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Spin fluctuations and relaxation of local moments in heavy-fermion systems

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Abstract. The Korringa relaxation-rate process is derived for a heavy-fermion system. The approach is based on a time-dependent perturbation theory, which avoids the use of the Ward identity and ignores possible effects due to 4f rare-earth concentration, employing instead the Keldysh formalism. The Yoshimori–Kasai Hamiltonian is used to model an electron spin resonance (ESR) experiment, and the spin relaxation rate is derived from Green functions obtained within the context of this model. The low-temperature equilibrium properties of the model are briefly discussed, and an expression for the enhanced mass is derived indicating the transition from the zero-temperature coherent state to the finite-temperature scattering state. The Korringa rate process is expressed as an integral equation, and the contribution due to the correlation of fluctuating impurity spins is identified as a resonance near the Fermi energy within the integrand of this equation. The results are compared to experimental ESR work on UBe_{13} , CeCu_2Si_2 and CeAl_3 , and found to have good qualitative agreement. The model connects the exchange coupling parameter J to the observation of temperature-dependent non-linear behaviour of the spin relaxation rate in heavy-fermion systems.

1. Introduction

For over a decade heavy-fermion compounds have generated a vast number of experimental and theoretical investigations. Compounds such as UBe_{13} , UPt_3 , U_6Fe and CeCu_2Si_2 are representative of this class of material, which exhibit enormous values of magnetic susceptibility and coefficient γ of the linear term in the electronic specific heat. CeCu_2Si_2 has been classified as a Kondo lattice, exhibiting, below the Kondo temperature T_K , a logarithmically increasing electrical resistance with decreasing temperature similar to dilute Kondo alloys. Also, the rare-earth ion of the intermediate-valence materials fluctuates between two charge states such as Ce^{4+} and Ce^{3+} (f^0 , f^1), other charge states being considered inaccessible in energy. Gandra *et al* (1985) measured the electron spin resonance (ESR) of UBe_{13} doped with local moments of various rare-earth impurities, but were unable to detect any significant anomalies in the thermal broadening of the ESR lines, which was surprising because of the expectation of an enhanced relaxation due to the unusual heavy-fermion ground state. Subsequently, Coldea *et al* (1987) discovered enhanced broadening of ESR lines of the Gd-doped Kondo-system isostructure $\text{Y}_{1-x}\text{Ce}_x\text{Al}_2$. A short time later, Schlott *et al* (1988) discovered large temperature-dependent anomalies in the Gd ESR relaxation rates around the Kondo temperature of Gd-doped CeCu_2Si_2 and CeAl_3 , and attributed this enhanced spin relaxation to Ce 4f spin fluctuations mediated by Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling.

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In an attempt to explain the strikingly normal behaviour in the studies of Gandra *et al* (1985), Simanek and Sasahara (1987) argue that the enhancement of the Korringa rate and its anomalous temperature dependence of heavy-fermion compounds is absent in ESR studies because the Korringa rate is unaffected by the heavy-fermion renormalizations. This proposal was made before the discovery of the non-linear temperature dependence of spin relaxation in $Y_{1-x}Ce_xAl_2$, $CeCu_2Si_2$ and $CeAl_3$, and unfortunately the theoretical approach is unable to account for these non-linearities. Simanek and Sasahara use the model of Yoshimori and Kasai (1983) to derive the spin relaxation rate from the transverse susceptibility by using the Ward identity. They argue that the vertex function associated with the local spin fluctuation is given by the sum of all Feynman graphs by inserting a free vertex in each bare conduction-electron line in the set of diagrams obtained by a perturbation expansion of the fully renormalized conduction-electron propagator. They contend that, because the self-energy of the dressed conduction-electron propagator does not contain any bare conduction propagator, there can be no vertex correction to the spin fluctuation propagator, and therefore the transverse susceptibility is simply given by the product of fully renormalized conduction-electron Green functions.

Implicit in this reasoning, however, is that the use of the Ward identities demands that certain conservation laws are preserved by the original Hamiltonian. Early theoretical work on dense Kondo systems (Read and Newns 1983) shows that the original Hamiltonian does not conserve particle numbers of f electrons on a given lattice site. This fact may cast doubt on the satisfaction of the Ward identities where the Green functions are evaluated using the Hamiltonian of Yoshimori and Kasai. Also, in order to calculate Green functions in a statistical system by the above approach, one must assume that the system is very close to thermodynamic equilibrium. In carrying out this programme, the deviation of the system from the Gibbs distribution must be small, i.e. external fields must be small. In a typical ESR experiment the external field, aside from the RF field, may be quite large, and one cannot assume the existence of an approximate thermodynamic equilibrium.

Ochi and de Menezes have proposed a model in which the non-linear temperature-dependent behaviour of the Korringa relaxation process could be described through a large change of the Fermi energy referred to the position of a 'hybridization gap' (Ochi and de Menezes 1988). This gap appears in the local density of conduction states at the impurity site due to the hybridization between the $4f$ states of neighbouring Ce ions with conduction states at the impurity site. In this model there arises a large variation with impurity concentration x of the Fermi energy position relative to the 'hybridization hole', and the model gives good agreement with many experimental ESR linewidth results on Ce-based intermediate-valence materials (Ochi and de Menezes 1991). This model, however, reproduces the conventional linear Korringa rate in the limit as x goes to zero, in which case ($x = 0$) there are no $4f$ rare-earth ions surrounding the impurity. The results of Gandra *et al* (1985) on UBe_{13} and the results of Gandra *et al* (1987) on UPt_3 indicate that these materials do not seem to provide a 'hybridization hole' correction to the ESR relaxation process. These experimental results lead to the conclusion that the coupling between the uranium ion and rare-earth ion through the s - f electron interaction is very small and that observation of the non-linearities in Korringa rate processes in these materials is not a function of rare-earth concentration but rather a function of interaction strength.

In this paper we shall take a completely different approach to the problem of computing the spin relaxation rate of heavy-fermion compounds in an ESR experiment by avoiding the use of the Ward identity and ignoring possible effects due to the concentration of rare-earth ions. Our approach will be closer to the actual experimental situation by rejecting the assumption that the system approaches thermodynamic equilibrium. The problem will be

treated by perturbation theory within the context of the Keldysh formalism. Keldysh (1965) provided an elegant method of applying Feynman diagrammatic techniques to quantum-statistical systems far from equilibrium. We will also adopt the time-dependent approach of Cini (1980) in computing the electron propagators that enter the theory. This time-dependent approach provides a simple technique for calculating transport properties such as the Korringa relaxation. The physical quantities such as magnetization and susceptibility are computed, as Simanek and Sasahara have done, using the model of Yoshimori and Kasai. This model is derived from the 'periodic Anderson model', but the assumption is made that one can neglect all contributions to the self-energy from perturbation terms that include inter-site propagators, and we base our approximation on the *ansatz* that each f state in the lattice is independent, i.e. there are no interatomic f - f correlations. Because we assume that the system does not undergo a magnetic transition, the question of f - f site correlations is not crucial to the model, but unfortunately the model cannot account for possible magnetic frustration effects in heavy fermions as proposed by Coles *et al* (1987) and Kugel and Khomskii (1985). In spite of several shortcomings, however, this model provides a representation of the many-body effects of the Coulomb repulsion between f electrons and the main features of dense Kondo systems.

In order to discuss the spin relaxation rate in rigorous quantum-mechanical terms, a model, following Yoshimori and Kasai, of a heavy-fermion compound subjected to an ESR experiment will be developed in the following section. The time-dependent problem of a dense Kondo system with embedded magnetic impurities will be formulated, and the magnetic susceptibility will be seen to be an expression in terms of retarded Green functions and the retarded s -electron self-energy for the Anderson model of a single magnetic impurity. Using a technique developed in a previous paper (Wright 1994), the retarded Keldysh self-energy is evaluated in the appendix. A by-product of evaluating this self-energy with the Keldysh formalism is that the temperature associated with the divergence of the vertex part (Kondo 1964) is pushed to infinity, and since this divergence is implicit in the equilibrium technique of Simanek and Sasahara at T_K , we see a further advantage of abandoning an approach based on free vertex insertions. The rate of the Korringa relaxation process will be obtained by using a procedure (Cini 1980) where the system Hamiltonian is assumed to evolve in time to a constant final value and the asymptotic form of the time-dependent Green function is obtained from its frequency-dependent Fourier transform. The low-temperature equilibrium behaviour of the model will be briefly discussed in terms of a Fermi liquid and Luttinger's theorem, and an expression for the enhanced mass will be derived that demonstrates the transition from the zero-temperature coherent state to the finite-temperature non-coherent scattering state. Upon evaluating the ESR relaxation process, we will see how non-linearities arise around the Kondo temperature and how the impurity spin probes the renormalized conduction-electron density of states.

2. Formulation

The model of Yoshimori and Kasai (1983) describes a dense Kondo system using the Hamiltonian

$$\begin{aligned}
 H_{YK} = & \sum_{k\sigma} \epsilon_{k\sigma} c_{k\sigma}^{\dagger} c_{k\sigma} + V \sum_{k\sigma} (c_{k\sigma}^{\dagger} f_{k\sigma} + c_{k\sigma} f_{k\sigma}^{\dagger}) + \sum_{k\sigma} \epsilon_V(k) f_{k\sigma}^{\dagger} f_{k\sigma} + U \\
 & \times \sum_i f_{i\uparrow}^{\dagger} f_{i\uparrow} f_{i\downarrow}^{\dagger} f_{i\downarrow}
 \end{aligned} \tag{1}$$

where $\epsilon_V(k)$ is the band energy of valence electrons, U is the on-site Coulomb energy and V is the conduction-electron-f-electron mixing parameter. Yoshimori and Kasai make the important assumption that the inter-site contributions to the self-energy of the one-particle Green function and to the vertex part can be rejected. They also ensure that the model describes a metal by giving a small dispersion to the f-band energy,

$$\epsilon_V(k) = \alpha \epsilon_k \quad \alpha \ll 1. \quad (2)$$

We modify the above Hamiltonian to include a small time-dependent perturbation due to the RF field of the ESR experiment,

$$\begin{aligned} H &= H_{YK} + H_{RF} + H_{Zeeman} \\ H_{RF} &= -\frac{1}{2} \gamma \hbar h (S_+ + S_-) \cos(\Omega t) \\ H_{Zeeman} &= \gamma \hbar \mathbf{S} \cdot \mathbf{H}_{ex} \end{aligned} \quad (3)$$

where γ is the magnetogyric ratio, h is the RF magnetic field magnitude, (S_+, S_-) are angular momentum raising and lowering operators that characterize a local moment of spin \mathbf{S} , Ω is the frequency of the linearly polarized RF field, and \mathbf{H}_{ex} is the externally applied steady magnetic field.

Using standard techniques we write for the retarded f-electron Green function and the retarded conduction-electron Green function, respectively,

$$\begin{aligned} G_f^r(\mathbf{r}, \mathbf{r}'; \omega) &= g_f^r(\mathbf{r}, \mathbf{r}'; \omega) V G_c^r(\mathbf{r}, \mathbf{r}'; \omega) \\ &+ \iint g_f^r(\mathbf{r}, \mathbf{r}_1; \omega) \Sigma_{sf}^r(\mathbf{r}_1, \mathbf{r}_2; \omega) G_f^r(\mathbf{r}_2, \mathbf{r}'; \omega) d\mathbf{r}_1 d\mathbf{r}_2 \end{aligned} \quad (4)$$

$$G_c^r(\mathbf{r}, \mathbf{r}'; \omega) = g_c^r(\mathbf{r}, \mathbf{r}'; \omega) + g_c^r(\mathbf{r}, \mathbf{r}'; \omega) V G_f^r(\mathbf{r}, \mathbf{r}'; \omega) \quad (5)$$

where the lower-case g represents a bare propagator of a non-interacting particle, the upper-case G represents a fully dressed propagator, and Σ_{sf}^r is the Keldysh retarded self-energy for a single impurity located at the origin of the coordinate system. Because the assumption is made that all contributions to the self-energy from perturbation terms that include inter-site propagators can be neglected, we follow Yoshimori and Kasai and assert that Σ_{sf}^r has the same form as that in the Anderson model of a single impurity. We have consigned the derivation of the Keldysh retarded self-energy for the Anderson model of a single magnetic impurity to the appendix. Using the following relations

$$g_c^{-1}(\mathbf{r}; \omega) g_c^r(\mathbf{r}, \mathbf{r}'; \omega) = \delta(\mathbf{r} - \mathbf{r}') \quad (6)$$

$$g_f^{-1}(\mathbf{r}; \omega) g_f^r(\mathbf{r}, \mathbf{r}'; \omega) = \delta(\mathbf{r} - \mathbf{r}') \quad (7)$$

$$g_c^{-1}(\mathbf{r}; \omega) = \omega + (\hbar^2/2m) \nabla_r^2 - H_{RF} + H_{Zeeman} \quad (8)$$

$$g_f^{-1}(\mathbf{r}; \omega) = \omega + \alpha(\hbar^2/2m) \nabla_r^2 - H_{RF} + H_{Zeeman} \quad (9)$$

we can write the Keldysh equation (Keldysh 1965) for the retarded conduction-electron Green function:

$$G_c^r(\mathbf{r}, \mathbf{r}'; \omega) = g_c^r(\mathbf{r}, \mathbf{r}'; \omega) + \iint g_c^r(\mathbf{r}, \mathbf{r}_1; \omega) \Sigma^r(\mathbf{r}_1, \mathbf{r}_2; \omega) G_c^r(\mathbf{r}_2, \mathbf{r}'; \omega) d\mathbf{r}_1 d\mathbf{r}_2. \quad (10)$$

Because the Zeeman term in equations (8) and (9) merely provides a constant displacement in energy, we have absorbed it in the definition of ω :

$$\omega \rightarrow \omega + H_{\text{Zeeman}}$$

Operating on both sides of equation (10) with g_c^{-1} we find:

$$(\omega - \nabla_r^2)G_c^r(\mathbf{r}, \mathbf{r}'; \omega) = \delta(\mathbf{r} - \mathbf{r}') + \int \Sigma^r(\mathbf{r}, \mathbf{r}_1; \omega)G_c^r(\mathbf{r}_1, \mathbf{r}'; \omega) d\mathbf{r}_1 \quad (11)$$

where

$$\Sigma^r(\mathbf{r}, \mathbf{r}_1; \omega) = \frac{V^2 \delta(\mathbf{r}_1 - \mathbf{r}')}{\omega - \alpha(\hbar^2/2m)\nabla_{r_1}^2 - \Sigma_{\text{sf}}^r(\omega) - H_{\text{RF}}} \quad (12)$$

and

$$\Sigma_{\text{sf}}^r(\omega) = \int_{-\infty}^{\infty} \Sigma_{\text{sf}}^r(0, \mathbf{r}_1; \omega) d\mathbf{r}_1. \quad (13)$$

Upon taking the Fourier transform of equation (11) with respect to \mathbf{r}' , we find

$$G_c^r(\mathbf{r}, \mathbf{k}; \omega) = \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{(2\pi)^{3/2}} \{\omega - a^2 - H_{\text{RF}} - V^2[\omega - \alpha a^2 - \Sigma_{\text{sf}}^r(\omega) - H_{\text{RF}}]^{-1}\}^{-1} \quad (14)$$

where

$$a^2 = (\hbar^2/2m)k^2. \quad (15)$$

For the sake of simplicity of calculation, the assumption is made that the lattice attains enough symmetry so that the following dispersion relation may be written:

$$\nabla_r^2 e^{-i\mathbf{k}\cdot\mathbf{r}} = -k^2 e^{-i\mathbf{k}\cdot\mathbf{r}}. \quad (16)$$

We are concerned with the calculation of the time-ordered finite-temperature Green function. Defined in the standard way it is

$$G^T(\mathbf{r}, t; \mathbf{r}', t') = -i \langle T[\psi(\mathbf{r}, t)\psi^\dagger(\mathbf{r}', t')] \rangle \quad (17)$$

where T represents the time ordering and ψ are Heisenberg electron field operators. The angular brackets denote a thermal average taken with an unperturbed density matrix and the number density is given by

$$\rho(\mathbf{r}, t) = -i \lim_{t' \rightarrow t+0} \lim_{\mathbf{r}' \rightarrow \mathbf{r}} G^T(\mathbf{r}, t; \mathbf{r}', t'). \quad (18)$$

In order to connect the time-ordered Green function to the retarded and advanced Green functions of the Keldysh formalism, one can show (Cini 1980) that, provided τ is any time earlier than t and t' ,

$$\begin{aligned} G^T(\mathbf{r}, t; \mathbf{k}', t') &= i\Theta(t' - t) \sum_q f_q G^r(\mathbf{k}, t; \mathbf{q}, \tau) G^a(\mathbf{q}, \tau; \mathbf{k}, t') - i\Theta(t - t') \\ &\times \sum_q (1 - f_q) G^r(\mathbf{k}, t; \mathbf{q}, \tau) G^a(\mathbf{q}, \tau; \mathbf{k}, t') \end{aligned} \quad (19)$$

where f_k is the Fermi function and $\Theta(t - t')$ is the Heavyside step function. We follow Cini (1980) and consider the initial state of conduction electrons to have an unperturbed system Hamiltonian H_{YK} . At time $t = 0$ we turn on the external fields ($H_{ex} \gg h$) and we are thus interested in G^T of equation (19) for $t > 0$ and $t' = t + 0$ and τ is taken to be just less than 0. We have $H \approx H_{YK}$ for $t < 0$ and $H = H_{YK} + H_{Zeeman} = H^f = \text{constant}$ for $t > 0$. A thermal average over the eigenstates of H^f yields the result

$$G^T(\mathbf{k}, t; \mathbf{k}', t') = G^{r,f}(\mathbf{k}, \mathbf{k}'; t) \quad (20)$$

where $G^{r,f}$ is a single-particle electron Green function calculated with the constant final-state Hamiltonian. The transverse component of the magnetization at position \mathbf{r} , and at time t , is

$$M_t(\mathbf{r}, t) = \text{Tr}_S \left(\gamma \frac{(S_+ + S_-)}{2} \sum_{\mathbf{k}} f_k |G^{r,f}(\mathbf{r}, \mathbf{k}; t)|^2 \right) \quad (21)$$

where Tr_S indicates a trace over angular momentum states.

Following Simanek and Sasahara (1987) the rate of the Korringa relaxation process is expressed in terms of the local transverse spin susceptibility by

$$1/T_1 = (Ck_B T / \Omega) \text{Im } \chi_r(\mathbf{r}, \Omega) \quad (22)$$

where Ω is the resonance frequency of the experiment and C is a constant that is proportional to the square of the exchange coupling parameter J used to evaluate s-f self-energy. We calculate the time-dependent transverse susceptibility by differentiating equation (21) and assuming that the RF field h is small:

$$\tilde{\chi}_t(\mathbf{r}, t) = \lim_{h \rightarrow 0} \frac{\partial}{\partial h} M_t(\mathbf{r}, t). \quad (23)$$

The frequency-dependent transverse susceptibility is then obtained from equation (23) by evaluating its Fourier transform:

$$\chi_t(\mathbf{r}, \Omega) = \int_{-\infty}^{\infty} \tilde{\chi}_t(\mathbf{r}, t) e^{i\Omega t} dt. \quad (24)$$

We see that, in order to obtain the Korringa relaxation process, $G^{r,f}(\mathbf{r}, \mathbf{k}; t)$ must be determined. If the assumption is made that the system Hamiltonian evolves to a constant value after the perturbing field is turned on, then the asymptotic form of the Fourier frequency transform of equation (13) may be used to obtain the magnetization of equation (21). This will be done in the following section.

3. The transverse susceptibility

In this section the magnetization of the spin-impurity system will be evaluated in three steps. First, the frequency-dependent retarded conduction-electron Green function (equation (14)) will be expanded to first order in the small perturbing RF field. Secondly, the asymptotic form of the time-dependent Green function used to calculate the magnetization of equation (21) will be obtained from the poles of the frequency-dependent Green function by the method of

'Fourier transforms of generalized functions'. The third step will be to write equation (21) in terms of an integral equation of this time-dependent Green function.

The Green function of equation (14) is expanded to first order in the RF field and found to be:

$$G_c^r(\mathbf{r}, \mathbf{k}; \omega) = \frac{e^{i\mathbf{k}\cdot\mathbf{r}}[\omega - \alpha a^2 - \Sigma_{sf}^r(\omega) - H_{RF}]}{[a^2 - f_+(\omega)][a^2 - f_-(\omega)]} + e^{i\mathbf{k}\cdot\mathbf{r}} H_{RF} \\ \times \left(\frac{[2\omega - (1 + \alpha)a^2 - \Sigma_{sf}^r(\omega)][\omega - \alpha a^2 - \Sigma_{sf}^r(\omega)]}{[a^2 - f_+(\omega)]^2[a^2 - f_-(\omega)]^2} \right) \quad (25)$$

where

$$f_{\pm}(\omega) = \frac{(1 + \alpha)\omega - \Sigma_{sf}^r(\omega) \pm \{[(1 - \alpha)\omega - \Sigma_{sf}^r(\omega)]^2 + 4\alpha V^2\}^{1/2}}{2\alpha}. \quad (26)$$

We seek the Fourier frequency transform of equation (25), but unfortunately the transform of this function is not known. Since we are interested in the long-time expansion of the Green function, we may approximate the Fourier transform by an asymptotic expression. This is accomplished by evaluating the Fourier transform in the vicinity of the poles of equation (25) (Lighthill 1959). We approximate equation (25) by

$$G_c^r(\mathbf{r}, \mathbf{k}; \omega) \simeq \frac{g_1(\mathbf{r}, \mathbf{k}; \omega)}{(\omega - \zeta_1)(\omega - \zeta_2)} + \frac{g_2(\mathbf{r}, \mathbf{k}; \omega)}{(\omega - \zeta_1)^2(\omega - \zeta_2)^2}. \quad (27)$$

In this form the asymptotic expression for the Fourier transform of equation (26) can be easily found. The poles of equation (27), ζ_1 and ζ_2 , are complex numbers obtained by expanding $f(\omega)$ about a^2 :

$$f_{\pm}(\omega) \simeq f_{\pm}(a^2) + (\omega - a^2)[df_{\pm}(\omega)/d\omega]_{\omega=a^2}. \quad (28)$$

The poles of equation (27) are thus

$$\zeta_{1,2} = a^2 - \left(\frac{R_1 e^{i\theta_1} \pm R_2 e^{i\theta_2}}{(\lambda e^{i\phi} + \alpha) \pm (R_1 e^{i(\theta_1 - \theta_2)}/R_2)(\lambda e^{i\theta} - \alpha)} \right) \quad (29)$$

where we have used the definitions

$$R_1(a^2)e^{i\theta_1(a^2)} = (1 - \alpha)a^2 - \Sigma_{sf}^r(a^2) \\ R_2(a^2)e^{i\theta_2(a^2)} = (R_1^2 e^{2i\theta_1} + 4\alpha V^2)^{1/2} \\ \lambda(a^2)e^{i\phi(a^2)} = 1 - [\partial \Sigma_{sf}^r(\omega)/\partial \omega]_{\omega=a^2}. \quad (30)$$

Because α is a small fraction of unity, we can simplify ζ with the following approximation:

$$\zeta_{1,2} = a^2 \pm [R_2(a^2)/\lambda(a^2)]e^{i[\theta_2(a^2) - \phi(a^2)]}. \quad (31)$$

Also, the second term in equation (27) is dropped because it provides a correction of $O(\alpha^2)$ to the first term.

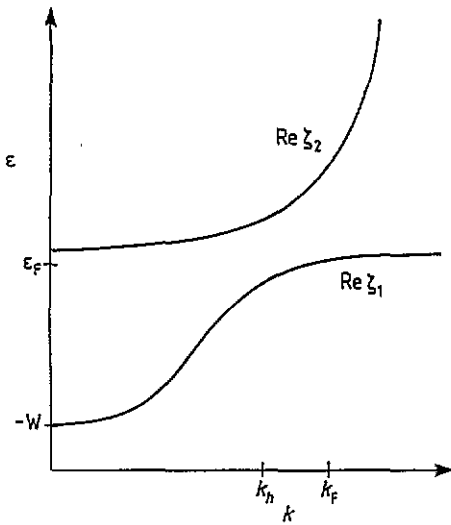


Figure 1. Sketch of energy ϵ versus momentum k of $\text{Re } \zeta_{1,2}$: k_h is the electron momentum corresponding to the minimum separation in energy between the quasiparticle bands; ϵ_F is the Fermi energy; and k_F is the quasiparticle momentum at the Fermi level.

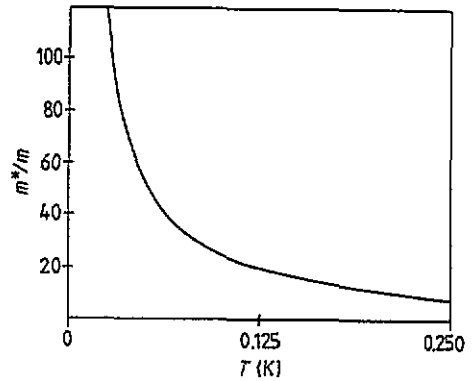


Figure 2. The mass enhancement factor as a function of temperature and evaluated with the parameters $V = 1.8$ eV and $\rho J/N = 0.50$.

We sketch in figure 1 the real parts of ζ_1 and ζ_2 versus the electron momentum k . We note that the real parts of the poles of the conduction-electron Green function with no applied external fields describe quasiparticle hybridized bands with $\text{Re } \zeta_1$ giving the dispersion relation for electrons in the lower band and $\text{Re } \zeta_2$ giving the dispersion relation for electrons in the upper band. The energy where the band gap is a minimum is given the symbol ϵ_h and its associated momentum is called k_h .

We must determine ϵ_F , which is a function of temperature. Because our energies are measured relative to the Fermi level, we have

$$\text{Re } \zeta_1(\epsilon_F) = 0. \tag{32}$$

One can show that, in the absence of self-energy corrections, $\epsilon_F = V$ in this model, and thus ϵ_F can be obtained by expanding $\text{Re } \zeta_1$ in a Taylor series about V :

$$\epsilon_F \simeq V - \frac{\text{Re } \zeta_1(\epsilon)}{\partial \text{Re } \zeta_1(\epsilon) / \partial \epsilon} \Big|_{\epsilon=V}. \tag{33}$$

Using the prescription of Lighthill the time-dependent conduction-electron propagator is found by evaluating the asymptotic form of the Fourier transform of equation (27):

$$\begin{aligned} G_c^T(\mathbf{r}, \mathbf{k}, t) = & \frac{-i\alpha e^{i\mathbf{k}\cdot\mathbf{r} + i \text{Re}(\zeta_1)t - \text{Im}(\zeta_1)t} \Theta(\text{Im}(\zeta_1)t) R_2(\epsilon_k) e^{i[\theta_2(\epsilon_k) - \phi(\epsilon_k)]}}{2(2\pi)^{3/2} V \lambda(\epsilon_k)} \\ & \times \left(R_1(\epsilon_k) e^{i\theta_1(\epsilon_k)} - \frac{R_2(\epsilon_k) e^{i[\theta_2(\epsilon_k) - \phi(\epsilon_k)]}}{\lambda(\epsilon_k)} - H_{\text{RF}} \right) \\ & + \frac{i\alpha e^{i\mathbf{k}\cdot\mathbf{r} + i \text{Re}(\zeta_2)t - \text{Im}(\zeta_2)t} \Theta(\text{Im}(\zeta_2)t) R_2(\epsilon_k) e^{i[\theta_2(\epsilon_k) - \phi(\epsilon_k)]}}{2(2\pi)^{3/2} V \lambda(\epsilon_k)} \\ & \times \left(R_1(\epsilon_k) e^{i\theta_1(\epsilon_k)} + \frac{R_2(\epsilon_k) e^{i[\theta_2(\epsilon_k) - \phi(\epsilon_k)]}}{\lambda(\epsilon_k)} - H_{\text{RF}} \right). \end{aligned} \tag{34}$$

The magnetization is found by using equation (32) and equation (3) in equation (21):

$$\begin{aligned}
 M_t(\mathbf{r}, t) = & \frac{S(S+1)(2S+1)}{16V^2} \frac{\alpha^2 \gamma^2 \hbar^3}{(2\pi)^3} \left(\frac{2m}{\hbar^2} \right)^{3/2} h \int_{-w}^{\infty} d\epsilon f_{\epsilon} \sqrt{\epsilon} \left(\frac{R_2(\epsilon)}{\lambda(\epsilon)} \right)^2 \cos(\Omega t) \\
 & \times \left[e^{-2\text{Im}(\zeta_1)t} \Theta(\text{Im}(\zeta_1)t) \left(R_1(\epsilon) \cos[\theta_1(\epsilon)] - \frac{R_2(\epsilon)}{\lambda(\epsilon)} \cos[\theta_2(\epsilon) - \phi(\epsilon)] \right) \right. \\
 & + e^{-2\text{Im}(\zeta_2)t} \Theta(\text{Im}(\zeta_2)t) \times \left(R_1(\epsilon) \cos[\theta_1(\epsilon)] + \frac{R_2(\epsilon)}{\lambda(\epsilon)} \right. \\
 & \left. \left. \times \cos[\theta_2(\epsilon) - \phi(\epsilon)] \right) \right]. \tag{35}
 \end{aligned}$$

We differentiate equation (35) with respect to h and take the Fourier transform according to equation (24) to find the imaginary part of the transverse susceptibility:

$$\begin{aligned}
 \text{Im } \chi_t(\mathbf{r}, \Omega) = & \left[\frac{S(S+1)(2S+1)}{8V^2} \frac{\alpha^2 \gamma^2 \hbar^3}{(2\pi)^3} \left(\frac{2m}{\hbar^2} \right)^{3/2} \Omega \int_{-w}^{\infty} d\epsilon f_{\epsilon} \sqrt{\epsilon} \left(\frac{R_2(\epsilon)}{\lambda(\epsilon)} \right)^2 \right. \\
 & \left. \times \frac{R_1(\epsilon) \cos[\theta_1(\epsilon)]}{|\text{Im}(\zeta_1)|^2 + \Omega^2} \right] \tag{36}
 \end{aligned}$$

where

$$\text{Im}(\zeta_1) = -[R_2(\epsilon)/\lambda(\epsilon)] \sin[\theta_2(\epsilon) - \phi(\epsilon)] \tag{37}$$

and we have used the fact that $\text{Im}(\zeta_1) = -\text{Im}(\zeta_2)$. We label the energy corresponding to the solution $\text{Im}(\zeta_1) = \text{Im}(\zeta_2) = 0$, by ϵ_{δ} .

4. Enhanced mass

It is well known that the low-temperature behaviour of heavy fermions, at sufficiently long wavelengths and low frequencies, can be described by a Fermi liquid with a highly enhanced density of states so long as the systems remain normal (Lee *et al* 1986). It may be of interest, therefore, at this stage in the development of our model, to consider the equilibrium properties of the low-temperature state. It has been emphasized by Schweitzer and Czycholl (1991) that it is important for any approximation that reproduces the Fermi-liquid behaviour within the context of the periodic Anderson model to satisfy Luttinger's theorem (Luttinger and Ward 1960, Luttinger 1961).

Luttinger's theorem requires the self-energy imaginary part to vanish at the Fermi energy at zero temperature. As a consequence, the quasiparticle excitations have infinite lifetime on the Fermi surface. Away from the Fermi surface at $T = 0$, however, the quasiparticles have finite lifetimes and the self-energy imaginary part attains finite values as the temperature is increased from zero. From equation (14) we identify the frequency-dependent self-energy:

$$\Sigma^r(\mathbf{k}, \omega) = V^2 / [\omega - \epsilon_F - \Sigma_{\text{sf}}^r(\omega)]. \tag{38}$$

The self-energy imaginary part is

$$\text{Im } \Sigma^r(\mathbf{k}, \omega) = \frac{V^2 \text{Im } \Sigma_{\text{sf}}^r(\omega)}{[\epsilon_F + \text{Re } \Sigma_{\text{sf}}^r(\omega)]^2 + [\text{Im } \Sigma_{\text{sf}}^r(\omega)]^2}. \tag{39}$$

We approximate equation (A35) for $0 < \omega \ll 1$ and $0 < T \ll 1$:

$$\begin{aligned} \operatorname{Re} \Sigma_{\text{sf}}(\omega) &\simeq \frac{9\rho}{32\pi} \left(\frac{J}{2N}\right)^3 \left[\frac{1}{2} + \frac{1}{\pi} \ln \left(\frac{D^2}{\omega^2 + (k_{\text{B}}T)^2} \right) \right] \\ \operatorname{Im} \Sigma_{\text{sf}}(\omega) &\simeq -\frac{27\rho}{32\pi} \left(\frac{J}{2N}\right)^3. \end{aligned} \quad (40)$$

Using this approximation in equation (39) we find, for low temperature and ω very close to the Fermi level, that

$$\operatorname{Im} \Sigma(\mathbf{k}, \omega) \simeq \frac{4}{\rho V^2} \frac{(2\pi\epsilon_{\text{F}})^3}{[\omega\kappa(\omega)]^2} \left(\omega^2 + \frac{(k_{\text{B}}T)^2}{\kappa(\omega)} \right) \quad (41)$$

where

$$\kappa(\omega) = \ln(D^2/\omega^2)$$

and where $2D$ is the width of the conduction band and we have used equation (A1).

This result is qualitatively similar to the analysis of Schweitzer and Czycholl (1990) where they use a self-consistent approach to the second-order perturbation theory of the periodic Anderson model and find that the Luttinger theorem is satisfied by $\operatorname{Im} \Sigma(\omega) \sim \omega^2 + (\pi T)^2$.

The low-temperature Fermi-liquid state is also characterized by a pronounced enhancement of the effective mass, and it is interesting to investigate the transition from a zero-temperature state with heavy quasiparticles to a non-coherent scattering high-temperature regime with no mass enhancement. We specify the enhanced mass in terms of the quasiparticle amplitude Z , neglecting for simplicity the k dependence of the self-energy (Varma 1985):

$$\frac{m^*}{m} = Z^{-1} = 1 + \left. \frac{\partial \operatorname{Re} \Sigma^r(\mathbf{k}, \omega)}{\partial \omega} \right|_{\omega=\epsilon_{\text{F}}} \quad (42)$$

The enhanced mass in our model, without using the low-temperature approximation, becomes

$$\begin{aligned} \frac{m^*}{m} &= 1 - V^2 \left[\left(1 - \left. \frac{\partial \Sigma_{\text{sf}}^r(\omega)}{\partial \omega} \right|_{\omega=\epsilon_{\text{F}}} \right) \{ [\epsilon_{\text{F}} + \operatorname{Re} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}})]^2 - [\operatorname{Im} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}})]^2 \} - 2[\epsilon_{\text{F}} + \operatorname{Re} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}})] \right. \\ &\quad \times \left. \operatorname{Im} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}}) \left. \frac{\partial \operatorname{Im} \Sigma_{\text{sf}}^r(\omega)}{\partial \omega} \right|_{\omega=\epsilon_{\text{F}}} \right] \{ [\epsilon_{\text{F}} + \operatorname{Re} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}})]^2 + [\operatorname{Im} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}})]^2 \}^{-2} \end{aligned} \quad (43)$$

where

$$\begin{aligned} \operatorname{Im} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}}) &= \frac{3w(\beta)}{[4 + w^2(\beta)][4/9 + w^2(\beta)]} \\ \operatorname{Re} \Sigma_{\text{sf}}^r(\epsilon_{\text{F}}) &= \frac{\frac{1}{3}w^2(\beta)[4/9 - w^2(\beta)]}{[4 + w^2(\beta)][4/9 + w^2(\beta)]} \\ w(\beta) &= \frac{\rho V}{2\pi\epsilon_{\text{F}}} \ln \left(\frac{(\beta D)^2}{4} \right) \end{aligned} \quad (44)$$

In figure 2 we sketch the enhanced mass as a function of temperature. The enhancement factor is reduced from a large value at $T = 0$ to essentially no enhancement at $T = 1$ K. This indicates a smooth transition from the zero-temperature coherent state to the high-temperature scattering state.

5. Korringa relaxation processes

The integrand of equation (36) contains a resonance centred at ϵ_δ , a few meV from ϵ_F . We attribute this contribution to the integrand to spin fluctuations of the heavy-fermion ground state. If one considers the conduction electron as a quasiparticle then, from equation (34), the quantity $\text{Im}(\zeta_1)$ determines the rate at which spin correlations decay in the interacting system. The bare quasiparticle of energy ϵ_δ becomes renormalized by dressing itself in a self-energy cloud composed of fluctuating valence states and its spectral density is most sharply defined where $\text{Im}(\zeta_1) = 0$. Thus, the impurity spin probes the renormalized s-electron density of states at the characteristic energy ϵ_δ .

This resonance is sketched in figure 3. The parameter ρ is the density of states in the band, assumed constant, and N is the number of electrons in the system. Each of the three curves is evaluated for a different temperature, and the width of each peak corresponds to the spin correlation decay rate of fluctuating f states.

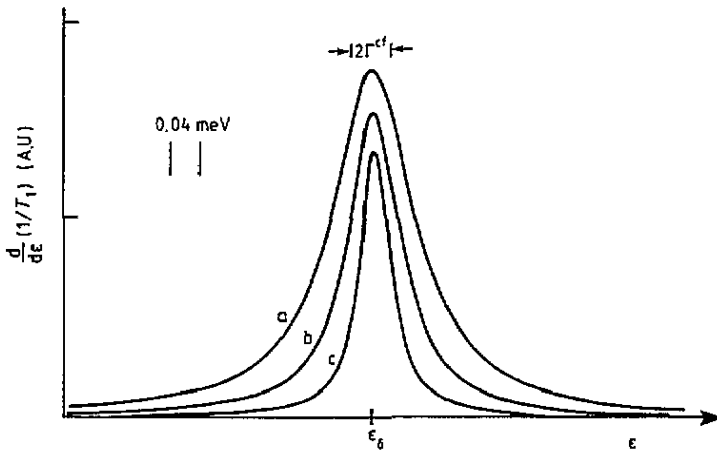


Figure 3. Integrand of equation (40) near the energy ϵ_δ evaluated for three different temperatures: Γ^{cf} is the inverse of the impurity spin correlation time. Curves a, b and c were evaluated with temperatures $T = 20$ K, $T = 15$ K and $T = 10$ K respectively.

Separating out the contribution due to spin fluctuations, we write the relaxation rate from equation (22) as

$$\begin{aligned} \frac{1}{T_1} \simeq & C' k_B T \left(\int_{-W}^{\epsilon_\delta - 2\Omega/\sqrt{3}} + \int_{\epsilon_\delta + 2\Omega/\sqrt{3}}^{\infty} \right) f_\epsilon \sqrt{\epsilon} \left(\frac{R_2(\epsilon)}{\lambda(\epsilon)} \right)^2 \frac{R_1(\epsilon) \cos[\theta_1(\epsilon)]}{|\text{Im}(\zeta_1)|^2 + \Omega^2} d\epsilon \\ & + \frac{\sqrt{3} C' k_B T}{2\Omega^2} f_{\epsilon_\delta} \sqrt{\epsilon_\delta} \left(\frac{R_2(\epsilon_\delta)}{\lambda(\epsilon_\delta)} \right)^2 R_1(\epsilon_\delta) \cos[\theta_1(\epsilon_\delta)] \end{aligned} \quad (45)$$

where an approximate value for ϵ_δ can be determined by expanding $\text{Im}(\zeta_1)$ about ϵ_F and setting $\text{Im}(\zeta_1) = 0$. We find

$$\epsilon_\delta \simeq \epsilon_F - \frac{\text{Im}(\zeta_1)}{\partial \text{Im}(\zeta_1)/\partial \epsilon} \Big|_{\epsilon=\epsilon_F} \quad (46)$$

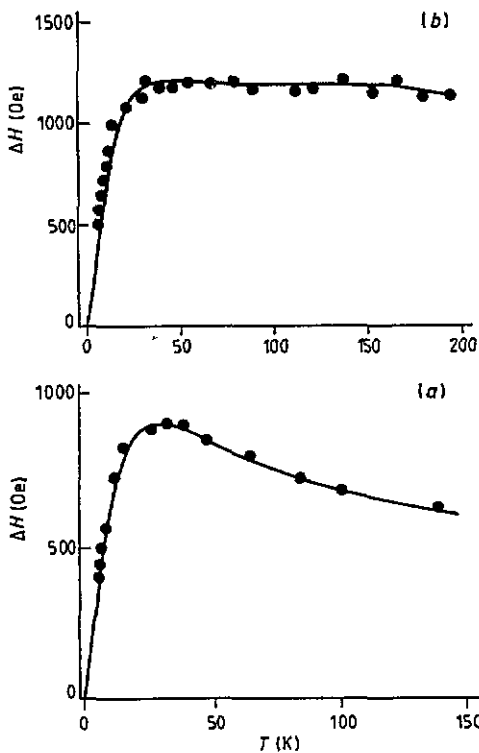


Figure 4. The temperature dependence of the contribution to the Gd ESR linewidth in (a) CeCu_2Si_2 and (b) CeAl_3 due to impurity spin fluctuations. The full circles are the experimental data (Schlott *et al* 1988) and the full curve denotes the theoretical curve obtained from the second term in equation (42) evaluated with the parameters (a) $\rho J/N = 0.560$, $V = 2.40$ eV, $\alpha = 0.003$ and (b) $\rho J/N = 0.537$, $V = 2.01$ eV, $\alpha = 0.003$.

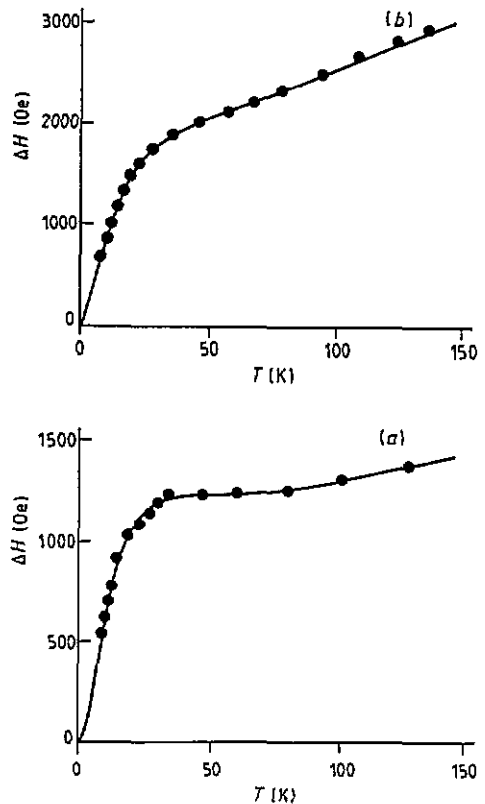


Figure 5. The temperature dependence of the Korringa relaxation process (Gd ESR linewidth) in (a) CeCu_2Si_2 and (b) CeAl_3 . The full circles are the experimental data (Schlott *et al* 1988) and the full curve denotes the theoretical curve obtained from equation (42) evaluated with the parameters (a) $\rho J/N = 0.560$, $V = 2.40$ eV, $\alpha = 0.003$ and (b) $\rho J/N = 0.537$, $V = 2.01$ eV, $\alpha = 0.003$.

The last term in equation (45), the f-spin fluctuation term, is plotted as a function of temperature in figure 4. This plot is presented with the contribution due to Ce 4f-spin fluctuations deduced in the ESR spin relaxation experiments of Schlott *et al* (1988) on CeCu_2Si_2 and CeAl_3 .

Equation (45) was evaluated numerically, and in figure 5 we plot the ESR linewidth as a function of temperature and compared with the results of Schlott *et al* (1988). The trend in figure 5 indicates that observation of non-linear relaxation behaviour around the Kondo temperature in heavy-fermion compounds is highly dependent on the magnitude of the s-f interaction strength. We find that, as the parameter $\rho J/N$ becomes smaller, decreasing from 0.60 to 0, the curvature in the Korringa rate process about the Kondo temperature becomes smaller. With $\rho J/N \sim 0.50$, equation (45), as plotted against temperature, becomes linear, corresponding to the conventional Korringa rate.

The curves in figure 5 were obtained using $J > 0$, indicating ferromagnetic coupling. Qualitatively similar results can be obtained for $J < 0$, antiferromagnetic coupling; however,

the non-linearities of the relaxation rate around the Kondo temperature are greatly enhanced compared to the ferromagnetic case.

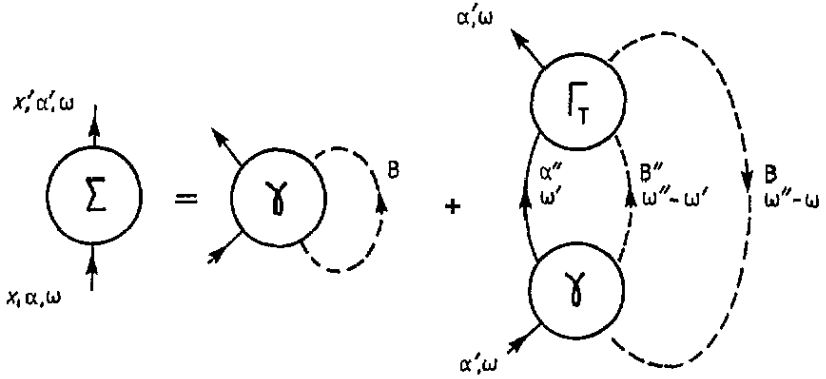


Figure 6. The s-electron self-energy Σ in terms of the total proper Keldysh vertex part Γ_T and the Keldysh vertex γ . The solid lines are bare s-electron propagators and the dotted lines are bare f-spin propagators. The Keldysh indices are not shown.

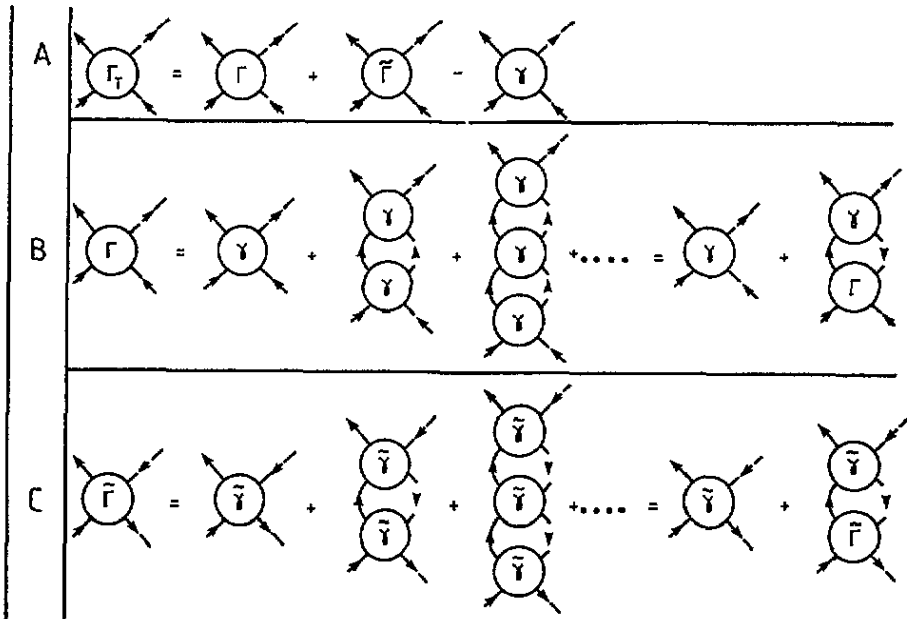


Figure 7. The s-f Keldysh vertex part in the ladder approximation. (A) The total proper vertex part Γ_T in terms of the proper particle-particle part Γ , the proper time reversed particle-hole part $\tilde{\Gamma}$ and the Keldysh vertex γ . (B) and (C) The ladder sum and integral equation for Γ and $\tilde{\Gamma}$ respectively in terms of γ and the time-reversed Keldysh vertex $\tilde{\gamma}$.

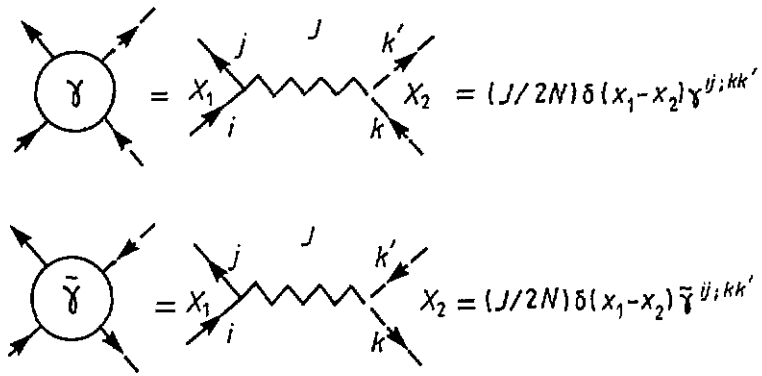


Figure 8. The upper diagram is an abbreviated form of the s-f interaction at the Keldysh vertex, with the long form of the interaction (represented by the wavy line) shown immediately to its right-hand side. To the right of the long form of the interaction is the mathematical representation of the diagram as the product of the Keldysh tensor with the Dirac delta-function and the interaction strength. The lower diagram is the interaction at the time-reversed Keldysh vertex.

6. Conclusion

In this paper I have sought an expression for the Korringa relaxation processes in a heavy-fermion compound. I attempted to derive the spin relaxation rate by using a time-dependent approach for evaluating the system Green functions within the context of the Keldysh formalism. The derivation was microscopic from first principles using the Hamiltonian of Yoshimori and Kasai. The spin relaxation rate was expressed as an integral equation containing two terms, a term involving the conventional relaxation processes and a term making non-linear contributions associated with rare-earth spin fluctuations. Comparison between a plot of equation (45) and the experimental results of Schlott *et al* (1988) shows good agreement.

The model developed in this paper provides for the non-linearities of the Korringa relaxation process arising around the Kondo temperature in heavy-fermion compounds by introducing a dependence on the exchange coupling parameter. As the magnitude of *J* decreases in this model, the temperature dependence of the Korringa rate assumes a linear behaviour.

Appendix

In this appendix the Keldysh retarded self-energy $\Sigma_{sf}(0, \omega)$ will be evaluated for a single impurity located at the origin of coordinates. The interaction Hamiltonian we will use is the same as that of the Anderson model for a single magnetic impurity. After performing a Coqblin and Schrieffer transformation (Coqblin and Schrieffer 1969), the total time-independent Hamiltonian for a single spin impurity is given by

$$H_{sf} = \sum_{k\alpha} \epsilon_k c_{k\alpha}^+ c_{k\alpha} + \sum_{\beta} \epsilon_F c_{f\beta}^+ c_{f\beta} - \frac{J}{2N} \sum_{\alpha\alpha'\beta\beta'kk'} (\sigma_{\alpha'\alpha} \cdot S_{\beta'\beta}) c_{f\beta'}^+ c_{k'\alpha}^+ c_{k\alpha} c_{f\beta} \tag{A1}$$

where *N* is the number of atoms; *J* is the s-f coupling constant

$$J/2N = V k^2 / \epsilon_F$$

ϵ_k and ϵ_F are the bare s-f electron energies relative to the Fermi level; $c_{k\alpha}^+$, $c_{k\alpha}$, $c_{f\beta}^+$, $c_{f\beta}$ are creation and annihilation operators for s, f electrons; and $\sigma_{\alpha\alpha}$, $2S_{\beta\beta}$ are the Pauli matrices for s, f electrons. Henceforth we shall drop the {sf} subscript on the self-energy and all subscripts on the self-energy and Green functions will indicate elements of a Keldysh 2×2 matrix.

As Abrikosov points out (Abrikosov 1965), the representation of the spin operator in terms of second quantized operators yields for spin other than 1/2, extra 'unphysical states'. If, however, we take ϵ_F to be zero (Cheung and Mattuck 1970), then the average number of f electrons is equal to one, and the only effect of the unphysical states is to introduce a normalization factor of 2. We will, therefore, calculate the Keldysh s-f scattering amplitude Γ and the Keldysh self-energy Σ_{sf} in terms of the 'exchange' $(J/2N)\sigma \cdot S$ interaction by setting $\epsilon_F = 0$ and assume, in the end, that these calculated quantities are valid for ϵ_F attaining a small but finite value. The correspondence between J and the parameters of the Anderson Hamiltonian, provided by the Coqblin and Schrieffer transformation, is thus assumed to be preserved independently of the details of the self-consistent perturbation treatment of the Keldysh self-energy.

The self-energy is evaluated by performing the sum and integrations represented by figure 6. The vertex part can be computed from figure 7, and used to evaluate the self-energy in a self-consistent manner.

The perturbation expansion for the s-f vertex part, in the parquet series, is the sum of an electron-electron Γ and an electron-hole part γ , as shown in figure 7 (the roman superscripts will denote the Keldysh indices). The diagrams in figure 7 represent integral equations, which may be translated into functions by associating with each solid line one of three Keldysh s-electron propagators:

$$g^t(\mathbf{r}, \mathbf{r}'; \omega) = \lim_{\delta \rightarrow 0^+} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\rho(\mathbf{r}, \mathbf{r}'; \omega') d\omega'}{\omega - \omega' + i\delta} \quad (\text{A2})$$

$$g^a(\mathbf{r}, \mathbf{r}'; \omega) = \lim_{\delta \rightarrow 0^-} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\rho(\mathbf{r}, \mathbf{r}'; \omega') d\omega'}{\omega - \omega' - i\delta} \quad (\text{A3})$$

$$g^F(\mathbf{r}, \mathbf{r}'; \omega) = 2g^\pm \mp (g^r - g^a) = -i \tanh(\beta\omega/2)\rho(\mathbf{r}, \mathbf{r}'; \omega) \quad (\text{A4})$$

where

$$g^\pm(\mathbf{r}, \mathbf{r}'; \omega) = \pm i f(\pm\omega)\rho(\mathbf{r}, \mathbf{r}'; \omega) \quad (\text{A5})$$

and where $\rho(\mathbf{r}, \mathbf{r}'; \omega)$ is the s-electron spectral density, $f(\omega)$ is the Fermi function and $\beta = 1/k_B T$. In likewise fashion, each dotted line is associated with one of three f-spin propagators,

$$f^t(\mathbf{r}, \mathbf{r}'; \omega) = \lim_{\delta \rightarrow 0^+} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\sigma(\mathbf{r}, \mathbf{r}'; \omega') d\omega'}{\omega - \omega' + i\delta} \quad (\text{A6})$$

$$f^a(\mathbf{r}, \mathbf{r}'; \omega) = \lim_{\delta \rightarrow 0^-} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\sigma(\mathbf{r}, \mathbf{r}'; \omega') d\omega'}{\omega - \omega' - i\delta} \quad (\text{A7})$$

$$f^F(\mathbf{r}, \mathbf{r}'; \omega) = -i \tanh(\beta\omega/2)\sigma(\mathbf{r}, \mathbf{r}'; \omega) \quad (\text{A8})$$

where $\sigma(\mathbf{r}, \mathbf{r}'; \omega)$ is the spectral density of the impurity f spin.

The Keldysh γ diagram is an abbreviated form of the s-f interaction and the Keldysh vertex as shown in figure 8. The value $(J/2N)(\sigma_{\alpha'\alpha} \cdot S_{\beta'\beta})\gamma^{ijkl}$ is associated with each circle, with γ^{ijkl} being a bare Keldysh vertex matrix, and where the Greek subscripts are associated with a Pauli matrix spin index. The Keldysh vertex matrix is given by

$$\gamma^{ijkl} = \delta_{ij}\delta_{jk}\sigma_{kk}^z \quad (\text{A9})$$

where σ_z is the third Pauli spin matrix. The vertex is a unit tensor in the i, j indices, reflecting that the electron, at the vertex, enters and leaves the same space-time point. A change in $\{k, k'\}$ will change the sign of the tensor, reflecting that the point at the other end of the f-spin propagator can reside on either the upper or the lower branch of the Keldysh time contour (Rammer and Smith 1986). The $k-j$ indices of the unit tensor reflect the assumption that we are dealing with a delta-function contact interaction between the s conduction electron and the f spin impurity. On performing a transformation (Keldysh 1965) in the Keldysh space, the Keldysh vertex function is written as

$$\gamma_{mn}^{11} = \gamma_{mn}^{22} = \sigma_{mn}^x = \tilde{\gamma}_{mn}^{12} = \tilde{\gamma}_{mn}^{21} \quad (\text{A10})$$

$$\gamma_{mn}^{12} = \gamma_{mn}^{21} = \delta_{mn} = \tilde{\gamma}_{mn}^{11} = \tilde{\gamma}_{mn}^{22} \quad (\text{A11})$$

where the tilde indicates a time-reversed impurity spin matrix.

Let us examine the electron-electron vertex part first. Figure 7(B) yields

$$\begin{aligned} & \Gamma_{\alpha\beta\alpha''\beta''}^{ij;kl}(\mathbf{r}, \mathbf{r}'; \omega', \omega'', \omega^{\text{III}}) \\ &= \frac{J}{2N}(\sigma_{\alpha\alpha''} \cdot S_{\beta\beta''})\gamma^{ij;kl} + \frac{J}{2N} \sum_{mnop, \alpha'\beta'} \iiint d\mathbf{r}_1 d\mathbf{r}_2 d\omega \gamma^{im;ko} g_{mn}(\mathbf{r}_1, \mathbf{r}_2; \omega) \\ & \quad \times f_{op}(\mathbf{r}_1, \mathbf{r}_2; \omega'' - \omega) \Gamma_{\alpha'\beta'\alpha''\beta''}^{nj;pl}(\mathbf{r}_1, \mathbf{r}'; \omega, \omega', \omega''). \end{aligned} \quad (\text{A12})$$

The spin impurity is assumed to be located at $\mathbf{r} = 0$.

Writing Γ as the sum of a scalar and vector part in the spin indices (Abrikosov 1965),

$$\Gamma_{\alpha\beta\alpha''\beta''}^{ij;kl} = {}^0\Gamma^{ij;kl} \delta_{\alpha\alpha''} \delta_{\beta\beta''} + {}^1\Gamma^{ij;kl} \sigma_{\alpha\alpha''} \cdot S_{\beta\beta''} \quad (\text{A13})$$

and utilizing the fact that

$$\sum_{\alpha'\beta'} (\alpha_{\alpha\alpha'} \cdot S_{\beta\beta'}) (\sigma_{\alpha'\alpha''} \cdot S_{\beta'\beta''}) = \frac{3}{4} \delta_{\alpha\alpha''} \delta_{\beta\beta''} - \sigma_{\alpha\alpha''} \cdot S_{\beta\beta''} \quad (\text{A14})$$

yields the coupled equations

$$\begin{aligned} & {}^0\Gamma^{ij;kl}(\mathbf{r}, \mathbf{r}'; \omega', \omega'', \omega^{\text{III}}) \\ &= \frac{3}{8N} \iiint d\mathbf{r}_1 d\mathbf{r}_2 d\omega \gamma^{im;ko} J(\mathbf{r}_1) g_{mn}(\mathbf{r}_1, \mathbf{r}_2; \omega) f_{op}(\mathbf{r}_1, \mathbf{r}_2; \omega^{\text{III}} - \omega) \\ & \quad \times {}^1\Gamma^{nj;pl}(\mathbf{r}_2, \mathbf{r}'; \omega, \omega'', \omega^{\text{III}}) \end{aligned} \quad (\text{A15})$$

and

$$\begin{aligned} & {}^1\Gamma^{ij;kl}(\mathbf{r}, \mathbf{r}'; \omega', \omega'', \omega^{\text{III}}) \\ &= \frac{J(\mathbf{r}')}{2N} \gamma^{ij;kl} + \frac{1}{2N} \iiint d\mathbf{r}_1 d\mathbf{r}_2 d\omega \gamma^{im;ko} J(\mathbf{r}_1) g_{mn}(\mathbf{r}_1, \mathbf{r}_2; \omega) \\ & \quad \times f_{op}(\mathbf{r}_1, \mathbf{r}_2; \omega^{\text{III}} - \omega) [{}^0\Gamma^{nj;pl}(\mathbf{r}_2, \mathbf{r}'; \omega, \omega', \omega'') - {}^1\Gamma^{nj;pl}(\mathbf{r}_2, \mathbf{r}'; \omega, \omega'', \omega^{\text{III}})] \end{aligned} \quad (\text{A16})$$

where a sum is implied over repeated Keldysh indices.

Substituting equation (A16) into equation (A15), noting that ${}^1\Gamma$ is independent of \mathbf{r} and ω' so that it can be factored out of the integral, we write ${}^1\Gamma$ as a column vector:

$$({}^1\Gamma) = \frac{J\delta(\mathbf{r})}{2N}(\gamma) + \frac{1}{2N}\mathbf{A}({}^0\Gamma) - \frac{1}{2N}\mathbf{A}({}^1\Gamma) \quad (\text{A17})$$

$$({}^0\Gamma) = \frac{3}{8N}\mathbf{A}({}^1\Gamma) \quad (\text{A18})$$

where \mathbf{A} is a 16×16 matrix, and where the first entry in the Γ column is $\Gamma^{11;11}$, the second entry is $\Gamma^{12;11}$, etc.

Before computing the elements of \mathbf{A} , we note that they are sums of terms of the form

$$\iiint d\mathbf{r} d\mathbf{r}' d\omega J(\mathbf{r})g_{ij}(\mathbf{r}, \mathbf{r}'; \omega)f_{kl}(\mathbf{r}, \mathbf{r}'; \omega' - \omega) \quad (\text{A19})$$

where the spectral density of the s conduction electron, $\rho(\mathbf{r}, \mathbf{r}'; \omega)$, is assumed to be confined to a band of width $2D$. Evaluating g^{Γ} with this density we find

$$g^{\Gamma} = g^{a*} = \lim_{\delta \rightarrow 0^+} \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{[\theta(\omega' + D) - \theta(\omega' - D)] d\omega'}{\omega - \omega' + i\delta} = i \frac{e^{\eta\omega}}{\pi} \sinh(\eta D). \quad (\text{A20})$$

Because η is infinitesimal g^{a*} is infinitesimal. Since $g_{11} = 0$, only terms involving $g_{22} = g^{\Gamma} = -i\rho(\mathbf{r}, \mathbf{r}') \tanh(\beta\omega/2)$ are considered when evaluating the elements of \mathbf{A} .

The spectral density of the impurity is a delta-function, and on evaluating the frequency integration in equation (A19), we see that the number of terms needed to evaluate each element of \mathbf{A} is reduced from 16 to two. These terms are

$$z_1(\omega') = \frac{1}{2N} \iiint J(\mathbf{r})g_{22}(\mathbf{r}, \mathbf{r}'; \omega)f_{12}(\mathbf{r}, \mathbf{r}'; \omega' - \omega) d\mathbf{r} d\mathbf{r}' d\omega \quad (\text{A21})$$

and

$$z_2(\omega') = \frac{1}{2N} \iiint J(\mathbf{r})g_{22}(\mathbf{r}, \mathbf{r}'; \omega)f_{21}(\mathbf{r}, \mathbf{r}'; \omega' - \omega) d\mathbf{r} d\mathbf{r}' d\omega. \quad (\text{A22})$$

We introduce a local impurity density of states and assume that

$$\sigma(\mathbf{r}, \mathbf{r}'; \omega) = \sigma(\mathbf{r}', \mathbf{r}'; \omega) = \sigma(\mathbf{r}'; \omega) \quad (\text{A23})$$

which obey the sum rule

$$\int_{-\infty}^{\infty} \sigma(\mathbf{r}'; \omega) d\mathbf{r}' = \sigma(\omega) = \delta(\omega - \epsilon_{\text{F}}) = \delta(\omega). \quad (\text{A24})$$

Evaluating equation (A22) using equation (A24) and equation (A5), we find

$$\begin{aligned} z_1(\omega) &= \frac{1}{2N} \iiint d\mathbf{r} d\mathbf{r}' d\omega J(\mathbf{r})g_{22}(\mathbf{r}, \mathbf{r}'; \omega)f_{21}(\mathbf{r}, \mathbf{r}'; \omega' - \omega) \\ &= -i \frac{\rho J}{2N} \int_{-D}^D \frac{d\omega' \tanh(\beta\omega'/2)}{2\pi(\omega' - \omega)} - \frac{\rho J}{4N} \tanh\left(\frac{\beta\omega}{2}\right) \simeq \frac{\rho J}{4N} \tanh\left(\frac{\beta\omega}{2}\right) + i \frac{\rho J}{4\pi N} \\ &\quad \times \ln\left(\frac{D^2}{\omega^2 + (2/\beta)^2}\right) = z(\omega) \end{aligned} \quad (\text{A25})$$

where we have used the assumption that $\rho(\mathbf{r}, \mathbf{r}') = \rho$, a constant. The remaining term needed to evaluate the elements of \mathbf{A} is computed in similar fashion, and found to be

$$z_2(\omega) = \frac{1}{2N} \int \int \int d\mathbf{r} d\mathbf{r}' d\omega J(\mathbf{r}) g_{22}(\mathbf{r}, \mathbf{r}'; \omega) f_{12}(\mathbf{r}, \mathbf{r}'; \omega' - \omega) \\ \simeq -\frac{\rho J}{4N} \tanh\left(\frac{\beta\omega}{2}\right) + i\frac{\rho J}{4\pi N} \ln\left(\frac{D^2}{\omega^2 + (2/\beta)^2}\right) = -z^*(\omega). \quad (\text{A26})$$

We now evaluate the elements of \mathbf{A} . Substituting equation (A18) into equation (A19), and solving for the column vector ${}^1\Gamma$, we find that

$${}^1\Gamma^{11;11}(\mathbf{r}, \omega) = {}^1\Gamma^{21;21}(\mathbf{r}, \omega) = {}^1\Gamma^{22;22}(\mathbf{r}, \omega) = \left(\frac{J}{2N}\right) \delta(\mathbf{r}) \\ \times \frac{(J/2N)z(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]}{1 - (J/2N)z^*(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]} \\ {}^1\Gamma^{12;11}(\mathbf{r}, \omega) = {}^1\Gamma^{11;12}(\mathbf{r}, \omega) = {}^1\Gamma^{12;12}(\mathbf{r}, \omega) = {}^1\Gamma^{22;21}(\mathbf{r}, \omega) = \left(\frac{J}{2N}\right) \delta(\mathbf{r}) \\ \times \frac{1}{1 - (J/2N)z^*(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]} \\ {}^1\Gamma^{12;21}(\mathbf{r}, \omega) = {}^1\Gamma^{21;12}(\mathbf{r}, \omega) = {}^1\Gamma^{22;11}(\mathbf{r}, \omega) = {}^1\Gamma^{11;22}(\mathbf{r}, \omega) = 0 \\ {}^1\Gamma^{22;12}(\mathbf{r}, \omega) = {}^1\Gamma^{11;21}(\mathbf{r}, \omega) = {}^1\Gamma^{21;11}(\mathbf{r}, \omega) = {}^1\Gamma^{12;22}(\mathbf{r}, \omega) = {}^1\Gamma^{21;22}(\mathbf{r}, \omega) \\ = (J/2N)\delta(\mathbf{r}). \quad (\text{A27})$$

Similarly, we evaluate ${}^1\tilde{\Gamma}$ using figure 7(B), and find that

$${}^1\tilde{\Gamma}^{21;11}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{22;12}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{11;21}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{12;22}(\mathbf{r}, \omega) = \left(\frac{J}{2N}\right) \delta(\mathbf{r}) \\ \times \frac{(J/2N)z(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]}{1 - (J/2N)z^*(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]} \\ {}^1\tilde{\Gamma}^{22;11}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{12;21}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{11;22}(\mathbf{r}, \omega) = \left(\frac{J}{2N}\right) \delta(\mathbf{r}) \\ \times \frac{1}{1 - (J/2N)z^*(\omega)[1 + \frac{3}{4}(J/2N)z^*(\omega)]} \\ {}^1\tilde{\Gamma}^{12;11}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{11;12}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{22;21}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{21;22}(\mathbf{r}, \omega) = 0 \\ {}^1\tilde{\Gamma}^{11;11}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{12;12}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{21;12}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{21;21}(\mathbf{r}, \omega) = {}^1\tilde{\Gamma}^{22;22}(\mathbf{r}, \omega) \\ = (J/2N)\delta(\mathbf{r}). \quad (\text{A28})$$

The conduction-electron retarded self-energy may be obtained from the vertex part as shown in figure 6. From the figure, we see that the outgoing spin must always be the same

as the incoming spin, and hence, following the analysis of Cheung and Mattuck (1970), the spin sum for figure 6 is

$$\sum_{\beta\beta''\alpha''} [\Gamma^0 \Gamma_{\Gamma}^{ij:kl} \delta_{\alpha''\alpha''} + {}^1\Gamma_{\Gamma}^{ij:kl} (\sigma_{\alpha''\alpha''} \cdot \mathbf{S}_{\beta\beta''})] \times (\sigma_{\alpha''\alpha''} \cdot \mathbf{S}_{\beta''\beta}) = \frac{3}{2} {}^1\Gamma_{\Gamma}^{ij:kl} \delta_{\alpha''\alpha''} \quad (\text{A29})$$

where

$${}^1\Gamma_{\Gamma}^{ij:kl}(\mathbf{r}, \omega) = {}^1\Gamma^{ij:kl}(\mathbf{r}, \omega) + {}^1\tilde{\Gamma}^{ij:kl}(\mathbf{r}, \omega) - \frac{J\delta(\mathbf{r})}{2N} \gamma^{ij:kl}. \quad (\text{A30})$$

Expanding figure 6 in the Keldysh indices gives

$$\begin{aligned} \Sigma_{ij}(0, \mathbf{r}; \omega) &= \frac{3}{2} \frac{J\delta(\mathbf{r})}{2N} \iiint \int d\mathbf{r}' d\mathbf{r}'' d\omega' d\omega'' \delta(\mathbf{r}') \left(\sum_{nn'oo'tm} \gamma^{il;nn'} g_{lm}(\mathbf{r}', \mathbf{r}'', \omega') \right. \\ &\quad \times f_{n'o}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega') f_{n'o'}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega) [{}^1\Gamma^{oo';mj}(\omega'') + {}^1\tilde{\Gamma}^{oo';mj}(\omega'')] \\ &\quad \left. - (J/2N) \gamma^{oo';mj} \right). \end{aligned} \quad (\text{A31})$$

Noting that terms in the sum involving f_{22} integrate to zero, we find that

$$\begin{aligned} \Sigma_{12}(0, \mathbf{r}; \omega) &= \Sigma^{\Gamma}(0, \mathbf{r}; \omega) \\ &= \frac{3}{2} \frac{J\delta(\mathbf{r})}{2N} \iiint \int d\mathbf{r}' d\mathbf{r}'' d\omega' d\omega'' \delta(\mathbf{r}') \{g_{22}(\mathbf{r}', \mathbf{r}'', \omega') f_{12}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega') \\ &\quad \times f_{21}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega) + g_{22}(\mathbf{r}', \mathbf{r}'', \omega') f_{21}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega') f_{12}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega) \\ &\quad \times [{}^1\Gamma^{22;11}(\omega'') + {}^1\tilde{\Gamma}^{22;11}(\omega'')]\}. \end{aligned} \quad (\text{A32})$$

Also

$$\begin{aligned} \Sigma_{22}(0, \mathbf{r}; \omega) &= 0 \\ &= \frac{3}{2} \frac{J\delta(\mathbf{r})}{2N} \iiint \int d\mathbf{r}' d\mathbf{r}'' d\omega' d\omega'' \delta(\mathbf{r}') \{g_{22}(\mathbf{r}', \mathbf{r}'', \omega') \\ &\quad \times f_{12}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega') f_{12}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega) [{}^1\Gamma^{22;12}(\omega'') + {}^1\tilde{\Gamma}^{22;12}(\omega'')] \\ &\quad + g_{22}(\mathbf{r}', \mathbf{r}'', \omega') f_{21}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega') \\ &\quad \times f_{21}(\mathbf{r}', \mathbf{r}''; \omega'' - \omega) [{}^1\Gamma^{22;21}(\omega'') + {}^1\tilde{\Gamma}^{22;21}(\omega'')]\}. \end{aligned} \quad (\text{A33})$$

We subtract equation (32) from equation (A33), and perform the spatial integration to find

$$\begin{aligned} \Sigma^{\Gamma}(0, \mathbf{r}; \omega) &= \frac{3}{2} \frac{\rho J^2 \delta(\mathbf{r})}{2\pi(2N)^2} \left(\frac{i}{2\pi} \int_{-D}^D \int_{-\infty}^{\infty} d\omega' d\omega'' \frac{\tanh(\beta\omega'/2)}{(\omega'' - \omega')(\omega'' - \omega)} \right. \\ &\quad + 2 \int_{-D}^D \int_{-\infty}^{\infty} d\omega' d\omega'' \frac{\delta(\omega'' - \omega) \tanh(\beta\omega'/2)}{(\omega'' - \omega')} \\ &\quad \times [{}^1\Gamma^{22;22}(\omega'') + {}^1\tilde{\Gamma}^{22;22}(\omega'') - {}^1\Gamma^{22;11}(\omega'') - {}^1\tilde{\Gamma}^{22;11}(\omega'')] \\ &\quad + \frac{2i}{\pi} \int_{-D}^D \int_{-\infty}^{\infty} d\omega' d\omega'' \delta(\omega'' - \omega') \delta(\omega'' - \omega) \tanh(\beta\omega'/2) \\ &\quad \left. \times [{}^1\Gamma^{22;22}(\omega'') + {}^1\tilde{\Gamma}^{22;22}(\omega'') + {}^1\Gamma^{22;11}(\omega'') + {}^1\tilde{\Gamma}^{22;11}(\omega'') - \frac{1}{2}(J/2N)] \right). \end{aligned} \quad (\text{A34})$$

These integrations are elementary, and we find Σ^f :

$$\Sigma_{\text{sr}}^f(0, r; \omega) = \frac{\frac{3}{2}(\rho/8\pi)(J/N)^2\delta(r)}{1 - z^*(\omega)[1 + \frac{3}{4}z^*(\omega)]} \left[[z(\omega) - z^*(\omega)][1 + \frac{3}{4}z^*(\omega)] \right. \\ \left. \times \ln \left(\frac{D^2}{\omega^2 + (2/\beta)^2} \right) + i\{1 + z(\omega)[1 + \frac{3}{4}z^*(\omega)] \tanh(\beta\omega/2)\} \right]. \quad (\text{A35})$$

This is the self-energy, which we use to evaluate equation (30).

References

- Abrikosov A A 1965 *Physics* 2 5
 Cheung C Y and Mattuck R D 1970 *Phys. Rev. B* 7 2735
 Cini M 1980 *Phys. Rev. B* 22 5887
 Coldea M, Schaeffer H, Weissenberger V and Elschner B 1987 *Z. Phys. B* 68 25
 Coles B R, Oseroff S and Fisk Z 1987 *J. Phys. F: Met. Phys.* 17 L169
 Coqblin B and Schrieffer J R 1969 *Phys. Rev.* 185 847
 Gandra F G, Pontes M J, Schultz S and Oseroff S B 1987 *Solid State Commun.* 64 859
 Gandra F, Schultz S, Oseroff S B, Fisk Z and Smith J L 1985 *Phys. Rev. Lett.* 55 2719
 Keldysh L V 1965 *Sov. Phys.-JETP* 20 1018
 Kondo J 1964 *Prog. Theor. Phys.* 32 37
 Kugel K I and Khomskii D I 1985 *Proc. 17th Soviet Conf. on the Physics of Magnetic Phenomena (Donetsk)* p 107
 Lee P A, Rice T M, Serene J W, Sham L J and Wilkins J W 1986 *Commun. Condens. Matter Phys.* 12 98
 Lighthill M J 1959 *Introduction to Fourier Analysis and Generalized Functions* (Cambridge: Cambridge University Press) pp 44–75
 Luttinger J M 1961 *Phys. Rev.* 119 1153
 Luttinger J M and Ward J C 1960 *Phys. Rev.* 118 1417
 Ochi M M and de Menezes O L T 1988 *Solid State Commun.* 68 1035
 ——— 1991 *Physica B* 171 320
 Rammer J and Smith H 1986 *Rev. Mod. Phys.* 58 323
 Read N and Newns D M 1983 *J. Phys. C: Solid State Phys.* 16 3273
 Schlott M, Elschner B, Herrmann M and Assmus W 1988 *Z. Phys. B* 72 385
 Schweitzer H and Czyczoll G 1990 *Z. Phys. B* 79 377
 ——— 1991 *Phys. Rev. Lett.* 67 3724
 Simanek E and Sasahara K 1987 *J. Low Temp. Phys.* 66 249
 Varma C M 1985 *Phys. Rev. Lett.* 55 2723
 Wright F 1994 *J. Phys.: Condens. Matter* 6 223
 Yoshimori A and Kasai H 1983 *J. Magn. Magn. Mater.* 31–34 475