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Title:
Quadrupole moments in metallic systems studied by inelastic light scattering

Abstract:
Long-range ordering of local moments is characteristic of solid-state systems. Such ordering gives rise to a wide range of symmetry-broken states, which serve as the basis of electronic devices such as transducer and memory. Although it is desired to control the local moments by current, magnetic dipole moments do not directly coupled to current while electric dipole moments exist mostly in insulating systems. One promising route to further advance the industry of electronics is to explore higher-order moments. Electric quadrupole moments are compatible with metallic states, and this compatibility opens up a diversity of new physics and application. The key concept here is the interaction between local quadrupoles and itinerant electrons. On the one hand, consider the induced interaction between electrons mediated by the exchange of virtual collective fluctuations. For instance, superconducting pairing is induced by the exchange of phonons in the case of conventional s-wave superconductors. Other collective modes, spin fluctuations as an example, could result in high-temperature and heavy-fermion superconductors. Hence quadrupolar fluctuations hold the potential to support new superconducting states. On the other hand, consider the feedback effect of the itinerant electrons in the field of local quadrupoles. The conducting electrons could not only screen the electric fields of quadrupoles, but mediate the coupling between these local moments forming a variety of ordered states [1].

To investigate the quadrupolar fluctuations and the influence on quadrupolar interaction from itinerant electrons, we identify inelastic light scattering as a suitable experimental technique. This method offers the ability to disentangle the long-wavelength excitation spectra into individual symmetry channels. By virtue of this advantage, we can separate out the quadrupolar fluctuations and study the relationship between these fluctuations and other degrees of freedom. We choose two 4f-electron metals, CeB_6 and $YbRu_2Ge_2$, to study quadrupolar ordering at finite wavevector and zero wavevector, respectively. The two compounds not only have high lattice symmetry (cubic for CeB_6 and tetrahedral $YbRu_2Ge_2$), but the 4f ions (Ce^{3+} and Yb^{3+}) have a simple electronic configuration ($4f^1$). Thus, without undesired complexity, these two materials could illustrate in a clear way the physics of quadrupoles. CeB_6 enters an antiferro-quadrupolar (AFQ) state at T_{AFQ}=3.2K [2]. We study the temperature dependence of the low-energy Raman response for all Raman-allowed symmetry channels, and find that the temperature dependence of the static Raman susceptibility in the magnetic channel is consistent with the previously-reported magnetic susceptibility data. Such behavior implies that above T_{AFQ}, the tendency towards AFQ ordering induces ferromagnetic correlations which manifest as long-wavelength magnetic fluctuations [3]. $YbRu_2Ge_2$ develops a ferro-quadrupolar (FQ) phase below T_{FQ}=10K [4]. We show that the static Raman susceptibilities in quadrupolar channels exhibit nearly Curie law behavior, because exchange interactions between local moments are weak due to the compactness of *f* orbitals. It is the relatively strong coupling between the local quadrupole moments and the lattice strain fields in the B_{1g} channel that enhances the vanishingly small Weiss temperature to the temperature of quadrupole phase transition at 10K [5].

The observed close relationship between quadrupolar and magnetic fluctuations in CeB_6 broadens the scope of magneto-electric coupling. Next we will study systems with coexisting quadrupolar and superconducting phases, in which the superconductivity is likely mediated by quadrupolar fluctuations. Although the coupling between local quadrupoles and itinerant electrons in $YbRu_2Ge_2$ is weak, quadrupolar systems with larger orbital extension, such as 5f-electron metals, could give rise to larger coupling and deserve future study. Regarding methodology, our research demonstrates inelastic light scattering as an effective way to probe the electronic quadrupolar fluctuations and in turn capture the interacting strength between quadrupole moments and conducting electrons.

References:
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