Magnetoresistance in heavy-fermion alloys

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(Received 27 July 1987)

The spin-flip scattering of an electron by a pair of adjacent magnetic impurities is calculated in the presence of a magnetic field. In the limit of zero field the pair scattering yields a sharp drop in the low-temperature resistivity whose strength is inversely proportional to T. This process provides an explanation for the anomalous drop in resistivity below $T \leq 100$ K for UPt_3 and many other heavy-fermion alloys. An external magnetic field is shown to quench this spin-flip contribution, resulting in a positive magnetoresistance for UPt_3 at low temperatures. In conjunction with the single-impurity Kondo scattering, the pair process yields an interesting field dependence for the magnetoresistance at various temperatures and may cause a change of sign in the magnetoresistance as well as in the Hall effect.

I. INTRODUCTION

The "heavy-fermion" alloys are notable for extremely large magnetic susceptibilities and a sharp increase in the specific heat at low temperature. If one extrapolated the specific heat to zero temperature and invokes a conventional free-electron energy $\varepsilon = p^2/2m^*$, then a value of $m^* \sim 200 m_e$ is needed to describe the data. Despite the evident inadequacy of the free-electron model for these materials, the heavy-fermion nomenclature has persisted.

The discovery of superconductivity in $CeCu_2Si_2$ by Steglich *et al.*[1] challenged the foundations of condensed-matter physics, since this heavy-fermion compound is characterized by highly localized $4f$ electrons which would normally favor magnetism. Furthermore, their find of an enormous specific-heat discontinuity and a bulk Meissner effect indicated that the "heavy" electrons are indeed the ones that are superconducting. Since then, other heavy-fermion superconductors have been found, while many examples of magnetic compounds have also been formed. It is noteworthy that most of the heavy-fermi systems include rare-earth atoms whose localized electrons may tend to create a magnetic moment. This situation can result in the spin-flip scattering of conduction electrons and consequently may yield a strong temperature variation to the resistivity as we shall see below. Generally, the presence of magnetic moments tends to have a destructive influence on superconductivity, and thus the discovery of heavy-fermion superconductors has stimulated interest in spin-fluctuation mechanisms for novel forms of superconductivity.

A comprehensive review of experimental data on heavy-fermion compounds has been compiled by Stewart.[2] The magnetic-susceptibility measurements reveal a Curie-Weiss law, indicating strong magnetic moments and occasional magnetic ordering at low temperatures.

A strong temperature variation of the resistivity originally motivated theories to examine a Kondo lattice of magnetic impurities since a variation of the resistivity, $\rho_K \sim \ln T$, was observed in many cases in the range $50 < T < 300$ K. Since magnetic impurity interactions suppress the Kondo effect[3] at relatively low concentrations, say $x > 0.5$ at. %, in $CuMn_x$ and other conventional Kondo dilute alloys the observation of a $\ln T$ variation in the rare-earth alloys, such as UBe_{13}, is surprising. Furthermore, in virtually all of the heavy-fermion alloys studied so far, there is an anomalous drop in the resistivity below $T \sim 100$ K, which is quite different from the conventional saturation of the Kondo scattering at zero temperature. One proposed explanation for the anomalous drop to nearly zero resistivity at low T is the concept of a coherent state envisioned by Nozières[4] and discussed at length elsewhere.[5] The coherent state evolves for a system of independent, i.e., uncoupled, magnetic impurities and is expected to occur at very low temperature, $T < 1$ K.

At intermediate temperatures, $1 < T < 100$ K, we have found a strong temperature variation caused by the spin-flip scattering of an electron by a pair of magnetic impurities which are coupled by an interaction I whose origin is likely to be the indirect-exchange or Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism.[6] Together with the Kondo scattering, the RKKY term can create a resistivity maximum whose position is determined by the concentration of impurities, as in the case of $Ce_xLa_{1-x}Cu_2Si_2$, for example.[7]

The physical origin of the unusual temperature variation of the Kondo, as well as the RKKY, scattering is in the elastic nature of the spin flip of an electron by an impurity spin. Hence a perturbation-theory analysis will yield vanishing energy denominators whose sums contribute divergent cross sections at low temperature. For an electron coupled to the impurity by an exchange interaction J, the first divergence from a single impurity occurs in terms of order J^3 and gives the Kondo resistivity

$$\rho_K = -xA \ln T \ , \tag{1}$$

where x is the impurity concentration and $A \propto J^3$ is also dependent on the impurity spin and the electron density of states at the Fermi energy. Higher-order contributions from a single impurity indicate a breakdown of the perturbation analysis and novel methods have been used to overcome this difficulty to achieve exact solutions[8] at zero temperature which are in accord with the observed saturation in $\rho_K(T=0)$ in classic cases like $CuMn_x$.

However, heavy-fermion alloys often display a resistivity maximum near $T_m \sim 50$ K, and a sharp drop in ρ at lower temperatures, in contrast to Kondo behavior. We have shown that the drop in ρ and the associated maximum near T_m can be attributed to the consecutive spin-flip scattering of an electron by two adjacent impurities coupled via the RKKY interaction: The resulting resistivity is of the form[7]

$$\rho_{\rm RKKY} = -\frac{x^2 B(I,S)}{T} , \tag{2}$$

where the coefficient B depends on the rare-earth concentration x, the impurity spin S, and the electron density of states. The RKKY coupling I is highly sensitive to electronic structure and accordingly this resistivity contribution is strongly influenced by alloying, pressure, and sample quality. The RKKY contribution, together with the Kondo term, yields a good fit to the resistivity data of various heavy-fermion alloys.[7]

The purpose of the present work is to examine the influence of an external magnetic field on the impurity pair contribution to the electron scattering. Sufficiently high fields should inhibit the spin-flip dynamics and therefore yield a *positive* magnetoresistance at low temperature where the term $\rho_{\rm RKKY}$ may dominate the resistivity. An example of this situation is the resistivity of UPt_3,[9] which is shown in Fig. 1. Unlike many other heavy-fermion cases, $\rho(T)$ for UPt_3 does not show a $\ln T$ variation on a resistivity maximum. Instead, the data demonstrate a rapid drop in $\rho(T)$ below 100 K. The impurity-pair contribution $\rho_{\rm RKKY}$ yields a good fit to the data shown by the solid curve in Fig. 1 even when the Kondo scattering is neglected (a small phonon contribution DT is included in the fit). The Kondo scattering elevates the resistivity as shown by the dashed curve. At the lowest temperatures, the perturbation analysis becomes invalid and a deviation from the experiment is evident near $T \sim 5$ K. Hence the UPt_3 system, as well as UBe_{13} and others, provides a convenient challenge for the magnetoresistance and Hall-effect calculations considered here.

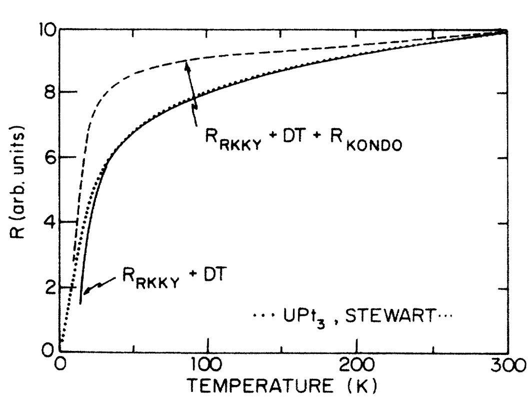

FIG. 1. Resistivity of UPt_3 as a function of temperature, with the data of Ref. 9 shown by the dots. The impurity-pair spin-flip scattering term $R_{\rm RKKY}$ yields a good fit to the data when a small phonon contribution DT is included. Kondo scattering by single impurities would elevate the curve at low temperatures as shown by the dashed curve.

Spin-flip scattering from a lattice of magnetic impurities has been invoked to explain the very origin of heavy-fermion phenomena, and the reported deviations from the dilute impurity Kondo behavior have been examined by means of several elegant theoretical approaches. At the heart of these approaches is the compensation of the impurity spin by the polarization of the conduction electrons in the zero-temperature limit. In the case of an isolated magnetic impurity, the low-temperature divergence in the electron cross section leads to a resonance at the so-called Kondo temperature T_K,[10] and it has been suggested that this resonance occurs at the Fermi energy E_F with a very narrow width of order T_K.[11] Such a resonance would yield a sharp peak in the electron density of states at E_F and therefore provides a tempting explanation for the anomalous low-temperature rise in the specific heat of heavy-fermion compounds.

An elegant theory of coherent electronic behavior in the limit of zero temperature has been proposed by Nozières.[4] He begins with the one impurity case in the $T \to 0$ limit, such that the electron spins are strongly polarized to compensate the impurity spin because of the extraordinary strong renormalization of the exchange interaction J. By means of a Fermi-liquid expansion, Nozières derives the specific heat, magnetic susceptibility, and the resistivity in the low-temperature regime. His result for the conductivity, $\sigma(T)=\sigma_0+\alpha T^2$, has been sometimes invoked in connection with the heavy-fermion data, and subsequently the extreme low-T behavior $T(\leq 0.5$ K) of the observed resistivity has been attributed[9] to "coherence" effects. These refer to the spatial extent of the conduction-electron-spin polarization which may range to 10 000 Å and therefore require sophisticated many-body treatments. Previous efforts to explain the resistivity of heavy fermions have neglected the RKKY coupling between impurities, and thus our work poses a further dilemma in the study of coherence at the lowest temperatures. If, in fact, the RKKY coupling is responsible for the precipitous drop in the resistivity below the maximum ρ region, then low-temperature treatment would seem to require the calculation of the electron-spin polarization caused by a *pair* of strongly interacting magnetic impurities. Thus this issue presents a central problem in the heavy-fermion systems and also poses an interesting challenge for the Bethe-ansatz methods which have been remarkably successful in achieving exact solutions of the single-impurity Kondo problem.[8]

Before turning to the formalism and our calculations, it seems appropriate to mention certain difficulties in determining the temperature regimes which may be appropriate to coherence, Kondo, and RKKY influences.

Looking at the resistivity of UPt_3 in Fig. 1 for guidance, we may infer that the Kondo temperature T_K is low, but it is difficult to be more precise because the low-temperature behavior of $\rho(T)$ is so different from the conventional dilute Kondo systems. On the other hand, magnetic susceptibility data are often compared to the Curie-Weiss law $\chi=C/(T+\Theta)$, and sometimes the parameter Θ has been attributed to the Kondo effect and thereby chosen to estimate $T_K \simeq \Theta$. The latter procedure should be applied with caution, however, since the Curie-Weiss parameter Θ may be influenced by many corrections,[12] including the RKKY coupling. Unfortunately, conclusive evidence on the value of T_K is lacking and we proceed to treat the Kondo scattering and the RKKY coupling on an equal footing, with a view toward learning about their relative strength from a comparison of our work to experimental data.

The magnetic field dependence of the Kondo scattering is also examined in the present work, following the earlier derivations of Kondo,[3] Abrikosov,[13] and Béal-Monod and Weiner,[4] but emphasizing the novel feature of consecutive flip of an electron spin by an impurity pair.

The magnetoresistance and Hall-effect contributions considered here originate from two sources: (a) the field and temperature dependence of the electron-scattering amplitudes, and (b) freezing out of the impurity-spin degrees of freedom by the external field as well as the mean field of nearby impurity spins.

The formalism based on the pseudofermion method is developed in Sec. II, and the key results for the magnetoresistance and other transport properties are presented in Sec. III. In Sec. IV we make a comparison to experiments on UBe_{13} and predict the field variation of UPt_3 on the basis of our calculations. Finally, in Sec. V we note some conclusions and promising directions for future research.

II. FORMALISM

We begin with a gas of electrons interacting with an array of magnetic impurity spins distributed over a lattice. The Hamiltonian of the system is taken as

$$H=H_0+H_1 , \tag{3}$$

where the unperturbed system conists of a free-electron energy $E_k=k^2/2m$ and the part of the impurity-impurity interaction that conserves spin, namely

$$H_0=\sum_{k,\sigma} E_k C^\dagger_{k\sigma} C_{k\sigma} - I \sum_{\substack{i,j \\ i\neq j}} S_i^z S_j^z , \tag{4}$$

where $C^\dagger_{k\sigma}$ is the creation operator for an electron in momentum state k and spin σ, $C_{k\sigma}$ is the analogous destruction operator, and the impurity spin at site $\mathbf{R}_j$ is denoted by S_j. The heavy-fermion systems are noted for very localized spin states corresponding to the rare-earth atoms and therefore it is reasonable to expect the coupling I between impurities to be dominated by the RKKY indirect-exchange mechanisms mediated by the conduction electrons. The perturbation of interest is the spin-flip scattering of electrons by the impurities since the spin-conserved processes are analogous to ordinary potential scattering. Here we write the interaction Hamiltonian as

$$H_1=-2J\sigma(\mathbf{r})\cdot \sum_j \mathbf{S}_j \delta(\mathbf{r}-\mathbf{R}_j) - \frac{I}{2} \sum_{\substack{i,j \\ i\neq j}} (S_i^+ S_j^- + S_i^- S_j^+) , \tag{5}$$

where J is the exchange coupling of the conduction-electron-spin density $\sigma(\mathbf{r})$ to the impurity spin, and $S_j^{\pm}=S_j^x \pm i S_j^y$ denotes the standard raising and lowering operators for the impurity spin. For a perturbation expansion it is convenient to rewrite the interaction Hamiltonian in terms of the Fourier transform of the spin density, which leads to

$$\begin{aligned} H_1=-J \sum_{\mathbf{k},\mathbf{k}'} \sum_j \big[& (C^\dagger_{k,\sigma} C_{k',\sigma} - C^\dagger_{k,-\sigma} C_{k',-\sigma}) S_j^z \\ & + C^\dagger_{k,\sigma} C_{k',-\sigma} S_j^- + C^\dagger_{k,-\sigma} C_{k',\sigma} S_j^+ \big] \\ & \times \exp[-i(\mathbf{k}-\mathbf{k}')\cdot \mathbf{R}_j] \\ & - \frac{I}{2} \sum_{\substack{j,l \\ j\neq l}} (S_j^+ S_l^- + S_j^- S_l^+) . \end{aligned} \tag{6}$$

This interaction Hamiltonian provides the basis for the Kondo effect when $I=0$, and now we can proceed further to calculate the impurity-pair contribution to the electron scattering.

A stumbling block in the problem arises because the angular-momentum commutation relations of the spin operators are not fermion or boson in character. Thus the usual diagram rules cannot be applied to make a systematic perturbation scheme. To overcome this difficulty, Abrikosov developed the pseudofermion method[10] by introducing virtual fermion field operators $b^\dagger_{jm}$ and b_{jm} which represent the impurity spin via the definition

$$\mathbf{S}_j = \sum_{m,m'} b^\dagger_{jm} b_{jm'} S_{mm'} , \tag{7}$$

where $S_{mm'}$ are the spin matrix elements with indices spanning the $2S+1$ spin projections $-S,-S+1,\ldots,S-1,S$. These field operators obey normal fermion anticommutation rules:

$$[b^\dagger_{jm}, b_{lm'}]_+ = \delta_{jl}\delta_{mm'} , \tag{8a}$$

$$[b^\dagger_{jm}, b^\dagger_{lm'}]_+ = [b_{jm}, b_{lm'}]_+ = 0 , \tag{8b}$$

where δ_{lj} is the Kronecker δ symbol. These operators allow the application of conventional diagram techniques with an important constraint. The physical meaning of the operator $b^\dagger_{jm}$ is that it creates an impurity state with spin projection m at site R_j, and, therefore, a mechanism is needed to exclude unphysical states where the number of states at a given impurity site is artificially at odds with the single-impurity value. This difficulty is avoided by assigning each impurity an energy $\lambda \gg T$, normalizing the computed results by a factor $\exp(\lambda/T)/(2S+1)$ and then taking the limit λ going to infinity.[10] We shall demonstrate this procedure below.

Now the perturbation expansion is conveniently expressed in terms of the Green's function for the localized impurity state

$$D(\omega)=\frac{1}{i\omega-\lambda} \tag{9}$$

and the electron propagator

$$G(\mathbf{k},\omega)=\frac{1}{i\omega-E_k} . \tag{10}$$

In the absence of an external field, the scattering relaxation time τ has been calculated previously to various orders of J. The lowest order gives the Born-approximation result

$$\frac{1}{\tau_B}=\pi J^2 N(0) S(S+1) , \tag{11}$$

and the leading divergent term found by Kondo is

$$\frac{1}{\tau_K}=4\pi J^3 N^2(0) S(S+1)\ln(T/D) , \tag{12}$$

where S denotes the impurity spin, $N(0)$ is the electron density of states at the Fermi level, and D is an energy-cutoff parameter which is required to compensate for assuming J to be independent of momentum. Higher-order terms in J which also diverge as powers of $\ln T$ lead to a resonance below the Kondo temperature T_K, and this resonance at the Fermi energy has been invoked as an explanation for the heavy character of the fermion specific heat. Using that approach as a guide, $T_K \leq 10$ K in many heavy-fermion alloys, whereas the region of interest for the present calculation is $10 \leq T \leq 100$ K.

Impurity-pair contributions to the electron-scattering rate give a strong temperature variation from the spin-flip process even at a low order of J^4, or equivalently IJ^2. The result is generally anisotropic and given by[7]

$$\frac{1}{\tau_{\rm RKKY}}=\frac{2\pi I J^2 N(0)[S(S+1)]^2 f^t}{9T} , \tag{13}$$

where the transport average over impurity sites f^t is determined by the distribution of impurity spins. For the example of $CeCu_2Si_2$ we found[7]

$$f^t_{\perp}(x)=\frac{2\sin(x/2)}{x} , \tag{14a}$$

for the electron momentum $\hat{\mathbf{k}}$ perpendicular to the square lattice of impurities, where $x=2k_F a$ is the product of the Fermi momentum k_F and the lattice spacing a. By comparison, for $\hat{\mathbf{k}}$ parallel to the impurity lattice.

$$f^t_{\parallel}(x)=\sin(x/2)[x+2x\cos(x/2) -2\sin(x/2)]/x^2 . \tag{14b}$$

Higher-order processes will become significant as $T \to 0$, and the treatment of these corrections in conjunction with the comparable Kondo terms is a challenging problem beyond the scope of the present work.

III. MAGNETORESISTANCE AND THE HALL EFFECT

In the presence of a magnetic field the conduction electrons experience a Zeeman energy sP which includes the external field $\frac{1}{2}g_e s\mu_B H$ in addition to the mean field of the polarized impurities $J\langle S\rangle$. Hence the electron Green's function becomes

$$G_s(\mathbf{k},\omega)=\frac{1}{i\omega-E_k-sP} , \tag{15}$$

where the spin index refers to $s=+1$ or -1, and g_e denotes the electron g factor with μ_B the Bohr magneton. The Zeeman energy of the impurity is likewise influenced by the appropriate impurity g factor g_i, plus the mean field of the conduction electrons $J\langle\sigma\rangle$, and the neighboring impurity field $I\langle S\rangle$. Writing their total as Q, we express the impurity Green's function as

$$D_m(\omega)=\frac{1}{i\omega-\lambda+mQ} . \tag{16}$$

The diagrams corresponding to the field-induced corrections are shown in Fig. 2. In principle, the equations for the field-dependent response of the coupled electron-impurity system could be solved self-consistently. However, a lower-order approximation will suffice to exhibit a strong field and temperature variation of the electron-scattering rate. Hence we write

$$P=\mu_B H + xJSB_S(Q/T) , \tag{17a}$$

where B_S is the Brillouin function. At low temperatures we may use

$$P \cong \mu_B H + JS , \tag{17b}$$

and then check the computed results using the Brillouin

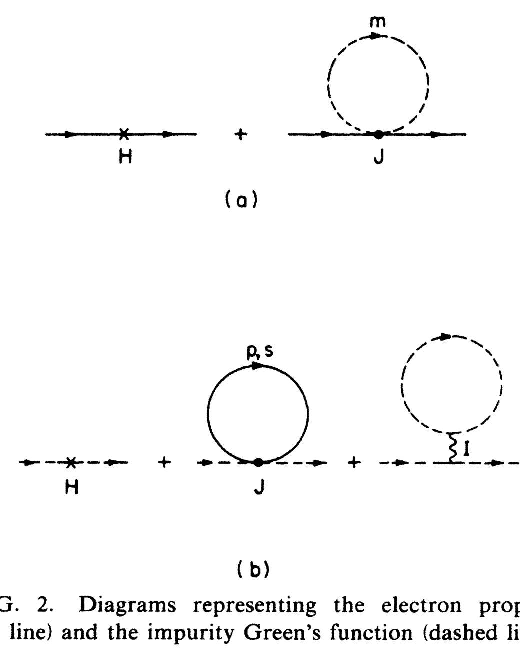

FIG. 2. Diagrams representing the electron propagator (solid line) and the impurity Green's function (dashed line). A cross represents an external field H and the dot refers to the exchange interaction J. The wavy line gives the RKKY coupling I. These graphs indicate the effective field contributions.

function as well. For convenience we consider the mean field of z nearest-neighbor impurities, and write

$$Q \cong g_i H[1+JN(0)]+zIS \ . \tag{18}$$

Within the pseudofermion method, it is important to use the renormalization $\exp(\lambda/T)/Z$, where

$$Z=\sum_{m=-S}^{S}\exp(mQ/T) \ , \tag{19}$$

where the limit $\lambda \to \infty$ is taken in evaluating the diagrams.

As a first step we compute the field-dependent version of the Born approximation shown in Fig. 3(a). The self-energy is

$$\Sigma_B(\omega)=-\lim_{\lambda\to\infty}\frac{\exp(\lambda/T)}{Z}\times J^2\sum_{m,s'}|L_{ms,m's'}|^2\times T^2\sum_{\mathbf{P},\omega_1,\omega_2}G_{s'}(\mathbf{P},\omega+\omega_1-\omega_2)\times D_m(\omega_1)D_{m'}(\omega_2) \ , \tag{20}$$

where the spin-interaction matrix element is

$$L_{m's',ms}=\langle m's'|2\sigma\cdot\mathbf{S}|ms\rangle = ms\delta_{s,s'}\delta_{m,m'} + [S(S+1)-m(m+s)]^{1/2}\delta_{s,-s'}\delta_{m+s,m'} \ . \tag{21}$$

Performing the indicated sums and integrations, we obtain

$$\frac{1}{\tau_B(H)}=-2\,\mathrm{Im}\,\Sigma_B(0) =\pi J^2N(0)[\langle m^2\rangle+2\langle s(s+1) -m(m+s)\rangle n_F(-sH)] \ , \tag{22}$$

where n_F denotes the Fermi function. The $H=0$ limit reduced to Eq. (11) and the limit $H/T\to\infty$ gives $\tau_B^{-1}(\infty)=2\pi J^2N(0)S^2$, which demonstrates that the

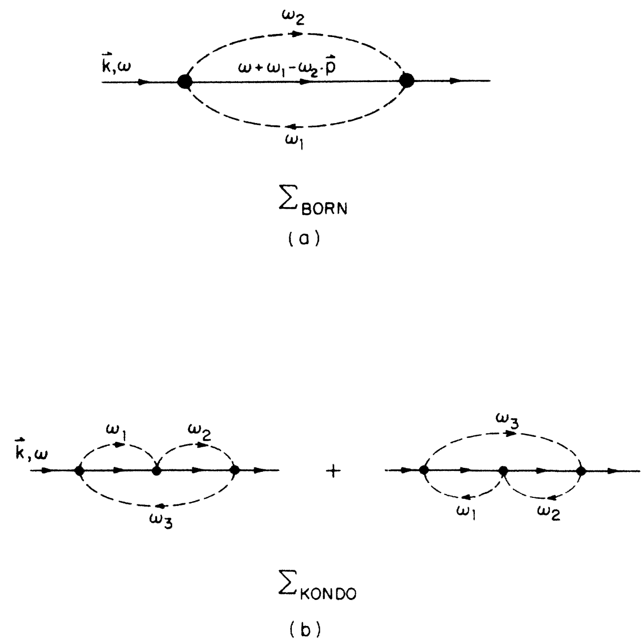

FIG. 3. Self-energy diagrams for (a) the lowest-order Born approximation and (b) the Kondo scattering.

Born approximation gives a negative magnetoresistance with a mean temperature variation originating from the thermal spin averages in Eq. (22).

A. Kondo scattering

The Kondo effect in the presence of a magnetic field has been considered by Kondo,[3] Abrikosov,[13] and Béal-Monod and Weiner.[14] However, an analytic solution for all magnetic fields has not been achieved previously. Since we wish to compare the Kondo with the RKKY contribution, we reexamine the Kondo self-energy shown in Figs. 3(b) and 3(c) to derive a useful approximate form for the magnetoresistance. The details of the computation are given by Sheng.[15]

If the spin of the electron is conserved in the Kondo processes of Figs. 3(b) and 3(c), the two diagrams give canceling contributions, thus eliminating terms which may be temperature or field dependent. Just as in the zero-field case, however, the spin-flip dynamics yield a logarithmic T divergence which is suppressed by a magnetic field. Performing the spin and frequency summations, we obtain the spin-flip terms

$$\frac{1}{\tau_K}=-8\pi J^3N^2(0)\Big[\langle[S(S+1)-m(m+s)](m+s)(-s)\rangle+n_F(-sH)\int dE(-E)^{-1}n_F(-E-sH) + \langle[S(S+1)-m(m+s)]ms\rangle\Big[n_F(-sH)\int dE(-E+sH)^{-1}n_F(-E+sH) +\tfrac{1}{2}\int dE(-E)^{-1}n_F(-E-sH)\Big]\Big] \ . \tag{23}$$

Now the divergent integral can be expressed as

$$\int_{-D}^{D} dE(-E)^{-1}n_F(-E-sH)=\ln\left|\frac{T}{D}\right|+\gamma(sH/T)\ , \quad (24)$$

where

$$\gamma(x)\cong\int_{-\infty}^{\infty} dy\,\ln|y|\,\exp(y-x)[\exp(y-x)+1]^{-2}\ , \quad (25)$$

where the limit $D/T\rightarrow\infty$ is presumed as usual. We have evaluated the integral numerically but also found an analytic expression of reasonable accuracy, which may be useful in future analysis of experimental data. The "slanting" approximation for the Fermi function replaces $[\exp(z)+1]^{-1}$ by the following: $n_F=1$ for $E<-T$, $n_F=0$ for $E>T$, and $n_F=0.5-E/2T$ in the interval $-T<E<T$. With this approximation we obtain

$$\gamma(x)\simeq 0.5(1-x)\ln|1-x|+0.5(1+x)\ln|1+x|\ , \quad (26)$$

which overestimates $\gamma(x)$ by roughly 20% over a wide range of x in comparison to the numerical solution.[15] Nevertheless, this form yields the semiquantitative representation of the Kondo magnetoresistance by the result

$$\frac{1}{\tau_K(H)}=8\pi J^3N^2(0)(S(S+1)\ln|T/D|+[S(S+1)-\langle m^2\rangle]\gamma(x)$$
$$+s[1-\exp(x)][1+\exp(x)]\{\langle m\rangle\ln|T/D|+[\langle m^3\rangle-(S^2+S-1)\langle m\rangle]\gamma(x)\})\ , \quad (27)$$

where $x=sH/T$.

In the $H\rightarrow 0$ limit, Eq. (27) reduces to the standard Kondo result of Eq. (12). In the high-field limit $H/T\gg 1$ we have $\langle m\rangle=S$, $\langle m^2\rangle=S^2$, $\langle m^3\rangle=S^3$, and an analytic expression of $\gamma(x)$ gives

$$\frac{1}{\tau_K(H\gg T)}\cong 8\pi J^3N^2(0)S^2\left[1-\left[\frac{T}{H}\right]^2+\ln\left[\frac{H}{D}\right]\right]\ . \quad (28)$$

Thus a strong magnetic field eliminates the $\ln T$ divergence by disallowing spin-flip processes. A similar effect occurs when the mean field of nearest-neighbor magnetic impurities inhibits the spin dynamics and usually suppresses the Kondo divergence at very low impurity concentrations, say $x<0.5$ at.% for $AgMn_x$.[6] By comparison, the persistence of a Kondo $\ln T$ contribution to the resistivity of many heavy-fermion alloys is mysterious in view of the large rare-earth content.

The temperature variation of the Kondo resistivity is shown in Fig. 4 at different values of the external field. At the lower temperatures, where the $\ln T$ behavior is dominant, a negative magnetoresistance is found, whereas only small variations in τ_K are evident at higher temperatures.

B. Impurity-pair processes

Physically, the process of interest here is the spin flip of an electron in a state $(\mathbf{k},\sigma)$ to an intermediate state $(\mathbf{q},-\sigma)$ by the first impurity and a second spin flip by another impurity to the original state. At low temperatures this process yields a $1/T$ divergence in the low-order IJ^2 of the perturbation analysis,[6,7] and its origin can be traced to the elastic nature of the spin-flip process.

Other nondivergent terms involving the RKKY interaction J include the distribution of spin-singlet and -triplet impurity states,[14] but these yield a smooth temperature variation and are not considered here.

The self-energy diagram corresponding to the consecutive spin-flip term is shown in Fig. 5. Following the rules of the pseudofermion technique we derive

$$\Sigma_{\rm RKKY}=IJ^2\lim_{\lambda\rightarrow\infty}[\exp(\lambda/T)/Z]^2\times\sum_{m,n'} l_2\sum_{p} d_I\langle\exp[i(\mathbf{k}-\mathbf{p})\cdot(\mathbf{R}_1-\mathbf{R}_2)]\rangle\ , \quad (29a)$$

where

$$l_2\equiv\langle mn's\,|\,2\boldsymbol{\sigma}\cdot\mathbf{S}_2\,|\,mns'\rangle\times\langle mns'\,|\,S_1^+S_2^-+S_1^-S_2^+\,|\,m'n's'\rangle\times\langle m'n's'\,|\,2\boldsymbol{\sigma}\cdot\mathbf{S}_1\,|\,mn's\rangle\ , \quad (29b)$$

and

$$d_I=T^3\sum_{\omega_1,\omega_2,\omega_3} G(\mathbf{p},\omega_3)D_m(\omega_1)D_{m'}(\omega+\omega_1-\omega_3)\times D_n(\omega_2)D_{n'}(\omega_2+\omega_3-\omega)\ , \quad (29c)$$

where m and n are spin quantum numbers for the first and second impurities situated at R_1 and R_2, respectively. The spin matrix l_2 reduces to the thermal average $\langle S(S+1)-m(m+s)\rangle^2$, and the limit $\lambda\rightarrow\infty$ yields

$$\Sigma_{\rm RKKY}=IJ^2l_2T\sum_{p,\omega_3}\frac{1}{i\omega_3-E_p-sH}\times\frac{1}{(i\omega_3-\omega-sH)^2}n_B^{-2}(sH)\times\langle\exp[i(\mathbf{k}-\mathbf{p})\cdot(\mathbf{R}_1-\mathbf{R}_2)]\rangle\ . \quad (30)$$

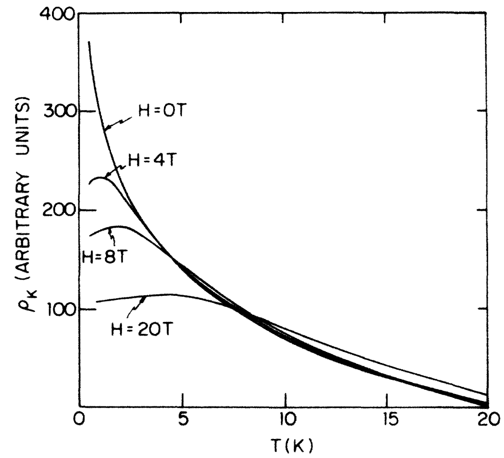

FIG. 4. Kondo magnetoresistance as a function of temperature, showing the conventional $\ln T$ behavior at zero field. A magnetic field H suppresses the spin-flip process and thus causes a negative magnetoresistance as shown for various H values, at low temperatures.

It is interesting to distinguish the elastic and inelastic contributions since only the elastic terms survive in the $H=0$ case. For the elastic case, $\omega_3=\omega$, the momentum sum is performed by taking the average over the impurity sites $\mathbf{R}_i$. Thus we obtain

$$\frac{1}{\tau_{\mathrm{RKKY}(\omega=\omega_3)}}=2\pi IJ^2N(0)l_2 f'(2k_F R)n_B^{-2}(sH)T/H^2 \ , \tag{31}$$

where n_B is the Bose-Einstein function and the spatial function $f'(2k_F R)$ is given in Eqs. (14a) and (14b). In the zero-field limit, the elastic-scattering term reduces to the previous results[7] given in Eq. (13), because

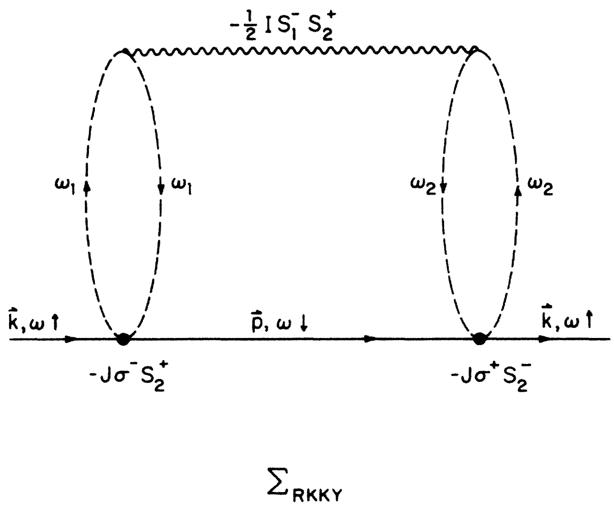

FIG. 5. Self-energy diagram for the spin-flip scattering of an electron by a pair of adjacent magnetic impurities with spin S_1 and S_2. The RKKY interaction is represented by a wavy line and the dots show the exchange coupling.

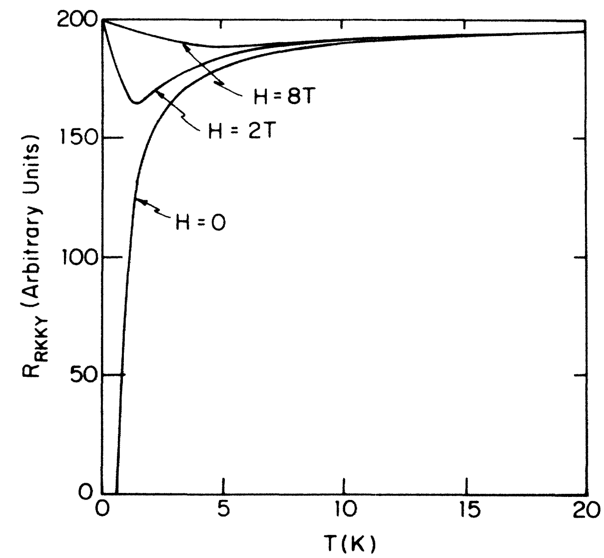

FIG. 6. Magnetoresistance of the impurity-pair contribution as a function of temperatures showing the suppression of the $1/T$ behavior at finite fields. As $T\to 0$ the analysis becomes invalid due to neglect of higher-order contributions, but nevertheless the trend of a positive magnetoresistance seen here may be relevant to UPt_3 and other alloys.

$n_B^{-2}(sH)TH^{-2}$ reduces to $1/T$. As expected, the limit of very large fields, $H/T \gg 1$, eliminates the $1/T$ divergence and yields

$$\frac{1}{\tau_{\mathrm{RKKY}}(\omega=\omega_3,\, H \gg T)}=2\pi IJ^2N(0)4S^2 f'T/H^2 \ . \tag{32}$$

Hence the freezing of the spin degrees of freedom eliminates the spin-flip transitions which are responsible for the $1/T$ behavior, and a similar restraint is imposed by the mean field of other impurities at sufficiently low T.

Inelastic terms in the spin-flip scattering have $\omega\neq\omega_3$, and vanish in the $H=0$ case. Mathematically, some complexity arises in the sum

$$M=T\sum_{\omega_3\neq\omega}\frac{1}{i\omega_3-E_p-sH}\frac{1}{(i\omega_3-\omega-sH)^2} \ . \tag{33}$$

The details may be obtained from Ref. 15, and the resulting scattering rate becomes

$$\frac{1}{\tau_{\mathrm{RKKY}(\omega\neq\omega_3)}}=2\pi IJ^2N(0)l_2 f' \times\{T^{-1}\exp(x)-n_B^2(x)T/H^2 +T^{-1}[n_F(x)/n_B(x)]^2\exp(x)\} \ , \tag{34}$$

where $x\equiv sH/T$. As $H\to 0$ the inelastic term vanishes as expected. Furthermore, for $H\gg T$ the inelastic contribution precisely cancels the elastic contribution of Eq. (32).

Adding elastic and inelastic contributions derived in Eqs. (31) and (34) gives the final result

$$\frac{1}{\tau_{\text{RKKY}}} = 2\pi I J^2 N(0) f' \langle S(S+1) - m(m+s) \rangle^2 \times T^{-1}[1 + n_F^2(x)/n_B^2(x)]\exp(x) \ . \qquad (35)$$

This scattering rate is shown as a function of temperature for various H values in Fig. 6. It is interesting to note how the field produces a positive magnetoresistance at low temperatures by eliminating the strong negative contribution of the RKKY scattering amplitude in this case. The sign of the impurity-pair resistivity is determined by the sign of the RKKY coupling I as well as the phase factor $2k_F R$ which enters in the definition of f'. A comparison to experiment requires the combined effect of the Kondo and impurity-pair scattering, which may lead to surprising variations in the resistivity, as we show in the next section.

IV. HEAVY-FERMION MAGNETORESISTANCE

The resistivity is related to the electron relaxation times by the Einstein formula

$$\rho = \frac{m^*}{ne^2}\left[\frac{1}{\tau_K} + \frac{1}{\tau_{\text{RKKY}}}\right] + C \ , \qquad (36)$$

where n is the conduction-electron density and C represents other nonspin contributions, such as potential and phonon scattering. An important point to notice is that the effective mass m^* becomes heavy in the sense of representing a giant heat-capacity intercept which occurs only at the lowest temperatures $T < T_K$,[16] whereas our calculation is valid in higher-T range, say $10 < T < 100$ K; thus we are compelled to use the free-electron mass for m^* in estimating the parameters.

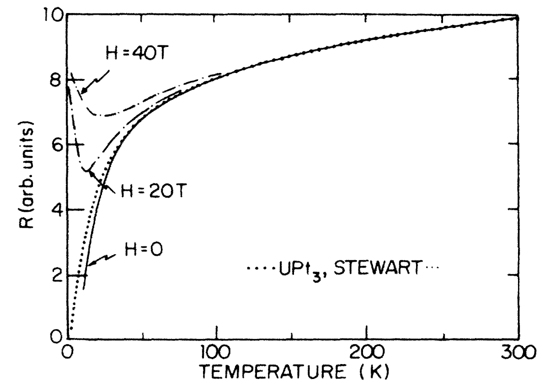

FIG. 8. Resistivity of UPt_3 as a function of temperature. The zero-field data points are from Ref. 9, and the solid curve is the fit using the impurity scattering plus a small phonon term DT, neglecting Kondo scattering. At finite magnetic fields H, a positive magnetoresistance is predicted as shown by the dotted-dashed curves.

In the zero-field limit, the combined Kondo and RKKY terms in Eq. (36) give a good fit to the resistivity of various heavy-fermion alloys and they provide a simple explanation for the resistivity maximum arising from these competing contributions with different sign and temperature variations.[7]

A magnetic field provides a good test of these ideas because it will suppress the Kondo and the RKKY scattering at different rates. The case of UBe_{13} demonstrates a shift in the resistivity maximum at a field $H=8$ T as shown in Fig. 7 in comparison to the experimental data of Ref. 17. At higher temperatures $T \sim 20$ K the calculated curves fall below the data, probably because of phonon scattering. However, in the region near the

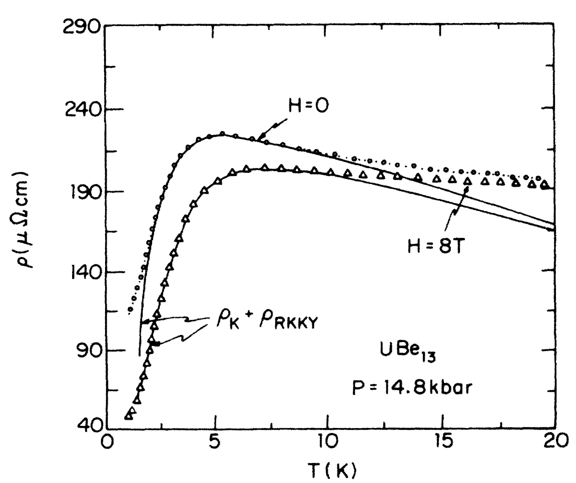

FIG. 7. Resistivity of UBe_{13} as a function of temperature is shown by dots for $H=0$ and triangles for $H=8$ T using the experimental data of Ref. 17. A good fit to the zero-field data is achieved using the Kondo and impurity-pair scattering terms of Eqs. (27) and (35), respectively, and reasonable parameters given in the text. At finite $H=8$ T the same parameters give an excellent fit to the low-temperature region with an internal field $JN(0)=0.5$.

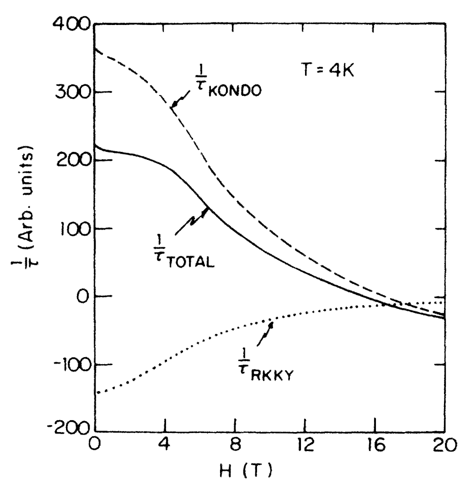

FIG. 9. Magnetic field variation of the spin-flip scattering is shown at a low temperature $T=4$ K. Both the Kondo and the impurity-pair scattering are suppressed by the field, but their relative decreases depend strongly on the temperature. Here the calculated curves presume a dominant Kondo contribution and, therefore, a negative magnetoresistance, in contrast to the case shown in Fig. 8.

maximum at $T \sim 4$ K a surprisingly good fit is achieved using $A = 100\ \mu\Omega$ cm and $B = 560\ \mu\Omega$ cm. Using a free-electron mass and $S = \frac{5}{2}$ gives a reasonable estimate for $J = 0.1$ eV and the RKKY coupling $I = 0.001$ eV. However, the estimation should be regarded as crude and the values are "effective" coupling parameters which represent a more complete expression derived by Coqblin and Schrieffer.[18] Nevertheless, it is gratifying that our effective parameter estimates are in line with their estimates for Ce alloys.[18] A significant feature of our analysis for Ube[13] is an estimate of the internal mean-field correction $JN(0) \simeq -0.5$ which reduces the field at the impurity site by 50% in comparison to the external field H. Thus, at the low temperature considered here, the analysis indicates a considerable spin polarization of the conduction electrons by the exchange coupling.

Another interesting case is UPt_3 whose resistivity does not show a Kondo behavior, but nevertheless exhibits a sharp drop at low temperatures as seen in Fig. 1. Our analysis predicts a positive magnetoresistance in this heavy-fermion alloy as shown in Fig. 8. It should be noted that the low-temperature resistivity is particularly sensitive to the magnetic field because of its spin-flip origin, and this feature may allow a separation of the RKKY-induced scattering from the Fermi-liquid model of the coherent state.

A sampling of experimental data for UPt_3 shows a positive enhancement of the resistivity near $T \sim 6$ K for $H = 5$ T, and 3 times that enhancement for $H = 8$ T,[19] in qualitative agreement with our calculated curves in Fig. 8, which did not include internal mean-field corrections. More complete magnetoresistance studies of UPt_3 and various alloys such as Pd substituted for Pt (Ref. 20) should pinpoint the relative influence of the RKKY interactions in these alloys.

The magnetoresistance data on Kondo lattices such as $CeAl_3$ and $CeCu_2Si_2$ and similar alloys exhibit a change in sign at various temperatures[21] and has been used to investigate the onset of the expected Fermi-liquid behavior at very low temperatures. However, the competition between Kondo and RKKY scattering may provide an alternate explanation as illustrated in Fig. 9. In this case the Kondo scattering was presumed to be dominant at $T = 4$ K, giving the negative magnetoresistance shown at various fields. However, at lower temperatures, the impurity-pair contribution may overcome the Kondo term (as seen in the extreme example of UPt_3), and then a positive magnetoresistance is expected.

The Hall effect may also be strongly influenced by the field and temperature variation of the spin-flip scattering rates, and a change in the sign of the Hall constant is possible. However, a direct comparison of our results to experiments is not yet feasible because of a lack of information concerning the Fermi-surface topology. Thus we note that the expressions for the relaxation rates τ_K and τ_{RKKY} given above may be useful in future studies.

Another complicating feature of the Hall-effect measurements is the strong influence of skew-scattering mechanisms[22] which also yield a strong temperature variation of the Hall constant of mixed-valence systems.

V. CONCLUSIONS

Spin fluctuations are a key feature of heavy-fermion compounds, and our results suggest the relative importance of RKKY interactions between impurities may have a strong influence on the transport properties. Previously, many studies disregarded RKKY coupling terms in part because the Kondo model of independent impurities was supported by a $\ln T$ term in the resistivity, and also because it provided an elegant description of the low-temperature specific-heat anomaly in terms of a narrow resonance whose origin is attributed to the spin-exchange interaction of a conduction electron scattering from isolated impurities. Extensions to Kondo-lattice models and the interesting concept of a coherent Fermi-liquid state at the lowest temperatures also avoided complexities relating to RKKY coupling. Nevertheless, estimates of the RKKY coupling strength in these materials are sparse, and the presumed weakness of such interactions remain mysterious.

The clear deviations of the heavy-fermion resistivity from conventional Kondo behavior at low temperatures provides a good testing ground for the RKKY processes considered here. The case of UPt_3 is particularly unusual, since it does not exhibit any hint of a $\ln T$ variation, whereas the sharp drop in its resistivity (Fig. 1) is similar in form to many other systems such as UBe_{13}, $CeCu_2Su_2$, $CeAl_3$, etc. We have shown that this anomalous drop in the resistivity follows readily from the consecutive spin flip of a conduction electron by two nearby impurities. Furthermore, the estimated magnitude of the RKKY coupling required to fit the drop is small, but nevertheless the $1/T$ variation of this scattering process allows it to overwhelm the Kondo resistivity at low temperatures. In the case of UPt_3, our results suggest that the RKKY scattering dominates the Kondo $\ln T$ term over the entire measured temperature range, but the analysis allows a weak Kondo effect to be present nevertheless. However, this example is particularly challenging since the heavy-fermion behavior of the specific heat should require a more complete analysis including the influence of RKKY coupling on the Kondo-Abrikosov-Suhl resonance, which is widely considered as the basis for the heavy character of the electron response at very low temperatures.

Magnetic fields suppress the spin-flip dynamics, which is necessary for both the Kondo and the RKKY-induced impurity-pair scattering. Thus our calculated results provide an independent means of determining the relative importance of the contributions. Of particular interest is the shift of the resistivity maximum in UBe_{13} at a field of 8 T, which follows directly in our calculations using the same exchange J and RKKY parameters used in the zero-field case. Furthermore, the good description of the magnetoresistance over a wide temperature range is gratifying.

In view of the competition between Kondo processes that give a negative magnetoresistance and the impurity-pair scattering, which generally contributes a positive magnetoresistance, the total resistivity may increase with magnetic field at low temperatures, but revert to a negative magnetoresistance at high temperatures, $T \sim 100$ K. Similarly, changes in the sign of the Hall effect are anticipated at different temperatures since the Hall constant is particularly sensitive to the electron relaxation time.

At the lowest temperatures our perturbation analysis breaks down and we are faced with higher-order contributions in league with the Kondo scattering. Innovative exact treatments, such as the Bethe ansatz, which has been successful in the single-impurity problem, may provide some insight into the pair-scattering terms as well. On the other hand, it would be worthwhile to extend the concepts of a Fermi-liquid coherent state to the Kondo lattice with weak interactions between impurities. In particular, the influence of a magnetic field on the proposed coherent state may indicate the realms of temperature and impurity concentrations where the RKKY interactions may be expected to provide a substantial influence.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge stimulating discussions with M. B. Maple, H. R. Ott, B. Shivaram, and G. R. Stewart. We appreciate the hospitality of our colleagues at Harvard University during our sabbatical. This research was supported by the U.S. Department of Energy under Grant No. DEFG05-84 ER-45113.

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