ABSTRACT OF THE DISSERTATION

Inelastic light scattering study of correlated electron systems

By MAI YE

Dissertation Director:

Girsh Blumberg

My thesis study focuses on exploring correlated electron systems by means of inelastic light scattering. Specifically, I investigate quadrupolar fluctuations and crystal-field (CF) excitations in $4f$-electron systems, and the nature of unconventional insulating phases in $5d$-electron systems. The light-scattering experimental technique offers both high energy resolution and the ability to disentangle the long-wavelength excitation spectra into individual symmetry channels. Such advantages are particularly useful to study the questions focused by my thesis.

Heavy-fermion metal CeB_6 exhibits an antiferroquadrupolar ordering of T_{2g} (xy type) symmetry below 3.2 K. We perform a comprehensive spectroscopic study of this material, discussing its electronic, magnetic, and phononic excitations [1]. In particular, we find the spectral linewidth of the CF excitations is broadened by Kondo effect, and the tendency towards finite-wavevector quadrupolar ordering induces ferromagnetic correlations which manifest as long-wavelength magnetic fluctuations.

Another heavy-fermion metal, $YbRu_2Ge_2$, enters a ferroquadrupolar phase of B_{1g} (x^2-y^2 type) below 10 K. We study the quadrupolar fluctuations, CF excitations, and lattice

dynamics of this compound [2]. We show that the electronic static Raman susceptibilities in quadrupolar symmetry channels follow nearly Curie-law behavior; the relatively strong coupling to the lattice in the B_{1g} channel enhances the vanishingly small electronic Weiss temperature to the temperature of quadrupole phase transition at 10 K.

Ferroelectric insulator $TbInO_3$ has been proposed to be a spin-liquid candidate. To understand its ground-state property, we study its low-energy CF excitations [3]. We show that the ground state of the Tb^{3+} ions is a non-Kramers doublet. We also demonstrate that the obtained CF level scheme is consistent with specific heat data. In addition, we observe hybrid excitations involving coupled CF and phonon modes, suggesting strong spin-lattice interaction. The complex spin Hamiltonian of $TbInO_3$ renders this material a suitable platform to investigate the effects of non-Kramers doublet ground state on a triangular magnetic lattice.

Paramagnetic insulator $Ba_5CuIr_3O_{12}$ hosts face-sharing Ir octahedra forming quasi-one-dimensional chains. We explore the electronic structure of this system [4]. We show that the insulating mechanism of this iridate cannot be described by the commonly-adopted J_{eff}=1/2 local moment picture. Instead, the shorter Ir-Ir distance in face-sharing geometry leads to strong covalency between neighboring Ir^{4+} ions; this strong covalency results in the formation of molecular orbits as the low-energy electronic degree of freedom. To further illustrate the nature of the insulating state of $Ba_5CuIr_3O_{12}$, we also study the thermodynamic properties of this compound [5]. While the temperature dependence of the magnetic susceptibility and specific heat suggests weak antiferromagnetic correlations, the magnetization does not saturate up 59 T. This phenomenon can be understood in the framework of random singlet state, and we obtain the exchange coupling distribution from the magnetization data.

Zero-gap semiconductor Ta_2NiSe_5 is one promising candidate of excitonic insulator, a coherent electronic phase resulting from the formation of a macroscopic population of excitons. We study its critical excitonic fluctuations and emergent coherence [6]. The

quadrupolar excitonic mode exhibits significant softening close to the phase transition, and its coupling to noncritical lattice modes enhances the transition temperature. On cooling, we observe gradual emergence of coherent superpositions of band states at the gap edge. Moreover, we explore the effect of sulfur doping [7, 8]. We find that the critical excitonic fluctuations diminish with the sulfur doping, and eventually shift to high energies, characteristic of a quantum phase transition. However, a symmetry-breaking transition at finite temperatures is detected at all doping level, exposing a cooperating lattice instability that takes over for large doping level. The study therefore reveals a failed excitonic quantum phase transition, masked by a preemptive structural order.