Magnetic phase transitions and anomalous transport properties in Ca-doped Eu hexaborides

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EuB₆ compound has been known as a ferromagnetic compound, showing two phase-transitions at low temperatures (T₁ = 15 K, and T₂ = 12 K)[1]. A small doping of Ca for Eu sites in Eu₀.₈Ca₀.₂B₆ induced a dramatic variation of carrier density in terms of temperature and applied magnetic fields in spite of an iso-electronic incorporation of Ca [2]. Hence, it will be interesting to study the systematic Ca doping effect on the series compounds of Eu₁₋ₓCaₓB₆. Here we present the temperature- and applied magnetic field-dependent magnetization M(T,H), electronic transport ρ(T,H), and Hall resistivity ρₓᵧ(T,H) for series compounds of Eu₁₋ₓCaₓB₆ (x = 0.0, 0.2, 0.4, 0.6, and 0.9). It was found that the electronic and magnetic properties of the compounds are strongly correlated through the charge of carrier density and its mobility. The small Ca doping of Eu₀.₈Ca₀.₂B₆ induces the shift of ferromagnetic transition temperature Tₖ from 12 K for EuB₆ to 5.5 K. The sharp drop of ρ(T) was occurred near the transition temperature for the ferromagnetic compounds of Eu₁₋ₓCaₓB₆ (x = 0.0, and 0.2). Interestingly, the antiferromagnetic transition was observed for the compounds of Eu₁₋ₓCaₓB₆ (x = 0.4, and 0.6), rather than ferromagnetic ordering. The AF ordering temperatures are T_N = 4.5 K for Eu₀.₆Ca₀.₄B₆ and T_N = 3 K for Eu₀.₄Ca₀.₆B₆. For Eu₀.₁Ca₀.₉B₆, the paramagnetic state persists down to T = 2 K. The two AF materials (x = 0.4, and 0.6) showed an exotic temperature and field dependence of ρ(T) at low temperatures (T ≤ 20 K). From the measurements of Hall resistivity, the anomalous increase of ρ(T) with increasing magnetic fields (H ≥ 2 T) can be understood as the decrease of Hall mobility |μH| and the rapid increase of carrier density nₑₑ(T,H) with applied magnetic fields. The unusual change of magnetic ground states from ferromagnetic to AF, the simultaneous exotic change of electric transport and their correlation will be discussed in detail.