Electronic transport and Kondo effect in $La_{1-x}Ce_x$ films

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The electrical resistivity ρ of thin $La_{1-x}Ce_x$ films ($50\ \text{Å} \leqslant d \leqslant 4000\ \text{Å}$) has been measured in the temperature range $1.4\ \text{K} \leqslant T \leqslant 10\ \text{K}$ and in magnetic fields B up to 5 T. The films grow in the La dhcp structure. Films with $x = 0.01$ show the superimposed effect of superconducting fluctuations and Kondo scattering, yielding a maximum in the magnetoresistance. For $x = 0.02$, the determined Kondo temperature for $d = 100$ Å is in good agreement with bulk data. In films with $x = 0.04$ where superconductivity is completely suppressed, evidence of spin-glass order is found from the negative deviation of ρ towards low T compared to the Kondo $-\ln(T/T_K)$ behavior. In large magnetic fields where the spin-glass correlations are suppressed, the magnetoresistivity $\rho(B)$ shows single-ion Kondo behavior. The exchange interaction J between Ce moments and conduction electrons extracted from $\rho(B)$ is independent of film thickness d down to 100 Å. [S0163-1829(96)08329-4]

I. INTRODUCTION

Although the Kondo effect of dilute magnetic impurities in metallic hosts has been studied for decades, the question of a possible geometry dependence has been addressed only fairly recently. A weakening of the Kondo effect could occur in thin films when the thickness d becomes much smaller than the radius R_K of the ''Kondo cloud'' of conduction electrons necessary to screen the magnetic moment of the impurity. R_K is given by $R_K \approx \hbar v_F / 2\pi k_B T_K$ where v_F is the Fermi velocity of the host metal and T_K is the Kondo temperature.[1,2] This argument led Chen and Giordano[2] to interpret the observed decrease of the logarithmic temperature dependence of the resistivity in thin AuFe films with 10–100 ppm Fe, attributed to the Kondo effect via $\Delta\rho \sim -\ln(T/T_K)$, as arising from a weakening of the Kondo effect ($\Delta\rho$ denotes the T-dependent part of the resistivity). However, subsequent experiments on AuFe and CuFe films revealed that the relevant length scale is not directly connected with T_K (Refs. 3,4). In addition, Bergmann showed that the notion of a Kondo cloud is in a certain sense misleading, since the apparent geometry dependence arises from the special spherical symmetry assumed.[5] Previous work by Peters *et al.*[6] and Van Haesendonck *et al.*[7] had indicated that the Kondo effect can be identified even in very thin films by exploiting the capability of weak localization to determine conduction-electron scattering times, e.g., the spin-flip scattering time.

DiTusa *et al.*[8] observed different temperature dependences for $Cu_{1-x}Cr_x$ films ($x = 0.001$) of different widths w between 0.16 and 35 μm. On the other hand, Chandrasekhar *et al.*[9] showed for very dilute $Au_{1-x}Fe_x$ films ($x = 5\times10^{-5}$) of largely varying widths that the slope of the logarithmic T dependence of ρ is independent of w for $0.038\ \mu\text{m} \leqslant w \leqslant 105\ \mu\text{m}$ when electron-electron interactions (EEI) are properly taken into account.

Up to now, all studies concerning a size dependence of the Kondo scattering have been carried out on noble-metal–transition-metal systems. For a complete understanding of the Kondo scattering phenomena occurring in thin films it is worthwhile to extend the investigations to rare-earth systems for which no studies have been reported yet, to our knowledge. The advantage of employing rare-earth alloys like $La_{1-x}Ce_x$ is that the Kondo effect can be modeled assuming independent moments up to rather high concentrations (several at. %) compared to transition-metal systems.

$La_{1-x}Ce_x$ is a well established Kondo system.[10] In the present work we determine the exchange interaction J between conduction electrons and magnetic moments from the magnetoresistivity of $La_{1-x}Ce_x$ films. In brief, we find that J is independent of film thickness d between 100 and 4000 Å. However, the complication arises that pure La is a superconductor [$T_c = 4.87$ K for dhcp La (Ref. 11)]. Therefore, we also studied the superconducting properties of pure La films. On the other hand, the destruction of superconductivity by spin-flip scattering offers an independent possibility to determine J, which can be compared with values deduced from the magnetoresistivity measurements.

II. EXPERIMENTAL DETAILS

The samples were prepared by evaporation onto sapphire substrates in $(11\bar{2}0)$ orientation held at room temperature in a UHV chamber (background pressure $<10^{-10}$ mbar). Polished substrates ($1 \times 15\ \text{mm}^2$ for all films) were etch-cleaned, annealed at 750 °C in UHV, sputtered with 1-keV Ar^+ ions, and annealed again at 1000 °C to reduce sputtering defects. Starting materials were La (99.98%, Alfa Products, Karlsruhe) and Ce (99.99%, Ames Laboratories, Ames, Iowa). Pure La films were deposited from a high-temperature effusion cell, $La_{1-x}Ce_x$ films were obtained by electron-beam evaporation of a $La_{1-x}Ce_x$ alloy prepared in an Ar arc furnace. After deposition at room temperature for all films no further annealing treatment was done. The films were covered with a 100-Å or 300-Å Ge (99.999%, Alfa Products, Karlsruhe) protective layer to prevent oxidation during transfer for the *ex situ* resistivity measurements. The concentration of the $x = 0.04$ ($\pm$ 0.005) films was checked by electron microprobe analysis. The (nominal) film thicknesses

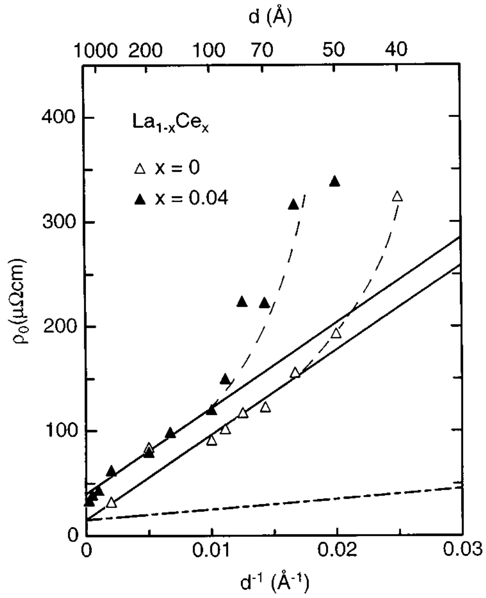

FIG. 1. Low-temperature resistivity ρ_0 for pure La and $La_{0.96}Ce_{0.04}$ films vs inverse film thickness d. Solid lines indicate fits to the Mayadas-Shatzkes model (Ref. 12), dashed-dotted line is the Fuchs-Sondheimer expectation for pure La. Dashed lines are guides to the eye. See text for the determination of ρ_0.

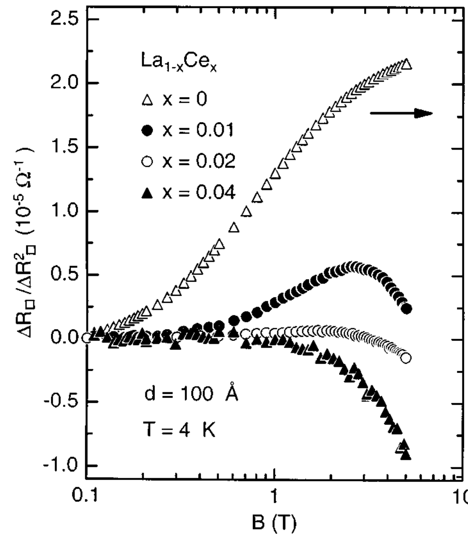

FIG. 2. Magnetoresistance $\Delta R_\square/R_\square^2=[R_\square(B)-R_\square(0)]/R_\square^2$ of $La_{1-x}Ce_x$ films of thickness $d=100$ Å vs magnetic field B (log-scale). Note the different vertical scales for pure La and $La_{1-x}Ce_x$.

were determined by quartz oscillators.

Reflection high-energy electron-diffraction (RHEED) measurements showed partly three-dimensional reflection patterns indicating considerable surface roughness. This was confirmed by taking an *in situ* scanning tunnel microscopy (STM) image on one of the $La_{0.96}Ce_{0.04}$ films ($d=100$ Å) which revealed a maximum surface roughness (peak to peak) of 30 Å. Auger depth profiling of a 200-Å La film (which was previously exposed to ambient air for several hours) showed no traces of oxygen until the sapphire substrate was exposed by sputtering, indicating the efficiency of the Ge protective layer. For $d \geqslant 200$ Å the crystal structure was determined by x-ray diffraction to be La dhcp with a fiber texture of [0001] along the growth direction.

The resistivity measurements were performed with standard four-probe methods using either silver paint or spring-loaded needles for contacts. The magnetic field was applied perpendicular to the film plane.

III. RESULTS AND DISCUSSION

A. Overview over the results

Before discussing our results in detail, a few general features will be presented in this section. Figure 1 shows the low-temperature resistivity ρ_0 as a function of inverse film thickness for $x=0$ and $x=0.04$. [Because of the different $\rho(T)$ behavior observed for different films we define ρ_0 as follows: for films exhibiting a resistance minimum, we take $\rho_0=\rho_{min}$, for the very thin films ($d \leqslant 80$ Å) where we found a negative $d\rho/dT$ up to room temperature, we take $\rho_0=\rho(15\text{ K})$, for the other (superconducting) films we take the residual resistivity in the normal state. We use ρ_0 only as a qualitative measure. The exact value of ρ_0 does not at all affect our analysis of the following sections.] ρ_0 depends much stronger on d^{-1} than expected from the classical Fuchs-Sondheimer theory (cf. dashed-dotted line in Fig. 1) which assumes boundary scattering of electrons, with bulk scattering being independent of the film thickness. In addition, grain-boundary scattering has to be taken into account.[12] In general, the average grain size decreases with decreasing d, leading to a shorter mean free path for thinner films. Assuming a constant ratio $q=3.8$ between film thickness and average grain size and a reflection coefficient $p=0.37$ from the grain boundaries, both $x=0$ and $x=0.04$ sets of data can be described quite well for $d>50$ and 90 Å, respectively (cf. solid lines in Fig. 1). The systematic difference between both data sets is, of course, due to the additional scattering by the Ce impurities. For lower d we observe strong deviations from the Mayadas-Shatzkes model[12] which probably must be attributed to the inhomogeneous film growth with a roughness of ~ 30 Å as mentioned above. Because of the strong influence of the changing morphology for thin d, samples with $d \leqslant 90$ Å ($x=0.04$) and $d \leqslant 50$ Å ($x=0$) are excluded from the following analysis and discussion.

As an introduction to the following discussion, we give in Fig. 2 an overview over the magnetoresistance $\Delta R_\square/R_\square^2=[R_\square(B)-R_\square(0)]/R_\square^2$ for 100-Å films of different concentrations at $T=4$ K. $R_\square=\rho/d$ is the square resistance. (Actually $\Delta R/R^2$ is the negative magnetoconductance.) For $x=0$ we observe a very strong positive magnetoresistance (MR) due to the suppression of superconducting fluctuations ($T_c=3.65$ K for this film). For $x=0.01$ these fluctuations are much weaker in our T range ($T_c=1.57$ K) and a maximum occurs because of the negative MR at large B arising from the suppression of Kondo scattering. This weak maximum is still visible for $x=0.02$ until finally for $x=0.04$ the MR is negative in the whole field range. Thus, as we shall see, only for this concentration the MR can be used to investigate the Kondo scattering without the complication of superconducting fluctuations.

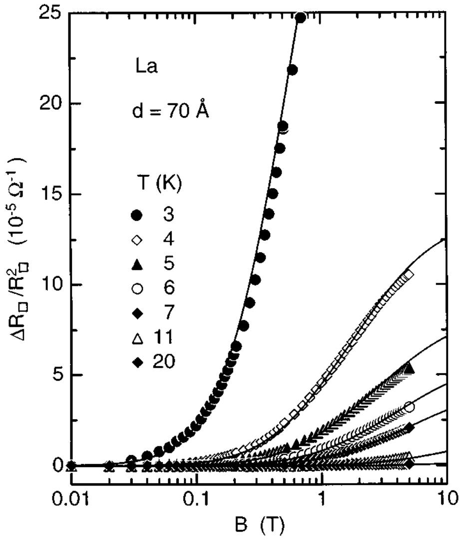

FIG. 3. Magnetoresistance $\Delta R_\square / R_\square^2$ of a 70-Å La film at various temperatures T. Solid lines indicate fits to Eq. (1) as described in the text.

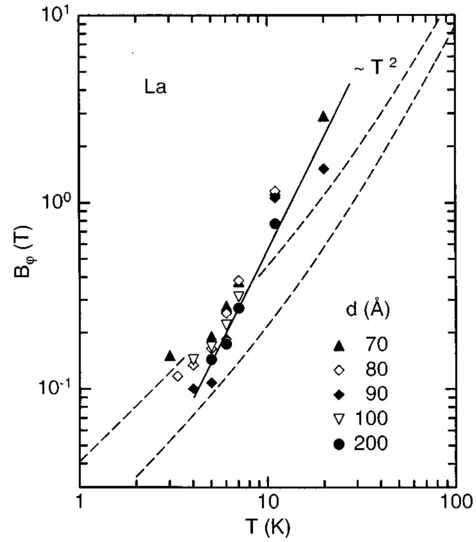

FIG. 4. Temperature dependence of the characteristic field B_ϕ of La films determined from fits as in Fig. 3 for various film thicknesses d. Dashed lines show the theoretically expected dependence for $d = 70$ Å (upper curve) and $d = 100$ Å (lower curve).

B. Resistivity of pure La films

Figure 3 shows the magnetoresistance $\Delta R_\square / R_\square^2$ of a 70-Å La film as a function of magnetic field B for various temperatures. The data are analyzed with the expression

$$\frac{\Delta R_\square}{R_\square^2} = -\Delta L_{\mathrm{WL}}(B) - \Delta L_{\mathrm{AL}}(B) - \Delta L_{\mathrm{MT}}(B), \qquad (1)$$

where $\Delta L_{\mathrm{WL}}, \Delta L_{\mathrm{AL}}$, and ΔL_{MT} describe the magnetoconductivity contribution due to destruction of weak localization (WL) and suppression of the Aslamazov-Larkin (AL) and Maki-Thompson (MT) contributions to the fluctuation conductivity of a superconductor above T_c, respectively. The individual contributions are given by[13–15]

$$\Delta L_{\mathrm{WL}}(B) = L_{00}\left[f\left(\frac{B_1}{B}\right) - \frac{3}{2} f\left(\frac{B_2}{B}\right) + \frac{1}{2} f\left(\frac{B_3}{B}\right) \right], \qquad (2)$$

$$\Delta L_{\mathrm{AL}}(B) = L_{00} \frac{\pi^2}{4} \frac{1}{\ln(T/T_c)} \left\{ \frac{2B_4}{B} - \left(\frac{2B_4}{B}\right)^2 \times \left[\Psi\left(1 + \frac{B_4}{B}\right) - \Psi\left(\frac{1}{2} + \frac{B_4}{B}\right) \right] - \frac{1}{2} \right\}, \qquad (3)$$

$$\Delta L_{\mathrm{MT}}(B) = L_{00} \frac{\pi^2}{4} \frac{1}{\ln(T/T_c)} \frac{B_4}{B_4 - B_3} \left\{ f\left(\frac{B_4}{B}\right) - f\left(\frac{B_3}{B}\right) \right\}, \qquad (4)$$

with $L_{00} = e^2/2\pi^2\hbar$, $f(B_i/B) = \Psi(\frac{1}{2} + B_i/B) - \ln(B_i/B)$ and Ψ the digamma function, $B_1 = B_0 + B_{\mathrm{so}} + B_s$, $B_2 = B_\phi + (4/3)(B_{\mathrm{so}} - B_s)$, $B_3 = B_\phi = B_{\mathrm{in}} + 2B_s$, $B_4 = 2k_B T \times \ln(T/T_c)/\pi e D$. These characteristic fields B_x correspond to the scattering rates τ_x^{-1} via $B_x = \hbar/4eD\tau_x$ where D is the electron diffusion constant. τ_0 denotes elastic scattering, τ_{in} inelastic scattering, τ_{so} spin-orbit scattering, and τ_s spin-flip scattering, respectively.

For the fits of the above expressions to our data the following parameters are used: $B_0 = 112$ T corresponding to $\tau_0 = 3/e^2 v_F^2 N(E_F)\rho_0$ with $v_F = 1.3 \times 10^5$ m/s, $N(E_F) = 2.86$ (eV atom)$^{-1}$ for La,[16,17] and our measured ρ_0 values, $B_{\mathrm{so}} = 0.01$ T, $B_s = 0$. For all films, B_ϕ was determined in the same way by using the corresponding input parameters. For a measured MR curve at a given T only B_ϕ was varied until a best fit was obtained. The example of Fig. 3 shows the quality of the fits. The resulting $B_\phi(T)$ is shown for different films in Fig. 4. The data can be compared with the theoretically expected $B_\phi(T)$ for our La films, taking into account the effect of electron-phonon interaction and EEI as done previously by Bergmann[18] for Cu, Ag, and Au films. Although $B_\phi(T)$ is compatible with a T^2 dependence expected for electron-phonon-scattering[18] the measured $B_\phi(T)$ values strongly deviate from the theoretical curves at high T. Since the difference between the data and theory is smaller at lower temperatures, a transition from two- to three-dimensional behavior with respect to the wavelength of dominant phonons might be responsible for the discrepancy. Furthermore, enhanced electron-phonon interaction in disordered films and proximity of the unoccupied $4f^0$ level to the Fermi level, which is held responsible for the anomalous $\rho(T)$ dependence of La,[20] might be important. In the above analysis, we have not included the influence of EEI on the magnetoresistance due to spin-splitting effects[19] which are estimated to be of the order of $10^{-7}\ \Omega_\square^{-1}$.

Equations (3) and (4) are valid only if $\hbar/k_B T\tau_{\mathrm{in}} \ll 1$ and $4eDB/k_B T \ll 1$. An extension of the theory to a larger range of parameters,[21] limited only by the condition $4eDB \ll \hbar/\tau_0$, yields essentially the same fits to our data. One can also reasonably well describe the temperature dependence of the resistivity above T_c (not shown) with AL and MT contributions.

We mention that in our La films the T_c depression with increasing $R_\square$ cannot be described with reasonable values of the coupling constant $g_1 N(E_F)$ in the Maekawa-Fukuyama theory.[22] Probably, the changing film morphology with thickness d already inferred above from the RHEED, STM,

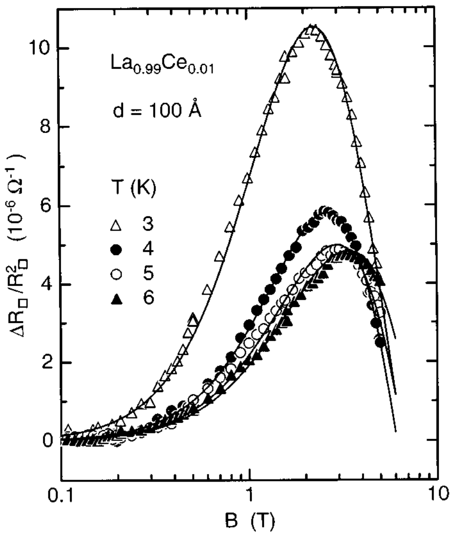

FIG. 5. Magnetoresistance of a $La_{0.99}Ce_{0.01}$ film at various temperatures T. Solid lines indicate fits of Eq. (6) to the data.

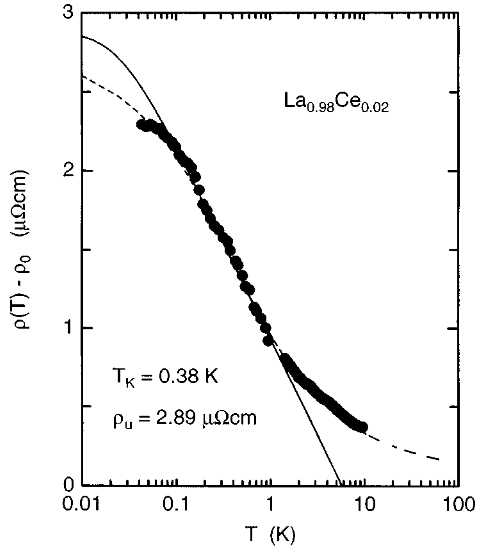

FIG. 6. Resistivity $[\rho(T)-\rho_0]$ vs ln T for a $La_{0.98}Ce_{0.02}$ film with $d=100$ Å. Solid line is an empirical fit using Eq. (7), dashed curve shows the behavior expected from the Suhl-Nagaoka theory.

and residual-resistivity data is responsible for the large apparent coupling constant $g_1N(E_F) = 5$ found in our films. In contrast to previous experiments on a-Mo-Ge,[23] $[\rho_0(d=\infty)-\rho_0(d)]/\rho_0^2(d)$ is not constant for our samples. Thus, the ''bulk'' T_{c0} by itself may change by the gradual change in the microstructure, leading to an overestimation in $g_1N(E_F)$.

C. Superconducting fluctuations and Kondo effect for $x=0.01$

As already mentioned in Sec. III A, superconducting fluctuations and Kondo effect superimpose to give a maximum in the magnetoresistance $\Delta R_\Box/R_\Box^2$ vs B for $x = 0.01$. Figure 5 shows the experimental data for several temperatures. The maximum shifts systematically to higher fields with increasing T. This confirms the above interpretation since a maximum resulting from a superposition of weak localization and weak antilocalization would be independent of B or shift to lower B with increasing T, depending on the presence or absence of magnetic scatterers.[13] In addition, the magnetic scattering rate[24] which is related to the characteristic field B_s

$$\tau_s^{-1}=4eDB_s/\hbar=xN(E_F)|J|^2S(S+1)/\hbar, \qquad (5)$$

yields roughly $B_s=0.5$ T when using $|J|\approx 0.1$ eV as deduced below. J is the exchange interaction between the conduction electrons and the localized $4f$ moments. (For this estimate and in the following, we neglect the weak logarithmic T dependence of the magnetic scattering rate.) These large magnetic scattering rates are expected to destroy the phase coherence necessary for WL and antilocalization completely.

The fit in Fig. 5 describes

$$\frac{\Delta R_\Box}{R_\Box^2}=-\Delta L_{\mathrm{AL}}-\Delta L_{\mathrm{MT}}+\frac{\Delta\rho_K}{R_\Box^2 d}, \qquad (6)$$

where the Kondo magnetoresistivity $\Delta\rho_K=\rho_K(B)-\rho_K(0)$ has been calculated by Beal-Monod and Weiner [Eq. (25) in Ref. 25] where J enters as an important parameter. Although the application of Eq. (4) for the MT contribution in the presence of magnetic scatterers might be questionable, the fits work very well. With $T_c = 1.57$ K as determined resistively, we obtain $J=-0.15$ eV from the fit using Eq. (6). The characteristic fields B_ϕ obtained from the MT term can be described by $B_\phi=2B_s+B_{\mathrm{in}}=2B_s+\lambda T^2$, with $B_s=0.24$ T and $\lambda=6\times10^{-3}T/K^2$. The value of B_s inferred gives $|J| = 0.072$ eV via Eq. (5). The difference between the two values of J thus obtained could be due to the application of Eq. (4) on a superconductor with magnetic impurities. Another determination of J for this concentration is available from the T_c depression.[26] Assuming an initial linear depression we obtain, with $T_{c0} = 3.65$ K for the 100-Å pure La film, $\Delta T_c/\Delta x = 2.1$ K/at. % yielding $|J| = 0.10$ eV [Eq. (5)]. $\Delta T_c/\Delta x$ is only in poor agreement with data for $T_c(x)$ of bulk dhcp $La_{1-x}Ce_x$ alloys[10] where $\Delta T_c/\Delta x = 1.22$ K/at. %. A possible origin for the discrepancy is the proximity to the suppression of superconductivity due to incipient localization, although this speculation needs further investigation. In estimating $|J|$ from $\Delta T_c/\Delta x$ we have used the simple expression Eq. (5) valid for temperature-independent scattering which is a good approximation down to $\sim T_{c0}/2$ if $T_{c0}\gg T_K$ as in the present case. In summary, the values of J obtained from the Beal-Monod and Weiner theory and the T_c depression are in reasonable agreement with $|J|\approx 0.12$ obtained from MR data for $x=0.04$ (see below).

D. Determination of the Kondo temperature for $x=0.02$

In order to determine the Kondo temperature T_K, the resistivity of a $x = 0.02$ film with $d=100$ Å was measured down to 40 mK. The result is shown in Fig. 6, together with a fit described by the empirical formula[27]

$$\rho_K(T)=\frac{\rho_u}{2}\left\{1-\frac{\ln[(\theta^2+T^2)/T_K]}{2\pi\sqrt{S(S+1)}}\right\} \qquad (7)$$

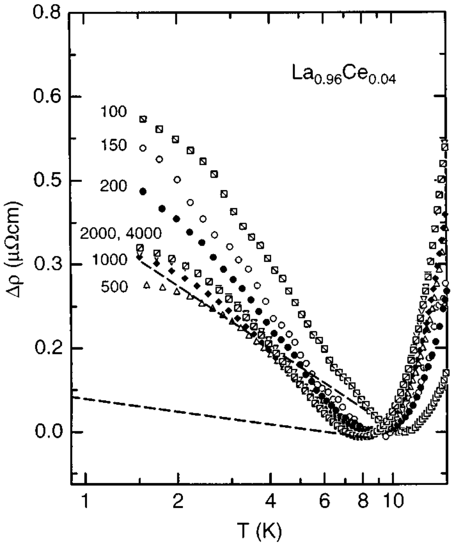

FIG. 7. Resistivity $\Delta\rho=[\rho(T)-\rho_0]$ vs temperature T for $La_{0.96}Ce_{0.04}$ films of various thickness d. Dashed lines show the estimated contribution of EEI for d = 100 Å (upper curve) and d = 200 Å (lower curve).

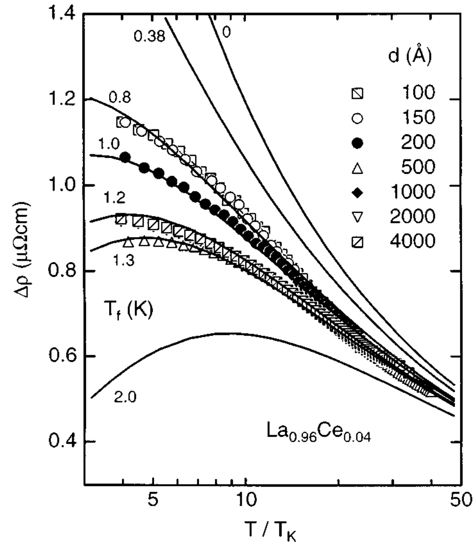

FIG. 8. Resistivity contribution $\Delta\rho=\rho-\rho_{ph}$ vs reduced temperature T/T_K for $La_{0.96}Ce_{0.04}$ films of various thickness d. See text for the determination of the electron-phonon resistivity ρ_{ph}. Solid lines indicate fits of Eq. (9) for different T_f.

for $S=1/2$, where $\ln(\theta/T_K)=-\pi\sqrt{S(S+1)}$ (Fig. 6, solid line). The phonon contribution which becomes important above ~ 8 K, was determined by assuming the same T dependence as measured for a pure La film with the same resistivity ratio, i.e., the same degree of structural disorder. In addition we plot the behavior of ρ_K from the Suhl-Nagaoka theory[27] (dashed line) which is valid for temperatures above T_K, without taking potential scattering into account.

The fit yields the unitarity limit $\rho_u=2.89$ $\mu\Omega$ cm and T_K = 0.38 K. The latter value is in reasonable agreement with other determinations of T_K in $La_{1-x}Ce_x$ bulk alloys which fall in the range of 0.15 to 0.60 K as obtained by different methods.[24,28,29] Taking the T_c depression of the x = 0.01 film ($d=100$ Å) with respect to the pure La film of the same thickness, one obtains $T_K=0.44$ K in good agreement with the above value, but somewhat higher than obtained from the T_c depression in bulk $La_{1-x}Ce_x$ alloys[10] as noted above.

Whereas an influence of WL can be neglected due to the high concentration of magnetic impurities the contribution from EEI on $\rho(T)$ has to be considered. In principle this could be estimated from magnetoresistance data. However, due to the presence of residual superconducting fluctuations in the magnetoresistance data (Fig. 2) we are not able to extract an estimate for the magnitude of $\rho_{ee}(T)$. In any case, ρ_u is comparable to the value 5.13 $\mu\Omega$ cm measured for bulk $(La_{0.982}Ce_{0.008})B_6$ (Ref. 30).

E. Resistivity and magnetoresistivity for $x=0.04$

Figure 7 shows the resistivity of all investigated $La_{0.96}Ce_{0.04}$ films with $\rho_0=\rho_{min}$ substracted. For $d<500$ Å we observe a strong *increase* of the T dependence with decreasing thickness d. For $d>500$ Å the thickness dependence of $\Delta\rho$ is much weaker, here $\Delta\rho$ tends to decrease with decreasing d. The leveling off of the logarithmic resistivity increase $d\rho/[d\ln(T/T_0)]$ towards low T for the films with $d\geqslant 100$ Å can be attributed to interaction between Ce moments eventually leading to spin-glass behavior. Effects of WL to $\rho(T)$ can be ignored due to the high concentration of magnetic impurities as mentioned above for $x=0.01$. The contribution from EEI, $\Delta\rho_{ee}(T)=-AL_{00}R_{\square}\rho_0\ln(T/1\ \mathrm{K})$ (Ref. 19), has been roughly estimated for some samples with $A=1$ (dashed lines). For $d\geqslant 200$ Å EEI can be neglected but might give a considerable contribution for thinner films. We repeat however, that the EEI contribution to the magnetoresistivity is very small.

A description of $\rho(T)$ for spin glasses with Kondo impurities has been worked out by Larsen who finds[31,32]

$$\rho_{SG}(T)\sim\left\{1+\left[\ln\left(\frac{2\pi T}{T_K}\right)+\Psi\left(\frac{1}{2}+\frac{\Delta}{2\pi T}\right)\right]^2 \Big/ P\left(\frac{T}{\Delta}\right)\right\}^{-1} \tag{8}$$

for $S=1/2$. Here $\Delta(T)$ describes the average coupling between the Kondo impurities. $P(T/\Delta)$ is a lengthy expression which can only be evaluated numerically.[31] In the spin-glass regime the spin-glass freezing temperature $T_f\approx\Delta(0)$ is related to the temperature T_{max} of the maximum of the resistivity ρ_{SG}. Figure 8 shows the magnetic contribution $\Delta\rho$ to ρ together with numerical calculations of Eq. (8) for different values of T_f vs reduced temperature T/T_K. We have used T_K = 0.38 K as determined in Sec. III D and estimated T_f from T_{max} with an expression given by Larsen.[31] The phonon-scattering contribution to $\rho(T)$ was determined in the same way as described in Sec. III D. For the thickest films where localization and interaction effects should be negligible, we infer $T_f\approx$ 1.2 K from $T_{max}\approx$ 1.5 K which is in good agreement with bulk data, where T_{max} = 3.15 K for x = 0.1 and T_{max}=5.1 K for x = 0.2 was found.[33]

For thinner films with $d<500$ Å the data can be well described by the calculations down to $d=100$ Å yielding a T_f that decreases with decreasing d. Although this can be qualitatively explained by the usual mean-field decrease of

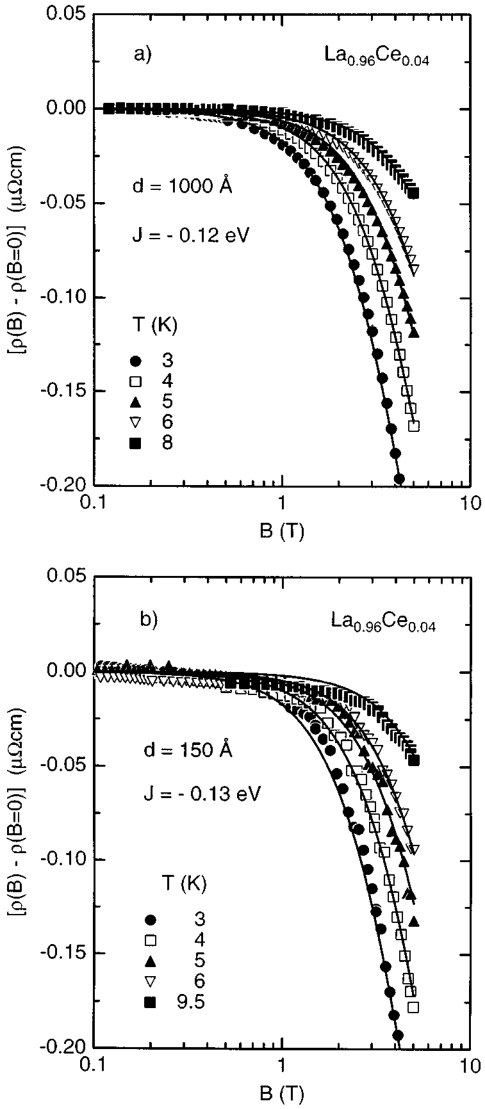

FIG. 9. Magnetoresistivity $\Delta\rho=\rho(B)-\rho(0)$ for $La_{0.96}Ce_{0.04}$ films with d = 1000 Å (a) and 150 Å (b). Solid lines indicate fits to the Beal-Monod–Weiner theory (Ref. 25).

the interaction with decreasing number of "neighbors," a decrease of T_f because of the decreasing electron mean free path (with decreasing d) is more likely.[34]

We now turn to the magnetoresistance data, two examples of which are shown in Fig. 9. As already anticipated in Sec. III A, $\Delta R_{\square}/R_{\square}^2$ is negative in the whole field range investigated. The fits obtained using the single-ion Kondo theory for the magnetoresistance[25] already employed for x = 0.01 (cf. Sec. III C) work remarkably well. The only free parameter in these fits is the Kondo exchange integral J. We used the following parameters: E_F = 6 eV, atomic volume 3.75×10^{-29} m^{-3} for La, number of conduction electrons per atom z = 3, and spin S = 1/2. For $\langle S_z\rangle$, the thermal average of the local moment, we took the Brillouin function. At first sight it might be surprising that we can describe our data so well with *independent* Ce moments, in view of the spin-glass order inferred above. However, we should note that our T range is still well above T_f for all films and, furthermore, a strong magnetic field $g\mu_B B > k_B T_f$ easily destroys the spin-glass correlations. The similarity of the magnetoresistance data for different film thicknesses (Fig. 9) is in contrast to a recent magnetoresistance study of thin AuFe films, where a suppression of $\Delta\rho=\rho(0)-\rho(4\ \mathrm{T})$ with film thickness d has been reported.[35]

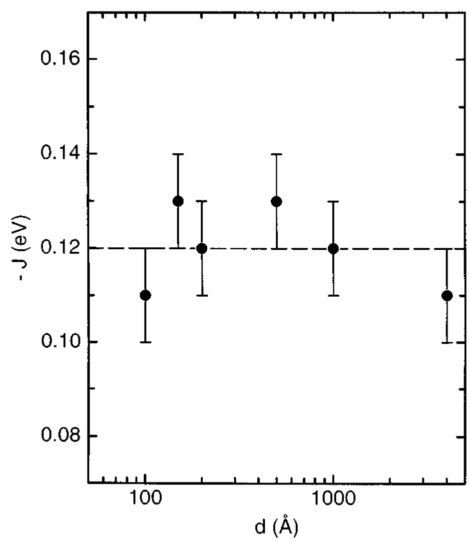

FIG. 10. Kondo exchange integral J as a function of film thickness d for $La_{0.96}Ce_{0.04}$ films.

Figure 10 shows J obtained from our fit as a function of d. Within the error bar, we find $J=-(0.12\pm0.01)$ eV for all investigated thicknesses from 4000 down to 100 Å , in good agreement with $J=-0.1$ eV for x = 0.01 (Sec. III C). This clearly demonstrates that J, being a local property, is independent of the film thickness. J lies between the values of earlier studies, where $J=-0.22$ eV was obtained from resistivity data,[28] and $J=-0.08$ eV inferred from the T_c depression in La-Ce alloys.[36]

In order to determine a possible influence of film thickness on the Kondo temperature from magnetoresistance data, one has to resort to more elaborate theories which go beyond perturbation theory. It has been shown[37] that the MR of a Kondo system with a given phase shift δ is a universal function of T/T_K and B/B_K where $B_K=k_B T_K/g\mu_B$ with the appropriate g factor (T_K is the zero-field Kondo temperature). Unfortunately, the numerical evaluation of the function is very involved. Furthermore, the phase shift δ and the resistivity $\rho(T=0,\ B=0)$ which are necessary for a comparison with the theory are not known for our samples. Thus, from the MR measurements we cannot unambiguously reveal a possible thickness dependence of the Kondo temperature.

IV. CONCLUDING DISCUSSION

For $La_{1-x}Ce_x$ we have shown that the Kondo temperature for a film with $d=100$ Å (x = 0.02) is in good agreement with bulk data. This thickness is well below $R_K=0.4$ μm or $R_K^*=\sqrt{R_K l_e}$ = 350 Å , supporting a thickness independent T_K. However, the analysis is complicated by the unknown contribution of EEI to $\rho(T)$ due to the high resistance of our films. In addition, in $La_{0.96}Ce_{0.04}$ films we have determined the Kondo exchange interaction J between the conduction electrons and the localized f moments from magnetoresistance data for which EEI contributions can be neglected. J is found to be independent of film thickness down to 100 Å. This nicely reflects the nature of J as a local property and must be contrasted with the strong thickness dependence of $\rho(T)$ as shown in Figs. 7 and 8. For very thin films ($d \leq 80$ Å) the strong disorder leads to a very high resistivity above 200 $\mu\Omega$ cm (cf. Fig. 1). The incipient electron localization leads to $d\rho/dT < 0$ up to room temperature (in agreement with the Mooij criterion[38]), and at low temperatures $\rho(T)$ is mostly governed by disorder and EEI. The decrease of $d\rho/dT$ with increasing d in the medium thickness range can be explained by the onset of spin-glass ordering in thicker films which weakens the Kondo scattering together with gradual decrease of EEI. Finally, the slight increase of $d\rho/dT$ between 500 and 4000 Å is what might be expected from a ''geometry dependence'' of the Kondo effect. We note, however, that in the same d range the magnetoresistance actually suggests a slight decrease of J, albeit within the error bars, cf. Fig. 10. Therefore, the apparent thickness dependence of $d\rho/dT$ in this d range *cannot* be attributed to a size dependence of the Kondo resistivity contribution as observed for CuFe and AuFe films.[2–4]

In the CuFe and AuFe system there is a length scale $l_K \approx 1500$ Å below which the Kondo resistivity $\Delta\rho_K$ is suppressed whereas T_K itself is independent of film thickness.[2–4] It has been shown that R_K does not control the Kondo behavior, neither in the ballistic nor in the diffusive regime [$R_K \approx 0.1$ μm (CuFe) and 2 μm (AuFe)]. For films, contributions from WL and EEI were neglected because of the small values of $R_\square$. For wires of width $w < 1000$ Å the EEI contributions were of the order of ρ_K and prevented a detailed analysis. In contrast, other experiments on AuFe wires[9] confirmed that the slope of ρ_K was independent down to widths of $w = 380$ Å. The temperature dependence of the resistivity of Kondo alloys with restricted geometries might not be a good probe to look for a geometry dependence unless the effects of disorder, WL and EEI effects are properly accounted for as has been done in Ref. 9.

Recently, an *increase* of the Kondo temperature has been reported for CuMn point contacts with decreasing point-contact diameter, prepared by the break-junction technique.[39] The apparent increase of T_K as inferred from the increasing width of a zero-bias anomaly in the differential resistance dV/dI vs voltage V was observed over a range of point-contact diameters between 1.5 and 50 nm. A difference to our work is that the (dimensionless) conductance G as measured in units of $e^2/\hbar$ is certainly very large in our films, even for the thinnest films of 100 Å used for the Kondo analysis, compared to the point contacts where the electron motion is more strongly confined. Thus, again, we expect EEI and localization effects to be more important in the latter. Indeed, a stronger increase of the resistivity with decreasing T for films of smaller width w was observed in $Au_{1-x}Fe_x$ films.[9] For films with $w = 38$ nm ($d_{\mathrm{eff}} = \sqrt{dw} = 33$ nm) the slope of $d\rho/d\ln T$ was by a factor of ~ 1.5 larger than for $w = 115$ nm ($d_{\mathrm{eff}} = 59$ nm). For larger w no big changes were observed in $d\rho/d\ln T$. These findings are overall consistent with the changes of $d\rho/d\ln V$ in the point-contact measurements, with the same diameter range as d_{eff}, suggesting that EEI are important. Further work has to investigate the transition from thin films of narrow width to point contacts in more detail. In particular, the applied voltage drops over a small distance in the point contact (of the order of the diameter), leading to a stronger local deviation from equilibrium than in the wires investigated in Ref. 9 and the films investigated in the present work. Another possibility that has to be investigated is the evolution of a nonmagnetic Kondo effect due to the fluctuations of scattering centers (''two-channel Kondo effect'').

Furthermore, one has to consider the different atomic environment for Ce impurities located near the film boundary compared to those in the inner part which may lead to a distribution of Kondo temperatures.[40] Very recently, it has been shown theoretically that the occurrence of a surface-induced perpendicular magnetic anisotropy is able to explain the experimental data for $Au_{1-x}Fe_x$ films and wires.[41] This anisotropy leads to a freezing out of an $S = 5/2$ or $S = 2$ spin into an $S_z = 1/2$ or $S_z = 0$ state with decreasing T, thus reducing the spin-flip scattering. However, this effect should not be present in $La_{1-x}Ce_x$ alloys with a doubly degenerate ground state ($S = 1/2$).

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