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Optical Conductivity of Skutterudite Kondo Insulators

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The filled-skutterudite compounds show various physical properties. Among them, we focus on the optical conductivity of the semiconducting materials, since it is a good tool to investigate electronic structures and correlation effects in Kondo insulators. Our aim is to analyze the experimental data on some materials showing the semiconducting behavior at low temperatures and shed light on them from the view point of the Kondo insulators by comparing with the known Kondo insulators like YbB_{12}.

Amongst the filled skutterudite compounds RT_4X_{12} (R=rare earth, T=Fe, Ru, Os and X=P, As, Sb), CeT_4P_{12}'s (rather large gap of 400$\sim$ 1500 K), $CeFe_4As_{12}$ and $CeOs_4Sb_{12}$ ($E_g \sim 100$K) exhibit semiconducting behaviors.

Optical conductivity $\sigma(\omega)$ of $CeOs_4Sb_{12}$ and $CeRu_4Sb_{12}$ are measured by Matsunami, et al. The former showed semiconducting behavior and the latter exhibited the Drude behavior at low frequencies. But the band calculation by Harima resulted in a metal for the former and an opening of a rather large gap of about $E_g = 0.1$ eV at the Fermi energy for the latter. Thus the semiconducting behavior of the former is considered to be due to some magnetic order at $T \sim$2K. The metallic behavior of the latter may be due to some other reasons.

The optical conductivity data for these two materials, however, resemble with each other over the wide frequency range except the gap region in the insulating material. Namely, the steep rise (shoulder) of the spectra at $10 \sim 30$ meV and the existence of the mid-IR peak at $\sim$ 0.1 eV. Interestingly, these features are commonly observed in YbB_{12}, too.

Properties of the Kondo insulators have been often analyzed by using a rather simple-minded periodic Anderson model. The degeneracy of the f states as well as those of the conduction bands are often neglected for simplicity. The $\boldsymbol{k}$-dependence of the hybrydization matrix elements $V_{\mathbf{k}}$ are also neglected. We have recently investigated the latter effect and found that it affects the shape of the optical conductivity spectra. Namely, if one neglects the $\boldsymbol{k}$-dependence of $V_{\mathbf{k}}$, the gap has the same value at all the $\boldsymbol{k}$ point in the Brillouine zone, so that the gap edge diverges as $(E - E_{th})^{-1/2}$ at the threshold in both the density of states $D(E)$ and in $\sigma(\omega)$. These behaviors are never observed in the real materials. The $\boldsymbol{k}$-dependence of $V_{\mathbf{k}}$ modifies them strongly to yield the round-off of the divergence at the threshold and the peak is pushed off therefrom.

Kondo insulator can be understood as a band insulator with strong correlation. The most typical and intensively studied example is YbB_{12}. The conduction band is well represented by the simple tight-binding of 5d ϵ_g orbitals. The energy gap is formed through the hybridization with the 4f Γ_8 states. Thereby, the proper consideration of the orbital degeneracy of both the conduction bands and the 4f states is essential. These treatment reproduces the LDA+U band calculation rather well. Such an approach may be applied also to the skutterudite compounds.

In the skutterudites, the rare-earth ions are confined into the cage of X_{12} clusters. The hybridization of the 4f electron on R with each of X_{12} may be weak but it works rather strongly in total for twelve pnictogens. The crystal field may be close to spherical. From the LDA band calculations we infer that the bands just below the gap are the top most of the bands of X_{12} p-states, and those just above the gap mainly consists of the 4f $J = 5/2$ states (the degeneracy N=6). These may hybrydize with each other. For a starting model, we have constructed tight-binding f-bands on bcc lattice with (ffσ) from A_{2u}, T_{1u} and T_{2u} states, neglecting the spin-orbit interaction. The valence band is approximated by an (ffσ) band which consists of the X_{12} p states in the A_{2u} symmetry hybridizing with 4f A_{2u}. We found that these bands reproduce the LDA calculation rather well. Calculation of the optical conductivity and a comparison with experiments will be discussed.