Magnetoresistance of Kondo Lattices

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Magnetoresistance at 0 K is theoretically examined. Because Kondo temperatures must be very sensitive to local disorder of ligands around f ions as implied by their large Grüneisen constant, they can be random in the neighborhood of defects and dislocations. The disorder of Kondo temperatures gives positive magnetoresistance at low fields, but negative magnetoresistance at high fields; the magnetoresistance has a peak, when the magnetization is about half of its saturated value. If Kondo temperatures are distributed down to the zero energy, the magnetoresistance can be positively linear to fields at low fields.

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Anomalously large magnetoresistance (MR) is commonly observed in heavy-electron compounds. It is negative at high temperatures, while it is positive at temperatures much lower than the Kondo temperature (T_K).[1-5] It is too large to be explained by orbital effects of multicarriers in terms of $(\omega_c\tau)^2$, where ω_c is the cyclotron frequency of heavy electrons and τ is their relaxation time. Large MR has been observed in the region of $\omega_c\tau \ll 1$.

Because it can be observed even in nominally pure and undoped compounds, the positive MR may be considered as an intrinsic effect and evidence that heavy electrons are condensed into a coherent state at low temperatures.[5] Kawakami and Okiji proposed an intrinsic mechanism of a metal-insulator transition in one spin component.[6] It is well known that the hybridization between f bands and conduction bands produces a hybridization gap. They assumed that the chemical potential lies outside the gap in the absence of fields. Then they claimed that in the presence of fields the chemical potential of one spin component of quasiparticles should move relatively within the gap because of the Zeeman energy.

However, observed data can be fitted by a form

$$\rho(H,T)=\rho_0(H)+A(H)T^n \tag{1}$$

at low enough temperatures, where H is magnetic field and T is temperature. It is clear that $\rho_0(H)$ should vanish in ideally periodic systems. Therefore, the field dependence of $\rho_0(H)$ should be related to a certain kind of disorder. The Grüneisen constant of T_K is commonly large in heavy-electron compounds. For example, it is as large as 200 in $\mathrm{CeRu_2Si_2}$.[7,8] The large Grüneisen constant is one of the main origins of the Kondo volume collapse,[9,10] and the metamagnetism of $\mathrm{CeRu_2Si_2}$.[11] The large Grüneisen constant implies that T_K must be very sensitive to the configuration of the nearest-neighbor ligands around f ions. The present author proposed a mechanism of positive MR.[12] His main assumption is that T_K can be defined in each f-ion site, and that it is random in the neighborhood of defects and dislocations due to disordered ligands. Then he showed that

$$\Delta\rho_0(H)=x\alpha\rho_u\left(\frac{\delta T_K}{T_K}\right)^2\left(\frac{\mu_B H}{T_K}\right)^2 \tag{2}$$

at low fields, where x is the concentration of impurity sites with disordered ligands, $\alpha \approx O(1)$ $(\alpha > 0)$, ρ_u is the unitarity limit resistivity, δT_K is the magnitude of the disorder of T_K, and $\mu_B H$ is the Zeeman energy. Although the proportionality to H^2 in Eq. (2) is consistent with time-reversal symmetry, recent experiments show that $\Delta\rho_0(H)$ often seems to be proportional to H itself.[13,14] Furthermore, his examination was restricted to the low-field regime of $\mu_B H \ll T_K$. The purposes of the present Letter are to extend his theory, and to propose a mechanism of the linear positive MR.

One of the most characteristic features of Kondo lattices is the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction. If the Kondo effect is stronger than the RKKY exchange interaction, the single-site picture is expected to be valid in the zeroth approximation even at 0 K. Such a starting point is valid in the large-N limit, N being the degeneracy of f levels. According to the Friedel sum rule,[15] the phase shift is given by the number of localized electrons. Such a phase shift can describe many-body scatterings by a single f ion at 0 K, although the Kondo screening takes place over many unit cells. Our essential assumption is the same as that of Razafimandimby, Fulde, and Keller.[16] Kondo lattices at 0 K can be described by a set of phase shifts defined at each unit cell. Then a coherent Kondo state at 0 K, or a Fermi-liquid state, can be obtained by solving the scattering problem described by the phase shifts defined at each site.

Then let us calculate the phase shift or the number of localized f electrons by using an impurity Anderson model with infinitely large correlation. The energy gain due to the Kondo effect per unit cell is obtained as

$$E_s = -\frac{\Delta}{\pi}\ln\frac{D}{\Delta} - [T_K^2+(\mu_B H)^2]^{1/2},$$

with $T_K = (D\Delta)\exp[\pi(\epsilon_f-\mu)/2\Delta]$ the Kondo temperature in the $1/N$-expansion method, $N=2$ here, where Δ is the hybridization energy, D is the width of conduction bands, ϵ_f is the position of f levels, and μ is the chemical potential. Here more than one conduction band is assumed to make the system metallic.[17] The dependence of T_K on fields can be ignored, as long as $\Delta \gg \mu_B H$. By using the relation $n_\uparrow - n_\downarrow = -\partial E_s/\partial(\mu_B H)$, the spin polarization is calculated as

$$n_\uparrow - n_\downarrow = \frac{\mu_B H}{[T_K^2+(\mu_B H)^2]^{1/2}},$$

where n_σ is the number of f electrons with spin σ per unit cell. Let us assume $\pi|\epsilon_f-\mu|/2\Delta \ll 1$ so that the number of f electrons might be very close to one per site; $n_\uparrow + n_\downarrow \approx 1$. Then the phase shift can be easily calculated by the Friedel sum rule:[15]

$$\delta_\sigma(H;T_K) = \frac{\pi}{2}\left[1+\sigma\frac{\mu_B H}{[T_K^2+(\mu_B H)^2]^{1/2}}\right].$$

Let us examine a case where the distribution of T_K is given by $P(\epsilon)$; the distribution of T_K is mainly due to the distribution of hybridization energy Δ around dislocations and defects. Then the average phase shift

$$\langle\delta_\sigma\rangle = \int_0^\infty d\epsilon\, P(\epsilon)\delta_\sigma(H;\epsilon)$$

can be defined. Here and in the following, $\langle A(\epsilon)\rangle$ is the average of $A(\epsilon)$ with the weight of $P(\epsilon)$; $P(\epsilon)$ is normalized such as $\langle 1\rangle = 1$. Let us assume a virtual crystal which has a phase shift of $\langle\delta_\sigma\rangle$ on every f-ion site. If this scattering problem is solved, quasiparticles at the chemical potential can be correctly given. It is clear that the coherent scatterings described by $\langle\delta_\sigma\rangle$ at each site do not contribute to the residual resistivity; a coherent Kondo lattice is realized. Then one can argue that the difference of the phase shift from $\langle\delta_\sigma\rangle$ gives the residual resistivity. Because of the approximation of $n_\uparrow + n_\downarrow = 1$, the conductivity is the same between up and down spins, and the residual resistivity is simply given by

$$\rho_0(H) = \rho_\mu\langle\sin^2[\delta_\uparrow(H;\epsilon)-\langle\delta_\uparrow\rangle]\rangle. \qquad (3)$$

Although T_K can be different around impurity sites, the phase shift is $\pi/2$ at any site in the absence of fields because of the approximation of $n_\uparrow + n_\downarrow = 1$. Therefore, the residual resistivity vanishes in the absence of fields in the present approximation.

The residual resistivity in the presence of fields is numerically calculated by assuming a square distribution of T_K: $P(\epsilon) = P_s(\epsilon)$, where $P_s(\epsilon) = 1/2sT_K$ for $(1-s)T_K \leq \epsilon \leq (1+s)T_K$, but $P_s(\epsilon) = 0$ otherwise. Figure 1 shows $\rho_0(H)$ as a function of normalized fields defined by $h = \mu_B H/T_K$. Equation (3) is reduced to

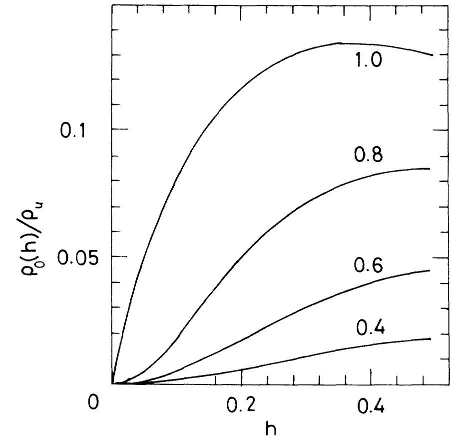

FIG. 1. $\rho_0(h)/\rho_u$ due to disordered ligands as a function of $h = \mu_B H/T_K$ for various s.

$$\rho_0(H) = \rho_u\frac{\pi^2}{4}\left[\frac{1}{1-s^2}-\frac{1}{2s}\ln\frac{1+s}{1-s}\right]\left[\frac{\mu_B H}{T_K}\right]^2$$

for $\mu_B H \ll (1-s)T_K$. The results are consistent with Eq. (2) at low fields except in the case of $s \approx 1$. However, $\rho_0(H)$ is proportional to H in the case of $s=1$, where T_K is distributed down to the zero energy and $P(\epsilon \to 0)$ is finite. Because a quadratic behavior is only expected for $\mu_B H \ll (1-s)T_K$, the quadratic region vanishes in such a case. Actually, for finite $P(0)$ Eq. (3) can be calculated as

$$\rho_0(H\to 0) = \beta\rho_\mu P(0)\mu_B H + O(H^2),$$

with $\beta \approx 2.622677\ldots$. On the other hand, Eq. (3) is reduced to

$$\rho_0(H) = \rho_u\frac{\pi^2}{12}s^2(1+\tfrac{1}{9}s^2)\left[\frac{T_K}{\mu_B H}\right]^4$$

for $\mu_B H \gg (1+s)T_K$. The resistivity decreases with increasing fields, and it vanishes in the limit of strong fields, because the phase shifts tend to π and 0 for up and down spins, respectively, at any site. Therefore, the MR is negative at high fields.

The RKKY exchange interaction has been ignored so far. As examined in Refs. 11 and 18, its lowest contribution can be included as a molecular field. In order to include the effects of molecular fields, Fig. 2 shows the residual resistivity as a function of the spin polarization defined by $m = (\langle\delta_\uparrow\rangle - \langle\delta_\downarrow\rangle)/\pi$ instead of fields; m should be determined by molecular fields as well as external fields. The MR has a maximum around $m \approx 0.5$ for any distribution of T_K. The dependence on m is nearly symmetrical around half of its saturated value.

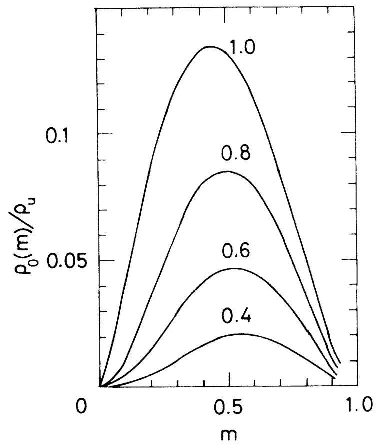

FIG. 2. $\rho_0(m)/\rho_u$ due to disordered ligands as a function of $m = M/\mu_B$ for various s.

Only the square-type distribution of T_K has been assumed so far. However, the following distribution of T_K is the most probable in actual compounds:

$$P(\epsilon) = (1-x)\delta(\epsilon - T_K) + xP_s(\epsilon) ,$$

where almost all sites have regular ligands, and there is a small number of impurity sites with the concentration of x $(0 < x < 1)$, for example, along dislocation lines.[19] The MR in such a case can be well described by multiplying the above results by the impurity concentration x.

As mentioned above, the residual resistivity vanishes in the absence of fields in the present approximation of $n_\uparrow + n_\downarrow = 1$. If the numbers of localized electrons are more precisely treated, the residual resistivity becomes finite, and the MR examined here should be proportional to the zero-field residual resistivity. However, the present theory predicts that large MR can be expected even if the residual resistivity is small. Because the positive MR examined here is not any intrinsic effect, it should depend on the sample preparation such as annealing time and radiation damage. It is interesting to examine the sample dependence.

At finite temperatures, inelastic scatterings become dominant in the Kondo effect. However, such inelastic scatterings cannot be fully described by any phase shifts. Therefore, the present theory can only be applied to the low-temperature regime as $T \ll T_K$ where elastic scatterings are dominant. When inelastic scatterings are dominant, however, the MR should be negative because magnetic fields depress such inelastic scatterings.

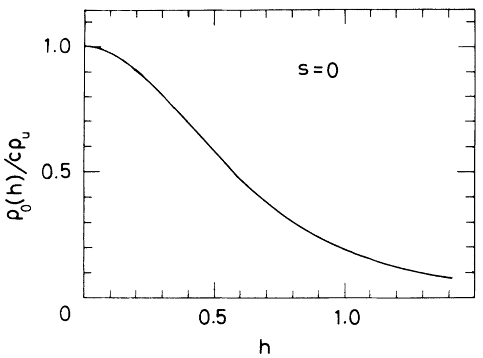

FIG. 3. $\rho_0(h)/c\rho_u$ due to Kondo holes as a function of h; c is the concentration of Kondo holes.

Besides f ions located in disordered ligands, other most probable impurities are the so-called Kondo holes; f ions, such as Ce ions, are replaced by non-f ions, such as La ions. Because the phase shift at non-f-ion sites is zero, the resistivity due to Kondo holes is easily calculated as $\rho_0(H) = c\rho_u \sin^2\langle\delta_\sigma\rangle$, where c is the concentration of Kondo holes. Figures 3 and 4 show MR due to Kondo holes; Fig. 4 can be regarded to include the effects of molecular fields. Because the results are very insensitive to s, the results for $s=0$ are only shown here. The dependence is the same as the negative MR of dilute Kondo alloys.

In actual compounds, two kinds of impurities must coexist such as disordered ligands and Kondo holes. In the case of disordered ligands, the MR has a maximum around $m \approx 0.5$ and the dependence on m is nearly symmetrical, as shown in Fig. 2. On the other hand, it is monotonically decreasing with increasing m in the case of Kondo holes. When $\rho_0(H)$, which can be obtained from an extrapolation of observed $\rho(H,T)$ according to Eq. (1), is plotted as a function of magnetizations, their shapes indicate the distribution of impurities. For example, if their shapes are symmetrical, one can expect that the main impurities are disordered ligands. There is no chance that positive MR can be still observed even when $m > 0.5$.

CeRu$_2$Si$_2$ shows large MR at low temperatures.[20] The MR changes its signs around $H=8$ T, where a metamagnetic transition occurs. Because the magnetization is about half of its saturated value at the metamagnetic point, the change of signs is consistent with the present prediction. It is interesting to plot the MR as a function of magnetizations instead of fields. Recently, positively

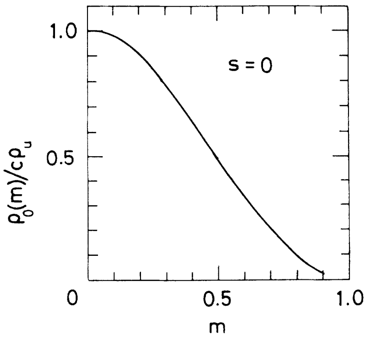

FIG. 4. $\rho_0(m)/c\rho_u$ due to Kondo holes as a function of m.

linear MR has been often observed at low fields.[13,14] One of the possible origins is the distribution of the Kondo temperatures down to very low energies. No quadratic region can be observed for $s=1$. Even for $s=0.4$ or 0.6, the quadratic dependence can be expected only at very low fields ($\mu_B H/T_K < 0.1$).

To summarize the present paper, the magnetoresistance of Kondo lattices is examined by assuming two kinds of impurities. Disordered ligands give positive magnetoresistance at low fields, and negative magnetoresistance at high fields. The magnetoresistance has a peak when the magnetization is about half of its saturated value. On the other hand, non-f ions replacing f ions give negative magnetoresistance at any fields. Therefore, positive magnetoresistance should vanish in any case, when the magnetization is larger than about half of its saturated value. When Kondo temperatures are distributed down to the zero energy due to disordered ligands, the positive magnetoresistance can be linearly proportional to fields at low fields.

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