Large low-temperature magnetoresistance in weakly spin-correlated $CeCu_2Sn_2$ and $CeNi_2Sn_2$

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Large negative magnetoresistance, $\Delta R(B)/R(0)$, of weak Kondo-like systems, $CeCu_2Sn_2$ and $CeNi_2Sn_2$, was observed in the vicinity of 2 K. At 8.3 T, it reached 7% and 23% for the former and latter materials, respectively. The measurements were taken up to 28 K where the magnetoresistance has diminished even at the highest magnetic fields used in our study. The results were interpreted in terms of relatively weak molecular fields which correlate the spin sublattice of these materials. Following the approach proposed by Abrikosov for calculating resistivity in weakly spin-correlated Kondo-like systems, we were able to fit successfully all our results. The fitting produced an estimate of the spin exchange integral for electron-impurity interaction J, which was found to be of the order of 0.36 meV for both systems.

I. INTRODUCTION

The ternary compounds RT_2X_2 where R is a rare-earth metal, T is a d-electron metal, and X is mostly Si or Ge, but also Sn, Ga, B, P, As, or Sb, have been extensively studied during the last decade. These materials exhibit a variety of ground states at low temperatures, e.g., magnetic ordering, Kondo and heavy-fermion behavior, or superconductivity. A group of ternary stannides CeT_2Sn_2 where T = Ni, Cu, Rh, Pd, Ir, or Pt has recently been synthesized[1,2] and their specific heat, magnetic susceptibility and resistivity have been studied.[2-8] Antiferromagnetic ordering is inferred from pronounced peaks in the specific heat for all compounds with $T_N=1.8$ K for $CeNi_2Sn_2$ and $T_N=1.6$ K for $CeCu_2Sn_2$. Values for the electronic contribution to the specific heat of $\gamma\sim 0.6$ J/mol K^2 for $CeNi_2Sn_2$ and $\gamma\sim 3.5$ J/mol K^2 for $CePt_2Sn_2$ have been observed[4] just above T_N. Ascribing the specific-heat enhancement just above T_N to the development of a heavy-electron state rather than critical fluctuations, Beyermann *et al.*,[4] using the single-ion relationship $\gamma T_K=0.68R$ (R is the gas constant), estimated Kondo temperatures of $T_K\sim 1.5$ K for $CePt_2Sn_2$ and $T_K\sim 10$ K for $CeNi_2Sn_2$.

Magnetoresistance measurements[9,10] make a valuable contribution to the study of transport phenomena in Kondo systems and such measurements are reported here for $CeCu_2Sn_2$ and $CeNi_2Sn_2$ samples. Specimens were prepared by arc-melting the constituents on a water-cooled copper hearth in an argon atmosphere. Metals with the following purities were used: Ce and Cu (99.99%), Ni (99.95%), and Sn (99.999%). Weight loss during melting was less than 1%. X-ray diffraction indicated good-quality single-phase samples of the primitive tetragonal $CaBe_2Ge_2$ structure, in agreement with previous studies.[1] The results to be reported in Sec. II were obtained on the as-prepared samples. Magnetoresistance results obtained on an annealed (at 800°C for 5 days) $CeCu_2Sn_2$ sample did not differ significantly from the as-prepared sample, although we notice that the (zero-field) temperature-dependent resistivity was changed notably with annealing.[8]

II. EXPERIMENTAL RESULTS

The study was conducted using a commercial 9 T Janis Superconducting Cryostat in the temperature- and magnetic-field-controlled regimes. Standard four-probe contact arrangements were used for current and voltage contacts to the samples. Copper wires (36 AWG) were attached to the sample using commerically available silver paste. This provided very reliable Ohmic contacts against the possible influence of thermal cycling at low temperatures. The data were collected in a discrete mode while regulating temperatures with a Lake-Shore temperature controller and using a persistent-current mode for the field. We applied a flipping current polarity technique to account for possible thermal voltage offsets in our potentiometric signals. For temperatures above 28 K the magnetoresistance was unresolved to within experimental uncertainty. Small negative magnetoresistance could be resolved for both materials at 23 K. The measurements were taken down to 2.4 K only due to the limiting capacity of our present cooling arrangement. As the temperature was lowered below 23 K, negative magnetoresistance values, $\Delta R(B)/R(0)$, were observed which reached large values of 7% and 23% at 8.3 T and 2.4 K for $CeCu_2Sn_2$ and $CeNi_2Sn_2$, respectively. In Fig. 1 the results obtained for both samples at the highest and lowest temperatures of our study are presented. The solid lines drawn in these figures are the best fits through the data points using Eq. (12) of Sec. III. In our fitting procedure we fixed the spin and gyromagnetic ratio at values of $S=\frac{1}{2}$ and $g=2$, while varying $\mu_{\rm eff}$, the effective magnetic moment of the impurity. We found that the least-mean-square fits produced a very strong temperature dependence of $\mu_{\rm eff}$ for both materials. The results are shown in Fig. 2 together with the calculated behavior suggested by Eqs. (2) and (3) of the next section. We now

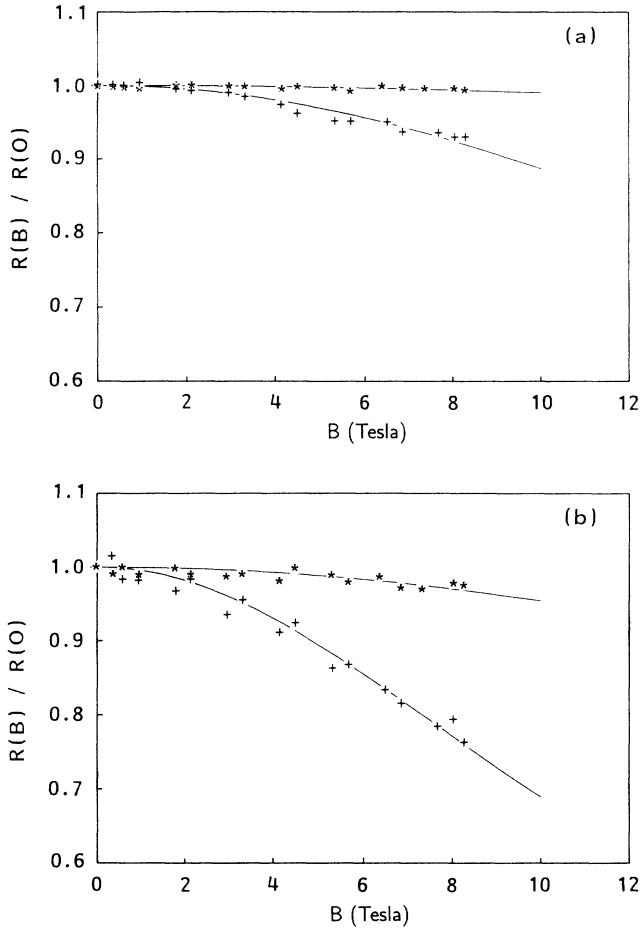

FIG. 1. Magnetoresistance $R(B)/R(0)$ of (a) $CeCu_2Sn_2$ and (b) $CeNi_2Sn_2$ at 2.4 K (+) and 23 K (*). Solid lines through the data are the best fits using Eq. (12) with fixed $S=\frac{1}{2}$, $g=2$; μ_{eff} was used as a free parameter found by the mean-square procedure.

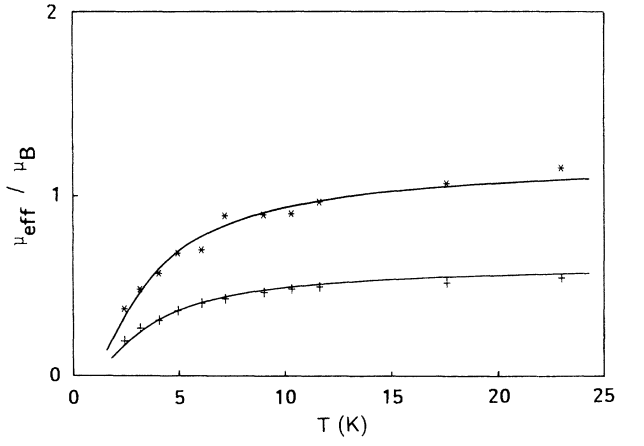

FIG. 2. Temperature dependence of μ_{eff}/μ_B for $CeCu_2Sn_2$ (+) and $CeNi_2Sn_2$ (*). The lines are the best fits to the data in terms of Eqs. (2) and (3). The coupling strength J/k_B was found to be 4.5 K for both materials.

turn to discuss in more detail our experimental results and their interpretation in the framework of weak impurity-spin exchange interaction in these compounds.

III. DISCUSSION

The presence in a metal of impurity atoms with unfilled inner shells (as in transition and rare-earth metals) which have a nonzero spin, may result in the so-called Kondo effect:[11] upon lowering of temperature the resistivity first passes through a minimum followed by a subsequent increase. The interaction energy of an electron with the atoms contains a term dependent on the conduction-electron spin σ and the localized impurity spin $\mathbf{S}$ and is given by[12]

$$V=-2J(\sigma\cdot\mathbf{S})\ . \tag{1}$$

Here J is the exchange energy, and σ are the Pauli matrices. The above expression corresponds to a scattering process in which the electron spin can flip with a simultaneous change in the orientation of the impurity spin. As is well known,[12] in the absence of magnetic impurities the action of an applied magnetic field on the resistance is associated with the curving of the electronic orbits. It always leads to an increase of electrical resistance, and in low fields one can expect that $\Delta\rho(B)\equiv\rho(B)-\rho(0)\propto(\Omega\tau)^2$, where $\Omega=eB/m$ is the Larmor frequency, and τ is the collision time. At the same time, in the presence of magnetic impurities, the magnetic field will polarize the spins of impurity atoms. Thus the direction of spins becomes fixed and the probability of spin-flip scattering declines. As a result, the resistivity falls off.

To interpret our experimental results, let us consider the resistivity contribution due to the spin electron-impurity scattering with the interaction potential (1) in an applied magnetic field B with the energy $H_0=g\mu_{\text{eff}}B$, assuming that the effective magnetic moment of the impurity, μ_{eff}, is defined, self-consistently, via the effective exchange interaction $V_{\text{eff}}\cong-2J\langle\sigma_z\rangle S_z=-JS_z$, namely,

$$\mu_{\text{eff}}(T)=(\tfrac{1}{2}-\langle S_z\rangle)\mu^*\ , \tag{2}$$

where

$$\langle S_z\rangle=\frac{\sum_{M=-1/2}^{1/2}e^{\beta MJ}\langle M|S_z|M\rangle}{\sum_{M=-1/2}^{1/2}e^{\beta MJ}}=\tfrac{1}{2}\tanh\left[\frac{\beta J}{2}\right]\ . \tag{3}$$

Here $|M\rangle$ and M are the eigenfunctions and eigenvalues of the S_z spin operator, respectively, so that $S_z|M\rangle=M|M\rangle$, g is the gyromagnetic ratio, $\beta\equiv1/k_BT$, and $\mu^*\equiv2\mu_{\text{eff}}(\beta\to0)$ is the high-temperature limit of the effective moment per Ce atom. For example, μ^* was found[4,6] to be $\simeq2.5\mu_B$ /Ce (μ_B is the Bohr magneton) for the high-temperature limit of the effective moment in $CeNi_2Sn_2$.

It is assumed that within the temperature and magnetic-field intervals under discussion the orbital

scattering is quenched ($L=0$) and the Ce spin is $S=\frac{1}{2}$ for both $CeCu_2Sn_2$ and $CeNi_2Sn_2$ compounds.

To calculate the magnetoresistivity, we shall follow the approach proposed by Abrikosov[13] for such systems. The main difficulty comes from the fact that the scattering of electrons is nonelastic. Since, however, the exchange interaction of electrons with impurities is usually much less than the potential (nonmagnetic) scattering, the inelastic part is only a small correction to the main elastic contribution. According to Abrikosov,[13] the kinetic equation for this case can be written in the form

$$e\mathbf{E}\left[\frac{\partial f_p}{\partial \mathbf{p}}\right]=-\frac{f_p-f_p^{(0)}}{\tau_{\rm tr}}+\int\frac{d^3p'}{(2\pi\hbar)^3}w_{pp'}\{-f_p(1-f_{p'})+f_{p'}(1-f_p)e^{-\beta\hbar\omega}\}\ , \quad (4)$$

where f_p is the distribution function, $f_p^{(0)}=1/\{1+\exp[\beta(\epsilon_p-\mu)]\}$ is the Fermi function, $\tau_{\rm tr}$ is the collision time due to the potential (non-spin) interaction with impurities, $w_{pp'}$ is the Born scattering probability from the $(\mathbf{p},\epsilon_p)$ to the $(\mathbf{p}',\epsilon_{p'})$ state corresponding to the exchange interaction of the electrons with magnetic impurities (1), $\hbar\omega=\epsilon_p-\epsilon_{p'}$, and μ is a chemical potential. For the reverse processes, the principle of detailed balance has been taken into account.

If $|i\rangle$ and $|k\rangle$ are the states of the scattering system corresponding to the energies E_i and E_k, so that $H_0|i\rangle=E_i|i\rangle$, then the Born scattering probability reads[13]

$$w_{pp'}\equiv w(\omega)=\frac{8\pi}{\hbar}\frac{n_m J^2}{n^2 Z_0}\sum_{ik}e^{-\beta E_i}\langle i|\boldsymbol{\sigma}\cdot\mathbf{S}|k\rangle\langle k|\boldsymbol{\sigma}\cdot\mathbf{S}|i\rangle\times\delta(E_i-E_k+\hbar\omega) \quad (5)$$

$$=\frac{4n_m J^2}{n^2\hbar^2}\int_{-\infty}^{\infty}dt\,e^{-i\omega t}\langle[\boldsymbol{\sigma}\cdot\mathbf{S}(0)][\boldsymbol{\sigma}\cdot\mathbf{S}(t)]\rangle\ , \quad (6)$$

where

$$\mathbf{S}(t)=e^{iH_0t}\mathbf{S}e^{-iH_0t}\ ,\qquad Z_0=Sp(e^{-\beta H_0})=\sum_i\langle i|e^{-\beta H_0}|i\rangle=\sum_i e^{-\beta E_i}\ . \quad (7)$$

Here n is the density of atoms of the host metal, and n_m is the concentration of impurity atoms.

Considering the second term in the right-hand side of Eq. (4) as a small correction to the first one, we can solve Eq. (4) by subsequent approximations. Finally, substituting the thus calculated distribution function f_p into the expression for the current density,

$$\mathbf{j}=e\sum_\sigma\int\frac{d^3p'}{(2\pi\hbar)^3}\mathbf{v}f_p\ , \quad (8)$$

we get for the resistivity[13]

$$\rho=\rho_n+\frac{6\beta n_m}{\hbar^2}\left[\frac{mJ}{nep_F}\right]^2\times\int d\epsilon\,d\epsilon' f^{(0)}(\epsilon)[1-f^{(0)}(\epsilon)]K(\omega)\ , \quad (9)$$

where

$$K(\omega)=\int_{-\infty}^{\infty}dt\,e^{-i\omega t}\langle\mathbf{S}(0)\cdot\mathbf{S}(t)\rangle\ . \quad (10)$$

Here $\rho_n=m/e^2n_e\tau_{\rm tr}$ is the resistivity due to potential (nonexchange) scattering, with $n_e=p_F^3/3\pi^2\hbar^3$ being the electron density, where $p_F=mv_F$ is the Fermi momentum. To pass from Eqs. (4)–(8) to Eq. (9), the usual substitutions $d^3p/(2\pi\hbar)^3\rightarrow\nu\,d\epsilon$ and $\partial f^{(0)}/\partial\mathbf{p}=\mathbf{v}\,\partial f^{(0)}/\partial\epsilon$ have been made, where $\nu=p_Fv_F/2\pi^2\hbar^3$ is the electron density of states at the Fermi level.

If the exchange energy is small enough, the electron scattering by the polarized (in applied magnetic field) impurity spins will entirely dominate the observed magnetoresistivity, so that

$$K(\omega)=\frac{2\pi}{Z_0}\sum_{M=-1/2}^{1/2}\sum_{M'=-1/2}^{1/2}e^{-\beta ME_M}\langle M|\mathbf{S}|M'\rangle\langle M'|\mathbf{S}|M\rangle\times\delta[\hbar\omega-(M-M')E_M]\ . \quad (11)$$

Here $Z_0=\sum_M\exp(\beta ME_M)$, and $E_M=g\mu_{\rm eff}B$ is the magnetic energy with the effective moment

$$\mu_{\rm eff}(T)=\tfrac{1}{2}[1-\tanh(\beta J/2)]\mu^*$$

[see Eq. (3)]. The matrix elements in Eq. (11) are well known,[14] and after some transformations we find for the magnetoresistivity of the impurity system[13]

$$\rho(T,B)=\rho(T,0)\left[1-\frac{1}{S+1}B_S(x)\frac{\sinh x-x}{\cosh x-1}\right]\ ,\qquad x\equiv\beta g\mu_{\rm eff}B\ , \quad (12)$$

where

$$\rho(T,0)=\rho_n+\frac{6\pi n_m}{\hbar}\left[\frac{mJ}{nep_F}\right]^2S(S+1)\ , \quad (13)$$

and

$$B_S(x)=\left[\frac{S+\frac{1}{2}}{S}\right]\coth[(S+\tfrac{1}{2})x]-\frac{1}{2S}\coth\left[\frac{x}{2}\right] \quad (14)$$

is the Brillouin function.

Figure 1 shows the best fit of our magnetoresistivity data according to Eq. (12) as a function of applied magnetic field for two representative temperatures for two samples. Figure 2 depicts the temperature behavior of the effective magnetic moment $\mu_{\rm eff}(T)$, calculated according to Eqs. (2) and (3), for two samples. The slopes of the curves yield an average exchange energy $J/k_B=4.5\pm0.4$ K, and values for the effective moments which at high temperatures approach

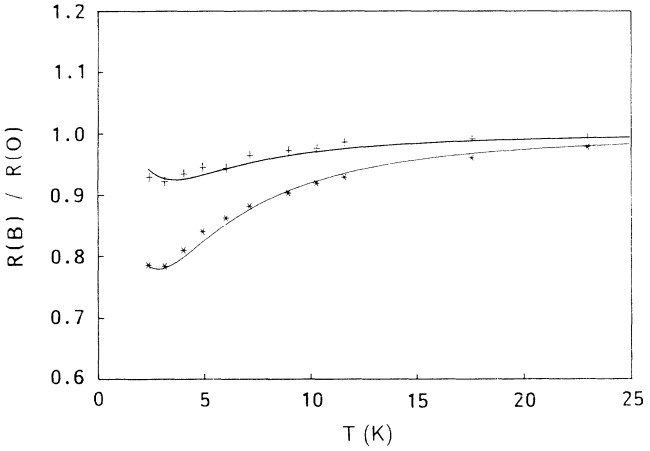

FIG. 3. Comparison of the observed temperature dependence of magnetoresistance for $CeCu_2Sn_2$ (+) and $CeNi_2Sn_2$ (*) with the calculated values (solid lines) obtained by using Eqs. (2) and (3) together with the J values derived from $\mu_{\rm eff}$ and using Eq. (12) for $R(B)/R(0)$ at the fixed value of $B=8.3$ T.

$$\mu^* \equiv 2\mu_{\rm eff}(\beta \to 0) \simeq (1.26 \pm 0.1)\mu_B$$

and $\mu^* \simeq (2.4 \pm 0.2)\mu_B$ for the $CeCu_2Sn_2$ and $CeNi_2Sn_2$ compounds, respectively. The latter estimate is consistent with the values reported[4,6] for this parameter. The small values of exchange energy J found from our magnetoresistance data support [via Eq. (13)] a weak Kondo-type behavior in these systems. Indeed, the Kondo-binding energy in Ce-based heavy-fermion materials was found[7,15,16] to be $\simeq 6-8$ K. Finally, Fig. 3 presents the plot of the magnetoresistivity data as a function of temperature together with the best fit according to Eq. (12) and using $\mu_{\rm eff}(T)$ as depicted in Fig. 2. As is seen, at the lowest temperatures achieved in our experiment, there are apparent precursors of the Kondo transition.

In summary, large low-temperature negative magnetoresistances were observed in $CeCu_2Sn_2$ and $CeNi_2Sn_2$, in the vicinity of 2 K, reaching (at 8.3 T) nearly 7% and 23% for the former and latter materials, respectively. Following the approach proposed by Abrikosov for calculating resistivity in weakly spin-correlated Kondo-like systems, we were able to fit successfully all our results and found an estimate of the spin exchange integral for the electron-impurity interaction J, which turned out to be of the order of 0.36 meV for both materials.

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