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Single-site effects of filled skutterudite compounds studied by using insulator hosts

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The characteristics of local f-electronic states in filled skutterudite compounds manifest themselves in some interesting phenomena such as the frequently observed multipole orderings, field-insensitive heavy-electron states, and a metal-insulator transition. In particular, it would be important to study the basic crystalline-electric-field (CEF) effects and their modulation due to the hybridization effects in a single icosahedron for deepening our understanding of the physics evolved in notable periodic systems. For this purpose, we notice that the insulators CeM_4P_{12} (M = Fe, Ru and Os) may serve as interesting hosts for the substitution of the f^n ($n \geq 2$) ions, i.e. Pr, Nd, Sm, etc., for Ce, where Kondo-type local quantum fluctuations due to the presence of Fermi surface could be completely eliminated. In the present contribution, we report the results of specific heat ($0.4\,\mathrm{K} \leq T \leq 40\,\mathrm{K}$; $\mathrm{B} \leq 12\,\mathrm{T}$) and magnetization ($2\,\mathrm{K} \leq T \leq 300\,\mathrm{K}$; $\mathrm{B} \leq 5.5\,\mathrm{T}$) measurements performed for polycrystalline samples of $Ce_{1-x}Pr_xM_4P_{12}$ (M = Fe, Ru; $x \leq 0.16$). For both the alloys, we observed that the magnetic susceptibility of doped Pr ions shows no tendency of saturation with decreasing temperature down to 2 K. The f-electronic contribution to the specific heat ($\Delta C/T$) also exhibits a significant upturn below ~ 2 K (Fig. 1). As magnetic field is applied, this upturn turns to a simple Schottky anomaly, from which we estimate the entropy of the local f states for the single Pr ion to be about $R\ln 4$ below ~ 20 K. On the basis of the detailed analyses on the field variations in $\Delta C/T$, we suggest that the low-energy CEF states of these alloys are both described by a triplet(0)-singlet(~ 20 K) level scheme involving a slight splitting of the lowest triplet, which is probably caused by the Jahn-Teller effects.

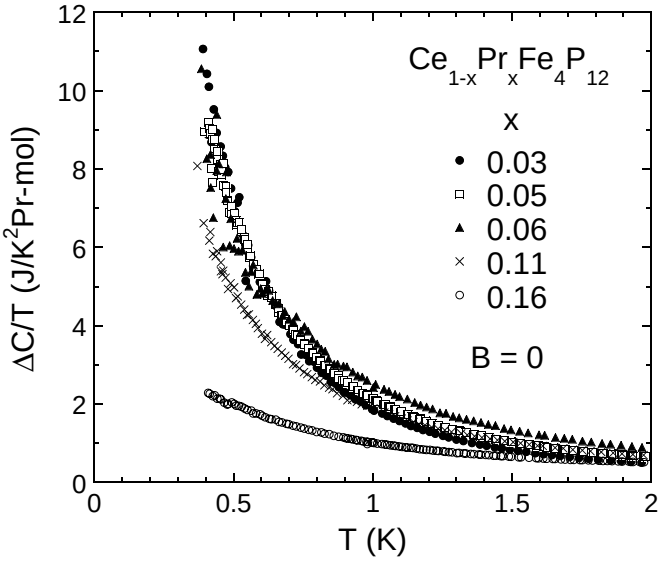

Figure 1: Low-temperature specific heat versus temperature for $Ce_{1-x}Pr_xFe_4P_{12}$ ($x \leq 0.16$).