

Onset of ferromagnetism in the band paramagnet LuCo_2 upon the replacement of cobalt by aluminum

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As the aluminum concentration in the $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ is increased, there are transitions from band paramagnetism to weak ferromagnetism and then to strong ferromagnetism. The evolution of the magnetic properties of this system can be explained in a qualitative way in a band model in terms of an increase in the state density at the Fermi level with increasing x .

The magnetic properties of d magnetic materials depend on the particular features of the energy band structure, primarily the shape of the curve of the electron state density versus the energy, $N(\epsilon)$, and the extent to which the band is filled by electrons (the position of the Fermi level, ϵ_F).¹ These characteristics are very sensitive to various external agents, particularly if the energy dependence $N(\epsilon)$ is sharp near ϵ_F . In particular, there may be a situation in which a partial replacement of the “magnetic” transition d metal by a nontransition metal converts the band paramagnet into a ferromagnetic state. Precisely this situation may in principle occur in mixed intermetallic compounds based on cubic Laves phases of LuCo_2 . In the intermetallic compound LuCo_2 , the d band is formed by hybridized $3d$ electrons of cobalt and $5d$ electrons of lutetium, and the Fermi level of the d band lies on a descending part of the $N(\epsilon)$ curve, i.e., we have $dN(\epsilon)/d\epsilon|_{\epsilon_F} < 0$. In pure LuCo_2 , Stoner's criterion for band ferromagnetism, $N(\epsilon_F)I \geq 1$ (I is the exchange integral), is not satisfied, and this compound is an exchange-strengthened paramagnet.^{2,3} If the d band in LuCo_2 is emptied,

and cobalt is replaced by an element with an empty d band, e.g., aluminum, with the electronic configuration $3s^23p^1$, the Fermi level ϵ_F will shift down the energy scale, and the state density $N(\epsilon_F)$ will increase [because of the relation $dN(\epsilon)/d\epsilon|_{\epsilon_F} < 0$]. If the increase in $N(\epsilon_F)$ overpowers the decrease in the exchange interaction because of the decrease in the number of d electrons, the exchange-strengthening factor $N(\epsilon_F)$ will increase upon the replacement, and Stoner's criterion may become satisfied in some concentration interval: The compounds will become ferromagnetic. In the mixed compounds $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$, there is thus the possibility of a seemingly paradoxical situation: The replacement of "magnetic" cobalt by "nonmagnetic" aluminum strengthens the magnetism and leads to a transition from a paramagnetic state to a ferromagnetic state.

To test these ideas, we have carried out measurements of the magnetization in pulsed fields up to 320 kOe, and we have also measured the lattice constants of the $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ system by an x-ray method over the temperature range 4.2–300 K. The polycrystalline samples are smelted by an induction method under quasilevitation conditions and then subjected to a homogenizing annealing. Measurements by x-ray and metallographic methods showed that the compositions with $x \leq 0.208$ are of a single phase and have a cubic crystal structure of the C15 type (the sample with higher aluminum concentration have the hexagonal C14 structure).

The measurements showed that the replacement of cobalt by aluminum in the system $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ leads to an increase in the paramagnetic susceptibility, and at $x \geq 0.078$ a ferromagnetic order appears. Figure 1 shows the x - T phase diagram of the $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ system, along with experimental curves of the field dependence of the magnetization at 4.2 K and of the temperature dependence of the lattice constant for some typical compositions. In terms of the nature of the magnetic behavior, the concentration interval $0 \leq x \leq 0.208$ can be partitioned into three regions.

1) Compositions with $0 \leq x \leq 0.07$. These compositions are paramagnetic. The temperature dependence of the lattice constant is described by the Debye law. The susceptibility increases with increasing aluminum concentration, and at $x = 0.06$ and $x = 0.07$ we see metamagnetic transitions to a ferromagnetic state in the external field. The field at which the transition occurs decreases with increasing x .

2) Compositions with $0.078 \leq x \leq 0.09$. A ferromagnetic order arises here. The magnetic moment is $(0.2-0.7)\mu_B$ per physical unit and increases with the aluminum concentration. The phase transition from the ferromagnetic state to the paramagnetic state is diffuse, and it is difficult to accurately determine the temperature of the magnetic ordering. The spontaneous magnetization of these compositions depends on their magnetic history. Deviations of the temperature dependence from Debye law are slight (below the sensitivity of the x-ray method). In an external field we observe a metamagnetic transition to a state with a larger magnetic moment.

3) Compositions with $0.095 \leq x \leq 0.208$. These compositions are also ferromagnetic, but their magnetic moment is substantially greater than $(0.7-1.2)\mu_B$ per physical unit. The transition from the paramagnetic state to the ferromagnetic state is accompanied by a magnetic-exchange anomaly [a deviation of $a(T)$ from the Debye law]. In the interval $0.095 \leq x \leq 0.11$ this transition is a first-order phase transition, as can be

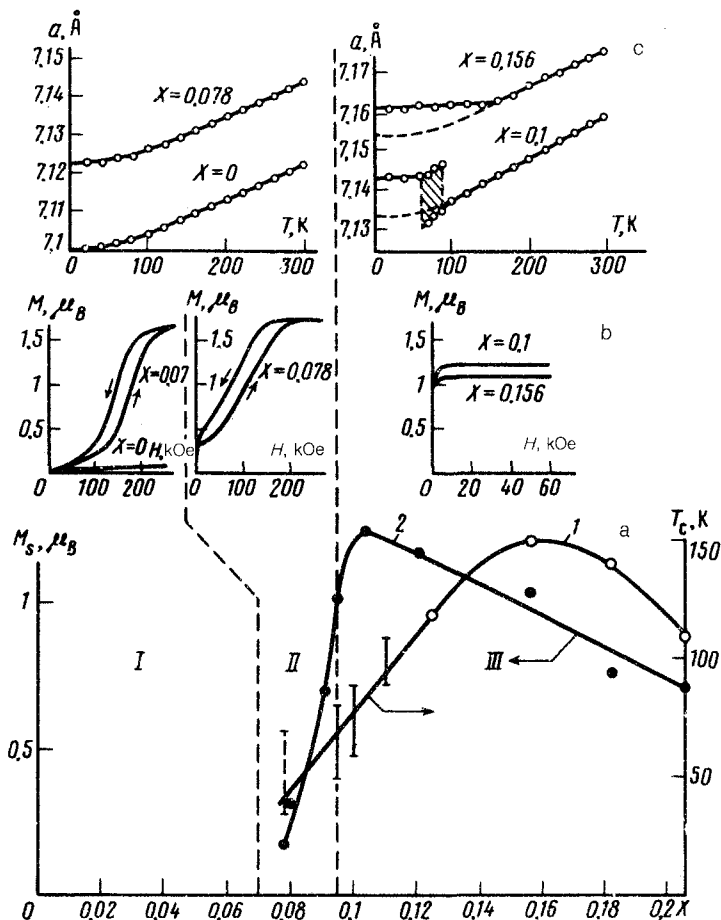


FIG. 1. Magnetic characteristics of the intermetallic compounds $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ ($x \leq 0.208$). a: Magnetic x - T phase diagram. I—Paramagnetic compositions; II—weakly ferromagnetic compositions; III—strongly ferromagnetic compositions. 1) Concentration dependence of the magnetic ordering temperature (the open circles represent second-order phase transitions; the vertical solid lines show regions of the coexistence of the different magnetic phases upon first-order transitions; and the vertical dashed lines show the region in which the magnetic order disappears); 2) concentration dependence of the spontaneous magnetization at 4.2 K (filled circles). b: Magnetization curves for typical compositions at 4.2 K. c: Temperature dependence of the lattice constant of typical compositions. The dashed lines show the Debye law.

seen clearly from the $a(T)$ curves. In the temperature regions ~ 20 – 30 K, near the magnetic-ordering temperature T_c , the paramagnetic and ferromagnetic phases coexist; as the temperature is varied, the relative fractions of these phases change in the volume of the sample.

These results confirm the suggestion that the Fermi level in LuCo_2 lies on a steeply descending part of the $N(\epsilon)$ curve. We wish to call attention to the circumstance that an external magnetic field induces a strongly ferromagnetic state in the

paramagnetic and weakly ferromagnetic phases. This behavior of a band system, which was first studied theoretically in Ref. 6, is evidence that the curve of $N(\epsilon)$ has a positive curvature near ϵ_F ($d^2N(\epsilon)/d\epsilon^2|_{\epsilon_F} > 0$).

At low aluminum concentrations, the behavior of the $\text{Lu}(\text{Co}_{1-x}\text{Al}_x)_2$ system is qualitatively the same as that of the $\text{Y}(\text{Co}_{1-x}\text{Al}_x)_2$ system,⁴ in which the d band is formed by a hybridization of cobalt $3d$ electrons and yttrium $4d$ electrons. Although a metamagnetism of d electrons is observed with increasing aluminum concentration in both systems, a strongly ferromagnetic phase does not form in $\text{Y}(\text{Co}_{1-x}\text{Al}_x)_2$ anywhere below the limiting concentration $x_{\text{lim}} = 0.2$. According to the information available^{3,5} on the energy structures of YCo_2 and LuCo_2 , these differences cannot be explained in terms of a rigid-band model. Effects stemming from an influence of changes in the interatomic distances on the band structure appear to become important in compositions with large degrees of replacement.

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