surface states. Monolayer WTe$_2$ has been predicted and experimentally confirmed to be a quantum spin hall insulator [51] [64] [65] [66] [52] [67] and exhibits gate-induced superconductivity [68] [69]. Bilayer WTe$_2$ has broken inversion symmetry and is known to be ferroelectric [50], yet experiments produce ambiguous results on whether its electronic structure is semi-metallic or semiconducting. Transport measurements support the semiconductor picture with a narrow electronic bandgap (<10 meV) [51]. Angle resolved photoemission spectroscopy (ARPES), however, revealed that bilayers could also be weakly semimetallic with a small negative gap [70]. A combination of inverted bands, strong spin-orbit coupling and low crystal symmetry also makes few-layer WTe$_2$ an ideal system for studying topological effects such as the nonlinear anomalous Hall effect [71] [47] [46] and various unusual photogalvanic effects [49] [48] [72]. The goal of the present study is to explore the evolution of the low-energy electrodynamics of WTe$_2$ from monolayer to few-layer variants (Figure 3.1a). We conclude that trilayer and thicker specimens are metallic and host surface plasmon polaritons (SPP) [73] [74] that dominate the response in the terahertz (THz) range. The metallic response is reduced in bilayer areas and disappears in monolayer regions.

Bulk WTe$_2$ exhibits high electronic mobility and its intraband (Drude) optical response is entirely contained in the THz region [44] [45]. Despite tremendous interest, the THz response of monolayer and few-layer samples remains unexplored. THz experiments on few-layer WTe$_2$
specimens are challenging because of the minuscule size of available samples typically under 10x10 µm². The wavelength of THz waves is of the order of ~300 µm and conventional diffraction-limited methods are inadequate for interrogating the THz response of WTe₂ microcrystals. In order to overcome the diffraction limit in THz, we utilize a scattering-type THz scanning near-field optical microscope (THz-SNOM) [1] [75] [34] [13] [76]. This technique is a hybrid of an atomic force microscope (AFM) with a pulsed THz source. AFM-based THz nanoscopy offers a robust experimental approach to investigate materials with sub-diffractional spatial resolution down to $\lambda/2000$ where $\lambda$ is the wavelength of the probe beam. THz-SNOMs are being successfully applied to an expanding list of materials and interesting problems. For example, THz-SNOM methods have provided insights into nano-scale studies of electronic phase separation in the vicinity of the insulator-to-metal transition in VO₂ [1], the plasmonic response of graphene [13] [77] [78], free carrier distributions in nanodevices [20] [21], and phonon resonances in multiferroic materials [79].

Here we report on near-field nano-optical experiments in THz range for WTe₂ conducted at cryogenic temperature. The nano-THz measurements reveal that trilayers of WTe₂ show metallic behavior and a plasmonic response consistent with the properties of bulk crystal, whereas bilayer samples exhibit weak semimetallic behavior.

3.2 Wight-light imaging of WTe₂ microcrystals

We investigated multi-terraced microcrystals of WTe₂ using a home-built apparatus enabling nano-THz experiments at cryogenic temperature [1]. The THz beam is focused onto an AFM tip with an 80 µm long shaft made of PtIr wire. The tip apex locally confines and enhances the THz electric field. The tip shaft functions as an antenna [80] and out-couples the near-field radiation into far-field radiation reaching the photoconductive antenna (PCA) detector. The
tapping of the tip modulates the near-field signal at ~70 kHz. We demodulated the amplitude of the tip-scattered electric field at the first ($S_1$) and the second ($S_2$) harmonics of the tip tapping frequency to suppress the undesired far-field background [1] [24].

**Figure 3.1:** Schematic of nano-THz experiments on multi-terraced crystals of WTe2. (a) Metallic AFM tip locally enhances the electric field and enables THz coupling to materials at length scales much smaller than the THz wavelength. The size of the focused THz beam in the schematic is much smaller than the real focus. (b) Optical microscope image of the WTe2 sample. Multi-terraced microcrystals of WTe2 are encapsulated on top and bottom with hexagonal boron nitride (hBN) and reside on a SiO2/Si substrate. Optical inspection reveals 1L, 2L and 3L regions. The red frame indicates the field of view used for temperature dependent study in Fig. 2. We demarcate the boundaries of terraces labeled by layer number (1L, 2L, 3L) with white dashed lines. (c) THz near-field signal $S_1$ at room temperature, showing much higher THz signal in the 3L region compared to 1L and 2L.

The exfoliated micro-crystals of WTe2 are encapsulated between 6 nm of hexagonal boron nitride (hBN) on top and 20 nm hBN on the bottom (Fig.1a). The exfoliated structure is assembled on top of a SiO2/Si wafer. This sample hosts terraces of mono-, bi-, and trilayer WTe2 within a 25×25 $\mu$m² area. These terraces are evident in both the optical inspection image (Fig.1b) and in the nano-THz scan displaying the contrast in the scattering amplitude of the THz signal (Fig. 1c). The topographic contrast of AFM scans has only limited utility in visualizing the terraces because this contrast is suppressed by the top encapsulating layer (see 3.7.1). We obtained the network of dashed lines in Fig. 1b and c using a combination of optical contrast and
nano-THz contrast. We remark that the top layer hBN is thin enough that the evanescent field from the sample is still detectable with the help of the AFM antenna tailored for the THz range. In nano-THz experiments, the near-field scattering amplitude is an observable carrying information on spatially localized electromagnetic response [24] [81] [82] [83]. The measured signal is denoted by $S_{1,2} \propto |\tilde{E}^{\text{NF}}|$ where $\tilde{E}^{\text{NF}}$ is the THz near-field electric field. We analyzed the so-called approach curves: the variation of the $S_{1,2}$ signal as a function of separation between the tip and the sample (see 3.7.2). This analysis confirmed that over 90% of $S_2$ originates from the near-field tip-sample interaction within 150nm above the sample surface [84]. Demodulation of the THz signal at higher harmonics is not practical in view of the rapidly diminishing signal-to-noise ratio already at the third harmonic. The far-field contribution is enhanced at higher optical frequencies outside of the THz range [24]. For that reason, nano-optical experiments conducted in the mid-IR and visible ranges typically require demodulation at the third, fourth or even fifth harmonics [24]. In our nano-THz experiments, the tip radius is $R = 150\sim200$ nm as determined by scanning electron microscopy. The tapping amplitude is $\sim150$ nm. The tip radius and the tapping amplitude govern the center momentum ($0.1/R\sim1/R$) for photon scattering by the tip [2] [85] and the achievable spatial resolution [84].

Here we report nano-THz imaging data collected in frequency-integrated mode at every pixel. The frequency range of the THz radiation in our experiments spans between 0.2 THz and 2.5 THz. Due to the antenna resonance effect of the tip, the near-field signal intensity is peaked at $\sim0.6$ THz [1] [86]. Our nano-THz apparatus is designed to produce hyperspectral images with frequency resolved information at every pixel by Fourier transforming the time-domain spectra [39]. However, energy-integrated or “white-light” (WL) THz imaging has an important advantage of significantly increasing the signal-to-noise required to produce high fidelity images.
of weakly absorbing few-layer WTe$_2$ samples (see figures). We accompany nano-THz data with images in the infrared range where we employ a monochromatic light source (see 3.7.5).

In Figure 3.2, we show the complete set of temperature dependent THz nano-imaging data. We plot the scattering amplitude signals $S_1$ and $S_2$ normalized by those of the SiO$_2$/Si
substrate $S_{\text{sub}}$, $S_1/S_{\text{sub}}^1$ and $S_2/S_{\text{sub}}^2$. The $S_1$ data have a roughly 2 times higher signal to noise ratio (SNR) than $S_2$. Both $S_1$ and $S_2$ images display the same salient features. Since $S_2$ has less contribution from far-field background, we rely on $S_2$ to quantify the temperature dependence of the near-field response in the analysis that follows. We confirmed that the near-field signal due to the SiO$_2$/Si substrate shows negligible temperature dependence. We therefore can use the signal produced by the bare substrate as a reference in our normalization procedure. In all THz images, we resolve a feature due to a $\sim$200nm wide topographic linear defect marked in the panel obtained at 100 K. This latter topographic feature confirms that the spatial resolution of our THz near-field imaging is well below $\sim$200 nm at all temperatures.

To analyze the contrast between terraces with different numbers of WTe$_2$ layers, horizontal line-cuts from the $S_1$ images are displayed in Figure 3.2d. The location of the line-cut is indicated with a dashed arrow in the 200 K image of Figure 3.2a. The line-cut shows evident plateaus corresponding to terraces with different numbers of layers. Regions with a higher number of layers exhibit higher near-field signal. The signal in the trilayer region increased substantially at lower temperature, typical of metallic responses. Monolayers (Figure 3.2c and see 3.7.3) are marginally distinguishable from the substrate, demonstrating a clear insulating response. Interestingly, the bilayer region produced an intermediate amount of signal. While the overall near-field signal is 10$\%$~15$\%$ higher than the insulating monolayer, the absence of any temperature dependence restricts the magnitude and sign of the bandgap, as we will discuss later.

In addition to $S_1$, the $S_2$ signal was analyzed in small areas at the center of three different regions. These areas are indicated as white dashed rectangles in the image in Figure 3.2b. The temperature dependence of nano-THz contrast extracted from this analysis is plotted in Figure
3.2e. The signal in the trilayer area increases by more than 40% between ambient and 44 K, whereas in bilayer and monolayer regions, the increase of signal at low temperature is absent.

It is instructive to compare the temperature dependence of the THz near-field contrast summarized in Figure 3.2d with DC transport data [51]. The DC conductivity of trilayer WTe₂ is metallic at all temperatures in agreement with the nano-THz trend we report in Figure 3.2d. For bilayer WTe₂, DC transport data indicates a semiconducting behavior with a narrow gap in the meV range [51]. Specifically, the DC conductivity drops significantly below 100 K [51]. In many conducting materials, the real part of the optical conductivity in the THz range matches the DC value. However, if a material has a THz-range gap, this will not be the case. Indeed, the temperature independent nano-THz response of the bilayer terraces contrasts with the drop of the DC conductivity in undoped bilayer WTe₂ at low temperature. We note that hBN encapsulated WTe₂ is normally found to be almost undoped and therefore extrinsic doping of this sample is unlikely [51].

Nano-THz imaging data presented in the form of two-dimensional maps in Figure 3.2a and b or line-cuts in Figure 3.2d reveal a significant spatial dependence of the scattering signal. This effect is manifested as a gradual change in both the $S_1$ and $S_2$ signal within a 2-3 µm vicinity of the boundaries of trilayer WTe₂ and across bilayer regions. We remark that the width of these transitional regions is significantly larger than the spatial resolution of our near-field imaging apparatus (~200nm), as well as the width of the physical boundary observed in Figure 3.1. Comparing the line-cut curves acquired at different temperatures in Figure 3.2e, the location and the width of the transitional region has no noticeable dependence on temperature. With the help of real-space near-field modeling of SPPs on the confined structure presented in the latter
part of the paper, we show that the gradual spatial variation of the signal arises from THz SPPs with long wavelength (6~20 µm on trilayer).

We now discuss our data in the context of recent observation of edge states in WTe$_2$ [64][66]. In Figure 3.2c, we further zoom in on the monolayer region at the lowest temperature, 44 K. If edge states produced contrast in the THz range, we would see signal near the boundary between the monolayer WTe$_2$ and hBN/SiO$_2$/Si substrate and possibly also at the boundaries between monolayer and bilayer terraces. Indeed, such signals near the boundary are seen in the GHz regime [64]. However, we observe no significant signal at the boundaries of the monolayer. This is likely due to that the conductance of the topological edge state is too low to induce observable contrast in near-field imaging. In addition, the contrast of the edge state could be suppressed if the width of the state is narrower than the resolution.

3.3 Lightning-rod modeling of nano-THz response

Figure 3.3: Electrodynamics and plasmonic response of 3L WTe2. Imaginary part of the momentum dependent p-polarized reflection coefficient $r_p(\omega, q)$ at (a) 44 K and (b) 290 K, based on bulk dielectric function data [44]. The red points located at $q = 3.3e3 \text{ cm}^{-1}$ ($\lambda = 19 \mu m$) and $q = 1.0e4 \text{ cm}^{-1}$ ($\lambda = 6.3 \mu m$) indicate the momentum of SPP at 0.6THz. Free space (FS) and SiO$_2$ (S) light lines are indicated with dashed lines, respectively. SPP dispersions are clearly observed, and the dispersion broadens with increasing temperature. (c) Spectra of the near-field scattering amplitude modeled following Ref. [56] at different temperatures based on calculated $r_p(\omega, q)$. The shaded area indicates the frequency spectrum of our THz source.
To understand the THz near-field contrast of WTe$_2$ microcrystals, we carried out modeling of the response associated with the trilayer region. We assumed that trilayer WTe$_2$ has the same relative permittivity as bulk WTe$_2$ [44]. This simple assumption allows us to determine the origin of the temperature dependence of the THz signal in the trilayer region. We will discuss the transition from trilayer to bulk WTe$_2$ in terms of THz near-field response later in the text. In our analysis we consider encapsulating hBN layers as well as the response of the SiO$_2$/Si substrate within the framework of the lightning-rod model (LRM), a multilayer model of the near-field response described in Ref. [56]. An implicit assumption of the model in Ref. [56] is that all layers in multi-layered structures are either isotropic or uniaxial with an out-of-plane optical axis. On the contrary, WTe$_2$ reveals notable in-plane anisotropy with distinct plasma frequencies between a and b axes within the WTe$_2$ plane [44] [45]. In the analysis that follows, we assumed that trilayer WTe$_2$ can be reasonably described as a uniaxial material with its in-plane relative permittivity represented by that of b-axis of bulk WTe$_2$. We also performed calculations with both a purely a-axis response and an effective dielectric function averaging between a-axis and b-axis data. All three methods produce qualitatively similar results (see 3.7.4).

We proceed with the quantitative analysis of the nano-THz response of WTe$_2$ trilayers by calculating the p-polarized reflectivity $r_p(\omega, q)$ following the procedure described in ref [56]. The imaginary part of $r_p(\omega, q)$ (Figure 3.3a) reveals a branch of strongly dispersing SPP. The three modes in Figure 3.3a and b are, from left to right, the free-space light line, the light line in SiO$_2$ and the SPP in trilayer WTe$_2$. The SPP is sharp at low temperatures, while at 290 K it is overdamped. This is due to reduced scattering of electrons at low temperature [87] [88].
The dispersion calculation in Figure 3.3a and b implies that the SPP wavelength is 6~20 μm in the THz range. Because the tips we utilize in nano-THz experiments have radii \( R = 150\text{~}200 \text{ nm} \), we gain access to the range of momenta peaked around \( 0.1/R \approx 5 \times 10^3 \text{ cm}^{-1} \) \([2][85][56]\). Since the THz intensity in our experiments is spread over 0.5~1.5 THz, we can extract the accessible range of wavelengths of the SPP modes from Figure 3.3a. This straightforward procedure suggests that the relevant modes occur between \( 4 \times 10^3 \text{ cm}^{-1} \) and \( 8 \times 10^3 \text{ cm}^{-1} \), implying that the wavelengths of these modes span the range between 6~20 μm. Our THz near-field tip is thus expected to efficiently couple to SPP modes in trilayer WTe_2.

Next, we calculated the near-field spectra of WTe_2 based on \( r_p(\omega, q) \) dispersion calculations. In Figure 3.3b, we show the near-field amplitude spectrum produced within the framework of the LRM \([56]\) at different temperatures. In the 0.5~1.5 THz range, the measured near-field signal is governed by the SPP of WTe_2. At low temperatures, plasmonic losses due to electron-phonon scattering are reduced and the SPP mode becomes more pronounced. By integrating the near-field signal at all frequencies investigated with our THz apparatus (shaded region in Figure 3.3c), we acquired the model near-field signal at all temperatures. The result is plotted in Figure 3.3e (red dashed line) along with the experimental data. This analysis captured the temperature dependence of the experimental data but produces higher signal level than the measurement. Therefore, we conclude that the temperature dependence of 3L WTe_2 is impacted by the SPP. The fact that the model signal is overall higher indicates that 3L WTe_2 is less metallic than the bulk.
Figure 3.4: Near-field electrodynamics of thermally activated carrier of few-layer WTe₂. (a) Temperature dependence of thermally activated carrier densities at different gap sizes calculated (see 3.7.6) based on the band structure investigated by ARPES [70]. (b) Near-field spectroscopic response of the thermally activated carriers of a model 2L WTe₂ with Δ = −10 meV. The green shaded region represents the power spectrum of the THz probe. (c) Right panel: Temperature dependent WL signal calculation based on LRM for 1L (black curve), 2L (blue curve) and 3L (red curve) WTe₂ with gap sizes 60 meV, -10 meV and -20 meV respectively. Along with the model, nano-THz data of 1L, 2L and 3L WTe₂ are displayed with squares. The error-bars are the standard deviations of the extracted data in regions indicated in Figure 3.2b. Both the model curves and experiment points are normalized to the substrate value. Left panel: The gap-size dependent near-field signal of 2L (blue) and 3L (red) at 44 K. The signal level is strongly suppressed when the gap is close to zero or positive. (d, e) Hypothetical band structure of semimetallic 2L WTe₂ (left) and insulating 1L WTe₂ [70] with a bandgap Δ > 60 meV (right).

While the presence of a large gap of >60 meV in monolayer WTe₂ is demonstrated by transport [51] and ARPES [70] measurements, the semiconductor versus semimetallic nature of the bilayer remains unclear. ARPES experiments on bilayer WTe₂ [70] indicate a vanishing, if not negative, gap (Figure 3.4d). Transport measurements indicate semiconducting/insulating behavior with a small positive gap (<10 meV) [51] (Figure 3.4e). Our local nano-THz
experiments provide a unique probe in the relevant frequency region, without complications from electrical contacts and inevitable defects. The pronounced temperature dependence observed in metallic trilayers is partially due to the impact of the SPP. The complete insulating behavior of monolayer areas is likely due to its large gap (>60 meV). On bilayer WTe$_2$, the fact that its near-field signal is higher than monolayer WTe$_2$ requires a weak metallicity, as we explain below.

Within the small gap or negative gap scenario, thermally activated carriers are the main contributor to the weak metallicity of bilayer WTe$_2$. In Figure 3.4, we theoretically investigated the temperature dependence of the near-field signal due to the thermally activated carriers in bilayer WTe$_2$ with different gap sizes (see 3.7.7). In Figure 3.4a, when the gap size is in the range of -10 meV to 10 meV, the carrier density at 44 K is as high as $n_2D = 0.2\sim 1\times 10^{12} \text{cm}^{-2}$, which is smaller than the value ($3.6\times 10^{12} \text{cm}^{-2}$) estimated in the ARPES experiment [70]. The temperature dependences of the scattering rate and of the carrier density dictate the temperature dependence of the near-field response. As is shown in Figure 3.4b, thermally activated carriers directly contribute to the signal measured in our experiment.

At $\Delta \sim -10$ meV, the simulated temperature dependence of WL near-field signal (blue curve in the right panel of Figure 3.4c) matches the experimental data well. In 3.7.7, the temperature dependence of the WL near-field signal corresponding to different gap sizes of bilayer WTe$_2$ are displayed. When the gap size is larger than 10 meV ($\sim 2.5$ THz), thermally activated carrier density is sufficiently low that the near-field response in our THz range (0.5~1.5 THz) resembles an insulator. In the right panel of Figure 3.4c, we modeled the temperature dependence for monolayer (black curve) with this “large gap” scenario ($\Delta = 60$ meV). When the gap size is reduced below +10 meV (see 3.7.7, Figure 3.12), the near-field signal at high
temperature gradually increases and is comparable to the experiment value. However, the carrier density (Figure 3.4a) at low temperature gradually vanishes, leading to a strong suppression of the near-field signal at low temperatures (Figure 3.12). The temperature independent behavior for bilayer WTe$_2$ observed in the experiment (orange square dots in Figure 3.4c) therefore calls for a finite carrier density even at the lowest temperature (44 K), which favors the semimetallic scenario. Once the gap size is reduced to -10 meV (overlapping conduction and valence band), the signal at low temperature becomes comparable to that at high temperature and better fits the experimental value (Figure 3.4c). In the left panel of Figure 3.4c, we summarized the gap-size-dependent near-field signal at the base temperature 44 K. Further increasing the absolute negative gap leads to an increase of near-field signal at low temperatures to the levels exceeding data for bilayer WTe$_2$, due to the abundance of carriers (Figure 3.12). Therefore, our observation of the temperature independent WL signal on bilayer WTe$_2$ favors the semimetallic nature with a small negative gap ($\Delta \sim -10$ meV).

We applied the same calculation to the trilayer WTe$_2$. In the left panel of Figure 3.4c, we summarized the gap-size-dependent near-field signal at the base temperature 44 K as well. Because of the thickness effect, the simulated near-field signal on trilayer is higher than bilayer with the same gap size. In the right panel of Figure 3.4c, the simulated temperature dependence of the WL signal on trilayer WTe$_2$ with $\Delta = -20$ meV (red curve) almost perfectly fits the experimental data. Therefore, thermally activated carriers with -20 meV gap better explain the trilayer near-field signal compared to the simulation using bulk WTe$_2$ optical constants (Figure 3.3).
Figure 3.5: Layer-dependent WL near-field signal on WTe$_2$. (a) Topography image of the sample at 44 K. The boundaries of different regions are highlighted with dashed lines. (b) Normalized WL signal $S_z/S^\text{sub}_z$ imaged simultaneously with the topography. (c) Layer dependent near-field signal on few-layer WTe$_2$. The experiment data are collected on two different samples. The data points extracted from Figure 3.2 (sample 1) are marked with crosses. The data points acquired on sample 2 are marked with squares. 3L WTe$_2$ is measured on both samples. High and low temperature data are displayed in red and blue points. The error-bars are the standard deviations of the extracted data in panel b and Figure 3.2b. The dashed lines are LRM near-field signal contributed by thermally activated carriers with a gap size $\sim$20 meV at 44 K (blue) and 295 K (red).

To illustrate how THz near-field signal evolves with thicker WTe$_2$, we prepared a different sample with 3L, 4L, 6L and 12L WTe$_2$ on which the same measurement was performed. Except for the difference in the thickness of WTe$_2$, the overall configuration of the sample is the same. In Figure 3.5c, the data extracted from Figure 3.2 (Exp 1) and the data extracted from Figure 3.5b (Exp 2) are displayed side-by-side. With the increase of the layer number, the near-field signal increases rapidly from 3L and the growth rate decreases with the increase of the layer number. For 12L at 44 K, the near-field signal level is 86% of the bulk WTe$_2$ calculated using bulk optical constants [44]. According to Figure 3.5, the growth continues at 12L, but the converging behavior is already obvious. By assuming 3L – 12L WTe$_2$ can still be described by a 2D band structure, we apply the same model described in Figure 3.4 on these thicknesses. Here, we fitted the 3L – 12L experimental data at high and low temperature using
LRM with only one free parameter, the gap size $\Delta$. When $\Delta \sim -20$ meV, the model results simultaneously matched the high and low temperature data. Therefore, from 3L to 12L, WTe$_2$ can be reasonably described as a semi-metal with a negative gap $\Delta \sim -20$ meV.

### 3.4 Modeling of polaritonic patterns in real-space

![Image](image-url)

**Figure 3.6:** Real-space modeling of SPP pattern. (a, b) Result of the model real-space near-field ($S_{2}/S_{2}^{sub}$) pattern associated with SPP at 295 K (a) and 70 K (b) on the investigated sample geometry. Dashed lines highlight the physical boundaries of all regions. Line-cuts across 2L and 3L WTe$_2$ are indicated by the solid line in both images. (c) Model $S_{2}/S_{2}^{sub}$ line-cuts extracted from a and b are plotted in solid lines. Along with the model result, experiment line-cuts are displayed in dashed lines and are shifted vertically for clarity.

With knowledge of the THz electrodynamic properties of mono-, bi- and trilayer regions on our WTe$_2$ microcrystal, it is now possible to model the real-space pattern (Figure 3.6) of the THz near-field based on the geometry of the sample shown in Figure 3.1c. Following the analysis in Figure 3.4, we assigned a semimetal model with a -10 meV gap for bilayer and a -20 meV gap for trilayer regions. We adopted the permittivity extracted from DFT calculation [89] for the monolayer region. The real-space modeling in Figure 3.6 considers the intrinsic SPP mode on the experimentally measured geometrical configuration of the microcrystals. Further details of this real-space calculation are provided in section 3.7.8.

The real-space near-field modeling results for few-layer WTe$_2$ (Figure 3.6a and b) are in excellent agreement with the experimental images (Figure 3.1c, Figure 3.2a and b). In Figure 3.6c, line-cuts were extracted at the same location with Figure 3.2 and are compared with the
experimental results. In the case of bilayer, the model shows the temperature independent behavior of the signal level as expected from the result in Figure 3.4. The slope of the signal from the substrate side to the trilayer side is also reproduced well. Importantly, the gradual transition of near-field signal on bi- and trilayer edges are present in both experiment and model results, proving that the blurred edges are caused by the long wavelength of the THz range SPPs. In the model result, a weak fringe pattern can be recognized on 3L WTe$_2$. In real samples, however, the fringe signature could be easily erased due to the lower quality factor of SPPs. As for bilayer WTe$_2$, despite the low carrier density, the near-field response of SPPs can be detected in THz frequencies and is strongly impacted by the thermally activated carriers. According to Figure 3.4c, a similar response is also expected in narrow gap semiconductors at even higher temperatures.

3.5 Conclusion

In conclusion, we investigated the low temperature nanoscale electromagnetic response of few-layer WTe$_2$ micro-crystals at THz frequencies. The low-temperature near-field signal has a strong dependence on the number of layers. The response of trilayer WTe$_2$ is clearly metallic as evidenced by the temperature dependence and is dominated by SPPs in the confined geometry of narrow terraces. The weak response of monolayer is consistent with an insulator with relatively large bandgap. Bilayer WTe$_2$ shows higher THz signal than insulating monolayers but the observed THz response is also independent of temperature from 250 K to 44 K. This latter behavior implies finite carrier density in bilayers down to the lowest temperature of this experiment (44 K). Comparison to our model suggests that the WTe$_2$ bilayer is a semimetal with a small negative gap $\Delta \sim -10$ meV for bilayer WTe$_2$. When the layer number is higher than three, the near-field signal continues growing and a negative gap $\Delta \sim -20$ meV can reasonably
describe 3L – 12L WTe₂. For 12L at 44 K, the near-field signal level is ~86% of the bulk WTe₂, calculated based on bulk optical constants. Finally, knowledge of the electrodynamics of mono-, bi- and trilayer WTe₂ in our sample allows for a direct real-space modeling of the THz near-field signal, which matches the experiment well. Our complete temperature dependent THz near-field images together with theoretical modeling paves the way for understanding the low energy electrodynamics of future quantum materials beyond the diffraction limit.

3.6 Method

3.6.1 THz scanning-type near-field optical microscope (THz-SNOM)

Both the AFM scanner and focusing optics of our apparatus (Figure 3.1a) are situated in an ultra-high vacuum (UHV) compartment. This allows for measurements at temperatures down to ~40K limited by the imperfect thermal contact of a sample carrier introduced through rapid access load locks into our UHV system.

In the experiment on sample 1 (Figure 3.1, Figure 3.2), we utilize a pair of low temperature-grown GaAs photoconductive antennas (PCA, Neaspec GmbH) as emitter and detector. We activate both PCAs with a 1550nm femtosecond fiber laser after doubling its frequency in a nonlinear crystal. In the experiment on sample 2 (Figure 3.5), we utilize optical rectification of a single pump beam for THz generation and electro-optic (EO) sampling for THz detection. By tilting the phase front of a 17 W, 1030 nm pump beam, we achieve the necessary phase matching condition to generate THz radiation via optical rectification in LiNbO₃ with an efficiency of 0.1 %. The scattered beam is routed to a ZnTe crystal for EO detection in the time domain using a delta-function like 800 nm gate beam with pulse duration of 20 fs.

In this experiment, we exploit the frequency-integrated (WL) signal to produce high fidelity images. When a THz pulse is scattered by the tip and reaches to the detector, we can
measure this pulse at different time point $t_m$. If we tune $t_m$ to the main peak of the detected pulse where the phases of all frequency components in the wave packet are roughly equal, the WL signal is acquired. For trilayer WTe$_2$, the near-field spectra are almost flat (Figure 3.3c). Therefore, WL images are suitable to track its temperature dependence. For bilayer and monolayer regions, because of the low signal level, WL images are needed to produce meaningful results.

### 3.6.2 Preparation of WTe$_2$ microcrystal

WTe$_2$ crystals are mechanically exfoliated onto highly p-doped silicon substrates consisting of 285 nm SiO$_2$ [90]. WTe$_2$ flakes of mono- to trilayers are optically identified and encapsulated within hBN flakes using standard polymer-based dry transfer technique. The top and bottom hBN flakes used for encapsulation are typically 5-7 nm thick and 12-30 nm thick, respectively. Both WTe$_2$ exfoliation and encapsulation processes are performed inside a nitrogen glovebox (oxygen and water vapor levels are less than 0.5 ppm). The polymer on top of the heterostructures are dissolved outside the glovebox before near-field optical measurements.

### 3.6.3 Lightning-rod model calculations of near-field signals.

We mainly follow the modeling procedure described in Ref. [56]. The modeling is based on reflection coefficient $r_p(\omega, q)$ of the layered structure of the sample. A numerical solution to the electric field distribution of a tip-sample system is used to calculate near-field signal. In this way, parameters like tip radius and tapping amplitude is considered in the modeling. However, because the model is based on a 19 $\mu$m long metallic tip with a cone structure. It does not account for the resonance of the 80 $\mu$m tip to THz beam in the experiment. Our solution is to manually multiply the model spectra with the spectra measured on Au and use it as an approximation to experiment result.
3.7 Supplementary

3.7.1 Comparison between topography and near-field images

![AFM image](image1)

![Near-field S1 image](image2)

Figure 3.7: Comparison between topography and near-field images. (a) The topography image of the field-of-view shown in Figure 3.1b and c. The dashed lines, indicating the boundaries between different regions, are determined by topographical contrast of monolayer (1L), bilayer (2L) and tri-layer (3L) regions. (b) The near-field S1 image taken simultaneously with the topography image.

A side-by-side comparison between topography and near-field images helps better locate different regions on the sample surface. The micrometer-sized white dots in topography images, corresponding to the black dots in the near-field images, result from bubbles between WTe$_2$ sample and hBN encapsulation layers. Because the existence of the top layer hBN and bubbles, locating different regions in atomic force microscopy (AFM) images is challenging. Nevertheless, we can still track the boundary by taking line-cuts and compare with the near-field image and the optical inspection image.
3.7.2 Spatial resolution and approach Curve of $S_1$ and $S_2$ near-field signal

Figure 3.8: Near-field Approach Curve of $S_1$ and $S_2$ signal. The approach curve is measured by varying the tip-sample distance. $S_1$ signal is mainly contributed by E field within 500nm above the sample surface, whereas $S_2$ signal is confined within 150nm above the sample surface.

To illustrate how localized the signal is above the sample surface, we measured the dependence of near-field signal on the tip-sample distance. In the Figure 3.8, $S_2$ signal is much more localized than $S_1$. More than 90% of $S_2$ signal is contributed within 150nm above the sample surface. For $S_1$ signal, this length scale of signal decay is ~500nm. In terms of full width half maximum, the decay length scale for $S_1$ and $S_2$ signal are <100nm and <50nm. The in-plane length scale of E-field localization of near-field signal equals that of out-of-plan [56]. Hence, the resolution of $S_1$ ($S_2$) image is no larger than 200nm (100nm).
3.7.3 Zoomed-in images of monolayer WTe2

Figure 3.9: Zoomed-in images of monolayer WTe2. (a) THz near-field S1 image of the whole sample area at room temperature. We mark the boundaries of terraces with different number of WTe2 layers (1L, 2L, 3L) with dashed lines. The red frame indicates the area where the monolayer images are taken. (b) Zoomed-in S1 image around monolayer region. (c) Zoomed-in S2 image around monolayer region.

Monolayer WTe₂ is confirmed to support quantum spin Hall state below 100K [91] [64] [65] [66] [92] [67] with edge conduction channels. At the lowest temperature of the experiment 44K, THz response near images around monolayer WTe₂ was measured in more detail (Figure 3.9). The location of the zoomed-in field-of-view is indicated in Figure 3.9a with a red frame. In this field-of-view, we can see the boundaries between monolayer WTe₂ and substrate and between monolayer and bi-layer WTe₂. With our current signal to noise ratio, no clear feature arising from the topological edge state is observed, despite previous observation of the edge state at DC [51] and microwave frequencies [64]. It is possible that the increase of conductivity due to edge state does not extend to THz range. However, the low conductivity [51] of the edge state is also challenging for nano-THz technology.
3.7.4 The influence of in-plane anisotropy of WTe$_2$ on near-field modeling.

Figure 3.10: (a) Temperature dependent normalized S2 from experiment and modeling. Experimental data points are indicated with solid squares. Near-field modeling are indicated with empty diamonds. Three different sets of parameters: $\varepsilon_{\parallel} = \varepsilon_b$, $\varepsilon_{\parallel} = \varepsilon_a$ and $\varepsilon_{\parallel} = (\varepsilon_a + \varepsilon_b)/2$ are configured for 3L WTe$_2$ modeling. (b) Normalized S2 signals averaged in the whole regions of 1L, 2L and 3L. The boundaries of corresponding regions are indicated in Figure 3.2a.

The near-field modeling mainly follows the procedure described in Ref. [56]. One assumption is that the material can be treated as isotropic or uniaxial material with a unique out-of-plane axis. On the contrary, WTe$_2$ reveals notable in-plane anisotropy with distinct plasma frequencies between a and b axes within the WTe$_2$ plane [44] [45]. We assumed that tri-layer WTe$_2$ can be reasonably described as a uniaxial material with its in-plane relative permittivity represented by that of b-axis of bulk WTe$_2$. Nevertheless, the finite in-plane anisotropy is evident only at much higher frequencies (~12 THz) [44] and gives rise to minor quantitative change in the observed near-field response, as demonstrated below. We first performed calculations with both a purely a-axis response and an effective dielectric function averaging between a-axis and b-axis data. In Figure 3.10, three different configurations are displayed. Modeling with $\varepsilon_{\parallel} = \varepsilon_b$ has the highest increase of near-field signal at low temperature. The increase is lower in $\varepsilon_{\parallel} =$
\((\epsilon_b + \epsilon_a)/2\) and the lowest in \(\epsilon_\parallel = \epsilon_a\), despite the overall differences are small enough to neglect.

The temperature dependent behavior is not unique to the field of view we chose to perform averaging. In Figure 3.10b, we display the near-field signal averaged within the entire regions of 1L, 2L and 3L indicated in Figure 3.2a. Because the edges with lower signal are included, the overall signal level is decreased. The inclusion of bubbles and other local defects contributes to slightly worse statistics. Except for the decreased signal level and influence due to defects, the temperature dependent behavior of each region is not changed.

### 3.7.5 Mid-IR response of monolayer and few layer WTe2 at the nanoscale.

Figure 3.11: Near-field nano-imaging on multi-terraced encapsulated WTe2 microcrystal in the mid-infrared. Image of the normalized near-field \(S_3\) signal (\(\omega=27\) THz) at (a) 250 K and (b) 57K. (c) Layout of the sample measured in the mid-IR near-field experiments. (d) Temperature dependence of mid-IR near-field signal for regions with different number of layers.
For completeness, we also acquired near-field data in the frequency range higher than the plasma frequency of bulk WTe$_2$. The experiment is carried out in the mid-IR range with a 27 THz CO$_2$ laser. Here, a third (S$_3$) or higher harmonic of the near-field signal must be measured to suppress the far-field contribution [24]. The WTe$_2$-based structure investigated in this experiment shares common elements with the devices in Figure 3.1a. The major difference is that there is another layer of graphite in between the SiO$_2$/Si substrate and bottom layer hBN. This latter architecture is only suitable for mid-IR experiments: the graphite beneath the sample saturates the near-field signal in THz range but not in mid-IR range. Images acquired at 250K and 57K are shown in Figure 3.11a and b. The temperature dependence of nano-IR signals is plotted in Figure 3.11d. WTe$_2$ shows a much weaker nano-IR signal compared to the signal produced by the platinum electrodes. Regions with more WTe$_2$ layers show systematically higher mid-IR signal levels and the temperature dependence for all layers is insignificant (Figure 3.11d). This temperature independent behavior is well explained by our model result shown in Figure 3.3d.

3.7.6 Near-field electrodynamics of thermally activated carriers of bi-layer and thicker WTe$_2$

In calculating the near-field signal of bi-layer WTe$_2$, we mainly consider the Drude response of its thermally activated carriers. The carrier density is computed assuming WTe$_2$ has quadratic bands with effective electron mass at the conduction and valence band edge:

\[
\begin{align*}
n_{2D} &= \int_{E_C-E_F}^{\infty} 2f_{FD}(E)g_{2D}(E)\,dE = \frac{2kTm^*_{C}}{\pi h^2} \ln \left( e^{\frac{E_C-E_F}{kT}} + 1 \right) \\
p_{2D} &= \int_{|E_D-E_F|}^{\infty} 2f_{FD}(E)g_{2D}(E)\,dE = \frac{2kTm^*_{V}}{\pi h^2} \ln \left( e^{\frac{|E_D-E_F|}{kT}} + 1 \right)
\end{align*}
\]

Eq. 3.1
Here, $m_C^*$ and $m_V^*$ are the effective electron mass at the band edge of conduction and valence band. $E_C$, $E_V$ and $E_F$ are the energy of conduction band edge, valence band edge and Fermi energy of bi-layer WTe$_2$. The Fermi energy is determined by the neutrality condition $n_{2D} = p_{2D}$ of the investigated system. The permittivity of the model bi-layer:

$$\epsilon = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega - i\gamma)}$$  \hspace{1cm} \text{Eq. 3.2}

Here $\omega_p$ is the plasma frequency determined by the thermally activated carrier density:

$$\omega_{p,n} = \sqrt{\frac{n_{2d}e^2}{d_m^*\epsilon'}} = \sqrt{\frac{2e^2kT}{d\pi\hbar^2\epsilon'\epsilon_0}} \ln \left( e^{\frac{E_C - E_F}{kT}} + 1 \right)$$  \hspace{1cm} \text{Eq. 3.3}

$$\omega_{p,p} = \sqrt{\frac{p_{2d}e^2}{d_m^*\epsilon'}} = \sqrt{\frac{2e^2kT}{d\pi\hbar^2\epsilon'\epsilon_0}} \ln \left( e^{\frac{|E_V - E_F|}{kT}} + 1 \right)$$

Here $d = 1.4 \text{ nm}$, $\epsilon' = 2.2$ is the dielectric constant of hBN at THz frequencies. The scattering rate $\gamma$ of the model bi-layer is assumed to be the average of the two scattering rates of two Drude components in the bulk [87] $\gamma = (\gamma_1 + \gamma_2)/2$.

For thicker samples, the carrier density multiplies due to the increase of the number of the electronic bands. In the calculation of $\omega_p$, the increase of carrier density cancels the increase of thickness. Therefore, the $\omega_p$ maintained the same value for WTe$_2$ of different thicknesses.
3.7.7 Temperature dependence of white-light near-field signal produced by LRM.

Figure 3.12: Near-field electrodynamics of thermally activated carrier of few-layer WTe2. In panel (c)) Right panel we calculated temperature dependent WL signal based on LRM for 2L (solid lines) and 3L (dashed lines) WTe2 with gap sizes ranging from -30 meV to +30 meV.

In Figure 3.12c, we show temperature dependence of near-field signal on 2L and 3L WTe2 with different gap-size produced by lightning-Rod model. For 2L (solid line in Figure 3.12c), if the gap size is positive and large, the signal at all temperature is close to the substrate. When the gap size is close to 0, the signal is still strongly suppressed at low temperature due to the low carrier density. When the gap becomes negative, the low temperature signal quickly increases and is even higher than high temperature when the gap size is below -10 meV. When the thickness increases from 2L to 3L (dashed line in Figure 3.12), the overall signal level is increased. With the increase of the gap size in the negative direction, the signal becomes stronger. We found Δ~10 meV and Δ~20 meV almost perfectly matches the 2L and 3L data, correspondingly.
3.7.8 Real-space near-field modeling of SPP structures

To generate predicted real-space images shown in the main text, we apply a semi-analytic method that approximates the near-field scattering signal from a 2D material as proportionate (to first-order) by the z-polarization of a polarizable dipole raster-scanned (at a height \( z = z_{dp} \)) some tens of nanometers over the surface of a sample (at \( z = 0 \)):

\[
S(\rho_{dp}) \sim p_z \approx \alpha E_{ref,z}(\rho_{dp}, z = z_{dp}).
\]

Eq. 3.4

Here \( \alpha \) denotes the dipole polarizability, \( E_{ref,z} \) denotes the z-component of the electric near-field reflected by the sample in response to the incident dipole field, and \( \rho_{dp} \) denotes evaluation at the in-plane coordinate of the probe. Although this expression represents only the first term in a Born expansion of the full self-consistent dipole polarization [93] a similar conceptual treatment was previously shown to faithfully replicate the polaritonic near-field response of two-dimensional materials as measured by scanning near-field optical microscopy [57]. Here we summarize the key points enabling our calculation of Eq. 3.4 in the quasi-electrostatic approximation and defer more detailed discussion to forthcoming work.

We recast Eq. 3.4 in a form reminiscent of the local photonic density of states [94] measured at the location \( r_{dp} = (\rho_{dp}, z_{dp}) \) of our dipole probe:

\[
S(\rho_{dp}) \sim \int_{z>0} dV \; j_{dp} \cdot \vec{E}_{ref} = \int_{z>0} dV \; \nabla \cdot j_{dp} \; \Phi_{ref} \propto \int_{z>0} dV \; -q_{dp} \cdot \Phi_{ref}
\]

Eq. 3.5

Here \( j_{dp} \) denotes the unit vector oriented along the direction of the point dipole current, \( q_{dp} \) denotes the instantaneous charge distribution associated with the dipole, and \( \Phi_{ref} \) is the electrostatic potential for the reflected field given by \( \vec{E}_{ref} = -\nabla \Phi_{ref} \). Now \( S(\rho_{dp}) \) can be evaluated entirely in the plane \( z = 0 \) by identifying the “incident” electrostatic potential generated by the dipole through \( q_{dp} = -\frac{1}{4\pi} \nabla^2 \Phi_{dp} \) and integrating Eq. 3.5 by parts, yielding:
\[
\int_{z>0} dV - \varrho_{dp} \cdot \Phi_{ref} = \frac{1}{4\pi} \left[ \int_{z=0^+} dA \left( -\hat{z} \cdot \nabla \Phi_{dp} \right) \Phi_{ref} - \int_{z>0} dV \nabla \Phi_{dp} \cdot \nabla \Phi_{ref} \right]
\]

\[= \frac{1}{4\pi} \int_{z=0^+} dA \left( \Phi_{dp} \partial_z \Phi_{ref} - \partial_z \Phi_{dp} \Phi_{ref} \right).\]

Eq. 3.6

Here we have applied the source-free condition \(\nabla^2 \Phi_{ref} = 0\) in the volume \(z > 0\). Eq. 3.6 represents an approximation for the signal \(S(\rho_{dp})\) when \(\Phi_{dp}\) is produced from a dipole-like probe at \(r_{dp}\). Further simplification is admitted by the fact that \(\Phi_{ref} = -\hat{R} \Phi_{dp} \equiv -\Phi_R\), with \(\hat{R}\) a generalized reflection operator. Moreover, for scalar potentials \(\Phi_{1,2}\) harmonic (viz. source-free) in the plane of integration, \(\int dA \Phi_1 \partial_z \Phi_2 = \pm \int d^2q |q| \tilde{\Phi}_1 \tilde{\Phi}_2\), where tilde quantities represent in-plane Fourier transforms with respect to the momentum \(q\), and \(\pm\) correspond to the cases where \(\Phi_2\) is sourced from \(z > 0\) or \(z < 0\), respectively. With these considerations, Eq. 3.6 reduces to:

\[S(\rho_{dp}) \sim \frac{1}{2\pi} \int d^2q |q| \tilde{\Phi}_{dp} \tilde{\Phi}_R = \frac{1}{2\pi} \int dA (q \ast \Phi_{dp}) \hat{R} \Phi_{dp}\]

Eq. 3.7

where \((q \ast \Phi_{dp})\) represents the incident scalar potential spatially convolved at \(z = 0^+\) with a sharpening function with Fourier kernel \(|q|\). Eq. 3.7 represents a norm of the function \(\Phi_{dp}\) in the plane \(z = 0\) with respect to the composite reflection operator \(q \ast \hat{R}\). By way of demonstration, we can consider cases where the reflected field is given by \(\Phi_R = r_p(q) \tilde{\Phi}_{dp}(q)\), with \(r_p\) the momentum-resolved Fresnel coefficient for e.g. a layered medium with in-plane translational invariance. Applying the in-plane Fourier transform of the dipole potential \(\tilde{\Phi}_{dp}(q) = e^{-aq_{dp}}\) at \(z = 0\), for such cases Eq. 3.7 evaluates to \(S \propto \int dq \ r_p(q) \ q^2 e^{-2aq_{dp}}\). This is indeed the first-order term in a Born series expansion of the point dipole model widely used to predict near-field observables in the case of multilayered systems [85] [95]. Meanwhile, whereas the real-space counterpart that we present in Eq. 3.7 remains underreported, it provides a powerful means to predict images recorded by scanning near-field optical microscopy.
We now briefly describe our method for evaluating $\hat{R}\Phi_{dp}$ in the case of a spatially inhomogeneous 2D material at $z = 0$ described by a (piecewise) optical conductivity $\sigma_{2D}(\rho)$ upon a substrate with isotropic reflectivity $\beta_{\text{subs}}$. We first consider the integro-differential equation for the scalar potential $\Phi_{\text{ref}}$ generated by $\sigma_{2D}$ in response to the potential $\Phi_{dp}$ of our quasi-dipolar probe [96], in absence of a substrate:

$$\left[ 1 + V \ast \sum_m \frac{1}{2\pi q_{p,m}} \nabla \cdot \tilde{\sigma}_m(\rho) \nabla \right] \Phi(\rho) = \Phi_{dp}(\rho), \text{ with } \Phi = \Phi_{dp} + \Phi_{\text{ref}}. \quad \text{Eq. 3.8}$$

Here $m$ indexes the piecewise homogeneous domains of our 2D material (i.e. in our case domains of mono-, bi-, and tri-layer WTe$_2$), $q_{p,m}$ denotes the complex plasmon wavevector associated with each domain, and $\tilde{\sigma}_m(\rho)$ are piecewise homogeneous functions equal to zero or 1 marking the lateral regions $\rho \in \Omega_m$ occupied by each domain. Meanwhile, $V(\mathbf{r}, \mathbf{r}') = 1/|\mathbf{r} - \mathbf{r'}|$ is the Coulomb kernel, and the asterisk ($\ast$) denotes spatial convolution over the in-plane coordinate $\rho = (x, y)$. We solve Eq. 3.8 by expanding $\Phi_{\text{ref}}(\rho) = \sum_{mn} \phi_{mn}^{\text{ref}} \Phi_{mn}(\rho)$ into an orthonormal basis of eigenfunctions specified on the domains $\partial\Omega_m$ by $\nabla \cdot \tilde{\sigma}_m(\rho) \nabla \Phi_{mn}(\rho) = -q_{mn}^2 \Phi_{mn}(\rho)$ and subject to the “zero current” boundary conditions $\mathbf{n} \cdot \nabla \Phi_{mn}$ on the domain edges $\partial\Omega_m$. These functions are obtained with the finite element solver FEniCs [97] after meshing the experimentally relevant domain configurations shown in Figure 3.6a.

Table 3.1: Parameters for modeling real-space near-field images of WTe$_2$ monolayer, bilayer, and trilayer domains at 70 K and 295 K. Layer thickness and the complex-valued optical permittivity $\varepsilon=\varepsilon_1+i\varepsilon_2$ determines the plasmon wave-vector. Permittivities are estimated according to discussion in the main test.

<table>
<thead>
<tr>
<th>Domain (indexed)</th>
<th>Thickness (nm)</th>
<th>Optical permittivity (THz)</th>
<th>Plasmon wave-vector (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Monolayer (70 K)</td>
<td>0.7</td>
<td>41</td>
<td>710000</td>
</tr>
<tr>
<td>Monolayer (295 K)</td>
<td>0.7</td>
<td>41</td>
<td>710000</td>
</tr>
<tr>
<td>Bilayer (70 K)</td>
<td>1.4</td>
<td>-463 + 1389i</td>
<td>3091 + 9252i</td>
</tr>
<tr>
<td>Bilayer (295 K)</td>
<td>1.4</td>
<td>-110 + 1362i</td>
<td>849 + 10420i</td>
</tr>
<tr>
<td>Trilayer (70 K)</td>
<td>2.1</td>
<td>-677 + 2031i</td>
<td>1409 + 4219i</td>
</tr>
</tbody>
</table>
Values for the plasmon wave-vectors are inferred from the respective layer thicknesses $d_m$ and complex in-plane optical permittivities $\varepsilon_m$ of each domain in $\Omega_m$ according to $q_{p,m} = 2(1 - \varepsilon_m)^{-1} d_m^{-1}$; see Table 3.1 for the values used in our simulations. Although WTe$_2$ is known to exhibit biaxial in-plane permittivity, for simplicity in our simulations we apply an isotropic approximation as earlier discussed in this supplement.

Assembling the expansion coefficients $\phi^\text{ref}_{mn}$ into a vector $\phi_{\text{ref}}$ (taking $mn$ as a composite index), we solve Eq. 3.8 by the matrix equation:

$$
\phi_{\text{ref}} = -\left[ -V \sum_m q_m^2 / (2\pi q_{p,m}) \right] \phi_{dp}
$$

Eq. 3.9

As with $\phi_{\text{ref}}$, here $\phi_{dp}$ represents the vector of expansion coefficients for $\Phi_{dp}(\rho)$. Meanwhile, each $q_m^2$ denotes a diagonal matrix of eigenvalues $q_{mn}^2$ acting in the vector subspace spanned by $\{\Phi_{mn} \forall n\}$ at fixed domain index $m$, and $V$ is the coulomb matrix whose elements are given by $V_{kl,mn} = \int_{z=0} dA \Phi_{kl}(\rho) V \Phi_{mn}(\rho)$. The term in brackets in Eq. 3.9 represents the generalized reflection operator $R$ for the system in the $\Phi_{mn}$ basis. (The denominator is understood in the sense of a matrix inverse applied before pre-multiplication by the numerator.)

While we defer the derivation to forthcoming work, this reflection operator generalizes to the case of our 2D materials upon a substrate with isotropic reflectivity $\beta_{\text{subs}}$ as follows:

$$
R = \beta_{\text{subs}} - V \sum_n q_n^2 / (2\pi \kappa q_{p,n}) \left/ \frac{1 - V \sum_n q_n^2 / (2\pi \kappa q_{p,n})}{1 - V \sum_n q_n^2 / (2\pi \kappa q_{p,n})} \right.
$$

Eq. 3.10

Here $\kappa = (\varepsilon_{\text{subs}} + 1)/2$ and $\beta_{\text{subs}} = (\varepsilon_{\text{subs}} - 1)/(\varepsilon_{\text{subs}} + 1)$, with $\varepsilon_{\text{subs}}$ the substrate permittivity; in this work the substrate includes 20nm hBN and bulk SiO$_2$. 

| Trilayer (295 K) | 2.1 | -134 + 1576i | 514+6000i |
We also define a symmetric matrix \( Q \) in the \( \Phi_{mn} \) basis corresponding to the spatial convolution in Eq. 3.7, with elements given by 
\[
Q_{kl,mn} = \int_{z=0} dA \, \Phi_{kl}(\rho) \, q \ast \Phi_{mn}(\rho).
\]
Since the \( \Phi_{mn} \) are orthonormal, Eq. 3.7 reduces to:
\[
S(\rho_{dp}) \sim \frac{1}{2\pi} \, \phi_{dp}(\rho_{dp})^T \, QR \, \phi_{dp}(\rho_{dp}).
\]
Eq. 3.11
This represents a vector norm of \( \phi_{dp} \) with respect to the matrix \( QR \).

In summary, after computing eigenfunctions \( \Phi_{mn} \) associated with our sample geometry, we compute symmetric matrices \( V \) and \( Q \) and the generalized reflectance operator \( R \). Then, in order to predict a spatial map \( S(\rho_{dp}) \), we simply project the incident potential emitted by our quasi-dipolar probe at each location \( \rho_{dp} \) into the \( \Phi_{nm} \) basis by evaluating the vector of coefficients 
\[
\phi_{dp,mn}(\rho_{dp}) = \int dA \, \Phi_{dp}(\rho) \Phi_{mn}(\rho)
\]
and successively applying Eq. S8. Although the eigenbasis \( \{\Phi_{mn}\} \) is of infinite size, projections into \( \phi_{dp} \) decay exponentially with \( n \) when \( \Phi_{mn} \) are sorted by increasing eigenvalue \( q_{mn}^2 \), so a truncated basis of size \( N \approx 10^3 \) is in our case sufficient for a converged map of near-field scattering amplitude \( |S(\rho_{dp})| \). In this way, the observables of near-field microscopy can be predicted entirely by evaluating functions in the plane of the sample \( (z = 0) \). This computational method may be suitable for qualitative and quantitative modeling of near-field response of other spatially inhomogeneous 2D heterostructures. Such applications and details of their unique numerical implementation will be reported elsewhere.
4.1 Introduction

Interactions between charge, spin, lattice and/or orbital degree of freedom have been the central topic in the research of metal-to-insulator transition (MIT) in transition metal oxides (TMO) [98] [99]. Cd$_2$Os$_2$O$_7$ [100] [101] [102] along with R$_2$Ir$_2$O$_7$ (R=Nd, Sm, Eu, Gd, Tb, Dy and Ho) [103] [104] belongs to a special group of TMO whose MIT is argued to be driven by the concurrent emergence of the magnetic ordering of their 5d electrons. In Slater’s theory [105], a second-order MIT in a magnetic system can be driven by the doubling of the unit cell due to the collinearly aligned opposite spins. Hints for the Slater transition were identified in early experiments on Cd$_2$Os$_2$O$_7$ [102]. However, instead of the collinear ordering, numerous experimental studies [106] [107] [108] [109] [110] [111] [112] confirmed that the magnetic ground state of pyrochlore magnetic insulators is all-in/all-out (AIAO) where 4 spins points inward/outward from the center of the corner-shared tetrahedral. AIAO ground state in 5d pyrochlore iridates was predicted to support topological Weyl semimetal phase [113] [114] [115] with a gapless surface state [113]. Therefore, a plethora of new states and phenomena [116] are expected in Cd$_2$Os$_2$O$_7$, especially considering the coexistence of the electron correlation. The undistorted AIAO ground state challenges the Slater scenario but can be better described in terms of the Lifshitz type transitions [108]. The latter scenario involves the continuous shift of conduction and valence band, which was supported by optical [117] and Raman spectroscopic studies [118] of Cd$_2$Os$_2$O$_7$.

Conducting domain walls (DWs) and charged line defects have been predicted in magnetic and antiferromagnetic insulators [119] [120] [121]. THz time-domain spectra on untrained and
trained pyrochlore iridates hinted a Drude-like conductivity residing in the antiferromagnetic domain walls between AIAO and AOAI domains [122]. Microwave impedance microscopy successfully captured the existence of the conducting DWs in Nd$_2$Ir$_2$O$_7$ and their disappearance under the high magnetic field [123]. Cd$_2$Os$_2$O$_7$, on the other hand, has been reported to exhibit controllable domain structures via cooling under the magnetic field [111]. Far-field optical studies also revealed an anomalous increase of THz optical conductivity from 150 K to 25 K [102]. This motivates the search for similar nano-scale evidence of the conducting DWs in Cd$_2$Os$_2$O$_7$ using low-energy probes.

To explore the nano-scale electrodynamics of the MIT and the DWs on Cd$_2$Os$_2$O$_7$, we utilized a scattering-type THz scanning near-field optical microscope (THz-SNOM) [1] [13] [21] [35] [76] [32] [38] [124]. This technique is a combination of atomic force microscopy (AFM) and THz spectroscopy where the AFM tip scatters the local electric field into detectable far-field radiation. A pair of GaAs photoconductive antennas (PCA) is activated by 780 nm femtosecond fiber laser to generate and detect the THz radiation. The scattered field is demodulated at the first- and second-harmonic of the tip-tapping frequency which results in scattering amplitude denoted by $S_{1,2} \propto |\vec{E}^{\text{NF}}|$. The near-field scattering amplitude carries information on the spatially localized electromagnetic response [24, 81, 82, 83]. By probing the scattering amplitude, we overcame the diffraction limit and managed to image the nano-scale textures of the sample, especially the assumed DWs. SNOM technique has been successfully applied to explore MIT in Mott materials like VO$_2$ [1] [125] [15], V$_2$O$_3$ [17] and NdNiO$_3$ [126]. In this experiment, we conducted nano-THz imaging from 230 K to 43 K to track the MIT in Cd$_2$Os$_2$O$_7$ and searched for evidence of the possible DW conductivity.
Here, we focus on the frequency-integrated (“white-light”) near-field signal within the 0.5 THz~1.5 THz range. Apart from the 100 cm⁻¹ (3 THz) and 200 cm⁻¹ (6 THz) phonons, the optical conductivity of Cd₂Os₂O₇ in the THz range is almost frequency independent [102, 117]. As a result, the “white-light” signal is sufficient to track the THz response of MIT in Cd₂Os₂O₇. Besides, high fidelity “white-light” images can be acquired within a reasonable time scale while the sample surface remains pristine. Therefore, we chose “white-light” signal instead of the time/frequency domain spectra [39] to present the near-field mapping of the sample surface. The scattered signal is modulated at the tip oscillation frequency and the demodulation at the first (S₁) and second (S₂) harmonics are collected. The typical signal-to-noise ratio (SNR) of S₁ images (~30:1) is triple of that of S₂ images (~10:1). In principle, higher-order demodulations better suppress far-field scattering [24] and therefore deliver near-field images with genuine local response and finer resolution. In nano-THz, however, the mismatch between the long wavelength of the THz beam and the short tip shank efficiently suppresses the far-field component [24, 83]. Over 90% of the signal in S₁ (S₂) results from the tip scattering within 500nm (150nm) above the sample surface. Therefore, we consider S₁ and S₂ both carry local sample responses at the vicinity of the near-field probe.

4.2 Result

Figure 4.1: Schematic of nano-THz experiments on Cd₂Os₂O₇ single-crystals. (a) Metallic AFM tip locally enhances the electric field and enables THz coupling to materials at
length scales much shorter than the THz wavelength. All nano-THz images are taken near the Au/Cd$_2$Os$_2$O$_7$ edge so that proper normalization of the near-field signal can be implemented. The schematic in the top-right shows the AIAO spin configuration on Osmium atoms. (b) A 30 µm × 30 µm topography image taken at the Au/Cd$_2$Os$_2$O$_7$ interface. The 20 nm thick gold thin film (left) is deposited on the top of Cd$_2$Os$_2$O$_7$. Straight diagonal lines across gold and Cd$_2$Os$_2$O$_7$ are caused by corrugations on the {111} crystal surface. (c) The near-field “white-light” signal (0.5 - 1.5 THz) was taken simultaneously with the topography image at 74K. The gold film exhibits ~10% higher signal than Cd$_2$Os$_2$O$_7$. The counterpart of diagonal topographic features is also evident in nano-THz data.

We investigated the THz near-field response of high-quality Cd$_2$Os$_2$O$_7$ single-crystals as a function of temperature across MIT. The measurement was performed on the as-grown {111} and {100} surfaces [111], on which 20 nm gold thin film is partially coated (Figure 4.1a). We treat the gold films as good THz reflectors [13] and used them to normalize the signal on Cd$_2$Os$_2$O$_7$ crystals for quantitative analysis [1]. Besides the edge of the gold film, the other prominent topographical feature in Figure 4.1b is the straight diagonal lines across both gold and Cd$_2$Os$_2$O$_7$, presumably arising from the corrugation of the crystal surface. These corrugations are widespread in our samples. In Fig. S2, we show that the topographic contrast at the corrugation is below 10 nm. Nevertheless, these features impact the near-field signal and yield dark diagonal lines in near-field images (Figure 4.1c). In some regions, these corrugations are too dense (Fig. S2) to achieve a proper characterization of the near-field response of the crystal. Hence, we specifically chose the region shown in Figure 4.1b and c, where the corrugation density is low, to perform the systematic near-field study. Smaller areas with uniform signals are chosen to carry out the statistics of the near-field signal for quantitative analysis.
Figure 4.2: Nano-THz response of Cd₂Os₂O₇ across the temperature-tuned metal-insulator transition. All nano-scropy images were collected for {111} facet and share the same scale bar shown in the topography image. The false-color scales are indicated on top of each panel. (a) Near-field images of the normalized amplitude contrast $S_1/S_1^{Au}$ of nearly identical regions are measured below the transition temperature $T_{MIT} = 225 K$. The topography image is shown in the top left. (b) Near-field images of the normalized amplitude contrast $S_2/S_2^{Au}$ taken simultaneously with the $S_1/S_1^{Au}$ images in (a). A photograph of the crystal is displayed in the top left, in which the scan area is marked.

The temperature-dependence of THz near-field response of Cd₂Os₂O₇ single crystals below the nominal transition temperature $T_{MIT} = 225 K$ [101] [108] is shown in Figure 4.2. At 220 K, the near-field signal of Cd₂Os₂O₇ is comparable to that on the Au thin film (Figure 4.2a and b). The high signal level persists down to 190K as shown in Figure 4.2a and b. At lower temperatures, $S_1$ ($S_2$) on Cd₂Os₂O₇ decreases gradually until reaching 92% (86%) of the Au signal level at $T = 43K$. We found that the signal is mostly uniform in Cd₂Os₂O₇ at all temperatures, except for the regions impacted by corrugations or defects. No obvious DWs can be observed.
4.3 Lightning-rod modeling of near-field signal

Figure 4.3: Quantitative analysis of nano-THz signal across the MIT in Cd$_2$Os$_2$O$_7$. (a) Mean value of $S_1/S_{1,Au}$ (diamonds) were calculated from data in Fig. 2a. Nano-THz signals was calculated using the lightning-rod model with DC conductivity extracted from Ref [101] as the input. (b) Same as panel (a) but using $S_2/S_{2,Au}$. (c) and (d) The distribution (pixel count) of temperature-dependent $S_1/S_{1,Au}$ and $S_2/S_{2,Au}$ of Cd$_2$Os$_2$O$_7$, respectively.

Quantitative analysis of the temperature dependence of the averaged near-field signal in Cd$_2$Os$_2$O$_7$ is displayed in Figure 4.3a and b. The histograms of the near-field signal are plotted in Figure 4.3c and d. A gradual decrease in the near-field signal at T < 190 K is clearly visible. The histogram helps to better understand the nature of the MIT transitions [17] [126] [1]. In general, a first-order phase transition is characterized by an abrupt change of near-field signals accompanied by the percolation of insulating or metallic phases [17] [126] [1]. In Figure 4.3c and d, we instead see a gradual evolution of near-field signal at THz frequencies, resembling a 2$^{nd}$ order phase transition [126].
We used the lightning rod near-field model to fit the data following the procedure described in Ref. [56]. Due to the lack of optical conductivity data at THz frequencies, we started with the direct-current (DC) conductivity of Cd$_2$Os$_2$O$_7$ [101]. We assumed a simple Drude response with DC conductivity $\sigma_0$ and scattering rate $\gamma$ at each temperature:

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega/\gamma}$$

The DC conductivity $\sigma_0$ is adopted from experiments [101] and the scattering rate $\gamma$ is assumed to be large compared to the investigated frequencies. As a result, $\epsilon_1 \approx \epsilon_{\text{inf}}$ and $\epsilon_2 \approx \frac{\sigma_0}{\omega}$ (4.6.3) at THz frequencies. This assumption is supported by the optical measurement [117] where the carrier density at the Fermi energy gradually changes due to the shift of the conduction and valence band. The result is indicated as solid lines in Figure 4.3a and b, which matches perfectly with the experiment result.

4.4 Discussion

We note that there is an apparent inconsistency between the lower “transition temperature” (~190 K) observed in the near-field experiments and the values reported by transport and far-field experiments. This can be well explained by the signal response function of near-field probes. Cd$_2$Os$_2$O$_7$ has a relatively high conductivity which yields a near-saturated near-field signal at high temperatures above 190 K. Only when the temperature is reduced below 190K, the conductivity of the material is low enough to yield a good contrast to the metallic phase. In Ref. [117], the plasma frequency $\omega_p$ extracted from the extended Drude model implies the existence of residual carrier density below the transition temperature $T_{\text{MIT}}$. This observation is consistent with the result in our experiment that the near-field signal is still high at the lowest temperature (>85% of the RT value).
Cd$_2$Os$_2$O$_7$, like Nd$_2$Ir$_2$O$_7$, is anti-ferromagnetic insulator at low temperature whose DW is claimed to be conductive [122] [123]. To search for the existence of the DWs, we performed nano-THz scans over a wide range of the sample surface. The Cd$_2$Os$_2$O$_7$ single-crystal holds stripe-like domains as wide as ~50 µm on a series of facets detailed in Ref. [111] when the crystal is field cooled, while other facets are single domain. The interaction between magnetic field and the magnetization located at the surface and domain boundary of AIAO ordering leads to the migration and annihilation of the DWs [127] [128] [129]. Without the external magnetic field, DWs are mainly trapped at crystalline defects [127]. Because of the current challenge in applying the high magnetic field with near-field methods, the chance of discovering the DWs mainly depends on the density of the crystalline defect. Our THz near-field measurements conducted on {111} and {100} Cd$_2$Os$_2$O$_7$ did not demonstrate any clear-cut features that can be ascribed to conductive DWs.

Two reasons can likely explain the absence of the DW contrast. Firstly, magnetic domain walls in pyrochlore iridates are theoretically predicted to host various kinds of quantum phases ranging from metal to insulator [130]. Therefore, Cd$_2$Os$_2$O$_7$, sharing the same crystal structure, is possible to host an insulating DW state. Secondly, the conductivity of Cd$_2$Os$_2$O$_7$ is high enough to generate >85% of near-field contrast of Au even at the insulating state below $T_{\text{MIT}}$ (Figure 4.3). The high conductivity overshadows the impact of the domain wall conductivity. On the other hand, in Nd$_2$Ir$_2$O$_7$, far field THz time-domain spectrum hinted that an appreciable near-field contrast can be observed due to the non-zero imaginary part of conductivity above 0.5 THz. Therefore, our experiment result calls for near-field experiment on Nd$_2$Ir$_2$O$_7$ and other candidates in the family of pyrochlore magnetic insulators with lower conductivity.
4.5 Conclusion

In conclusion, we have investigated the metal-to-insulator transition of Cd$_2$Os$_2$O$_7$ single crystals using THz range near-field technology. The evolution of nano-THz signal across the transition is in perfect quantitative arrangement with the DC resistivity measurements and optical measurement in Ref. [117]. The near-field signal across the transition is spatially uniform except for extended corrugation steps. Domain walls revealed in x-ray imaging do not produce a distinguishable nano-THz contrast.

4.6 Supplementary

4.6.1 Terahertz near-field Approach curve

![Terahertz near-field approach curve](image)

Figure 4.4: THz near-field approach curve. The near-field scattering amplitudes are measured as a function of distance between the tip and the sample. Over 90% of the $S_2$ signal is generated within 150 nm above the sample surface.

The in-plane resolution of the near-field signal is proportional to the out-of-plane decay length of the electric field. The decay length is quantified by measuring the dependence of near-field signal on the tip-sample distance (approach curve). In Figure 4.4, the approach curve of
near-field signal demodulated at the first \( (S_1) \) and second \( (S_2) \) harmonics of the tip-modulation frequency are displayed. Over 90\% of the \( S_2 \) signal is generated within 150 nm above the sample surface, which arises from pure near-field electrodynamics. In \( S_1 \), there are finite signal when the tip is 500 nm above the sample surface, proving that \( S_1 \) contains substantial amount of far-field contribution.

### 4.6.2 Surface corrugation of Cd\(_2\)Os\(_2\)O\(_7\) single crystal

![Image of surface corrugation](image)

**Figure 4.5:** Surface corrugation of Cd\(_2\)Os\(_2\)O\(_7\) single crystal. (a), (b) and (c) The topography and near-field signal are acquired at the same time. The scale bars in all images are 5 \( \mu \)m. The density of the surface corrugation varies continuously and in wide range on the surface of the single crystal. These corrugations induce observable impact on the near-field signal. d Three line-cuts are taken at the same location in Z(a), S1(b) and S2(c). The linecut results are compared side-by-side in (d). The height of the corrugation step is below 10 nm.

In Figure 4.5, we show the near-field measurement of a selected region with dense surface corrugation. The topography and near-field images are taken simultaneously. Line-cuts at the same location are extracted and the results are displayed in Figure 4.5d. Because the scanner is controlled using open-loop method, the curvatures shown in the images does not necessarily reflect the real curvatures of the corrugation profiles in the field of view. The topographical step size of the corrugation is on the order of several nanometers.
4.6.3 Surface corrugation of Cd2Os2O7 single crystal

Figure 4.6: Near-field modeling of Cd2Os2O7 single crystal. (a) The temperature dependence of resistivity extracted from Ref. [101]. (b), (c) Frequency dependent $\varepsilon_1$ and $\varepsilon_2$ based on the model described in the main text. (d), € Imaginary part of reflectivity $\mathcal{R}(r_p(\omega, q))$ at 20 K and 210 K. f Near-field spectra at different temperature calculated based on Ref. [17].

We follow the lightning-rod method (LRM) demonstrated in Ref. [17] to model the near-field signal. In LRM, the frequency and momentum dependent reflectivity $r_p(\omega, q)$ is used as input. In order to calculate $r_p(\omega, q)$, we assigned the dielectric function of Cd2Os2O7 based on the DC conductivity [101]. In the infrared optical conductivity published in Ref. [117], the frequency dependence below 500 cm$^{-1}$ is negligible except for phonon lines. Therefore, a simple Drude model is used to characterize the dielectric behavior of Cd2Os2O7,

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega/\gamma}$$
Here, $\sigma_0(T)$ is anchored to the DC conductivity and $\gamma = 1000 \text{ cm}^{-1}$, which is far larger than the frequency range of interest. As a result, $\varepsilon_1 \approx \varepsilon_{\text{inf}}$ and $\varepsilon_2 \approx \frac{\sigma_0}{\omega}$ in the THz frequencies, as is shown in Figure 4.6b and c.

Based on the dielectric function, we can calculate the $r_p(\omega, q)$ at different temperature and the imaginary part at 20 K and 210 K is shown in Figure 4.6d and e. By incorporating the LRM, we can calculate the near-field spectrum (Figure 4.6f). The overall contrast of the near-field spectrum at different temperatures explains the temperature dependence of the near-field white-light signal measured on the Cd$_2$Os$_2$O$_7$ single crystal.
Chapter 5: Imaging hydrodynamic electron flow via nano-
photocurrent measurement

5.1 Schockley-Ramo Formalism

The simulation of the spatial photocurrent pattern of a material with Ohmic transport
behavior is based on Shockley-Ramo (SR) theorem. In the early papers [131, 132], the theory
describes the current collected on the contacts of a capacitor when electrons move in the
capacitor. The similar concept is adapted [133] to calculate the current collected from a material
when photocurrent is locally excited within the material.

The calculation assumes an auxiliary electric potential field $\phi_{SR}$. The field is established
assuming that the current collection and the grounding contacts have 0V and 1V electric
potential,

$$\nabla \cdot \sigma \nabla \phi_{SR} = 0$$  \hspace{1cm} \text{Eq 5.1}

According to the continuity,

$$\nabla \cdot j = \nabla \cdot (j_{pc} + j_n) = 0$$  \hspace{1cm} \text{Eq 5.2}

Here, $j_{pc}$ is the photocurrent generated at a local point and $j_n$ is the normal current distribution
which compensates the photocurrent.

The SR formalism calculates the integral of the inner product of $j_{pc}$ and the auxiliary field $\nabla \phi_{SR}$,

$$\int_S j_{pc} \cdot \nabla \phi_{SR} d^2 r = \int_{\partial S} \phi_{SR} j_{pc} \cdot d\mathbf{n} - \int_S \phi_{SR} \nabla \cdot j_{pc} d^2 r$$
$$= \int_{\partial S} \phi_{SR} j \cdot d\mathbf{n} - \int_S \nabla \phi_{SR} \cdot \sigma \cdot \nabla \phi d^2 r$$  \hspace{1cm} \text{Eq 5.3}
$$= \sum_k I_k \phi_{SR,k} + 0$$
With the Ohmic transport property, the SR integral is directly connected to the current collected at each contact. By assigning potential as stated above, the current collected at one specific contact can be calculated.

5.2 Photothermal nano-photocurrent of an isotropic material

Photothermal effect is one important mechanism in explaining nano-photocurrent of graphene [134]. Due to the heating effect of a far-field illumination or the field enhancement due to a metal tip, the temperature on the graphene becomes inhomogeneous. The temperature gradient drives current near the center of the illumination due to Seebeck effect. The current can be expressed as $j_{loc} = \alpha \nabla \delta T = \sigma S \nabla \delta T$. According to Eq 5.3,

$$\int_S j_{pc} \cdot \nabla \phi_{SR} d^2r = \int_S \nabla \delta T \cdot \alpha \cdot \nabla \phi_{SR} d^2r$$

For isotropic material, $\alpha_{xx} = \alpha_{yy}$ and $\alpha_{xy} = -\alpha_{yx}$, therefore,

$$\int_S j_{pc} \cdot \nabla \phi_{SR} d^2r = \sum_{ij} \int_S \alpha_{ij} \nabla_i \delta T \nabla_j \phi_{SR} d^2r$$

$$= \int_S \alpha_{xx} \nabla \cdot (\delta T \nabla \phi_{SR})d^2r - \int_S \alpha_{xy} \nabla \times (\delta T \nabla \phi_{SR}) d^2r$$

$$= \int_{\partial S} \alpha_{xx} \delta T d\mathbf{n} \cdot \nabla \phi_{SR} - \int_{\partial S} \alpha_{xy} \delta T (d\mathbf{n} \times \nabla \phi_{SR})_z$$

Eq 5.4
Eq 5.4 indicates that photocurrent signal is non-zero only when the edge of the sample is heated. For non-chiral material measured under no magnetic field, the off-diagonal term of the thermoelectric tensor is zero $\alpha_{xy} = 0$. The inner product between the normal vector and the SR electric field is zero on non-contact edges because the SR electric field is parallel to these edges. On the contact, the signal is proportional to the diagonal term of the thermoelectric tensor (panel a and b of Figure 5.1). For chiral material or non-chiral material measured in non-zero magnetic field (panel c and d of Figure 5.1), the off-diagonal term of the thermoelectric tensor is non-zero $\alpha_{xy} \neq 0$. The SR electric field $\nabla \phi_{SR}$ is not parallel to neighboring edges. Both the first and second term in Eq 5.4 becomes non-zero. On non-contact edges the photocurrent is,

$$I_{pc} = \pm (\alpha_{xx}\sigma_{xy} - \alpha_{xy}\sigma_{xx}) \int_{\partial S} \delta T |\nabla \phi| dl$$

Eq 5.5

In the case of inhomogeneity, the conductivity and Seebeck coefficient has spatial dependence, $\alpha(r) = \sigma(r)S(r)$.

$$\int_S j_{pc} \cdot \nabla \phi_{SR} d^2r = \int_{\partial S} \alpha \delta T \nabla \phi_{SR} d\mathbf{n} - \int_S \delta T \nabla \phi_{SR} \cdot \nabla \alpha d^2r$$

Eq 5.6

The second term in Eq 5.6 contributes to a finite signal near the inhomogeneity, such as domain wall.

### 5.3 Photothermal nano-photocurrent of an anisotropic material

For anisotropic material, the diagonal components of thermoelectric tensor are different. An easy way to simplify the calculation is to decompose the thermoelectric tensor,
\[
\alpha = \begin{bmatrix}
\alpha_{xx} & 
\alpha_{xy} \\
\alpha_{xy} & 
\alpha_{yy}
\end{bmatrix}
= \frac{1}{2} \left( \begin{array}{cc}
\alpha_{xx} + \alpha_{xy} & 
1 \\
1 & 
\alpha_{xx} - \alpha_{xy}
\end{array} \right)
\]
Eq 5.7
The SR integral of the isotropic part follows Eq 5.4. The anisotropic part can be further calculated,
\[
\int_S \nabla \delta T \cdot \begin{bmatrix} 1 & -1 \end{bmatrix} \cdot \nabla \phi_{SR} d^2 \mathbf{r}
= \int_{\partial S} \delta T d \mathbf{n} \cdot \begin{bmatrix} 1 & -1 \end{bmatrix} \cdot \nabla \phi_{SR} + \int_S \delta T (\partial^2_x - \partial^2_y) \phi_{SR} d^2 \mathbf{r}
\]
Eq 5.8
The first term is integrated on the border the sample. Together with the first term in Eq 5.4, they combine into \( \int_{\partial S} \delta T d \mathbf{n} \cdot \alpha \cdot \nabla \phi_{SR} \). The second term is non-zero in the interior of the sample, which is the fundamental difference from the isotropic photocurrent.

5.4 Nano-photocurrent generated by shift current

![Figure 5.2: The shift current generated by a metal tip. When a metal tip is illuminated with external light, enhanced electric field is radiated radially near the tip location. Shift current along a and b axis are generated due to the non-zero second order nonlinear tensor components. In either a or b axis, the SR integral functions as a dipolar convolution of the SR electric field.](image)

In low-symmetry materials like TMD, second order nonlinear coefficient is non-zero. In TaIrTe4, experiments at microwave [135] and mi-IR frequencies [136] proves that the shift current is strong across a wide range of spectrum. Due to symmetry limitation, the non-zero second order coefficients are required to have two identical sub-indices, such as \( \sigma_{aacc} \) and \( \sigma_{bbbc} \).

We now consider the shift current with the form,
\[
j_{pc,l} = \sigma_{lich} E_l E_z
\]
Eq 5.9
Where the index \( i \) represents \( a \) or \( b \). Because the tip electric field on the sample surface is mainly confined within the circle centered at the tip location with a radius of several tip-sample distance, the local photocurrent is generated within this small range. The photocurrent generated along either \( a \) or \( b \) axis forms a dipole-like distribution. Inserting this photocurrent into the SR formula, the integral is a dipole-like convolution of the SR field. We can approximately represent the integral within one lobe of the dipole using one single point \( r_i^+ \), where the \( \pm \) sign represents the two different lobes. Therefore, the integral becomes,

\[
\int \mathbf{j}_{pc,i} \cdot \nabla \phi_{SR} \, d^2 \mathbf{r} \propto \sigma_{uc} \left( E_i E_c \nabla_i \phi_{SR} \big|_{r=r_i^+} + E_i E_c \nabla_i \phi_{SR} \big|_{r=r_i^-} \right)
\]

\[
\propto \sigma_{uc} \left( r_i^+ - r_i^- \right) \nabla_i \phi_{SR}
\]

**Eq 5.10**

Here, we assumed that the electric field radiated by the tip is isotropic, \( |E_{in-plane}| = f(r) \). The result of the SR integral along each axis is approximated by the second order derivative to the SR potential.

Like the previous section, we can also decompose the photocurrent along \( a \) and \( b \) axis into isotropic part \( \sigma_{aac} = \sigma_{bcb} \) and anisotropic part \( \sigma_{aac} = -\sigma_{bcb} \). For isotropic part, Eq 5.10 becomes

\[
\int \mathbf{j}_{pc} \cdot \nabla \phi_{SR} \, d^2 \mathbf{r} \propto \nabla^2 \phi_{SR} = 0
\]

**Eq 5.11**

Therefore, like the previous section, the isotropic part doesn’t generate photocurrent signal within in the sample. When the tip is close to the border, part of the electric field radiated by the tip is located outside the sample. As a result, the SR integral is not purely dipolar-like, but acquires a mono-pole component because the opposite lobes of shift current (Figure 5.2) are not
symmetrically generated. Hence, the isotropic part generates a signal close to the edge (roughly within 100 nm to the edge).

For the anisotropic part, Eq 5.10 can be approximately simplified as,

$$\int J \cdot \nabla \phi_{SR} d^2 r \propto (\partial_a^2 - \partial_b^2) \phi_{SR}$$

Eq 5.12

The result resembles the second term in Eq 5.8, which generates non-zero photocurrent signal within the sample. On the boundary, because part of the tip field is located outside the sample, the anisotropic part generates non-zero photocurrent signal near the edge. Like Eq 5.7, the decomposed anisotropic part has a pre-factor $\frac{\sigma_{aa} - \sigma_{bb}}{2}$. Hence, the photocurrent signal generated in the interior of any material is proportional to the degree of anisotropy, but the distribution of the signal depends not on the actual value of $J_{pc,a}$ and $J_{pc,b}$.

Figure 5.3: Nano-photocurrent experiment and simulation of TaIrTe$_4$. a, experimental data. b, simulation with $J_{pc,a} \neq J_{pc,b}$. c, simulation with $J_{pc,a} = J_{pc,b}$. d, the auxiliary field of the device. Here, we assume the in-plane DC conductivity ratio $\frac{\sigma_{0,a}}{\sigma_{0,b}} = 4$. 
In Figure 5.3, we exhibit the nano-photocurrent data and simulation of TaIrTe₄. TaIrTe₄ is a highly anisotropic material with the a- and b-axis DC conductivity ratio of 4. In the experiment (Figure 5.3a), we observed strong signal near the contacts, but the signal is not limited to region close to the contact. The signal around the contact has opposite signs depending on the position relative to neighboring contacts. If the local photocurrent is generated isotropically (Figure 5.3c), the photocurrent is limited to the contact and the signal around one contact is entirely positive or negative. If we adopt the assumption that the photocurrent is generated anisotropically (Figure 5.3b), the simulation matches the experimental data well.

Due to the similar mathematical representation of the nano-photocurrent generated by photothermal and nonlinear effect, the two mechanisms are difficult to differentiate. One potential method is to reshape the tip so that the electric field radiated by the tip is anisotropic. In deriving Eq 5.10, we assumed that the tip field has circular symmetry, hence the tip field is not an important factor in the subsequent derivation. However, if the tip field is anisotropic $\frac{E_a}{E_b} = \frac{\sigma_{abc}}{\sigma_{aac}}$, the anisotropy of the local shift current can be compensated. The resulting SR integration reduces to Eq 5.11. In the case of photothermal effect, the derivation of Eq 5.8 depends not on the shape of the tip field. Therefore, the photocurrent signal due to anisotropic photothermal effect cannot be compensated by the anisotropy of the tip field.

5.5 Photobolometric photocurrent

Photobolometric effect describes the change of the conductivity of the material due to heat under illumination. With the existence of a metallic AFM tip near the sample surface, the lightning-rod effect helps heat up the sample locally. Considering the cooling length of the material, the size of the hot region can be on the order of hundreds of nanometers or even longer. Here we provide a mathematical description of the nano-photobolometric signal,
We consider a metallic device is sourced at fixed voltage drop across the source and drain contacts. When the sample is illuminated, the conductivity is changed slightly. Correspondingly, the potential also needs to be modified. We assume the modification is small,

\[
\begin{align*}
\sigma &= \sigma^{(0)} + \sigma^{(1)} \\
\phi &= \phi^{(0)} + \phi^{(1)}
\end{align*}
\tag{Eq 5.13}
\]

The continuity equation gives,

\[
\nabla \cdot (\sigma \nabla \phi) = \nabla \sigma^{(1)} \nabla \phi^{(0)} + \sigma^{(0)} \nabla^2 \phi^{(1)} = 0
\tag{Eq 5.14}
\]

Consider integration,

\[
\int_S \sigma^{(0)} \phi^{(0)} \nabla^2 \phi^{(1)} = \sigma^{(0)} \int_{\partial S} \phi^{(0)} \nabla \phi^{(1)} \, dn - \sigma^{(0)} \int_S \nabla \phi^{(0)} \cdot \nabla \phi^{(1)} \, dr
\tag{Eq 5.15}
\]

Both terms in Eq 5.15 is contributed by integration on the contacts. On the drain (ground) contact, we have \(\phi^{(1)}_{\text{drain}} = \phi^{(0)}_{\text{drain}} = 0\) and we keep a constant voltage drop, \(\phi^{(1)}_{\text{source}} = 0\).

Therefore, the second term is always zero. The first term is non-zero only near the source contact and the value is \(\phi^{(0)}_{\text{source}} \delta I\). Here, \(\delta I\) is the photocurrent signal we can measure. Therefore,

\[
\phi^{(0)}_{\text{source}} \delta I = \int_S \sigma^{(0)} \phi^{(0)} \nabla^2 \phi^{(1)}
\tag{Eq 5.16}
\]

The RHS of Eq 5.16 can be further calculated based on Eq 5.14,

\[
\int_S \sigma^{(0)} \phi^{(0)} \nabla^2 \phi^{(1)} = - \int_S \phi^{(0)} \nabla \sigma^{(1)} \nabla \phi^{(0)} \, dr
\tag{Eq 5.17}
\]

Assume \(\sigma^{(1)}\) is zero at the contact, we have,

\[
\phi^{(0)}_{\text{source}} \delta I = \int_S \sigma^{(1)} (\nabla \phi^{(0)})^2 \, dr
\tag{Eq 5.18}
\]

Therefore, the nano-photobolometric signal scales with the square of the local current density.

The result is reasonable considering that the LHS and the RHS calculates the extra energy dissipated in the device due to bolometric effect. The same logic applies to the photothermal current Eq 5.3 as well. The external illumination generates a small local photocurrent \(j_{loc}\). The
energy dissipated by the extra current is an integral of the $j_{loc}$ with local auxiliary field. By maintaining the auxiliary voltage on the contacts, we can express the energy dissipated in the device as the measured current signal with the auxiliary voltage. In later chapters, we discuss the photocurrent measurement in hydrodynamic systems. In hydrodynamic systems, the Shockley-Ramo formalism no longer holds accurate. However, we approximate the final result by balancing the total energy dissipated in the device and the energy injected through the contacts.

### 5.6 Photocurrent simulation of hydrodynamic electron system

In hydrodynamic electron systems, the transport equation has an additional diffusion term comparing to the Ohmic transport equation [137],

$$-\sigma \nabla \phi + D_v^2 \nabla^2 J - J = 0$$

Eq 5.19

The diffusion parameter $D_v$ characterizes the mean free path of electrons. In Ohmic systems, the momentum of the electron is relaxed via interaction between electron-phonon interaction or scattering due to disorder. In hydrodynamic system, the momentum relaxing process is less frequent than the momentum exchange via electron-electron interaction. The momentum relaxing mean free path is longer than the channel width of the device. As a result, the momentum of the electron is mainly relaxed on the edge of the device. The diffusion parameter is an average between the channel width $W$ and the electron-electron interaction mean free path $D_v = \frac{W_{ee}}{2}$. The width of the boundary layer and the size of the vortex can be characterized by $D_v$.

For hydrodynamic systems, the SR formalism are not guaranteed to be accurate. In the last line of Eq 5.3, the conductivity in the second term is replaced by the hydrodynamic transport operator,
\[
\int_S \nabla \phi_{SR} \cdot \frac{\sigma}{1 - D^2 v^2} \cdot \nabla \phi d^2 r \neq \int_{\partial S} \nabla \phi \cdot \frac{\sigma}{1 - D^2 v^2} \nabla \phi_{SR} d\mathbf{n} = 0
\]  
\text{Eq 5.20}

The hydrodynamic operator \(\frac{1}{1 - D^2 v^2}\) is not guaranteed to be Hermitian, therefore the left side of Eq 5.20 is not always 0.

Figure 5.4: Nano-photocurrent simulation of an anisotropic sample. The contact of the device is the upper and lower edge of the rectangle sample. a and b, the material has Ohmic transport behavior. The angle between the a-axis and the horizontal axis is 0 degree (panel a) and 5 degrees (panel b). c and d, the material has hydrodynamic transport behavior. The angle between the a-axis and the horizontal axis is 0 degree (panel c) and 5 degrees (panel d). The diffusion parameter \(D_v = 0.35 \mu m\).

To simulate the photocurrent signal of hydrodynamic system, we numerically solved Eq 5.19, assuming a tiny current source mimicking the tip generated local photocurrent. Because the hydrodynamic equation Eq 5.19 is linear, we can decompose the local current source into isotropic and anisotropic source. The simulations are calculated for both kinds and recomposed according to the local photocurrent generated along each axis. In Figure 5.4, we simulated nanophotocurrent of Ohmic and hydrodynamic materials with anisotropic electrodynamic properties. When the angle between the a-axis and the horizontal axis is \(\theta_a = 0^\circ\) (Figure 5.4a and c), the photocurrent signal is limited to regions close to the contacts. In the hydrodynamic case, the signal extends more into the sample because of the diffusion term. When \(\theta_a = 5^\circ\), opposite signals develop on the left and right edges of the sample. The signal now occupies almost the
entire sample in the hydrodynamic case, whereas the signal in the Ohmic case is still limited to the edge of the sample.

To better understand the origin of the nano-photocurrent signal in hydrodynamic systems, we can make approximations to the hydrodynamic equation and apply the Shockley-Ramo formalism. We assume that at small $D_v$, SR formalism still holds. For anisotropic photocurrent, we have

$$I_{pc} \sim \left( \partial_a J_{SR,a} - \partial_b J_{SR,b} \right) = (\partial_x \partial_y) R^T \begin{bmatrix} 1 \\ -1 \end{bmatrix} R \begin{bmatrix} J_{SR,x} \\ J_{SR,y} \end{bmatrix}$$

Eq 5.21

Here, the auxiliary field is simulated using the hydrodynamic equation Eq 5.19. We replaced the auxiliary electric field $\nabla \phi_{SR}$ in Eq 5.12 with the SR current $J_{SR}$ because the current explicitly exhibit the no-slip boundary effect of the wall. In a pipe flow, we can reasonably assume $J_{SR,x} \approx 0$ and $\partial_y J_{SR,y} \approx 0$. Therefore,

$$I_{pc} \sim (\partial_x \partial_y) R^T \begin{bmatrix} 1 \\ -1 \end{bmatrix} R \begin{bmatrix} J_{SR,x} \\ J_{SR,y} \end{bmatrix} = \sin 2\theta_a \cdot \partial_x J_{SR,y} \neq 0$$

Eq 5.22

Only the x derivative of the y component of the SR current remained. The direct effect of a hydrodynamic boundary layer is the non-zero signal in the wide range near the edge of the sample if $\sin 2\theta_a \neq 0$. 

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Figure 5.5: Nano-photocurrent simulation of vortices in viscous fluid. The sample displayed in all panels consist of a vertical channel and a circular chamber on the right side of the channel. The sample is anisotropic, and the a-axis of the material is aligned with the x-axis of the coordinate. a, b, if the material is Ohmic, no vortex is formed in the chamber. We observe non-zero signal inside the chamber. c, d, if the material is hydrodynamic, vortex is formed inside the chamber. The flow direction inside the chamber is opposite compared to the Ohmic scenario. Hence the signal has opposite signs compared to Ohmic scenario.

Nano-photocurrent is also sensitive to the vortex flow pattern of the auxiliary current in an anisotropic material. Eq 5.12 and Eq 5.21 indicate that the photocurrent signal measures the geometrical property of the auxiliary field or current. Boundary layer of hydrodynamic system has a signature flow pattern that the flow speed is damped by the wall of the sample. Vortex is also a characteristic phenomenon of viscous fluid which has a special flow pattern compared to the non-viscous fluid. In Figure 5.5, we simulated the nano-photocurrent in a specially designed sample. The sample has a circular chamber on one side of its main flow channel. Depending on whether the sample is hydrodynamic or not, vortex can form within the chamber. Comparing the
Ohmic and the hydrodynamic scenarios, the flow direction inside the chamber is opposite. Therefore, according to Eq 5.21, we should observe opposite signal in the chamber. The vortex core has zero flux speed, which manifests as a special point in Figure 5.5d.

Figure 5.6: Nano-photobolometric simulation in hydrodynamic and Ohmic regime. The material is homogeneous, and the shape is outlined with the dashed line. The device consists of a rectangular main flow channel and the circular side chamber. The source and drain contacts are on the top and bottom edge. We simulated the nano-photobolometric signal of a portion of the material, including the right half of the main channel and the whole side chamber. In panel a and b, the material is in hydrodynamic regime. The simulation in a and b is identical, we only changed the color scale. In panel c, the material is in Ohmic regime.

We also anticipate seeing hydrodynamic effect induced signal in nano-photobolometric. According to Eq 5.18, the signal directly scales with the square of the local current density. The nano-photobolometric signal should be sensitive to the boundary layer and the vortex core, where both locations have lower or even zero current flow. In Figure 5.6, we simulated on a sample device. The device consists of a rectangular main flow channel and a circular side chamber. The source and drain is located on the top and bottom edge. In panel a and b, the material is in the hydrodynamic regime. Both panels are identical simulation, and the only difference is the color scale. Because of the boundary layer, the signal is lower on the edge of the main flow channel compared to the interior. In the side chamber, the current density is much
lower, contributing to weak signal. The signature of the vortex core is clear. Because there is no current density in the vortex core, the nano-photobolometric signal is low as well. In panel c, the material is in the Ohmic regime. The current density is uniform in the main flow channel, contributing to a uniform signal distribution. Near the side chamber, the current flows into the chamber, creating two current density hot spots on two sides of the opening to the chamber.
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