Spin Relaxation and Transport in Local-Moment Systems*

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Using a kinetic equation, the effect of Kondo-type anomalies on spin relaxation and transport in local-moment systems is studied. For zero wave number of the external magnetic field a modified Hasegawa equation can be derived. With increasing wave number a diffusion regime is reached. The diffusion constant can be related to the electrical resistivity. Further increase of the wave number yields a collisionless regime for the conduction electrons. In this regime, the spin relaxation becomes essentially unbottlenecked, and the local-moment relaxation proceeds with the full (anomalous) Korringa value.

I. INTRODUCTION

An exchange interaction $J\mathbf{s}\cdot\mathbf{S}$ between the conduction-electron (s) spin $\mathbf{s}$ and local-moment (d) spin $\mathbf{S}$ in a metal leads to an anomalous temperature dependence of the s-d scattering amplitude $t+\tau\mathbf{s}\cdot\mathbf{S}$, if the sign of J is antiferromagnetic.[1] The anomalies in the scattering lead to a number of peculiarities in the linear response of local-moment systems to electrical, thermal, and magnetic external disturbances. There exist[2] a maximum in the electrical and in the thermal resistivity at $T=0$, a maximum in the impurity part of the specific heat near the so-called Kondo temperature T_K, and deviations from the Curie law of the static impurity susceptibility.

The theory of magnetic response is more difficult than the theory of nonmagnetic response. Nonmagnetic response can be calculated from the non-spin-flip part $t(\epsilon)$ of the scattering amplitude alone. For instance,[1–3] electrical and thermal transport coefficients depend essentially only on $\mathrm{Im}t(\epsilon_F)$, and the specific heat only on $t(\epsilon)$ for large ϵ.[4] For the calculation of these quantities, the two main approximation schemes—the S-matrix approach of Suhl and the Green's-function decoupling procedure of Nagaoka—work equally well and yield identical results.[4,5]

On the other hand, $t(\epsilon)$ is not sufficient to calculate magnetic response. Aside from literature on the static susceptibility[6,7] and spin relaxation[8–11] in which the s-d scattering is treated using perturbation theory, there exist calculations using the full information contained in Suhl's spin-flip amplitude τ,[12–15] or else Nagaoka's spin-flip Green's function Γ.[16] As a consequence of different approximations used in the two[14–16] approaches, the results for the static susceptibilities disagree. Thus, the precise nature of information about spin flip which can be obtained from experiments on the static susceptibility is unclear as yet.

Additional information can be obtained by looking at the frequency dependence of the suceptibility. In the following sections, we want to discuss this possibility and also investigate the wave number dependence of the susceptibility in more detail.

II. KINETIC EQUATION

We shall base our investigation on two kinetic equations for the coupled s-d system[13]:

$$\left(\frac{\partial}{\partial t}+\frac{\partial\epsilon_{p\sigma}}{\partial\mathbf{p}}\cdot\frac{\partial}{\partial\mathbf{r}}-\frac{\partial\epsilon_{p\sigma}}{\partial\mathbf{r}}\cdot\frac{\partial}{\partial\mathbf{p}}\right)f_{p\sigma}=I_{p\sigma}, \tag{1a}$$

$$\left(\frac{\partial}{\partial t}+\frac{\partial\omega_{k\kappa}}{\partial\mathbf{k}}\cdot\frac{\partial}{\partial\mathbf{r}}-\frac{\partial\omega_{k\kappa}}{\partial\mathbf{r}}\cdot\frac{\partial}{\partial\mathbf{k}}\right)n_{k\kappa}=I_{k\kappa}', \tag{1b}$$

where

$$\epsilon_{p\sigma}=\epsilon_p-\sigma h_s, \qquad \sigma=\pm\tfrac{1}{2},$$
$$\omega_{k\kappa}=\omega_k-\kappa h_d, \qquad \kappa=-S,\ \ldots,\ S.$$

As far as possible, we shall follow the notation of Ref. 4. In particular, we shall consider the "narrow impurity band limit," i.e., $\omega_k=k^2/2m-\mu_d$ and $m\to\infty$, and $h_s=\mu_B g_s B$, $h_d=\mu_B g_d B$. $f_{p\sigma}(r,t)$ and $n_{k\kappa}(r,t)$ are the distribution functions for the s and d electrons, respectively. I and I' are the collision integrals for s and d electrons. They have the usual Boltzmann-type gain and loss terms with scattering amplitudes proportional to $T=t+\tau\mathbf{s}\cdot\mathbf{S}$ and in addition two spin-lattice relaxation terms. Thus, we use as collision integrals

$$I_{p\sigma}=-V^{-2}\sum_{p',k;\sigma',\kappa,\kappa'}\langle\sigma\kappa\,|\,C_{pp'k}\,|\,\sigma'\kappa'\rangle-\alpha_s{}^0[f_{p\sigma}-f^0(\epsilon_{p\sigma})], \tag{2a}$$

$$I_{k\kappa}'=-V^{-2}\sum_{p,p';\sigma,\sigma',\kappa'}\langle\sigma\kappa\,|\,C_{pp'k}\,|\,\sigma'\kappa'\rangle-\alpha_d{}^0[n_{k\kappa}-f^0(\omega_{k\kappa})], \tag{2b}$$

where

$$\langle\sigma\kappa\,|\,C_{pp'k}\,|\,\sigma'\kappa'\rangle=|\,\langle\sigma\kappa\,|\,T\,|\,\sigma'\kappa'\rangle\,|^2\times2\pi\delta(\epsilon_{p\sigma}+\omega_{k\kappa}-\epsilon_{p'\sigma'}-\omega_{k'\kappa'})\times[f_{p\sigma}n_{k\kappa}(1-f_{p'\sigma'})(1-n_{k+p-p'\kappa'})-(1-f_{p\sigma})(1-n_{k\kappa})f_{p'\sigma'}n_{k+p-p'\kappa'}], \tag{3}$$

with

$$\langle\sigma\kappa\,|\,T\,|\,\sigma'\kappa'\rangle=t(\epsilon_F)\delta_{\sigma\sigma'}\delta_{\kappa\kappa'}+\tau(\epsilon_F)\,\mathbf{s}_{\sigma\sigma'}\cdot\mathbf{S}_{\kappa\kappa'}. \tag{4}$$

$\alpha_s{}^0$ and $\alpha_d{}^0$ are two phenomenological spin-lattice re-

laxation frequencies and $f^0(\epsilon)$ is the Fermi function. To obtain the linear response function, one linearizes in the external field and takes the Fourier transform with respect to $\mathbf{r}$, t. In doing so, one has to pay attention to the fact, that the magnetic field contained in the Zeeman energies of $\epsilon_{p\sigma}$ and $\omega_{k\kappa}$ not only occurs in the left-hand side of the kinetic equations but also in the collision terms (2a), (2b), and (3). This then leads to equations in which the magnetization of each spin species properly relaxes to its instantaneous local value.[9,17] Apart from this difference, which becomes relevant only for $g_s \neq g_d$, our equations agree in the long-wavelength limit with the equations given by Hasegawa.[18]

In order to exhibit the dependence on the g factors explicitly, we first consider angular momentum densities rather than magnetizations. In Eq. (1b), we also sum over k and take the limit of infinite impurity mass, i.e., we consider

$$n_p = \sum_\sigma \sigma f_{p\sigma}, \qquad n_p^{\rm loc} = \sum_\sigma \sigma [f_{p\sigma} - f^0(\epsilon_{p\sigma})];$$
$$n_s = V^{-1} \sum_p n_p, \qquad n_s^0 = \tfrac{1}{2}\rho_F h_s = r_s^0 h_s; \tag{5a}$$
$$n_d = V^{-1} \sum_{k\kappa} \kappa n_{k\kappa}, \qquad n_d^{\rm loc} = n_d - n_d^0;$$
$$n_d^0 = cN[S(S+1)/3T] h_d = r_d^0 h_d. \tag{5b}$$

We omit the details of the linearization and Fourier transformation procedure[13] and just give the final result for the Fourier components $n_p(q, \omega)$ and $n_d(q, \omega)$ of the angular momentum densities:

$$(\omega - \mathbf{v}_p \cdot \mathbf{q}) n_p - \mathbf{v} \cdot \tfrac{1}{2}\mathbf{q} (\partial f^0/\partial \epsilon_p) h_s$$
$$= -i\gamma^0 (n_p^{\rm loc} - \langle n_p^{\rm loc} \rangle) - i\gamma_s^0 \langle n_p^{\rm loc} \rangle$$
$$- i\gamma_d^0 (\partial f^0/\partial \epsilon_p) n_d^{\rm loc}/\rho_F - i\alpha_s^0 n_p^{\rm loc}, \tag{6a}$$
$$\omega n_d = i\gamma_s^0 n_s^{\rm loc} - i(\gamma_d^0 + \alpha_d^0) n_d^{\rm loc}. \tag{6b}$$

Here $\langle n_p \rangle$ is the average of n_p taken over the Fermi surface of the conduction electrons, and the various relaxation frequencies can be expressed in terms of t and τ as

$$\gamma_s^0 = \tfrac{2}{3}\pi\rho_F cN \mid \tau \mid^2 S(S+1) = 2\pi\rho_F T r_d^0 \mid \tau \mid^2, \tag{7a}$$
$$\gamma_d^0 = \pi\rho_F{}^2 T \mid \tau \mid^2 = 2\pi\rho_F T r_s^0 \mid \tau \mid^2, \tag{7b}$$
$$\gamma^0 = 2\pi\rho_F cN [\mid t \mid^2 + \tfrac{1}{4} S(S+1) \mid \tau \mid^2]. \tag{7c}$$

Regarding the q dependence of Eqs. (6), one can distinguish three different limiting cases. There is first a relaxation regime for $q \to 0$. With increasing q, a diffusion regime is reached and further increase of q finally yields a collisionless regime for the conduction electrons in which Landau damping is predominant. We first discuss the $q=0$ case.

III. RELAXATION REGIME

If one sets $q=0$ in (6a) and sums over p, one obtains together with (6b) two equations for the total angular momentum of each spin species:

$$[\omega + i(\gamma_s^0 + \alpha_s^0)] n_s - i\gamma_d^0 n_d = i(\gamma_s^0 + \alpha_s^0) n_s^0 - i\gamma_d^0 n_d^0, \tag{8a}$$
$$-i\gamma_s^0 n_s + [\omega + i(\gamma_d^0 + \alpha_d^0)] n_d = -i\gamma_s^0 n_s^0 + i(\gamma_d^0 + \alpha_d^0) n_d^0. \tag{8b}$$

Apart from the terms proportional to γ_s^0 and γ_d^0 on the right-hand sides of these equations, they agree with Hasegawa's equations[18] if $g_s = g_d$. If the g factors are not equal, one has to distinguish between equations for magnetizations and angular momenta. This leads to changes in the corresponding off-diagonal values of the relaxation frequencies in Eqs. (8). In our notation the two s-d relaxation rates, according to (7), are related by

$$\gamma_s^0 r_s^0 = \gamma_d^0 r_d^0 = \gamma_s^0 \chi_s^0 / g_s^2 = \gamma_d^0 \chi_d^0 / g_d^2, \tag{9}$$

which agrees with Hasegawa's detailed balance condition for equal g factors. Equations (8) satisfy three conditions:

(i) In the static limit $\omega = 0$, the solutions approach their static values n_s^0 and n_d^0.

(ii) The energy absorption is positive.

(iii) The total angular momentum $n_s + n_d$ is conserved if $\alpha_s^0 = \alpha_d^0 = 0$ (regardless of the g values).

Usually,[17,18] (i) and (ii) are satisfied by violating Eq. (9) and (iii). In order to determine the frequency dependence of the susceptibility, one has to solve (8) for n_s and n_d and multiply each by its g value to obtain the magnetizations $m_s = g_s n_s$ and $m_d = g_d n_d$. The susceptibility then is given as $\chi_{\rm tot}(\omega) = (m_s + m_d)/B$. The solution of (8) is

$$m_s = \left(1 - \frac{\omega^2}{D}\right) m_s^0 - \frac{\omega}{D} i \left\{ (\gamma_d^0 + \alpha_d^0) m_s^0 + \frac{\gamma_d^0 g_s m_d^0}{g_d} \right\}, \tag{10a}$$

$$m_d = \left(1 - \frac{\omega^2}{D}\right) m_d^0 - \frac{\omega}{D} i \left\{ \frac{\gamma_s^0 g_d m_s^0}{g_s} + (\gamma_s^0 + \alpha_s^0) m_d^0 \right\}, \tag{10b}$$

with the denominator

$$D = [\omega + i(\gamma_s^0 + \alpha_s^0)][\omega + i(\gamma_d^0 + \alpha_d^0)] + \gamma_s^0 \gamma_d^0. \tag{11}$$

In the limit of small spin-lattice relaxation frequencies $\alpha_{s,d}$ and $g_s \sim g_d$, Eq. (10) exhibits the familiar bottleneck: The absorptive part of $\chi_{\rm tot}$ is dominated by a narrow peak with a width controlled by the spin-lattice relaxation. This width does not depend on γ_s and γ_d separately but only on the ratio $\gamma_d/(\gamma_s + \gamma_d)$. In this ratio, the anomalous $\mid \tau \mid^2$ of (7) has dropped out.

In order to apply the results to EPR experiments, one has to include the dc field B^z, which adds the two Larmor precession terms

$$\omega_s = \mu_B g_s B^z = h_s{}^z, \qquad \omega_d = \mu_B g_d B^z = h_d{}^z \tag{12}$$

to the diagonal part of the equations for the transverse magnetization. Furthermore one has to take into account the "molecular field terms" by replacing the external field everywhere by the corresponding effective field

$$\mathbf{h}_s^{\text{eff}}=\mathbf{h}_s+\lambda_s\mathbf{n}_s+\lambda\mathbf{n}_d, \tag{13a}$$

$$\mathbf{h}_d^{\text{eff}}=\mathbf{h}_d+\lambda\mathbf{n}_s+\lambda_d\mathbf{n}_d. \tag{13b}$$

The diagonal terms λ_s, λ_d then have no effect on the Larmor frequencies but lead to an enhancement of the static susceptibilities and a "slowing down" of the *s-d* relaxation (for ferromagnetic sign). These effects can be taken into account by introducing the new quantities

$$\alpha_s=\alpha_s^0(1-\lambda_s r_s^0), \qquad \gamma_s=\gamma_s^0(1-\lambda_s r_s^0), \tag{14}$$

and

$$r_s=r_s^0/(1-\lambda_s r_s^0) \tag{15}$$

(and three similar quantities with $s\rightleftarrows d$). These quantities again satisfy the detailed balance condition (9). Furthermore, the diagonal exchange terms will renormalize the original exchange interaction of the Kondo problem. We will assume that in the calculation of t and τ entering the quantities (7), this renormalized value of J has been used already. The final result for the transverse magnetizations including off-diagonal exchange λ can be cast into a form similar to (10) by introducing the quantities[17]

$$\tilde{\omega}_s=\omega_s+\lambda n_d{}^z-i(\alpha_s+\gamma_s)-i\lambda r_d\gamma_d, \tag{16}$$

$$\zeta_s=\lambda n_s{}^z-i\gamma_d-i\lambda r_s(\alpha_s+\gamma_s), \tag{17}$$

$$\tilde{m}_s=(g_s{}^2r_s+\lambda g_s g_d r_s r_d)\mu_B B/(1-\lambda^2 r_s r_d) \tag{18}$$

(and three similar quantities with $s\rightleftarrows d$).

One then finds

$$m_s=\left(1-\frac{\omega^2}{D}\right)\tilde{m}_s+\frac{\omega}{D}\left\{\tilde{\omega}_d\tilde{m}_s+\frac{\zeta_s g_s\tilde{m}_d}{g_d}\right\}, \tag{19a}$$

$$m_d=\left(1-\frac{\omega^2}{D}\right)\tilde{m}_d+\frac{\omega}{D}\left\{\tilde{\omega}_s\tilde{m}_d+\frac{\zeta_d g_d\tilde{m}_s}{g_s}\right\}, \tag{19b}$$

with the denominator

$$D=(\omega-\tilde{\omega}_s)(\omega-\tilde{\omega}_d)-\zeta_s\zeta_d. \tag{20}$$

IV. GENERAL CASE ($q\neq 0$)

In the general case of nonzero q, one has to treat (6a) more carefully. The solution in this case (omitting dc and exchange fields) takes the form

$$n_s=[1-(\omega^2/D)(1-L)]n_s^0 - (\omega/D)i\{(\gamma_d^0+\alpha_d^0)n_s^0+\gamma_d^0 n_d^0\}(1-L), \tag{21a}$$

$$n_d=(1-\omega^2/D)n_d^0-(\omega/D)i\{\gamma_s^0 n_s^0(1-L) +[\gamma_s^0+\alpha_s^0+(\gamma^0-\gamma_s^0)L]n_d^0\}, \tag{21b}$$

with

$$D=\{\omega+i[\gamma_s^0+\alpha_s^0+(\gamma^0-\gamma_s^0)L]\} \times[\omega+i(\gamma_d^0+\alpha_d^0)]+\gamma_s^0\gamma_d^0(1-L). \tag{22}$$

The q dependence occurs via the Lindhard function

$$L\left(\frac{\omega+i(\alpha_s^0+\gamma^0)}{v_F q}\right)=\left\langle\frac{-\mathbf{v}_p\cdot\mathbf{q}}{\omega+i(\alpha_s^0+\gamma^0)-\mathbf{v}_p\cdot\mathbf{q}}\right\rangle_{|p|=mv_F} \tag{23}$$

The two limiting forms of this function for small and large values of q are

$$L(x)=-1/3x^2, \qquad v_F q\ll|\omega+i(\alpha_s^0+\gamma^0)| \tag{24}$$

and

$$L(x)=1+\tfrac{1}{2}i\pi x, \qquad v_F q\gg|\omega+i(\alpha_s^0+\gamma^0)|. \tag{25}$$

In particular, for $q\to 0$, $L\to 0$ and one recovers the results (10) of the relaxation regime.

For slightly larger values of q but still in the regime (24), one finds aside from relaxation widths a diffusion width proportional to q^2. For small values of α_s and $g_s\sim g_d$, the diffusion width, for instance, is given by $D_{\text{eff}}^0 q^2$ with[19]

$$D_{\text{eff}}^0=[\gamma_d^0/(\gamma_s^0+\gamma_d^0)](v_F^2/3\gamma^0). \tag{26}$$

The diffusion constant therefore is related to the total *s-d* scattering cross section contained in γ^0 and therefore shows the same kind of anomalies as the electrical and thermal conductivity.

Exchange corrections can be taken into account again in the molecular field approximation in analogy to (14) and (15). A nonzero conduction-electron exchange constant λ_s, for instance, changes (26) into

$$D_{\text{eff}}=[\gamma_d/(\gamma_s+\gamma_d)](v_F^2/3\gamma), \tag{27}$$

where γ_s, γ_d is again given by (14), whereas γ is given by

$$1/\gamma=(1-\lambda_s r_s^0)/\gamma^0. \tag{28}$$

Thus the diffusion pole also becomes narrower for ferromagnetic exchange.

An interesting regime is reached at even larger q values where (25) applies. One may then to lowest order replace L by 1. Looking at (21) and (22), this yields

$$n_s=n_s^0,$$

$$n_d=[i(\gamma_d^0+\alpha_d^0)/\omega+i(\gamma_d^0+\alpha_d^0)]n_d^0. \tag{29}$$

In this order, the s electrons are in equilibrium and the absorption takes place at the d electrons alone. In higher order, according to (25) there will be some absorption at the conduction electrons as well, but at low temperatures where n_d^0 is large, the local-moment absorption will dominate.

q values as large as in this regime are obtained, for instance, in inelastic neutron scattering. Experiments using such tools can therefore eventually yield

rather direct information about the spin-flip scattering cross section contained in γ_d.

V. VALIDITY OF KINETIC EQUATION

The kinetic equations (1) and (2) have been derived[13] using the method of Kadanoff and Baym.[20] There are several nontrivial points which have not been mentioned explicitly in Ref. 13. Here we want to discuss them briefly as far as they are related to the Kondo problem.

(a) In order to determine the collision integrals (2), one needs the collision rates $M_s^<$ and $M_d^<$ which are closely related to the self-energies of the s and d electrons. M_s can be expressed in terms of the scattering matrix $T(\epsilon)$, Eq. (4). In Suhl's approximation, T contains only single-particle intermediate-state cuts.[4] It can therefore be decomposed into $T(\epsilon)=T^p(\epsilon)+T^h(\epsilon)$ where $T^p(T^h)$ contains only single-particle (single-hole) intermediate states. The collision integral (2a) then contains only the scattering cross section which is proportional to $|T^p(\epsilon+i\eta)+T^h(\epsilon+i\eta)|^2$. If an analogous kind of "cut philosophy" is applied to the calculation of M_d, it turns out that the corresponding cross section is not exactly proportional to $|T(\epsilon+i\eta)|^2$ but rather to $|T^p(\epsilon+i\eta)+T^h(\epsilon-i\eta)|^2$. If this result were used to calculate the collision integral I' of (2b), one would therefore obtain a kinetic equation which would violate (iii) of Sec. III. We have therefore used $T(\epsilon+i\eta)$ in the calculation of both collision integrals (2) in order to have a transport theory consistent with conservation laws. Although a power expansion of $T^h(\epsilon+i\eta)-T^h(\epsilon-i\eta)$ in powers of J yields terms which are less singular in every order of J than the corresponding ones for $T^h(\epsilon\pm i\eta)$ separately, it seems difficult at present to find a straightforward diagrammatic derivation of kinetic equations consistent with conservation laws.

(b) The inclusion of a dc field by just adding Larmor precession (12) terms to the diagonal part of the equations for the transverse susceptibility neglects the frequency dependence of the scattering matrix which is produced by such a dc field. Effects of this kind have been considered[10] in second order of perturbation theory and neglecting off-diagonal terms in the kinetic equations. It would be interesting to look at these effects in a more general treatment.

(c) The static susceptibilities $\chi_s^0=g_s^2\gamma_s^0$ and $\chi_d^0=g_d^2\gamma_d^0$ occurring on the right-hand side of the kinetic equations according to (5) are just the ordinary Pauli and Curie susceptibilities. The static solution of (8) at $\omega=0$ thus shows no Kondo anomalies at all. This defect is due to the approximations inherent in the Boltzmann equation in which the equilibrium properties are generally treated on a lower level than the relaxation effects. Although it looks promising that reasonable equilibrium properties can be calculated from diagrams which are closely related[14,15] to our kinetic equations, and although it is known in principle[20] how the correct equilibrium behavior can be incorporated into the transport theory, it remains to be shown how this can be done in detail.

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