Exchange interaction and relaxation in $LuAl_2$:Ce and YAl_2:Ce intermetallic compounds*

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The spin-flip scattering of conduction electrons due to Ce ions was measured by breaking the ESR bottleneck present in $LuAl_2$:Gd and YAl_2:Gd with the addition of Ce. The exchange interaction between the Ce ions and the conduction electrons was extracted using the model of Cornut and Coqblin.

I. INTRODUCTION

This paper presents new measurements of the spin-flip relaxation rates of the conduction electrons due to cerium ions in the intermetallic compounds YAl_2 and $LuAl_2$. We were able to extract these relaxation rates by using the bottleneck behavior observed previously for[1] YAl_2:Gd and $LuAl_2$:Gd.[2] Addition of Ce, as a second impurity, yields an additional channel for conduction-electron spin-flip relaxation. This manifests itself in an appreciable increase in the EPR g shift and the thermal broadening of Gd upon Ce addition. It provides us with a measure of the spin-flip relaxation rate of the conduction electrons.

Assuming that the resonance scattering mechanism of Cornut and Coqblin[3] is the dominant mechanism for spin-flip relaxation,[4,5] we were able to extract an effective exchange interaction between the Ce ions and the conduction electrons. The values of the exchange observed have the same magnitude as in the "Kondo system" $LaAl_2$:Ce.[6] This indicates the close proximity of the Ce 4f level to the Fermi levels of $LuAl_2$ and YAl_2, allowing for large admixture of the conduction electrons into the Ce 4f level. The existence of large admixture can partially explain the "failure" to observe the ESR of Ce^{3+} in these systems. Thus, in the absence of direct measurement of the exchange interaction (by observing the resonance), our indirect method is the only powerful way to extract this important information.

Experimental results

The Gd ESR measurements were performed on powdered samples of $Gd_xCe_yLu_{1-x-y}Al_2$ and $Gd_xCe_yY_{1-x-y}Al_2$. The temperature was changed from 0.7 to 25 K. The resonance properties are very similar to those reported previously for $Gd_xLu_{1-x}Al_2$ and $Gd_xY_{1-x}Al_2$.[2] The high-temperature g shift and thermal broadening of Gd as functions of Ce concentration are exhibited in Figs. 1 and 2. The behavior observed characterizes a bottleneck system with an unbottlenecked g shift and thermal broadening of $\Delta g = 0.085$, $\Delta H/T = 72 \pm 10$ G/K, and $\Delta g = 0.07$, $\Delta H/T = 50 \pm 10$ G/K, for Gd in $LuAl_2$ and YAl_2, respectively.

In addition, the ESR of Er in $LuAl_2$ has been observed (Fig. 3) for Er concentrations of 1500 and 3000 ppm. The field for resonance is appropriate to a g value of 6.79 ± 0.05. This is very close to the value expected for an isolated crystal-field-split Γ_7 ground state. The ESR linewidth was fitted to the formula $\Delta H = a + bT$ with $a = 35$ G and $b = 8 \pm 2$ G/K. We found that the best experimental results on $LuAl_2$:Er were observed using powder and polycrystalline rods prepared by pulling from the melt, using a Chokralsky three-arc technique. No resonance associated with the Ce in $LuAl_2$ or YAl_2 was observed either in powdered samples or polycrystalline rods.

The effect of nonmagnetic impurities on a bottleneck system has been demonstrated previously.[2] In the case of magnetic impurities, additional interaction effects are expected. These interaction effects might manifest themselves by marked temperature dependences of the ESR g shift and linewidth, especially at low temperatures.

The experimental results indicate, however, no temperature dependence of the g shift and linewidth associated with interaction effects upon Ce addition. This is in contrast to the behavior observed upon addition of Er ions (magnetic) into $Gd_xLu_{1-x}Al_2$. Large temperature dependences of both the g shift and the linewidth were observed, indicating the dominance of interaction effects in this case.

II. ANALYSIS OF THE EXPERIMENTAL RESULTS

In the extreme bottleneck regime, if dynamic effects are neglected, the effective relaxation rate of the Gd localized moment to the lattice, δ_{eff}, can be written

$$\delta_{eff} = \delta_{ie}\,\delta_{eL}/\delta_{ei} . \quad (1)$$

Relation (1) holds provided that the Gd susceptibility dominates over the Pauli susceptibility. It can be understood by a two-step process in which, first, the local moment and the conduction electrons mutually flip their spins under the effect of the exchange (δ_{ie} and δ_{ei} being the corresponding spin-flip relaxation rates) and, second, the conduction electrons relax to the lattice with a rate δ_{eL}.[2] It is worthwhile, at this stage, to elucidate the origin of δ_{eL}. This relaxation rate originates with any mechanism that flips the spins of the conduction electrons without flipping the localized moment spin. More specifically, for the bottleneck systems $Gd_xCe_yB_{1-x-y}Al_2$ (B = Y, Lu, La) the mechanisms contributing to δ_{eL} can be summarized as follows:

(a) Spin-flip resonance scattering rate due to the 4f *resonance level of the* Ce ions, $\delta_{eL}^{(a)}$. Such a mechanism has been suggested previously by Cornut and Coqblin[3] for the interpretation of the spin-flip scattering rate of the conduction electrons by the resonant 4f level of Ce in $LaAl_2$. It takes into account both combined spin and orbit exchange scattering and the crystalline field of the Ce 4f level.

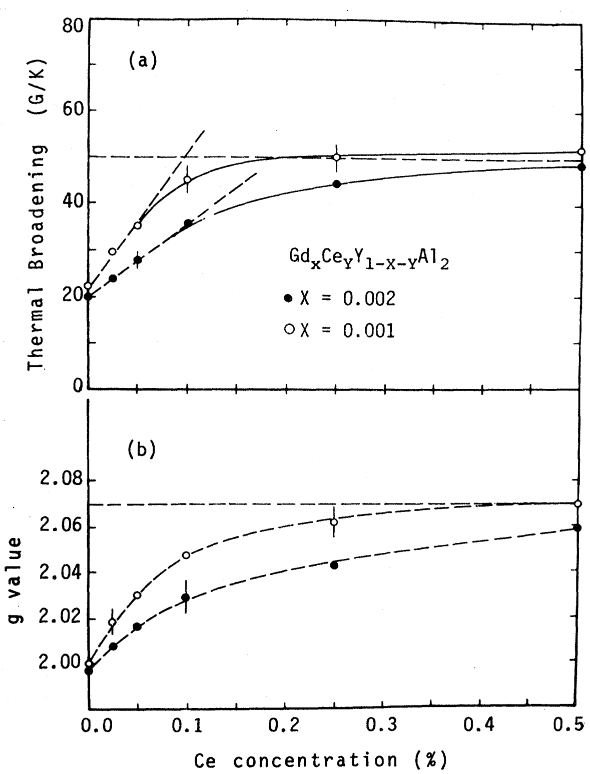

FIG. 1. High-temperature (a) thermal broadening, $\Delta H/T$, and (b) g value of Gd in $Gd_xCe_yY_{1-x-y}Al_2$ (x = 0.002, 0.001) as a function of Ce concentration. The horizontal dashed lines represent the unbottlenecked values of $\Delta H/T$ and g.

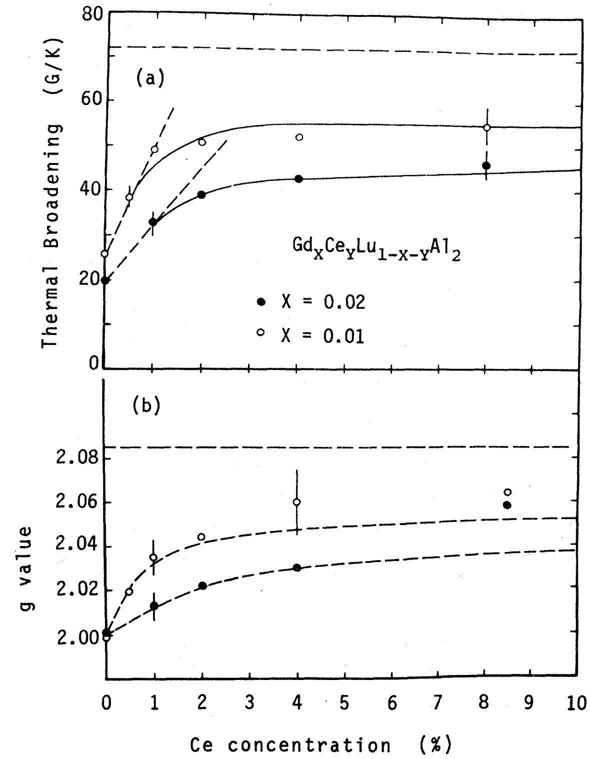

FIG. 2. High-temperature (a) thermal broadening, $\Delta H/T$, and (b) g value of $Gd_xCe_yLu_{1-x-y}Al_2$ (x = 0.02, 0.01) as a function of Ce concentration. The horizontal dashed lines represent the unbottlenecked values of $\Delta H/T$ and g.

(b) Spin-orbit-spin-flip scattering due to admixture of the conduction electrons with other nonmagnetic core states (p or d) on the Ce *site*. We shall denote the relaxation rate associated with this mechanism by $\delta_{eL}^{(b)}$.

(c) Spin-orbit spin-flip scattering due to admixture of the conduction electrons with nonmagnetic states localized on the Gd site, $\delta_{eL}^{(c)}$.

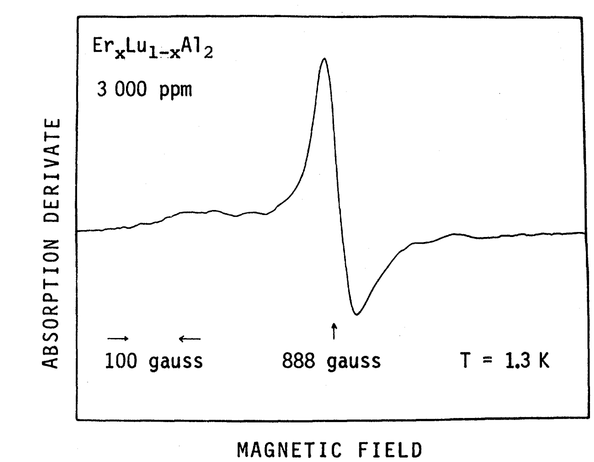

FIG. 3. EPR spectra of Er in $LuAl_2$ at 1.3 K. The Er concentration is 3000 ppm.

(d) Background relaxation, $\delta_{eL}^{(0)}$, associated with dislocations or other impurities present in the sample.

In the first approximation, δ_{eL} can be expressed as

$$\delta_{eL}=\delta_{eL}^{(0)}+\left(\frac{\partial\delta_{eL}^{(a)}}{\partial C}\right)_{Ce}C_{Ce}+\left(\frac{\partial\delta_{eL}^{(b)}}{\partial C}\right)_{Ce}C_{Ce}+\left(\frac{\partial\delta_{eL}^{(c)}}{\partial C}\right)_{Gd}C_{Gd}\,, \tag{2}$$

where C_{Ce} and C_{Gd} represent the concentrations of Ce and Gd, respectively, in BAl_2 (B=Y, Lu). The values of δ_{eff} and δ_{ie} in (1) are related to the experimental and the unbottleneck linewidths, respectively. Their ratio can be expressed as $\delta_{eff}/\delta_{ie}=\Delta H/\Delta H_K$, where $\Delta H_K/T$ is equal to 72 and 50 G/K for $LuAl_2$: Gd and YAl_2 : Gd, respectively. Thus, using (1) and (2), the initial slope of the experimental thermal broadening $\Delta H/T$ is given by

$$\frac{\Delta H}{T}=\frac{\Delta H_K}{T}\left[\delta_{eL}^{(0)}+\left(\frac{\partial\delta_{eL}^{(a)}}{\partial C}\right)_{Ce}C_{Ce}+\left(\frac{\partial\delta_{eL}^{(b)}}{\partial C}\right)_{Ce}C_{Ce}+\left(\frac{\partial\delta_{eL}^{(c)}}{\partial C}\right)_{Gd}C_{Gd}\right]\Bigg/\left(\frac{\partial\delta_{ei}}{\partial C}\right)_{Gd}C_{Gd}\,. \tag{2a}$$

This should then give

$$\frac{\partial}{\partial C_{Ce}}\left(\frac{\Delta H}{T}\right)_{C_{Ce}=0}=\frac{\Delta H_K}{TC_{Gd}}\beta\,, \tag{2b}$$

where the parameter β is defined as

$$\beta=\left[\left(\frac{\partial\delta_{eL}^{(a)}}{\partial C}\right)_{Ce}+\left(\frac{\partial\delta_{eL}^{(b)}}{\partial C}\right)_{Ce}\right]\Bigg/\left(\frac{\partial\delta_{ei}}{\partial C}\right)_{Gd}\,. \tag{3}$$

The parameters β for the various systems were extracted from the initial slopes in Figs. 1 and 2 (extreme bottleneck regime). Their values are tabulated in Table II. It should be stressed that β can be extracted (in principle) from the g-shift (Δg) behavior in the bottleneck regime. This, however, might yield large "error bars" because of the nonlinearity of Δg versus δ_{eL}/δ_{ei}.

The relaxation rate of the conduction electrons to the Ce ions (proportional to the parameter β) is determined by both mechanisms a and b. Thus an independent estimate of β is needed in order to determine the dominant mechanism. Such an estimate is possible in $LaAl_2$ because of additional available information (i.e., superconducting transition temperature data).

The resonance scattering mechanism of Cornut and Coqblin[3] yields the following expression for the conduction-electron spin-flip scattering rate per unit Ce concentration:

$$\left.\frac{\partial\delta_{eL}^{(a)}}{\partial C}\right|_{Ce}=\frac{2\pi}{\hbar}\,\eta(E_F)\langle J^2\rangle_{Ce}A_{00}\,, \tag{4}$$

where $\eta(E_F)$ is the density of states for one spin direction and A_{00} is a parameter which depends on the ground-state crystal-field splitting (assuming that only this state is populated) of the Ce $4f$ level. A_{00} is defined by Cornut and Coqblin.[3] The exchange spin-flip scattering rate due to Gd ions is given by

$$\left.\frac{\partial\delta_{ei}}{\partial C}\right|_{Gd}=\frac{4\pi}{3\hbar}\,\eta(E_F)\langle J^2\rangle_{Gd}S(S+1)\,. \tag{5}$$

In both (4) and (5) the electron-electron coulomb interaction responsible for the exchange enhancement of the host susceptibility was neglected. A modification of (4) and (5) to include this enhancement can be easily obtained, using partial-wave analysis.[7] The model of Cornut and Coqblin assumes that the exchange interaction $\langle J^2\rangle_{Ce}$ between the Ce ions and the conduction electrons is due to f-like covalent mixing between the $4f$ shell and the conduction electrons of f character. We shall therefore identify the exchange interaction $\langle J^2\rangle_{Ce}$ in (4) with the $L=3$ partial-wave amplitude. The enhancement factor in the $L=3$ partial-wave amplitude can be written

$$7\left\langle\left(\frac{p_3(1-q^2/2K_F^2)}{1-\upsilon\chi(q)}\right)^2\right\rangle\,, \tag{6}$$

where P_3 is the third-order Legendre polynomial, υ is the electron-electron Coulomb interaction, $\chi(q)$ is the q-dependent susceptibility of the conduction electrons, q is the momentum transfer vector, K_F is the Fermi wave vector, and $\langle\ \rangle$ indicates the normalized sum from $0\le|q|\le 2K_F$.

Similarly, the exchange interaction between the Gd and the conduction electrons originates mainly with s-wave scattering ($L=0$). The enhancement factor for ($L=0$) partial-wave scattering can be expressed as

$$\left\langle\left(\frac{1}{1-\upsilon\chi(q)}\right)^2\right\rangle\,. \tag{7}$$

The enhancement factors in (4) and (5) are obtained from (6) and (7), respectively, multiplied, however, by the factor $1-\upsilon\chi(0)$. It is clearly seen that the enhancement factors (6) and (7) are different, implying different enhancement corrections in (4) and (5). However, if $\chi(q)$ does not vary appreciably with q in the range $0\le q\le 2K_F$, we expect (6) to be very close to (7). This is due to the orthogonalization requirement of the Legendre polynomials together with the appreciable variation of P_3 with respect to $\chi(q)$. This last factor is not known experimentally. We shall therefore use a δ function for the Coulomb electron-electron interaction together with a free-electron value for $\chi(q)$,

$$\chi(q)=\frac{\chi(0)}{2}\left(1+\frac{4K_F^2-q^2}{4K_Fq}\ln\left|\frac{2K_F+q}{2K_F-q}\right|\right)\,. \tag{8}$$

In this approximation, we find (6) to deviate from (7) by a maximum value of 20% [for various values of $\alpha = \mathcal{V}\chi(0)$]. This is much smaller than the "error bars" in the measured values of the spin-flip scattering rates (approximately 30%). Thus, in analyzing our data, we shall consider the ratio between (4) and (5). This ratio is independent of the enhancement factor in our approximation. We shall therefore define the ratio γ, using (4) and (5), as

$$\gamma = \left(\frac{\partial \delta_{eL}^{(a)}}{\partial C}\right)_{Ce} \Bigg/ \left(\frac{\partial \delta_{el}}{\partial C}\right)_{Gd} . \qquad (9)$$

γ can be determined theoretically provided that the ratio $\langle J^2\rangle_{Ce}/\langle J^2\rangle_{Gd}$ is known. For the case of $LaAl_2$, the ratio $\langle J^2\rangle_{Ce}/\langle J^2\rangle_{Gd}$ can be easily obtained from the initial depression of the superconducting transition temperature by alloying $LaAl_2$ with Ce or Gd. According to Cornut and Coqblin,[3] the ratio of the initial depressions is given as

$$\left(\frac{\Delta T_c}{\Delta C}\right)_{Ce} \Bigg/ \left(\frac{\Delta T_c}{\Delta C}\right)_{Gd} = \frac{2}{189} \frac{\langle J^2\rangle_{Ce}}{\langle J^2\rangle_{Gd}} \frac{\lambda_0^2 - 1}{\lambda_0} , \qquad (10)$$

where λ_0 is the degeneracy of the crystal-field ground state of the Ce $4f$ level. The value of $\Delta T_c/\Delta C$ was measured by Maple[5,3] to be 3.79 and 2.56 K/at.% for Gd and Ce, respectively. Thus, from (10), $\langle J^2\rangle_{Ce}/\langle J^2\rangle_{Gd}$ was extracted and found to be

$$\frac{\langle J^2\rangle_{Ce}}{\langle J^2\rangle_{Gd}} = \begin{cases} 17 & \text{for a } \Gamma_8 \text{ ground state} \\ 42 & \text{for a } \Gamma_7 \text{ ground state} \end{cases} . \qquad (11)$$

By using (11) the ratio γ was found to be 0.51 for both Γ_7 and Γ_8 crystal-field-split ground states of Ce^{3+} in $LaAl_2$. This value of γ is very close to the value of β observed experimentally for $LaAl_2$ (see Table II). This indicates that the spin-orbit spin-flip scattering rate due to nonmagnetic p or d states on the Ce site [mechanism (b)] is probably much smaller than mechanism (a). Thus it is completely justified to analyze the experimental values of β (Table II) in terms of the Ce $4f$ resonance scattering model. Further support for this conclusion is provided by the small value of the conduction-electron spin-orbit spin flip scattering rate due to Gd impurities in $LaAl_2$. We found this value to be equal to $(1 \pm 0.7)\times 10^7$ sec^{-1}/ppm,[5] much smaller than the exchange spin-flip scattering rate (13×10^7 sec^{-1}/ppm) due to Gd impurities in the same host.[8] The small value of $(\partial \delta_{eL}^{(c)}/\partial C)_{Gd}$ is consistent with our conclusion reached for Ce-doped $LaAl_2$, provided that the conduction-electron scattering rate due to p or d states does not change appreciably across the $4f$ series.

We hoped to measure directly the spin-orbit spin-flip scattering due to nonmagnetic core states by using the resonance properties of Er in $LuAl_2$ as follows: The Er ESR thermal broadening ($\Delta H/T \simeq 8$ G/K) provides us with a measure of the conduction-electron spin-flip scattering due to the exchange interaction with the Er ions. The *total*

TABLE I. Crystal-field parameters and ground states for various rare-earth ions in BAl_2 (B = La, Y, Lu) as predicted by several experimental techniques.

Host	Crystal-field ground state	Crystal-field parameters $A_4\langle r^4\rangle$ (meV)	$A_6\langle r^6\rangle$ (meV)	Experimental technique and reference
$LaAl_2$:Tm	Γ_5	−3.85	−1.14	susceptibility[a]
$LaAl_2$:Tb	$\Gamma_3(x=-0.6)$	negative	negative	superconductivity critical field[b]
$LaAl_2$:Tb	$\Gamma_3(x=-0.6)$	negative	negative	thermoelectric power[c]
$LaAl_2$:Ce	Γ_7	···	···	susceptibility[d]
YAl_2:Tm		+2.04	−0.47	inelastic neutron scattering[e]
YAl_2:Tm		+1.63	−0.425	specific heat[f]
$LuAl_2$:Er	Γ_7	positive or negative	positive	ESR[g]

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[g]This work.

TABLE II. Exchange parameters $|J_{Ce}(\Gamma_8)|$ and $|J_{Ce}(\Gamma_7)|$ of Ce in BAl_2 (B = La, Y, Lu) assuming Γ_8 or Γ_7 crystal-field-split ground states, respectively.

Host	β (experimental)	Predicted ground state	$\|J_{Ce}(\Gamma_8)\|$ (eV)	$\|J_{Ce}(\Gamma_7)\|$ (eV)
$LaAl_2$	0.54 ± 0.1[a]	$\Gamma_7(\Gamma_8)$[b]	0.42 ± 0.15	0.65 ± 0.2
YAl_2	0.6 ± 0.2	Γ_7	0.35 ± 0.15	0.56 ± 0.2
$LuAl_2$	0.32 ± 0.15	Γ_7 or Γ_8	0.33 ± 0.15	0.5 ± 0.2

[a]Extracted from the g shift (Ref. 5).

[b]A Γ_7 ground state was measured by Maple for Ce impurities in $LaAl_2$. This is in disagreement with the crystal-field parameters for other rare-earth ions in $LaAl_2$ (see Table I).

(spin-orbit and exchange) spin-flip scattering rate due to Er in $LuAl_2$ can be measured by breaking the bottleneck present in $LuAl_2$: Gd upon Er addition. The difference of these two measured quantities yields the conduction-electron spin-flip scattering rate due to nonmagnetic core states of Er. Preliminary experiments in this direction indicate marked temperature dependence of the g shift and linewidth upon addition of 5000-ppm Er into $Gd_{0.01}Lu_{0.99}Al_2$. This indicates the dominance of interaction effects as explained above. Thus we were not able to extract the relaxation rate of the conduction electrons due to Er. The dominance of interaction effects indicates, however, that this relaxation rate is relatively small, as expected.

III. DISCUSSION

Under the assumption that the spin-flip scattering rate due to nonmagnetic core states is relatively small (i.e., the dominance of the Cornut-Coqblin mechanism), the value of β is very close to γ ($\beta \approx \gamma$).

Thus, by comparing the experimental values of β with (9), (4), and (5), the ratio of the exchange interactions $\langle J^2\rangle_{Ce}/\langle J^2\rangle_{Gd}$ can be extracted. This requires, however, the knowledge of the crystal-field ground state of Ce^{3+} in BAl_2 (B = Lu, Y, La). In the absence of direct measurement, one can use crystal-field parameters as measured on other rare-earth ions in the same hosts together with the assumption that these parameters do not change appreciably across the $4f$ series and at least retain their sign. This last assumption is supported by recent EPR measurements[9] on several systems. In the present case, however, it should be regarded with caution because of the large conduction-electron admixture. Table I exhibits the crystalline field parameters for various rare earths in BAl_2 (B = Lu, Y, La). Surprising enough, the signs of these parameters are completely different in the three systems, although the crystalline and band structures are expected to be similar.

In the absence of conclusive information about the crystal-field ground state of Ce^{3+}, we estimated $\langle J^2\rangle_{Ce}/\langle J^2\rangle_{Gd}$ for both Γ_7 and Γ_8 ground states. The value of $\langle J^2\rangle_{Gd}$ is known, however, from the Gd ESR thermal broadening.[2] This enables us to extract $|J_{Ce}| = (\langle J^2\rangle_{Ce})^{1/2}$. The values of $|J_{Ce}|$ for the various BAl_2 systems are tabulated in Table II. It is clearly seen that the $|J_{Ce}|$ for YAl_2 : Ce and $LuAl_2$: Ce have the same orders of magnitude as that for $LaAl_2$: Ce. This might indicate large admixture in the former. It would be extremely interesting to verify this conclusion by means of other experimental techniques. Resistivity measurements are presently in progress.

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