f-Electron antiscreening in the heavy-fermion system consisting of $CeAl_3$ and a magnetic impurity

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A study of the neutron spectra and specific heat of $Ce_{1-x}Nd_xAl_3$ has established that the large width of the transitions between the crystal-electric-field levels of *f* electrons in $CeAl_3$ is determined by Kondo scattering of conduction electrons. An antiscreening of the crystal electric field of $CeAl_3$ has been observed upon a strengthening of the exchange interaction of *f* and *s* electrons.

In research on heavy-fermion systems it is usually assumed that the coupling of the localized 4*f* electrons with conduction electrons (the *s-f* coupling) and their coupling with the crystal electric field are mutually independent.[1] The experimental widths of transitions between the crystal-electric-field levels of 4*f* electrons in heavy-fermion systems,[2] however, are large (several millielectron volts), comparable to the splitting of the ground multiplet in the crystal electric field. This circumstance may reflect changes caused in the electron subsystem by Kondo scattering and the formation of heavy fermions.

An experimental study of this question has been carried out on the heavy-fermion system $CeAl_3$ [$\gamma \approx 1.6$ J/(mole·K^2)], to which magnetic neodymium ions were added in order to alter the *s-f* coupling. In the crystal electric field of $NdAl_3$ (Ref. 3) the neodymium ions have a nonvanishing projection of the total angular momentum J_z, as

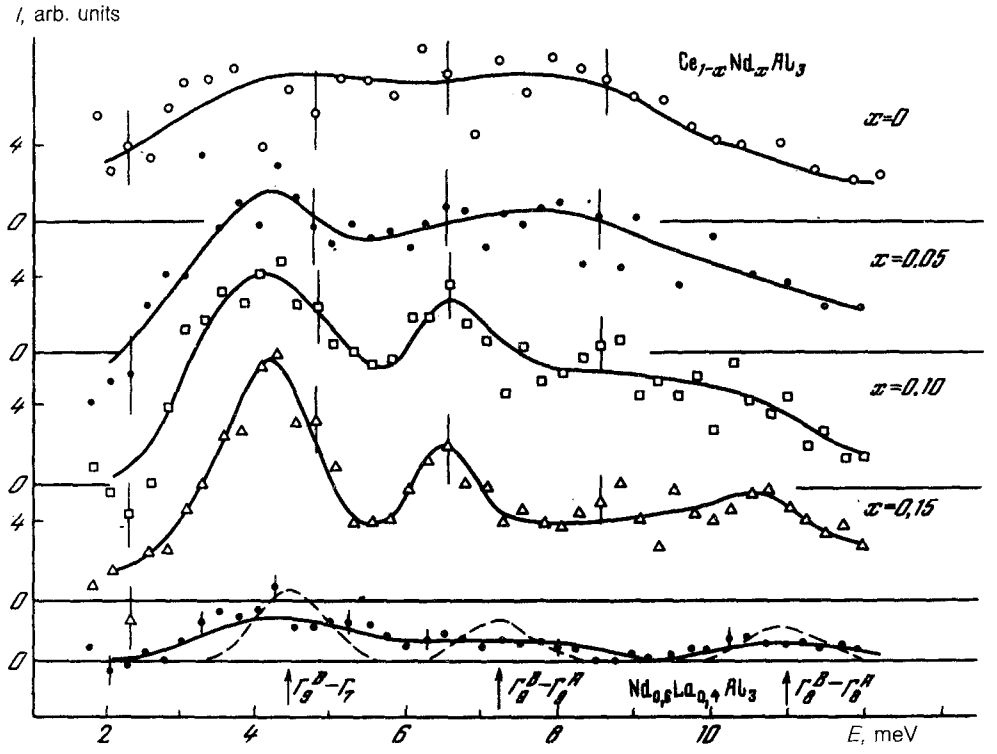

FIG. 1. Spectra of the inelastic magnetic neutron scattering at $T = 4.5$ K for $Ce_{1-x}Nd_xAl_3$ and $Nd_{0.6}La_{0.4}Al_3$. The dashed line shows the narrowing of the Nd^{3+} peaks which would be expected because of the decrease in the extent of disorder of the nearest neighborhood upon a decrease of the Nd concentration.

$T \to 0$ K (the magnetic moment of the ground state is $\sim 1\ \mu_B$). A known procedure[2] was used to synthesize several essentially single-phase samples of $Ce_{1-x}Nd_xAl_3$ ($x = 0$, 0.05, 0.10, and 0.15) and also an $Nd_{0.6}La_{0.4}Al_3$ sample, with lattice constants close to the values for $x = 0.10$. The inelastic magnetic neutron scattering by these compounds was measured in the IR-8 reactor (Kurchatov Institute of Atomic Energy) by the $Q = \text{const}$ method over the temperature interval $T = 4.5$–40 K. We also measured the specific heat of $Ce_{1-x}Nd_xAl_3$ in magnetic fields up to 8 T at $T = 2$–20 K.

Figure 1 shows the resulting spectra of the inelastic magnetic neutron scattering for $Ce_{1-x}Nd_xAl_3$ and of the magnetic scattering for $Nd_{0.6}La_{0.4}Al_3$, referred to the scattering by a sample with a 10% neodymium concentration (the dependence of the neutron absorption on the neodymium concentration was taken into account). The $CeAl_3$ spectrum has two rounded peaks with energies ≈ 4.5 and 8 meV, in agreement with Ref. 2. The neodymium impurity causes pronounced changes in the spectrum, consisting of the appearance, against the background of the $CeAl_3$ spectrum, of two narrow peaks, with energies lower than those in $CeAl_3$. The intensity of these peaks

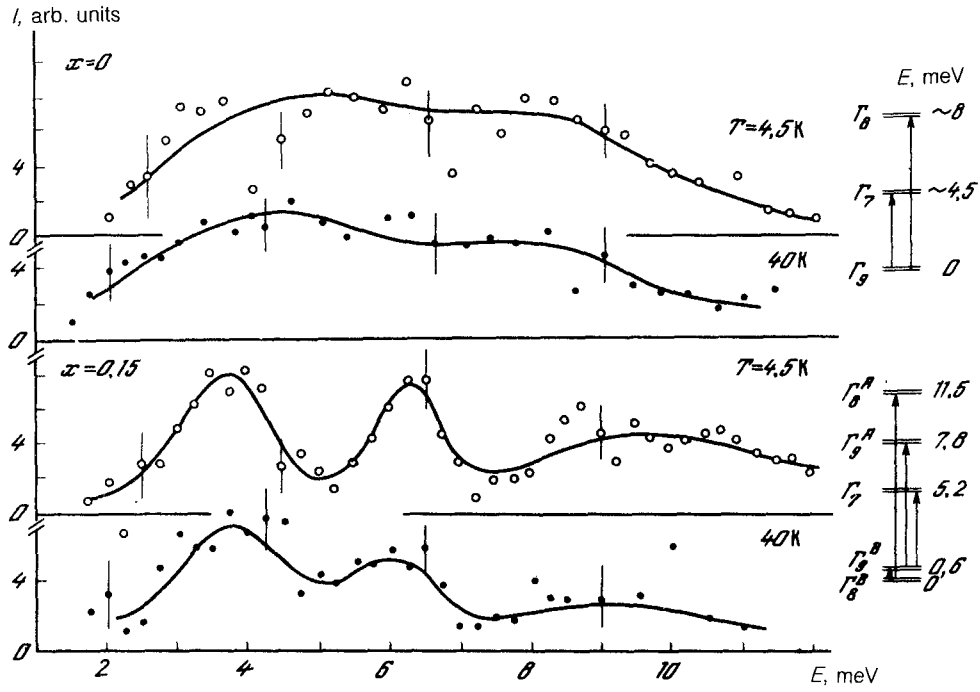

FIG. 2. Temperature dependence of the spectra of the inelastic magnetic scattering of neutrons for $Ce_{1-x}Nd_xAl_3$ ($x = 0$ and 0.15). The scattering by Nd has been eliminated. Shown at the right are crystal-electric-field levels of $4f$ electrons, for Ce^{3+} in $CeAl_3$ (Ref. 2) at the top and for Nd^{3+} in $Nd_{0.6}La_{0.4}Al_3$ at the bottom (the scaling according to the lattice constants based on data for $NdAl_3$; Ref. 3). The arrows show observable transitions.

increases with increasing x, and at $x = 0.15$ it becomes the major component ($\sim 80\%$) of the inelastic magnetic neutron scattering. Raising the temperature to 40 K reduces the intensities of the transitions somewhat for all of the samples, with all values of x (Fig. 2), in agreement with the nature of the changes in the population of the Ce^{3+} ground state.[2] An upper estimate of the intensity of the peaks from scattering by neodymium is lower by a factor of several units than the intensity observed for $Ce_{1-x}Nd_xAl_3$ (Fig. 1). The positions of the peaks from the transitions between the crystal-electric-field levels of $^{3+}$Nd ($E = 4.5$ meV, $E = 7.2$ meV) in $Nd_{0.6}La_{0.4}Al_3$ are not the same as the positions of the peaks in the $Ce_{1-x}Nd_xAl_3$ spectra ($x \neq 0$), but they do agree with the scaling in the lattice constants of the crystal-electric-field levels of $NdAl_3$ (Fig. 2). For $^{3+}$Nd in $Nd_{0.6}La_{0.4}Al_3$, we observe an increase in the intensities of these transitions as the temperature is raised to 40 K, because of an increase in the population of the level from which the transitions are observed (Γ_9^B in Fig. 2). These features suggest that the sharp peaks in the spectra correspond to a scattering of neutrons by cerium ions. Their appearance against the background of the broad $CeAl_3$ spectrum and their intensification with increasing x may be thought of as the result of a superposition of two types of spectral functions for cerium. One might thus suggest

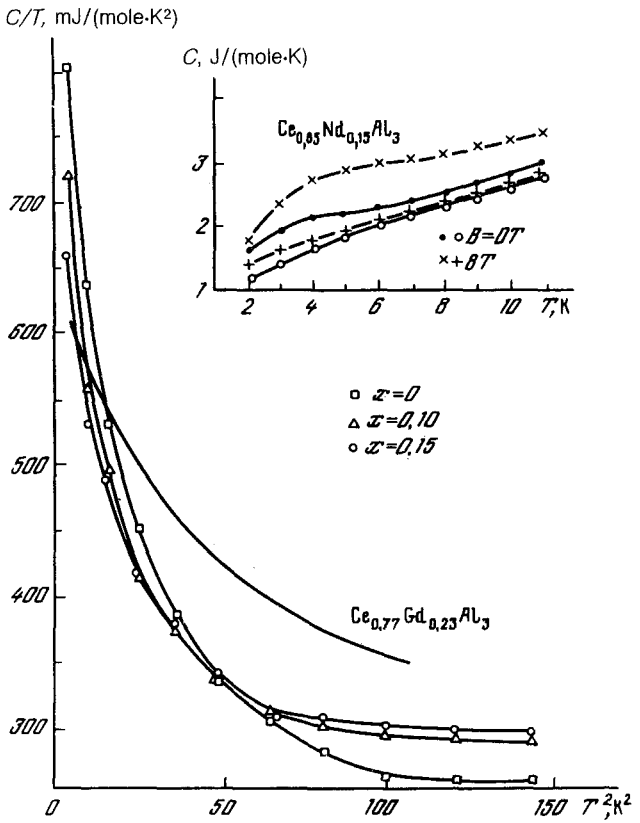

FIG. 3. Magnetic component of the specific heat of $Ce_{1-x}Nd_xAl_3$ ($x = 0$, 0.10, and 0.15) samples and $Ce_{0.77}Gd_{0.23}Al_3$ samples.[4] The Nd components have been eliminated. The results have been normalized to the sample with $x = 0$. Shown at the top are the specific heats of the samples before (●, ×) and after (○, +) the component due to the crystal-electric-field levels of Nd^{3+} was taken into account (Fig. 2) in fields of 0 and 8 T.

that there are finite regions in which the neodymium ions effect the cerium. Since we did not observe this transformation of the inelastic magnetic neutron scattering in experiments in which we replaced Ce by La in $CeAl_3$, it may be that the addition of ions with a magnetic moment, rather than the dilution, is responsible for the observed effects in the transition from the spectrum characteristic of a heavy-fermion system to a spectrum characteristic of an ordinary f-electron system.

Measurements of the specific heat of $Ce_{1-x}Nd_xAl_3$ (Fig. 3) reveal a significant decrease in γ at $T < 7$ K (by as much as 20% at $T = 2$ K). The effect of the magnetic field on the specific heat at these temperatures is due primarily to a splitting and a shift of the $^{3+}$Nd levels. Also shown in this figure are data[4] on the specific heat of $Ce_{0.77}Gd_{0.23}Al_3$. In addition to a radical decrease in γ, Edelstein *et al.*[4] observed the appearance of a state of a spin-glass type, i.e., a transition from a Kondo regime to a magnetic-ion regime. The similarity between the measured curves of the specific heat,

in light of the estimated values of the magnetic moments, suggests that the effect of Nd on $CeAl_3$ is similar to that of Gd. The decrease in the widths ($\Gamma \sim 1/\tau$) of the inelastic peaks in the inelastic magnetic neutron scattering by $Ce_{1-x}Nd_xAl_3$ can thus be regarded as an increase in the relaxation time (τ) of the f electrons due to a decrease in the Kondo scattering. In other words, the changes which occur in the specific heat and the spectra of the inelastic magnetic neutron scattering of $CeAl_3$ upon the addition of a magnetic ion (Gd or Nd) point to the appearance of an "antiscreening" (decancellation) of the magnetic moment.

Another characteristic feature of the inelastic magnetic neutron scattering by $Ce_{1-x}Nd_xAl_3$ is a decrease in the splitting in the crystal-electric-field levels of the $4f$ electrons of the cerium ions, along with a simultaneous decrease in Γ, upon the addition of Nd. (The inverse effect—a growth of the widths with increasing transition energy—was first observed experimentally[5] in $Ce_{0.9-x}La_xTh_{0.1}$ in a transition from a Kondo regime to a mixed-valence regime, which was linked with an increase in the s-f coupling.[1]) An interrelationship of this sort between the width and energy of the transitions, however, does not agree with the ideas of the relaxation model,[6] in which a decrease in the s-f coupling leads to a narrowing of the levels and some increase in the observed splitting of the crystal electric field. A possible explanation for this discrepancy is that the crystal electric field and the exchange interaction of the f electrons with the conduction electrons were treated as independent factors in the model of Ref. 6.

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