Topological Insulator Chalcogenides for Infrared Dielectric Metamaterials

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Abstract: We show that chalcogenide topological insulators have exceptionally high infrared refractive indices, enabling metamaterials with complex mode structures that could open up pathways to combine dielectric, plasmonic and magnetic metamaterials in a single platform.

The presence of time reversal symmetry-protected, highly conducting Dirac surface states have made topological insulator (TI) crystals an extremely attractive class of materials for electronic, spintronic and, more recently, photonic applications, where coupling of light to the topologically protected surface carriers may lead to propagating surface plasmon polaritons with very little scattering and other exotic phenomena. While there have been extensive studies on broadband electromagnetic properties of chalcogenide TI crystals and resonant nanostructure designs for enhancing interaction with the topological surface carriers in the THz and UV-visible frequencies, there have been hardly any studies on resonant TI structures at intermediate near- and mid-infrared frequencies, where the compositionally tunable refractive index is extremely high and optical conductivity from charge carriers in topological surface states becomes significant [1]. Within the family of chalcogenide crystals, we selected Bi_2Te_3 to demonstrate dielectric metamaterial structures in the technologically important near to mid-infrared frequency window. Bi_2Te_3 has refractive index values between 7 and 8 over the 2-10 μm spectral range, much larger than typical infrared dielectric materials like Si, Ge and PbTe. We exploited this exceptionally high refractive index to demonstrate resonant behaviour in dielectric nanoslit metamaterials, with distinct modes sustained deep into the mid-infrared region. Analysis of the mode structure suggests the existence of complex higher-order modes within the nanoslits, indicating that poloidal surface currents associated with these unconventional structural modes could be engineered to couple light with spin-polarized topological surface state carriers [2].

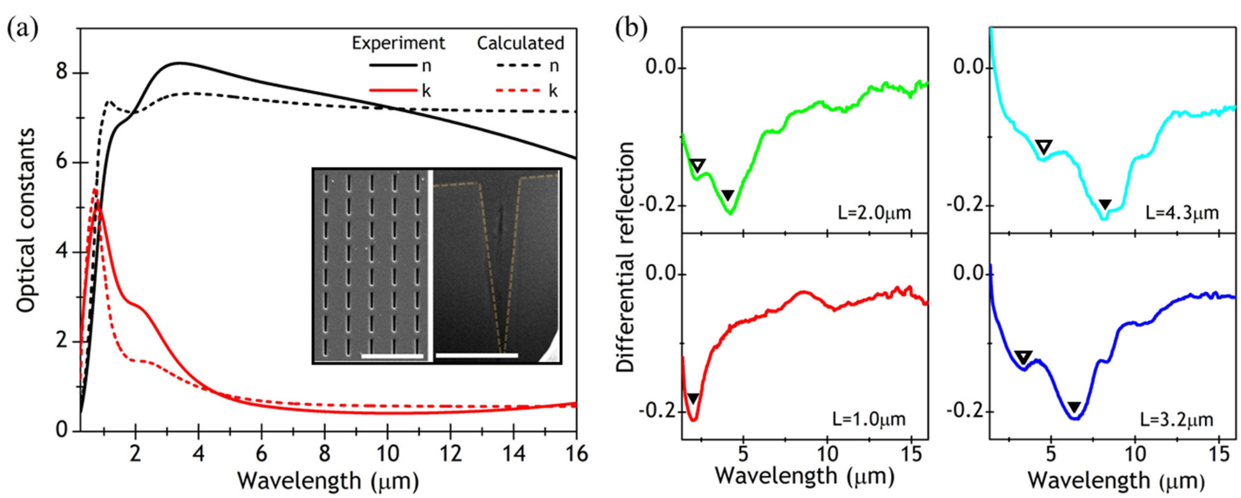

Fig. 1. (a) Dispersion of the experimental and calculated complex optical constants of crystalline Bi_2Te_3. Inset shows SEM images of nanoslit arrays fabricated in Bi_2Te_3, left image corresponds to the shortest slit length while the right one depicts cross-sectional image of the longest slit. Scale bar corresponds to 4 μm and 2 μm for the left and right images, respectively. (b) Representative experimental infrared differential reflection spectra from resonant slit arrays with slit lengths varying from 1.0 to 4.3 μm. Pronounced resonances, both first (▼) and second order (▽) are observed to red-shift with the slit length.

Figure 1a shows the optical constants of Bi_2Te_3 determined experimentally from spectroscopic ellipsometry and infrared reflection measurements compared with those determined using density functional theory (DFT) calculations.

They both feature absorption and negative permittivity at the shorter wavelengths, and strong dielectric behaviour in the infrared. Significantly, the material refractive index exceeds 7 in the 2-10 μm spectral range. The deviation from the calculated dispersion at longer wavelengths is due to a sharp decrease of the refractive index and an increase of the extinction coefficient induced by free bulk carriers from intrinsic doping which are not captured in the DFT calculations.

Optical materials with high refractive index and low losses such as Bi_2Te_3 in the mid-infrared are in great demand for dielectric metamaterials, as they can give rise to strong mode confinement and narrow resonances for small form factor devices. On this premise, we fabricated infrared nanoslit arrays of varying lengths from L = 1.0 μm to L = 4.3 μm via focused ion beam milling on the surface of exfoliated Bi_2Te_3 crystals. We measured their infrared reflection spectra for incident electric field polarized both parallel ($R_{E\parallel}$) and perpendicular ($R_{E\perp}$) to the length of the slits, plotted in differential form, $(R_{E\perp} \quad R_{E\parallel})/R_{E\parallel}$, in Figure 1b for some of the arrays. We observe two distinct resonances, the fundamental resonances (indicated by ▼) and, the second order resonances (indicated by ▽) which appear in the measured spectral region for slits longer than 1.5 μm.

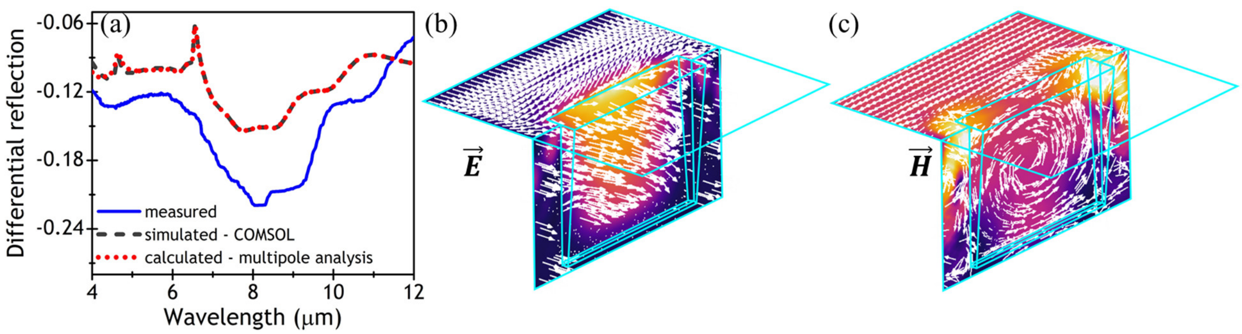

Fig. 2. (a) Experimental (solid blue curve), simulated (dashed grey curve) and calculated (dotted red curve) differential reflection spectra of Bi_2Te_3 metamaterial slit array of length 4.3 μm. Maps of (b) electric ($\bar{\boldsymbol{E}}$) and, (c) magnetic ($\bar{\boldsymbol{H}}$) fields determined by FEM simulations showing the nature of the mode at the fundamental resonance.

To gain a deeper understanding of the nature of the resonances, we simulated the optical response of the slit array using finite element methods. Figure 2a shows the simulated differential reflection spectrum of the longest of the slit arrays (L = 4.3μm) plotted together with the experimental spectrum showing good agreement between the two. Corresponding maps of the resonant electric and magnetic fields for incident electric field polarized perpendicular to the slit length are depicted in Figures 2b and 2c, respectively. While the vortex oscillating behavior of magnetic field in Figure 2c suggests that the mode at 8.55 μm is a toroidal dipole, results of multipole analysis (not shown here) point out that the situation is far more nuanced. The excitation in the metamaterial is not dominated by any particular mode, but instead is a combination of multiple modes such as the electric dipole (order 0), magnetic dipole and electric quadrupole (order 1), toroidal dipole, magnetic dipole and electric quadrupole (order 2) etc. The differential reflection spectrum calculated from multipole analysis is shown as dotted line in Figure 2a and agrees quite well with the simulated and experimental spectra. In the context of TIs these results, in particular the poloidal currents induced on the surface of the TI by the resonant fields, may be used to couple light to the spin-polarized Dirac carriers and gain optical access to the topological surface states. This becomes especially significant in the mid-IR region where chalcogenide crystals feature a combination of high refractive index and larger contribution of topological surface states to the optical conductivity. In addition, the very high refractive index values make the TI chalcogenide crystal family a highly versatile platform for low-loss dielectric metamaterial architectures.

In conclusion, we have shown that chalcogenide crystals are a compelling material platform for photonic applications in the infrared part of the spectrum, with exceptionally high refractive index values larger than 7 in the 2-10 μm range in the case of Bi_2Te_3. This, in turn, aids nanostructured resonances sustained deep into the mid-infrared, as well as the formation of poloidal surface currents that may enable coupling to the spin-polarized topological surface states.

[1] J. Yin, H. N. Krishnamoorthy, G. Adamo, A. M. Dubrovkin, Y. Chong, N. I. Zheludev, and C. Soci, "Plasmonics of topological insulators at optical frequencies," NPG Asia Mater. **9**, e425 (2017).
[2] H. N. S. Krishnamoorthy, G. Adamo, J. Yin, V. Savinov, N. I. Zheludev, and C. Soci, "Infrared dielectric metamaterials from high refractive index chalcogenides," arXiv:1911.09252 [physics.optics] (2019).