

THz Aperture Near-Field Spectroscopy of Dirac Plasmons in Topological Insulator Bi_2Se_3

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Abstract— We map the dispersion relation in ribbon gratings of topological insulator Bi_2Se_3 using both far-field and aperture-type near-field THz spectroscopy methods. The results are consistent with theoretical predictions of coupling between the optical phonon and massless Dirac plasmons.

I. INTRODUCTION

TOPOLOGICAL insulators such as Bi_2Se_3 support two-dimensional Dirac plasmons on their surfaces, which have attracted significant attention for applications such as imaging and sensing owing to their robustness, surface confinement and sensitivity to refractive index changes.

Whilst Dirac plasmons could offer a whole host of useful functionalities, previous spectroscopic investigations of Bi_2Se_3 have revealed very complex optoelectronic behaviour, with contributions to the spectroscopic signature not only from Dirac plasmons, but also plasmons of massive 2DEG and bulk states as well as coupled phonon-plasmon polaritons [1-4]. Disentangling these separate contributions is of great importance for using Bi_2Se_3 in practical devices.

In contrast to far field methods, near-field techniques have the potential to increase the sensitivity and contrast of Bi_2Se_3 plasmon measurements [1,2]. In particular, aperture near-field microscopy enables the probing of evanescent fields on the sample surface at a spatial resolution determined by aperture size [5]. Here, we use both aperture near-field THz time-domain spectroscopy (TDS) and standard far-field THz-TDS spectroscopy to measure thin-film Bi_2Se_3 ribbon gratings. By varying the ribbon width, different gratings are designed to support plasmons of varying frequency. The measured spectra can be used to experimentally map the dispersion relationship, which is compared to theoretical predictions in order to determine the nature of the supported plasmons. The aperture method is a complimentary technique to previously used far and near-field measurements of topological insulators [1-4] and provides interesting insight into the near-field behaviour of Bi_2Se_3 plasmons.

II. METHODS

Ribbon gratings made of 100 nm thick Bi_2Se_3 in stripes of widths, $W = 12 - 40 \mu\text{m}$, were fabricated on sapphire substrates (Fig. 1b). The grating pattern allows for momentum matching and therefore coupling between the incident field and surface plasmon modes. The frequency of the surface plasmon supported is dependent on the ribbon width, W [4].

A schematic of the aperture near-field measurement

technique is shown in Figure 1a. The entire sample area is illuminated by a uniform broadband THz plane wave from an InAs source. A metallic aperture is placed a few microns from the sample, isolating the THz near-field field on the sample surface in a subwavelength-sized region ($10 \mu\text{m}$). The aperture is directly integrated with a photoconductive antenna which detects the THz field that couples through the aperture. In this way, evanescent plasmon fields which decay exponentially with distance from the sample surface (and are therefore lost in the far field) are directly detected by the near field probe. The sample and near-field probe form the centre of a standard time-domain spectroscopy system, where femtosecond IR pulses from a Ti:sapphire laser are used to excite the source and near-field probe. By changing these time delay between pump and probe beams, the THz field is sampled in time allowing the full retrieval of THz field amplitude and phase. In addition, by spatially scanning the near-field probe, the THz field across the entire sample area can be retrieved the spatial resolution of the aperture probe.

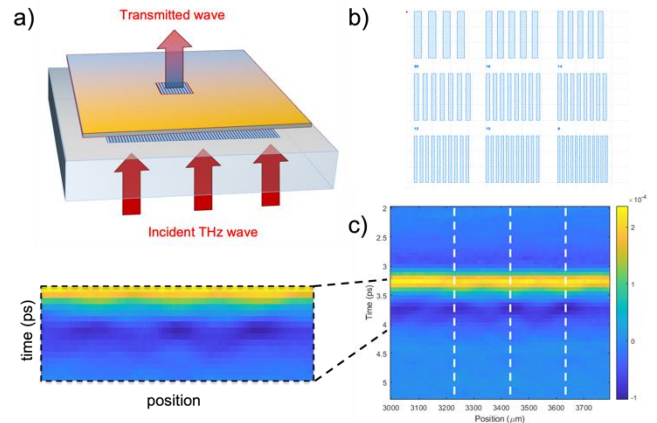


Fig. 1. Investigating Bi_2Se_3 ribbons with aperture near-field microscopy a) Schematic of near-field set-up measuring ribbon array. The ribbons are excited by the incident THz wave and surface plasmon waves are excited. Evanescent fields couple through the aperture to be detected. (b) Diagram of Bi_2Se_3 ribbon array containing ribbons of widths 12 - 40 μm . c) Near-field space-time map showing THz field measured across three ribbon arrays in positions shown by white dashed lines. Each vertical line corresponds to a time-domain waveform at a fixed position on the sample. Dark areas can be seen where the THz field is altered by the ribbon arrays (close-up).

III. RESULTS

Figure 1c shows an initial near-field measurement of the THz field across three ribbon gratings with ribbon widths 12 μm , 14 μm and 16 μm . Each vertical slice of the plot corresponds to the

THz waveform in time, measured at a particular location on the sample. There are three distinct darker regions on the plot which correspond to the positions of the three ribbon gratings, indicating that the THz pulse waveform is modified in these regions.

To investigate further, a Fourier transform of a time-domain trace at the centre of each array was taken. The spectra are normalised to the spectra from the substrate-only and the extinction spectra is calculated as $1 - P/P_{\text{Substrate}}$. Figure 2a shows this information plotted for the three ribbon arrays measured in the near field (lower panel), as well as for a 12 μm periodicity ribbon array measured with far-field THz-TDS (upper panel).

In the far-field, the ribbon array was measured with both TE (dashed line) and TM (solid line) incident THz polarisations. A clear difference can be seen between the TE and TM far-field data. For the TE polarisation, where plasmons cannot be excited by the incident field, a dip in extinction spectra is observed exactly at the frequency of the α phonon [4]. In contrast, for the TM polarisation, two dips are visible in the extinction spectra either side of the phonon line. This splitting of the phonon spectral peak suggests coupling to another nearby resonance. Given that this is only observed in the TM polarisation when surface plasmons can be excited, it is likely that the phonon is strongly coupled to a surface plasmon resonance, forming a plasmon-phonon polariton. The two spectral peaks therefore show the upper and lower polariton branches.

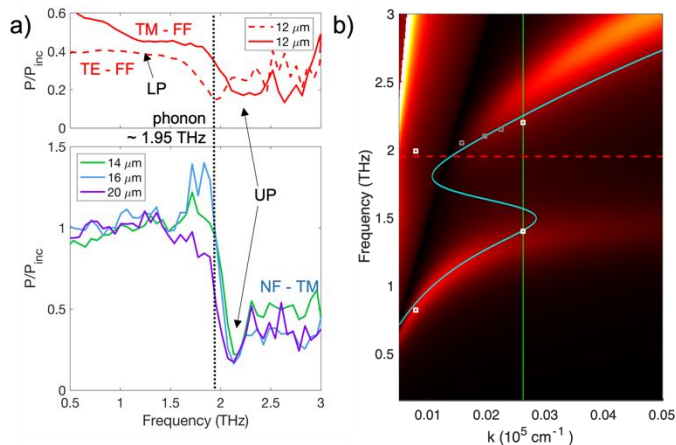


Fig. 2. a) THz spectra of Bi_2Se_3 ribbons: far-field measurements on 12 μm -width Bi_2Se_3 ribbon grating in TE and TM polarizations (red) and near-field measurements on gratings with ribbon widths 14 μm (purple), 16 μm (blue) and 20 μm (green) in the TM polarisation. Inset: schematic diagram of aperture-type near-field measurement of Bi_2Se_3 grating sample. (b) Calculated dispersion relationship of Dirac plasmon polariton with experimentally measured UP and LP branch frequencies overlaid. Measurements are taken from both far-field (white markers) and near-field (grey markers) data.

In the near-field, the upper polariton branch is also clearly visible and shifts in frequency with increasing ribbon width. However, interestingly the lower polariton branch appears less visible as the detector approaches the near field of the sample (5-10 μm away from the surface).

In order to determine the nature of the excited plasmon and confirm its coupling to the phonon, we experimentally map the

polariton branch frequencies with ribbon width. We observe a dependence of the polariton branch frequency on the square root of the ribbon width, \sqrt{W} , which is consistent with calculations of the Dirac plasmon behavior [3,4]. The dispersion relation of the hybridized Dirac plasmon-phonon polaritons is modelled using an analytical Fano model (Fig 2b. turquoise line) [3] and transfer matrix method (Fig 2b background plot) [5], which both consider the coupling between plasmons on the top and bottom surfaces. A good agreement can be seen between the experimental data and model, and we therefore we conclude that the plasmon is formed from light coupling to Dirac carriers in the surface, rather than bulk carriers.

IV. CONCLUSION

In summary, we have investigated Bi_2Se_3 ribbon arrays using near-field aperture and far-field time domain spectroscopy. By experimentally mapping the dependence of the spectra on ribbon width and comparing this to the theoretically calculated dispersion relationship, we conclude that we observe polaritons caused by strong coupling between Dirac plasmons and the Bi_2Se_3 phonon around 2 THz. Aperture near-field microscopy enables broadband measurement of THz plasmons in the 0.5-3 THz range and therefore is a complimentary measurement technique to previously used far and near-field methods to measure topological insulators.

REFERENCES

- [1] E.E.A. Pogna *et al.*, *Nature Communications*, vol.12, 6673, 2021.
- [2] Chen, S., Bylinkin, A., Wang, Z. *et al.*, *Nature Communications* vol. 13, 1374, 2022.
- [3] T. P. Ginley and S. Law, *Advanced Optical Materials*, vol. 6, no. 13, Jul. 2018.
- [4] P. di Pietro *et al.*, *Nature Nanotechnology*, vol. 8, pp. 556–560, 2013.
- [5] O. Mitrofanov *et al.*, *IEEE Transactions on Terahertz Science and Technology*, vol. 6, no. 3, pp. 382–388, 2016.
- [6] Z. Wang *et al.*, *Phys. Rev. Materials*, 4, 115202 (2020)

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