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Theory for Anomalous Phase Diagram, Thermoelectric Power and Optical Conductivity in $CeOs_4Sb_{12}$ and $CeRu_4Sb_{12}$

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The Ce-skutterudite compounds CeT_4X_{12} show a clear systematic trend. X = P and As compounds are insulators whereas X = Sb compounds are metals.[1] This is due to the increase in atomic size from P to Sb, and hence larger hybridization with the f-electron on Ce in the X_{12} cage. In fact, all X = P compounds are insulators with large energy gaps of the order of 0.1 to 0.03 eV, whereas $CeRu_4Sb_{12}$ is a semimetal at low temperatures according to the transport measurement[2, 3]. According to this trend, $CeOs_4Sb_{12}$ must be a metal, but it is an insulator (perhaps an SDW ordering occurs[1]). Moreover, T_N increases by a magnetic field though the ground state (Γ_7) of f electron does not have a quadrupole moment. In this paper, we investigate theoretically this anomalous phase diagram in $CeOs_4Sb_{12}$, and the transport properties of $CeOs_4Sb_{12}$[2, 3, 4, 5] by using the simplified band model with strong correlation.[6, 7]

The present band model consists of a single wide band (called c-band):

$$\varepsilon^c_{\mathbf{k}} = t_1 \cos(k_x/2)\cos(k_y/2)\cos(k_z/2) + t_2(\cos k_x + \cos k_y + \cos k_z) \tag{1}$$

on the bcc lattice, which represents the top-most p band of X_{12} clusters. For f-electrons, we assume that the the ground state is Γ_7 doublet and use the following simple f-band: $\varepsilon^f_{\mathbf{k}} = E_f + \alpha\varepsilon^c_{\mathbf{k}}$, where α is a small factor representing the width of the f-band dispersion. This may be a reasonable choice since both X_{12} and R occupy the same bcc sites, so that the f-band may have a dispersion similar to c. These c and f bands hybridize with each other. Using these bands, we construct the periodic Anderson Hamiltonian with a constant hybridization V. Actually, of course, the f-dispersion is created through the X_{12} clusters, so that the present treatment is phenomenological, but gives a reasonable band model. For treating the correlation effects on f electrons, we use the dynamical mean-field theory and the iterative perturbation theory.

$CeOs_4Sb_{12}$

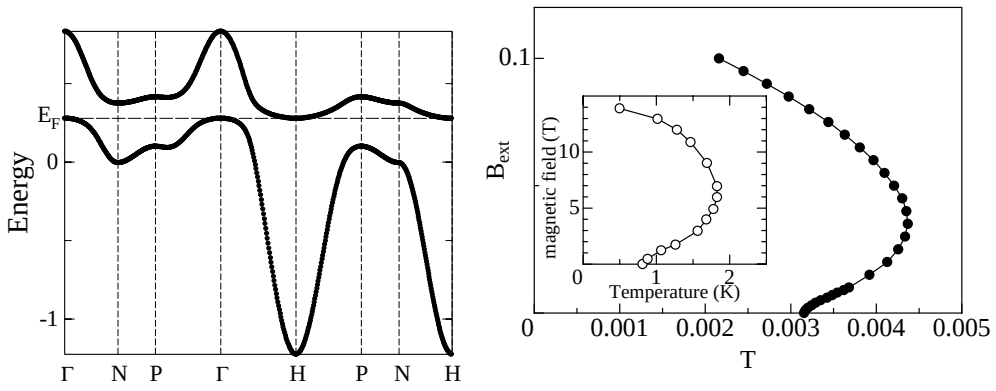

Figure 1: (a) The model band for $CeOs_4Sb_{12}$. (b) The temperature-field phase diagram of $CeOs_4Sb_{12}$ for $U = 0.2$. The inset shows the experimental result [1].

We assumed the band parameters as $t_1 = 1.0$, $t_2 = 0.07$, $\alpha = 0.02$, $E_f = 0.3$, $V = 0.15$ and the Coulom repulsion $U = 0.2$. The resulting band shown in Fig.1(a) has a semi-metallic

character because of the finite α. The top of the lower band at Γ point and the bottom of the upper band at H point slightly touch the Fermi level, and overlap by about 0.001. Although our band structure is quite simplified, it captures the essential part of the low-energy structure of the LDA band calculation[8]. This band has a nesting between the Γ point in the conduction band and H point in the valence band, which yields the SDW state at $T_N = 0.0032$ with $\boldsymbol{Q} = (001)$ at B=0. Then we applied the magnetic field B in z-direction, and calculated the parallel ($\chi_z(\boldsymbol{q})$) and the perpendicular ($\chi_{x,y}(\boldsymbol{q})$) magnetic susceptibility in RPA. We found that $\chi_z(\boldsymbol{q})$ decreases by the applied field but $\chi_x, y(\boldsymbol{q})$ is enhanced. The latter yields the increase of $T_N asisshowninFig.1$[6] and it explains the phase diagram found in the experiment.[1] In reality, there should be a small magnetic anisotropy, so that B might lead to a canting of the moments at B=0.

$\underline{\mathbf{CeRu_4Sb_{12}}}$

We assumed the band parameters as $t_1 = 1.0$, $t_2 = 0.1$, $\alpha = 0.02$, $E_{\rm f} = 0.3$, $V = 0.1$ and $U = 0.3$.[7] The band overlapping is about 0.03.

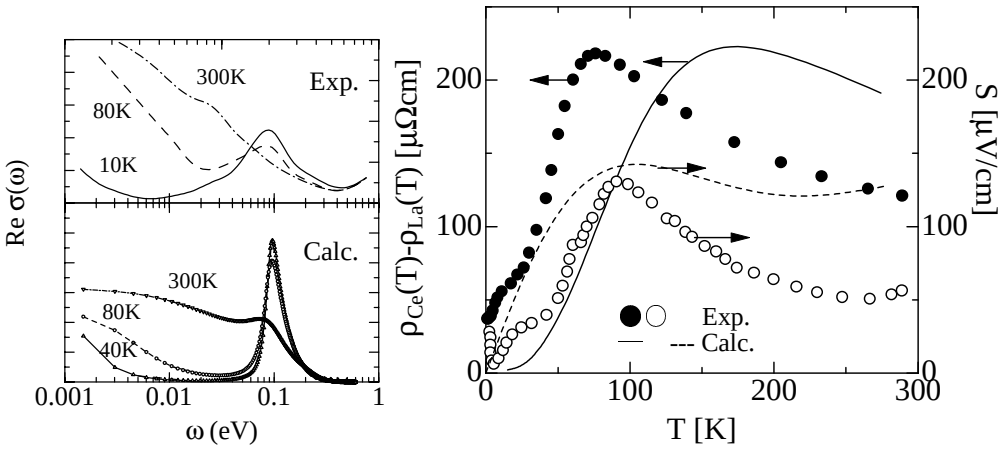

Figure 2: (a) The experimental[5] and theoretical results for the dynamical conductivity. (b)The calculated resistivity (the straight line) and the Seebeck coeffifient (broken line) compared with the experimental values (the filled and the open circles).

The optical conductivity was calculated by the linear response formula although the vertex correction was neglected (Fig.2(a)). By choosing the scale as $t_1 = 7000$ K, the overall structures and tepmerature-dependences are rather well reproduced by the present simple model.

The resistivity and Seebeck coefficient are compared with the experimental values in Figs. 2(b). The calculated peak position agrees with the experimental value in the case of Seebeck coefficient, but it is twice as high in the resistivity. Nevertheless, our results reproduce the global features rather well, except the shoulders at 20 K.

References

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