Visible Range Plasmons in Topological Insulators

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Abstract: Near-field imaging in the visible reveals the existence of localized and propagating plasmons on the surface of topological insulator. We also report ab-initio band structure calculations explain the origin of plasmonic response in chalcogenide compounds.

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The developments of nanophotonic applications for the visible and ultraviolet parts of the spectrum are hampered by the lack of low-loss plasmonic media. Chalcogenide topological insulators (TIs) were recently identified as plasmonic materials with highly conductive surface states, thus providing a new avenue for manipulating of light at nanoscale. Spectroscopic measurements in visible and terahertz range demonstrated that TI plasmons originate from metal-like surface states induced by strong spin-orbit interaction in the material. At the same time a new mechanism for visible and UV plasmonic response was identified in $Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2}$ (BSTS) single crystals as combination of surface optical conductivity residing in a nanoscale layer of topologically protected surface states and bulk optical conductivity related to the dispersion created by interband transitions in the medium.

In this work we present direct real-space observation of the visible range plasmons in BSTS topological insulators at optical wavelength, where bulk permittivity is positive and therefore plasmonic response can only be attributed to the conductive surface states of the material (Fig. 1).

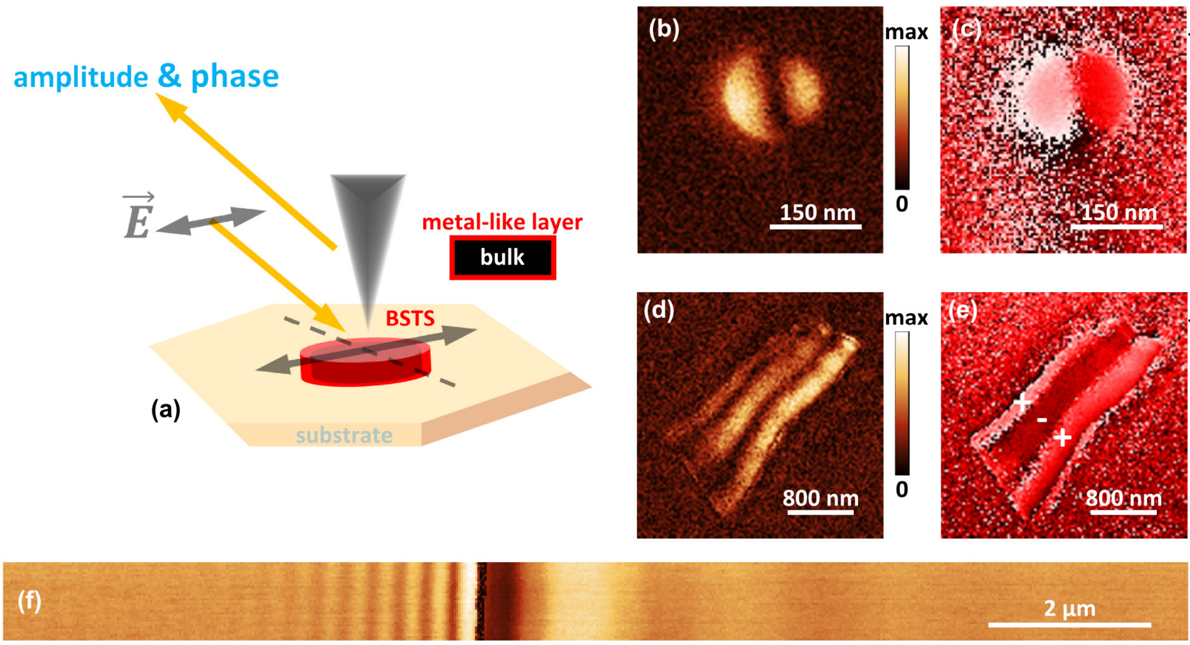

Fig. 1. (a) Sketch of the scattering-type scanning near-field optical microscopy experiment, realized in sp-geometry (s-polarization excitation, p-polarization detection); this configuration is used for localized plasmons mapping. (b,c) Near-field amplitude and phase images of BSTS nano-disk fabricated at $Si\text{-}SiO_2$ substrate by FIB milling. (d,e) Higher-order plasmon mode recorded in BSTS flake. (f) Propagating palsmons at the surface of optically thick BSTS microcrystal. The excitation wavelength is 633 nm.

Scattering-type near-field microscopy of the TI nanostructures (schematic in Fig 1a) at 633 nm wavelength revealed dipole and higher-order plasmon modes with well-defined field amplitude and phase profiles (Fig. 1b-e) supported by conductive surface of the material. This result provides an important and independent verification of plasmonic properties of the topologically protected surface state in this chalcogenide semiconductor. Experimental data are also supported by full wave numerical simulations.

We also observed propagating plasmons in both ultrathin and optically thick films of topological insulator crystals. We numerically calculated the dispersion of BSTS plasmons, and found a good agreement between predicted and extracted from near-field measurements values for the plasmon wavelength.

Table 1. The effective mass, carrier concentration of surface state and three-dimensional plasma frequency of 5 quintuple layers TI slabs.

Surface	**Effective Mass** $m^*(\times\, m_0 kg)$	**Carrier Concentration** n_e **(e/cm^2)**	**Plasma Frequency** $\omega_p\,(eV)$
Bi_2Se_3	0.158	1.09×10^{14}	5.76
Bi_2Se_2Te	0.191	3.44×10^{13}	3.20
Bi_2Se_2Te	0.191	5.08×10^{13}	3.78
Bi_2Te_3	0.172	7.84×10^{13}	4.61
Sb_2Te_3	0.164	6.02×10^{13}	4.21
$BiSbTeSe_2$	0.231	1.33×10^{14}	5.07
$BiSbTe_2Se$	0.330	1.58×10^{14}	4.49

Furthermore, we present a systematic study of the optical and plasmonic properties of BSTS topological insulator compounds by ab-initio density functional theory (DFT) modeling. Knowledge of surface charge distribution in thin TI slabs allows isolating the short-wavelength plasmonic contribution coming from surface electrons, which indicates that a conventional 3D Drude model for a ~2 nm metallic layer appropriately describes the optical response of the topological surface states in the spectral range of our studies.

We extract the effective mass and sheet carrier concentration from the band structure and surface charge distribution to derive the three-dimensional plasma frequency. As shown by consolidated results in Table 1, the quaternary TI materials ($BiSbTeSe_2$ and $BiSbTe_2Se$) have higher surface carrier concentration than the binary and ternary compounds. Nonetheless, irrespectively of the actual carrier concentration, the three-dimensional plasma frequencies inferred from DFT calculations all lay in the UV part of the spectrum, within 3.2 and 6 eV. To quantify the plasmonic contribution to the optical response of TI slabs, a Drude term was then added to the theoretical model of the optical transitions, using the parameters determined from DFT calculations. This analysis gives us confidence that the proposed TI slab model, in which the contributions of the conducting surface layer and the bulk are treated independently, can adequately describe the optical and plasmonic response of this family of TI materials. The results demonstrate the potential of DFT calculations to predict not only the electronic but also the optical properties of topological insulators, which can be used for effectively screening for low-loss TI plasmonic compounds.

Analysis of the calculated dielectric functions of TI films highlights the dependence of the optical bandgap and permittivity on composition, indicating that plasmonic behavior of these compounds in the UV-NIR spectral region arises from the interplay between a bulk negative permittivity and a contribution from metal-like surface states, where the bulk part may be switched off by tuning the excitation wavelength above interband transitions in the medium.

Our work reveals the existence of localized and propagating plasmons on the surface of topological insulators, elucidates the origin of bulk plasma properties and reveals the contribution of surface electrons to the optical response. These results will help designing new hybrid electro-optical and plasmonic devices that exploit plasmonic peculiarities of the band structure and topologically protected surface in the chalcogenide compounds.