Low-temperature nuclear orientation on the Kondo system $Ce_xNd_{1-x}Ni$

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The isotope ^{147}Nd has been oriented by means of low-temperature nuclear orientation on the pseudobinary series $Ce_xNd_{1-x}Ni$ for $x=0.0$, 0.1, 0.25, 0.5, and 0.8. The results show that the compounds are magnetically ordered due to Nd-Nd interactions and no magnetic moment exists on cerium. The Kondo effect is present (for $x\neq 0$) as revealed by measurements of electical resistivity and thermoelectric power.

I. INTRODUCTION

During the past decade there has been much interest in rare-earth intermetallic compounds containing the element Ce. It was discovered during the early eighties that this element causes anomalies in the magnetic and transport properties of these compounds, now known as concentrated Kondo systems.

There is a currently accepted classification based on the concentration (x) of the magnetic rare-earth impurity: for $x \ll 1$ the compounds are said to be on the *Kondo impurity* limit, and for $x \approx 1$ they are called *concentrated Kondo systems.* When $x=1$, when the magnetic rare earths form a periodic lattice, we refer to a *Kondo lattice.*

It is well known that Kondo systems can present magnetic order. This will depend upon the balance between the two opposing interactions: the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which favors magnetic ordering and the Kondo coupling, favoring a nonmagnetic ground state. The resultant state can be strongly dependent on the concentration of the magnetic impurity.[1] Thus, in a given system, much can be learnt by changing the concentration through the transition Kondo impurity $\rightarrow$ Kondo lattice.

Usually such studies are carried out on systems where the magnetically unstable element is replaced by a nonmagnetic one, as in $Ce_xLa_{1-x}Al_3$,[2] or $La_{1-x}Ce_xNi$.[3,4] However, to our knowledge there is no report in which an unstable rare earth is replaced by a magnetic one.

CeNi and NdNi compounds crystallize in the CrB-type orthorhombic structure, space group *Cmcm*.[5] CeNi is a Kondo lattice, extensively studied by Gignoux and coworkers.[6–8] NdNi is a simple ferromagnet with $T_c=28$ K and easy direction of magnetization at about 23° to the **a** axis.[9]

The nuclear-orientation technique can distinguish the magnetic states of cerium and neodymium ions by observing the anisotropy of the γ rays emitted by the isotopes ^{141}Ce and ^{147}Nd. In this work we report results of nuclear-orientation experiments in the pseudobinary series $Ce_xNd_{1-x}Ni$ for $x=0.0$, 0.1, 0.25, 0.5, and 0.8. Measurements of electrical resistivity and thermoelectric power were also performed for the same range of x.

II. EXPERIMENTAL

Single crystals of $Ce_xNd_{1-x}Ni$ were grown by the Bridgman method. For the nuclear-orientation experiment, samples weighing about 30 mg were cut with approximate dimensions $5\times2\times0.5$ mm^3 along **a**, **c**, and **b** directions, respectively. After cutting, the samples were irradiated with neutrons for about 7 h under a flux of 10^{12} neutrons cm^{-2} s^{-1}, producing typically 10–20 μCi of the isotopes ^{141}Ce and ^{147}Nd. The samples were soldered to a cold finger using pure indium and Baker's flux and loaded in a ^3He-^4He dilution refrigerator. An external field of 0.7 T was applied along the **a** axis of the crystals and the γ transitions (145 keV from the cerium isotope and 531 keV from the neodymium isotope) were detected along the main crystallographic axes using Ge(Li) detectors. The temperature dependence of the anisotropy of the γ emissions was monitored by using a $^{54}MnNi$ nuclear thermometer. The description of the procedure for fitting and extracting parameters is given in full detail in Ref. 10.

For electrical-resistivity and thermoelectric-power

measurements, samples were cut in a rectangular shape along the **a**, **b**, and **c** directions of the crystal with approximate dimensions of $6\times1\times1$ mm^3, respectively. The resistivity was measured by the dc four-terminal method with the current measured along the **a** axis. Thermopower results were obtained by measuring the voltage induced by a small temperature gradient along the **a** axis.

III. RESULTS

Figure 1 shows the temperature dependence of ^{147}Nd anisotropy in the $Ce_xNd_{1-x}Ni$ series for $x=0.00$ [Fig. 1(a)] and 0.80 [Fig. 1(b)]. Similar plots for $x=0.10$, 0.25, and 0.50 can be found in Ref. 10. The continuous line is a fit using the expression[11]

$$W(\theta)=1+f(A_2U_2Q_2B_2P_2(\cos\theta) + A_4U_4Q_4B_4P_4(\cos\theta)) , \quad (1)$$

where $A_\lambda U_\lambda$ are "nuclear parameters" describing the decay of the oriented state of the isotope. Q_λ are solid-angle correction factors. The values of these parameters are given in Ref. 10. B_λ are the "orientation parameters" and contain the hyperfine interaction; P_λ are Legendre polynomials describing the directional dependence of the anisotropy. θ is the angle between the axis of the detector and the direction of the orienting field (the **a** axis). The parameter f measures the fraction of nuclei which occupy a "good site" in the lattice. Knowing the nuclear parameters allows us to obtain f and B_{hf} from the fitting. In Fig. 1 the negative anisotropy corresponds to $\theta=0$ (**a** axis) and the positive to $\theta=90°$ (**b** and **c** axes). The fitted parameters are shown in Table I for the different cerium concentrations. Also shown in the table are estimates for B_{4f} arising from the orbital and spin contributions of the $4f$ electrons in the Nd^{3+} ions (see discussion below).

In all compounds, no significant effects could be observed from ^{141}Ce. At 13 mK, for $x=0.1$ we measured $W(\mathbf{a})=+0.32(34)\%$; for $x=0.25$, $+0.38(10)\%$; for $x=0.5$, $-0.05(12)\%$; and for $x=0.8$, $-0.15(10)\%$. These anisotropies are consistent with the fact that cerium does not have a magnetic moment for this range of concentration.

Although an electric quadrupolar interaction can be present at the neodymium site, the nuclear orientation is usually insensitive to it when a much stronger dipolar magnetic interaction is superimposed. Stone[12] has discussed the conditions in which quadrupolar interaction can be observed in nuclear-orientation experiments. Studies of the magnetic resonance of ^{143}Nd in NdNi by Shimizu[13] showed that the quadrupolar interaction contributes less than 0.5% of the total hyperfine splitting. Thus we can neglect electric quadrupolar effects in our fitting.

In Fig. 2 the electrical resistivity of $Ce_xNd_{1-x}Ni$ is shown for the range 2–300 K along the **a** direction. For $x=0$ a sharp cusp occurring at about 28 K can be observed, corresponding to the magnetic transition of NdNi. Above that, $\rho(T)$ increases linearly with T, in a typical metallic behavior. For $0<x\leq0.5$, the curves are similar to that of NdNi, but in the range $0.5<x<1.0$ they resemble the CeNi behavior, a typical Kondo lattice. We also note that no further anomaly is observed at low temperatures for any concentration.

Figure 3 shows the behavior of the thermoelectric power, measured along the **a** direction for the same range

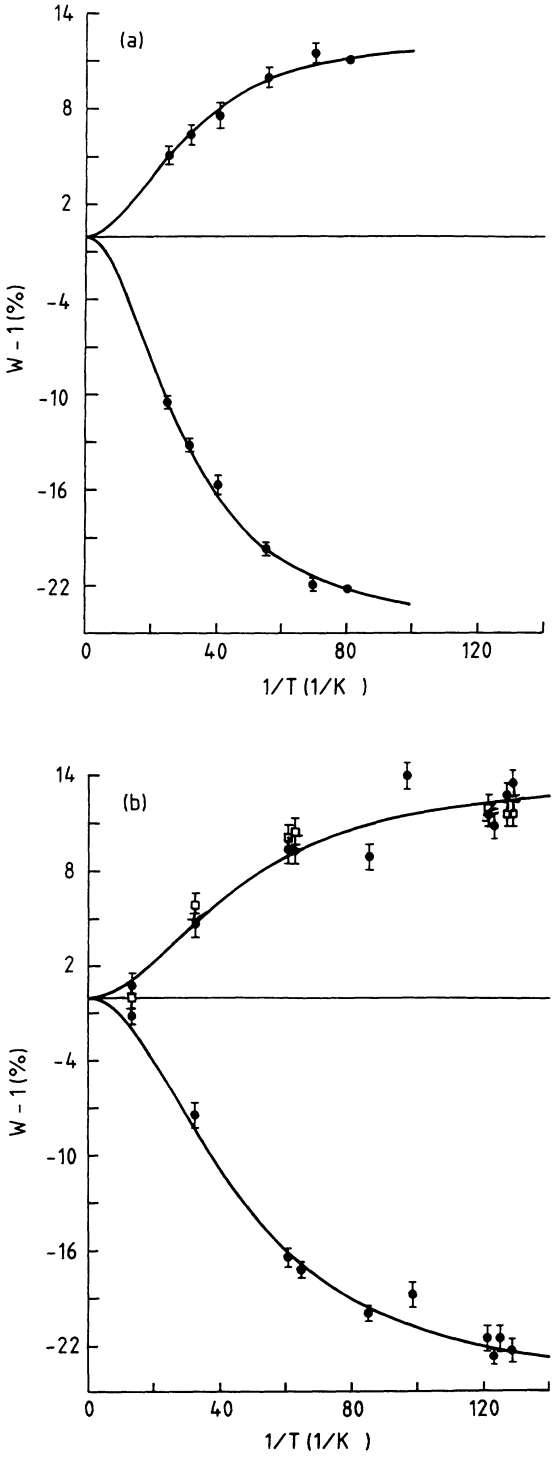

FIG. 1. Temperature dependence of the anisotropy in the γ transition at 531 keV from the ^{147}Nd oriented state for $x=0.0$ (a) and $x=0.8$ (b). The continuous line is a fitting using Eq. (1) (see text).

TABLE I. Measured hyperfine fields (B_{hf}) at the Nd site in $Ce_xNd_{1-x}Ni$ compounds. B_{4f} are estimates for the contributions to B_{hf} appearing from $4f$ electrons (see text).

Ce concentration (x)	B_{hf} (T)	B_{4f}	f (%)
0.00	418(12)	349(12)	88(3)
0.10	370(15)	301(15)	80(4)
0.25	373(15)	304(15)	95(5)
0.50	342(18)	273(18)	87(5)
0.80	272(18)	203(18)	90(3)

of temperature. We see that NdNi shows no anomaly in $S(T)$ but a broad peak is observed for all $x \neq 0$. The intensity of the maximum depends upon the concentration and it ranges from about 35 ($x=0.1$) to 50 $\mu V K^{-1}$ ($x=0.5$). It is also clear from this figure that there is a shift of the maximum toward lower temperature as the concentration decreases. These results show that cerium is in a Kondo state for $x \neq 0$.

IV. DISCUSSION AND CONCLUSION

It is known that in the RNi compounds ($R=$ rare earth) the $3d$ band is filled and no contribution appears from nickel to the magnetism.[9] Then, the total hyperfine field at Nd can be written as a sum of four terms,

$$B_{hf}=B_{4f}+B_{SP}+B_{NN}+B_{app} , \quad (2)$$

where $B_{4f}=A\langle J_z\rangle$ is the contribution appearing from the parent-ion $4f$ electrons; B_{SP} is caused by the conduction-band polarization due to the parent-ion magnetic moment ("self-polarization field"), B_{NN} is the "transferred field," appearing from conduction electrons polarized by Nd magnetic neighbors, and B_{app} is the applied field, usually negligible compared to the other terms. Although B_{NN} can be many teslas in some heavy rare-earth compounds, among the light ones it generally does not exceed 1.5 T. Assuming an isotropic interaction between Nd neighbors[14] and comparing with the results of Berthier, Devine, and Barbara[15] in $NdAl_2$, we can put

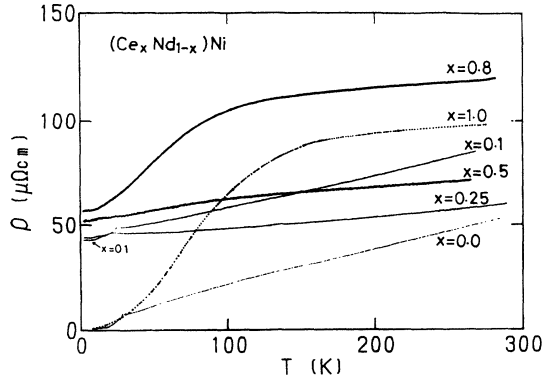

FIG. 2. Electrical resistivity measured along the a direction for different values of x.

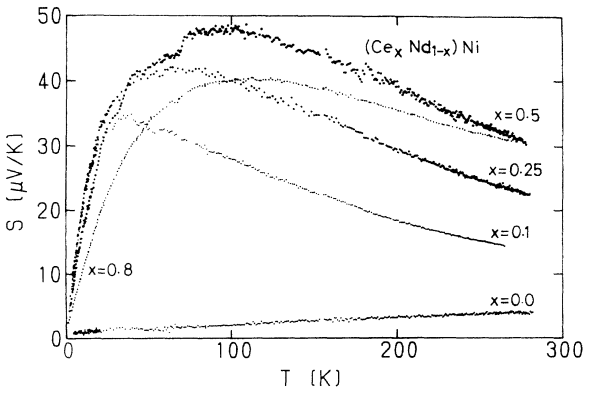

FIG. 3. Thermoelectric power measured along the a direction for different values of x.

an upper limit of 2.0(5) T for this term in NdNi. The effect of alloying with a nonmagnetic rare earth (like cerium in these compounds) will be a further decrease in this contribution. In the first term, A, the hyperfine coupling parameter, is known to vary little from free ions to insulators or metals. We can then use the value $A(Nd^{3+})=95.6(4)$ T given by Bleaney.[16] From magnetization measurements in NdNi,[9] the value $\langle J_z\rangle \approx 3.65$ can be deduced. We then have $B_{4f}=349(2)$ T. The reduction of $\langle J_z\rangle$ as compared to its value in the fully polarized ion is caused by partial quenching of the angular momentum by the crystal field. Substituting these values for B_{4f}, B_{NN}, and B_{app} in Eq. (2), we can deduce the value 67(12) T for B_{SP} in NdNi.

It would be reasonable to assume that alloying with a nonmagnetic rare earth would cause no significant change in B_{SP} with the concentration x. Allowing for the small contribution introduced by B_{NN}, we obtain the variation of B_{4f} with x shown in Table I. Such differences could be attributed to changes in the local molecular field caused by the introduction of a nonmagnetic element into the matrix.[15] However, it is not clear at the present if cerium could affect the transferred field in other ways.

The electrical resistivity and thermoelectric power of $Ce_xNd_{1-x}Ni$ can be compared to those of its nonmagnetic partner, $Ce_xLa_{1-x}Ni$ studied by Isikawa *et al.*[3] and Ohyama, Sakurai, and Komura.[4] Although the general pattern of the results is the same, some important differences seem to appear in the present case. First, no clear evidence of a transition to a concentrated Kondo system could be observed in $\rho(T)$ as occurs for $x \approx 0.4$ in the nonmagnetic case. Secondly, for $x=0.1$ our $S(T)$ curve is very different from that observed by Ohyama, Sakurai, and Komura.[4]

The main results of this work can then be summarized as follows: (i) the pseudobinary compounds $Ce_xNd_{1-x}Ni$ are magnetically ordered at low temperatures for the whole range of observed concentration x; (ii) the magnetic ordering is caused by Nd-Nd exchange interaction; (iii) no magnetic moment is observed in cerium; (iv) all the compounds for $x \neq 0$ show the Kondo effect. There are

clear differences in respect to the equivalent nonmagnetic $Ce_xLa_{1-x}Ni$ compounds. At the moment we are unable to give an explanation for these differences. Further experiments on specific heat, magnetization, magnetic susceptibility, and nuclear orientation for other values of x are being carried out in order to clarify the present results.

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