(8b4)

f-electron states of RT_4X_{12}(R=Ce∼Yb)

Takashi Hotta

Advanced Science Research Center, JAERI, Tokai, Ibaraki 319-1195

In order to clarify magnetic properties of filled skutterudites, we analyze the Anderson model including seven f orbitals hybridized with an a_u conduction band using a numerical technique. For n=2 corresponding to Pr-based filled skutterudites, where n is the local f-electron number, even if the ground state is a Γ_1 singlet, there remain significant magnetic fluctuations from a $\Gamma_4^{(2)}$ triplet state with a small excitation energy. This result can be understood by the fact that f-electron states are distinguished as itinerant Γ_7 and localized Γ_8 in the filled skutterudite structure, since the a_u conduction band with xyz symmetry is described by Γ_7. This picture also explains the complex results for f-electron magnetic susceptibility and entropy for n=1∼13.

In Figs. 1(a) and (b), we show the numerical results of susceptibility χ for the cases of n=1∼13. Note that we always use the same CEF parameters determined for n=2. For n=1, 3, and 5, the ground state is a Γ_{67} quartet in T_h, as confirmed from the residual entropy of log 4. For n=4 and 6, the ground state is a Γ_1 singlet. The excitation energy is large and both magnetic and orbital fluctuations should be rapidly suppressed with decreasing temperature. Thus, $T\chi$ immediately becomes zero at low temperatures for n=4 and 6. For $n \geq 7$, the absolute values of χ are much larger than those for $n<7$, since the total angular momentum J becomes large for $n \geq 7$ owing to Hund's rule coupling. Typically, at half-filling, total spin $S(=J)$ is equal to 7/2, and the Curie constant for an isolated ion is as large as $21\mu_\mathrm{B}^2/k_\mathrm{B}$. Over a broad temperature region, this value has been observed for n=7, indicating that the S=7/2 spin survives at relatively low temperatures. For n=8 and 12, the ground state is a Γ_1 singlet and the magnetic excited state energy is now large, in sharp contrast with the case of n=2. Thus, the susceptibility rapidly goes to zero. For n=9, 11, and 13, the local ground state is a Γ_5 Kramers doublet in T_h, which is the mixture of Γ_6 and Γ_7 of O_h. Since Γ_6 state does not hybridize with the a_u conduction band, the magnetic moment from Γ_6 still persists even in the low-temperature region. In fact, we observe a residual entropy of log 2 in these cases. For n=10, the local ground state is a Γ_4 triplet, but the local triplet seems to remain at low temperatures. This is easily understood, if we recall that Γ_4 triplets of T_h are given by mixtures of Γ_4 and Γ_5 of O_h. The Γ_5 triplet still persists even after hybridization, since it is composed of a couple of Γ_8 electrons and a Γ_8 electron does not hybridize with a Γ_7 conduction electron.

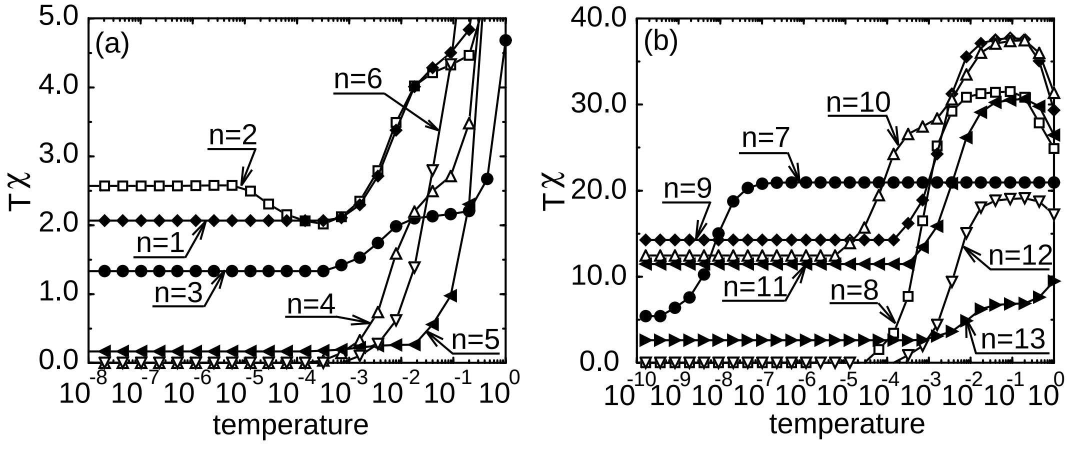

Figure 1: Magnetic susceptibility of f electron for (a) n=1∼6 and (b) n=7∼13.