ABSTRACT OF THE DISSERTATION

Collective excitations in the antisymmetric channel of Raman spectroscopy

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My thesis research focuses on using low temperature polarization resolved Raman spectroscopy to identify optically excited collective modes in strongly correlated electron systems and three-dimensional (3D) topological insulators. In particular, we are interested in collective modes presenting in the pseudovector symmetry channel, such as the A_{2g} representation of D_{4h} group, which is antisymmetric with respect to in-plane reflections. Previously, such excitations are primarily seen in magnetic materials, where the time reversal symmetry is broken. Here, we present two examples where A_{2g} collective modes arises from time reversal symmetry preserved ground states.

In the first example, we study a heavy fermion metal $URu_{2-x}Fe_xSi_2$, which holds long standing interest in the strongly correlated electron community due to an emergent long range order it exhibits at low temperature and $x < 0.1$, known as the "hidden order" (HO). By studying the temperature dependent Raman response function in all allowed symmetry channels for various iron concentrations x, we found evidences of broken symemtries and a possible order parameter. In the low doped HO phase, we observed a sharp in gap mode

with A_{2g} symmetry and small leakage into the A_{1g} symmetry. Our results show strong indication of the local reflection symmetries broken at the uranium site in the HO phase [Kung *et al.*, *Science* **347**, 6228 (2015)]. As we increase x, the energy of an A_{2g} collective mode decreases to almost zero at the phase boundary, and recovers in a wellknown AFM phase. This is a direct evidence that both HO and AFM phases are connected by an A_{2g} type order parameter, that arranges the uranium-5f orbitals into orders breaking either chirality or time reversal symmetry [Kung *et al.*, *Phys. Rev. Lett.* **117**, 227601 (2016)].

In the second example, we study a 3D topological insulator Bi_2Se_3, which has a rhombohedral lattice with the D_{3d} point group symmetry. At the crystal surface characterized by the C_{6v} point group, strong spin orbit coupling and time reversal symmetry conspire to form topologically protect Dirac states with chiral spin texture. While the existence and spin texture of the surface states are mostly understood for Bi_2Se_3 through photoemission and scanning tunneling spectroscopies, very few examples of collective excitations from the Dirac surface states have so far been identified, and therefore interactions between the Dirac particles are yet unclear. In our study, we tune the excitation energy into resonance between two surface Dirac cones about 1.8 eV apart to enhance the signal contributed by surface electrons. We observed a sharp collective mode with energy of about 150 meV appearing below the edge of a gapped continuum, in the pseudovector-like A_2 symmetry of the C_{6v} group. By comparing the data with calculations, we identify this peak as the transverse collective spin mode of surface Dirac fermions: a collective spin-flip excitation from the lower to upper Dirac cone [Kung *et al.*, *Phys. Rev. Lett.* **119**, 136802 (2017)]. This is a new and direct optical measurement of the dynamical response from the Dirac fermions in topological materials. The spin mode survives even at room temperature with slightly decreased intensity, which is likely due to the more available decay channels through interacting with surface phonons [Kung *et al.*, *Phys. Rev. B* **95**, 245406 (2017)]. The robustness of the chiral spin mode suggests potential applications in room temperature magnonics and optoelectronic devices.