Resonant Raman Scattering in the Giant Rashba System BiTeI

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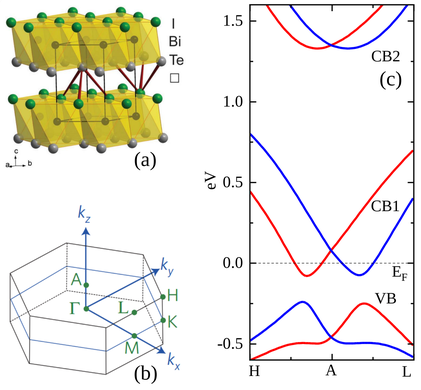
Abstract

BiTeI has recently seen a resurgence of interest following its discovery as a giant Rashba system. A major point of investigation has been to identify and analyze the collective mode excitations of this system. Using polarization resolved Raman spectroscopy, we have identified a previously unseen optical plasmon mode. We show by excitation dependence measurements that the mode undergoes resonant Raman signal enhancement over an incident energy region of less than 0.5eV. On this platform, we present the first Raman scattering study of BiTeI that has investigated its collective mode properties and unambiguously identified a collective mode appearing in the A_1 (charge) symmetry channel.

BiTeI Band Structure



- Bulk electronic bands at the A-point display intrinsic spin-polarization [1].
  - Electron spin is locked in perpendicular to momentum.
- Rashba-split conduction and valence bands results from:
  - Intrinsic electric field gradient
  - Inversion asymmetry
  - Strong spin-orbit coupling
- Results in two Fermi surfaces:
  - Outer: clockwise spin-orientation
  - Inner: switches from clockwise to counter-clockwise spin-orientation as chemical potential passes though “Dirac point”

Fig1: (a) Crystal structure of BiTeI. Trigonal non-centrosymmetric space group P3m1 [2]. Quasi-2D structure [1]: Covalently bonded $(BiTe)^+$ bilayers are ionically bonded to I layer. Layers along the c-axis are held together by weak van der Waals forces. Weak bulk k_z- electronic dispersion

Symmetry Channels

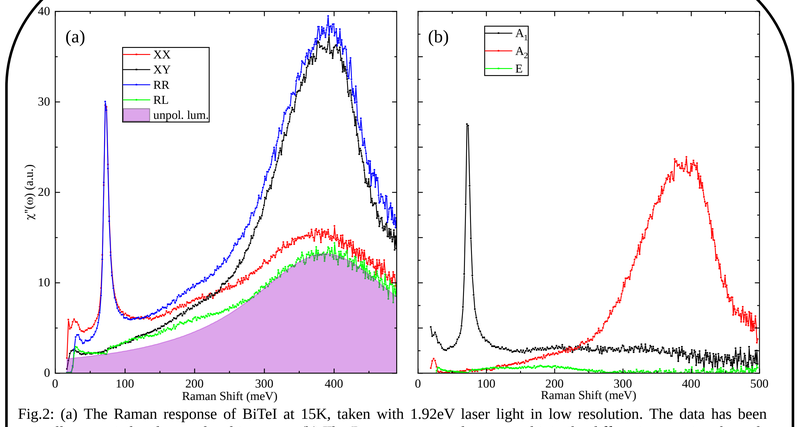


Fig.2: (a) The Raman response of BiTeI at 15K, taken with 1.92eV laser light in low resolution. The data has been optically corrected and normalized to power. (b) The Raman response decomposed into the different symmetry channels to allow for unambiguous identification the symmetry channels of different excitations. There is a non-vibrational collective mode located at 70meV in the A_1 channel.

Scattering Geometry	Irreducible Representations
XX	$A_1 + E$
XY	$A_2 + E$
RR	$A_1 + A_2$
RL	2E

- Polarization resolved Raman scattering allows for unambiguous identification of the symmetry of excitations
  - A_1 : fully symmetric
  - A_2 : transforms as a pseudovector along z-axis
  - E : double degenerate

Experimental Set Up

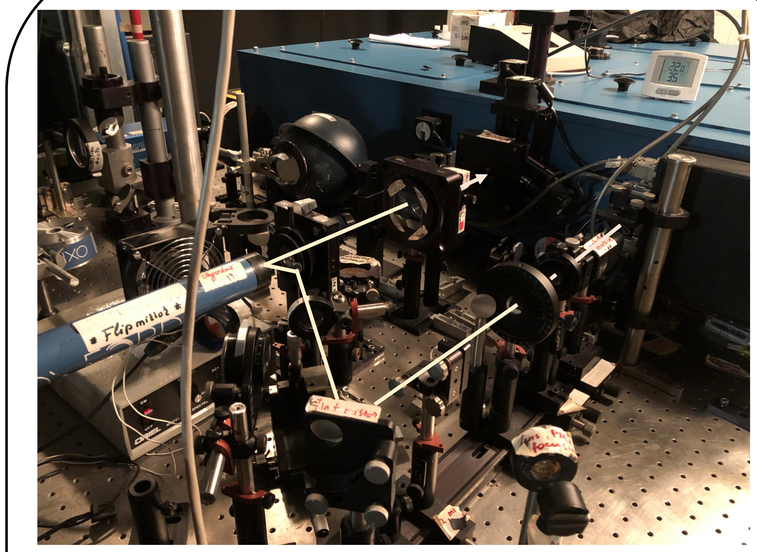
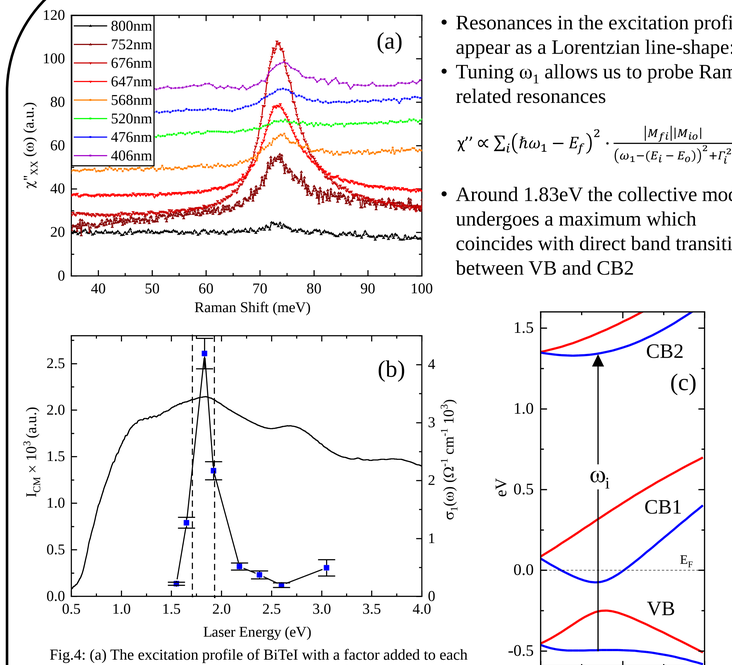


Fig3: Optical table setup for linearly polarized light. The beam path is visualized with arrows drawn into the photograph depicting an angled view of the actual setup.

- Samples cooled and placed in continuum He-flow optical cryostat.
- Kr^+ laser was used for excitation energies ranging from 1.55eV – 3.05eV.
- Triple-stage spectrometer setup was used to remove laser light from spectra.
- Control of the polarization of incident and scattered light entering the spectrometer allows for unambiguous identification of the symmetry of excitations.

Excitation Profile



- Resonances in the excitation profile appear as a Lorentzian line-shape:
- Tuning ω_1 allows us to probe Raman related resonances

$$\chi'' \propto \sum_i \left(\hbar\omega_1 - E_f\right)^2 \cdot \frac{|M_{fi}||M_{io}|}{\left(\omega_1 - (E_i - E_o)\right)^2 + \Gamma_i^2}$$

- Around 1.83eV the collective mode undergoes a maximum which coincides with direct band transitions between VB and CB2

Fig.4: (a) The excitation profile of BiTeI with a factor added to each spectra for clarity. (b) The integrated intensity of the plasmon mode as a function of laser energy. The real part of the optical conductivity, $\sigma_1(\omega)$, is overlaid to show that there is a slight peak in the excitation profile at that point. (c) Band structure of BiTeI along A-> L, which shows a possible resonant transition occurs from VB to CB2

Predicted Modes

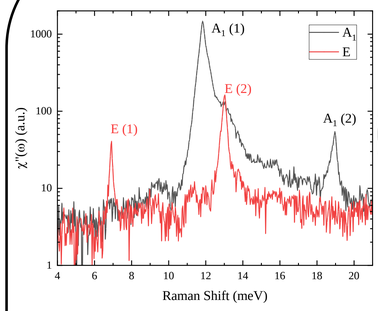


Fig5: Low energy Raman response in the XX scattering geometry, containing all predicted optical phonons.

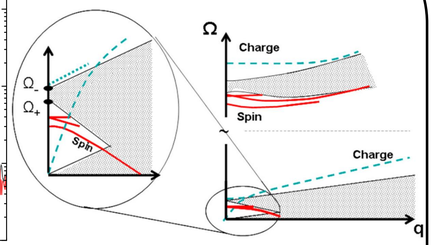


Fig6. Schematic picture of the single-particle continua (shaded region) and collective excitations (lines) with Rashba SOC in a 2D quantum well with the lowest sub-band occupied. (Right, top) Single-particle continua and collective excitations for the 3D case [3].

- Group theoretical calculations predict two A_1 and two E optical phonons
  - Polarization resolved Raman measurements reveal both sets of phonons.
- 2D acoustic mode is unobservable with Raman
- 2D optical mode is asymptotically close to the continuum even at q ~ 0.
- Working theory is that the collective mode is the 3D optical plasmon with finite energy at q ~ 0.

Carrier Concentration Dependence

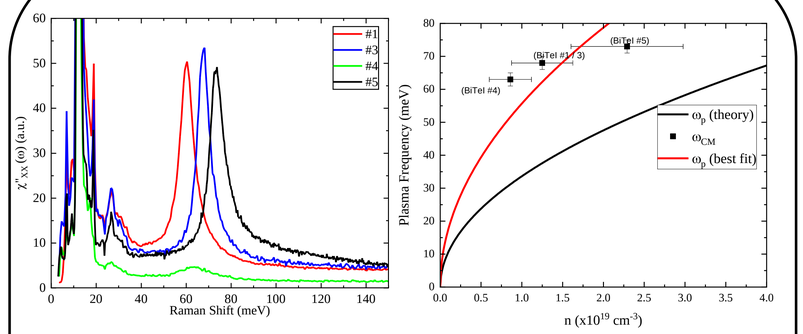


Fig 6: (left) Raman response of BiTeI for samples with different carrier concentrations (right) Energy of the collective mode overlaid with theoretical plasma frequency curve and curve of best fit

- Samples of BiTeI with different carrier concentrations were measured in the XX scattering geometry.
- The carrier concentration, n, was determined by Hall effect measurements.
- The energy of the mode does not follow the typical $\sqrt{n}$ energy dependence.

Conclusion

- Finite energy of the feature at q ~ 0 implies collective mode is 3D optical plasmon
- Unconventional carrier concentration dependence requires further examination.
- The absence of the resonance in optical conductivity measurements is a mystery.
- Experimental dispersion measurements would be greatly beneficial to clarify the nature of the mode.

References

[1] K. Ishizaka et al. Nature Materials, **10,** (2011)
[2] V. Gnezdilov et al. PRB, **89**, 195117 (2014)
[3] S. Maiti et al. PRB, 91, 035106 (2015)
[4] Band structure calculation provided by B. Monserrat

Acknowledgements

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