Mapping the Temperature Dependent Surface States of Bi_2Te_3 Topological Insulator Flakes by Ultra-cryogenic Terahertz Near-Field Nanoscopy

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Abstract—**Topological insulators (TIs) have drawn considerable attention for next generation high sensitivity, high efficiency devices thanks to their unique metallic surface conductivity via their topologically-protected surface states (TSSs). In this study, we utilized near-field nanoimaging and nanoscale spectroscopy in the Terahertz (THz) frequency range to explore the optical response of the surface layer from Bi_2Te_3 TI flakes. In all cases, we observe a higher near-field optical response from the TI flakes compared to a Si substrate. This contrast increases for more surface sensitive probing, which indicates the presence of a conductive surface state. In addition, we have performed temperature dependent THz nanoscopy and demonstrate a significantly enhanced near-field contrast for the TI flake at a sample temperature of 8.9 K compared to the Si substrate. This temperature-dependent behavior provides vital insight into the underlying mechanisms behind the observed conductive surface state in TI materials.**

I. INTRODUCTION

Topological insulators (TIs) have emerged as promising materials for next-generation electronic, thermoelectric and optical applications due to their unique surface states. While the bulk of a TI is insulating or semiconducting, the surface hosts topologically protected, gap-less conductive states known as topological surface states (TSSs) [1-3]. These states are characterized by a Dirac cone, displaying linear dispersion and metallic behavior, as well as by minimal carrier backscattering channels. In addition to TSSs, surface band bending can induce the formation of a two-dimensional electron gas (2DEG), further contributing to the high surface conductivity [4].

The experimental characterization of the topological surface layer is therefore crucial for distinguishing between topological and trivial surface states. However, for conventional characterization techniques, such as far-field optical spectroscopy, it is difficult to isolate the surface response due to the contribution from TI bulk carriers. Fortunately, an advanced technique, the scattering-type scanning near-field optical microscopy (s-SNOM), has been employed to study the surface states and polaritons of in TI materials, providing surface sensitive, high spatial resolution and non-destructive optical microscopic and spectroscopic measurements [5, 6].

In this study, we have employed the THz s-SNOM to study the surface layer of CVD-grown Bi_2Te_3 flakes on a Si substrate. Our THz nanoscopy and nanoscale THz-TDS measurements display a large depth inhomogeneity of the dielectric function within the Bi_2Te_3 flakes, which arises from a conductive surface layer. Moreover, we have performed temperature-dependent measurements, suggesting that the dielectric function of the surface conductive layer is less sensitive to temperature compared to conventional semiconductors, e.g., Si.

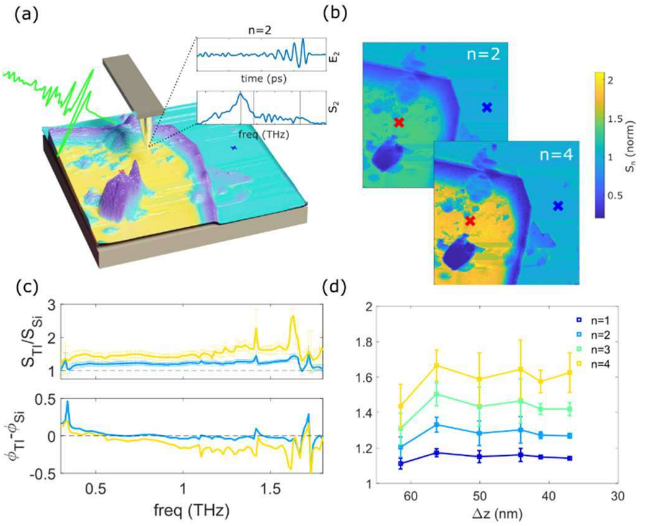

Fig. 1. (a) The schematic diagram of the SNOM measurement on a 35-nm-thick TI flake. Inset: The representative waveform and Fourier transform amplitude spectrum taken from specific position on the TI flake. (b-c) 2D images and spectra of the TI flake (taken at the position indicated by the red cross in (b)) at the 2nd and 4th demodulation order, normalized to the response of the Si substrate (blue cross in (c)). (d) Spectral averaged amplitude contrast at 0.5-1 THz plotted against the tapping amplitude Δz for demodulation orders n=1 to 4.

II. RESULTS

The cryogenic THz-SNOM (Neaspec cryo-neaSCOPE) utilizes two sets of photoconductive antennas as THz emitter and receiver, with femtosecond laser pulses utilized to generate THz radiation and act as gate beam for detection of the scattered THz waveforms (MenloSystems). The collimated THz radiation is focused onto the tip apex (Rocky Mountain Nanotechnology) via a parabolic mirror, then scattered back and collected by the same parabolic mirror. Because of the strong field confinement at the tip apex, the scattered signal carries the optical information which is only related to the small region under the tip for a small tip-sample distance. This information can be isolated via demodulating the detected signal at $n\Omega$, where Ω is the tip tapping frequency. This demodulated signal provides nano-scale surface sensitive optical response amplitude S_n and phase φ_n at the nth demodulation order. An example of a demodulated waveform taken from the TI sample is shown in the inset to Fig. 1a, along with the Fourier-transformed amplitude spectra, peaking at ~0.8 THz.

To study the surface properties of Bi_2Te_3 flakes, firstly, we have performed THz nanoscopy and nanoscale THz-TDS at room temperature to confirm the presence of the highly conductive surface layer. We have then performed THz

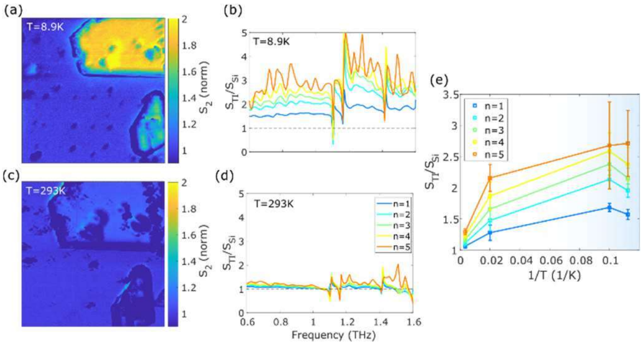

Fig. 2. (a, c) The near-field amplitude images S_2 and (b, d) amplitude spectra of the flake of interest at (a-b) a cryogenic temperature of 8.9 K and (c-d) at room temperature (293 K), normalized to Si substrate response. (e) The amplitude contrast values plotted as a function of the inverse temperature 1/T at different demodulation orders *n*.

nanoscopy and THz-TDS measurements at a corner of the flake of interest, as shown in Fig. 1b-c. The response from the Si substrate was used as reference for AFM and optical signal calibration. The frequency-averaged 2D amplitude images (Fig. 1b) show that the TI flake has a higher scattered amplitude THz signal S_2 and S_4 compared to the underlaying Si substrate. In addition, the higher demodulation order (S_4) shows stronger contrast compared to S_2. The dark edges can potentially be attributed to polaritons [3, 5], however, this is beyond the scope of this study and further verification is required. We focus on the response of the flake area away from the edges to extract the THz-TDS spectra. As shown in Fig. 1c, the TI shows overall a higher THz amplitude signal compared to the Si substrate ($S_{TI}/S_{Si}>1$), with higher contrast for higher demodulation order ($n=4$), correlating with the 2D images. The sharp peaks in the spectra at specific frequencies can be attributed to noise under low THz amplitudes. We attribute this demodulation order dependence to the depth inhomogeneity in the dielectric function due to the conductive surface layer.

To further investigate this demodulation order dependence, we performed nano-tomography on TI flake via altering the tapping amplitude Δz, as lower tapping amplitude provides stronger near-field spatial confinement, resulting in a higher surface sensitivity. The contrast values extracted for the THz frequency range from 0.5 to 1 THz are plotted against Δz for different demodulation order *n* in Fig. 1d. Stronger contrast is observed for higher demodulation order *n* and smaller tapping amplitude Δz, confirming the depth inhomogeneity due to the presence of a highly conductive surface layer. The parameters of this surface conductive layer, including thickness and dielectric function, can be potentially extracted via performing near-field simulations based on the finite dipole model for layered structures, which will be the our future goal [6].

To further confirm that the THz-SNOM signal has a significant contribution from the surface conductive layer, we next performed temperature-dependent THz-TDS s-SNOM measurements on another ~60-nm-thick TI flake. As shown in Fig. 2a-d, the 2D amplitude images and amplitude spectra taken at sample temperature of 8.9 K show a significantly stronger near-field contrast between the TI and the Si substrate, compared to the room temperature (293 K) measurements. The contrast values at different demodulation orders are plotted as a function of inverse temperature in Fig. 2e, showing an increase in contrast as the temperature decreases. We attribute this behavior to the robustness of the dielectric function of the TI surface layer against temperature compared to Si. At cryogenic temperatures, the free carriers in Si freeze out, resulting in a rapid drop of the scattered THz signal of the substrate [7, 8]. We expect that the carriers within the bulk states of the TI flake will also freeze out, reducing the contribution of the bulk to the near-field amplitude of the TI flake. While for the TSSs, i.e., gapless Dirac boundary states, we expect the carriers to be more robust to temperature change and to have a larger contribution to the measured near-field amplitude at low temperature.

III. SUMMARY

In conclusion, we employed cryogenic THz s-SNOM to study the electronic surface state of Bi_2Te_3 flakes. At room temperature, our 2D images and spectra show that the Bi_2Te_3 flake possesses higher near-field amplitude signal compared to a Si substrate. Moreover, the near-field amplitude contrast shows an increase with higher surface sensitivity, confirming the presence of a conductive surface state. At 8.9 K, the TI flake shows significantly stronger contrast against the Si substrate compared to room temperature measurements. We attribute this to the carrier freeze-out in Si substrate and the temperature insensitivity of the TSS carriers. Our results demonstrate that cryogenic THz-SNOM is an ideal tool for the study of TSSs on TI surfaces.

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