Electron spin resonance of a magnetic impurity in the resonant level model

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(Received 12 December 2000; published 7 August 2001)

We applied the renormalization group technique to calculate the spin relaxation rate of a well-defined magnetic moment in the neighborhood of a fluctuating valence ion. We have shown that for the resonance model the relaxation rate decreases in relation to the relaxation rate of a pure metal.

DOI: 10.1103/PhysRevB.64.092404 PACS number(s): 33.35.+r, 76.30.−v

I. INTRODUCTION

The electron spin resonance (ESR) of a magnetic probe embedded in an interconfiguration fluctuation (ICF) host has been used to investigate the effect of the valence fluctuation in the ESR linewidth. In order to explain the reduction of the linewidth of the ESR of a magnetic probe (Gd^{3+}) in host $CePd_3$, Gambke, Elschner, and Hirst[1] proposed a model where the dominant source of the relaxation is the coupling between the magnetic probe and the conduction band, given by $-2J_{k\mu}\vec{S}_{\mu}\cdot\vec{s}$, where $J_{k\mu}$ is the coupling constant, $\vec{S}_{\mu}$ is the magnetic probe spin and $\vec{s}$ is the conduction electron spin density. They represented the ICF host by the resonant level model and obtained the ESR linewidth of the magnetic probe in the zero temperature limit. Here we apply the renormalization group technique of Wilson[2] to that model to calculate the ESR linewidth for finite temperatures. For temperatures on the order of the resonant level energy we obtained that the relaxation rate divided by temperature as a function of the temperature presents a minimum due to the resonant coupling between the ICF orbital and the conduction band. In the limit of zero temperature the relaxation rate is proportional to the temperature and is lower than for a normal metal.

The ESR of a magnetic impurity embedded in a heavy fermion compounds results in an enhancement in the Korringa rate.[3] However, the results for the intermediate valence compounds are controverted.[4] Gambke *et al.*[1] associated the reduction in the relaxation rate of Gd in $CePd_3$ to the intermediate valence state. On the other hand, Heirinch and Meyer[5] observed the opposite phenomena in $CeBe_{13}$:Gd, as well Barberis *et al.*[4] in $CeIr_2$:Nd and Rettori *et al.*[6] in $YbInCu_4$:Gd.

II. THE MODEL

The ICF host is represented by the Hamiltonian of the resonance model, which consists of a conduction band whose electrons are hybridized with a resonance level:

$$H=\sum_{\vec{k},\mu}\varepsilon_{\vec{k}}C^{\dagger}_{\vec{k}\mu}C_{\vec{k}\mu}+\varepsilon_f C^{\dagger}_{f\mu}C_{f\mu}+V\sum_{\vec{k}}\,(C^{\dagger}_{\vec{k}\mu}C_{f\mu}+\text{H.c.}), \tag{1}$$

where $C^{\dagger}_{\vec{k}\mu}(C_{\vec{k}\mu})$ creates (annihilates) an electron in the conduction band with energy $\varepsilon_{\vec{k}}$, spin μ and momentum $\vec{k}$, $C^{\dagger}_{f\mu}(C_{f\mu})$ creates (annihilates) an electron with energy ε_f and spin μ in the ICF orbital and V is the hybridization interaction between this orbital and the conduction band.

The magnetic probe ion is sited at $\vec{R}_i$ and interacts with the conduction electrons via the Hamiltonian

$$H_x=-A[\Psi^{\dagger}_{\uparrow}(\vec{R}_i)\Psi_{\downarrow}(\vec{R}_i)I_{-}+\text{H.c.}], \tag{2}$$

where A is the coupling constant, I_{-} is the lowering spin operator of the magnetic probe ion, and the field operator $\Psi_{\mu}(\vec{R}_i)=\Sigma_{\vec{k}}e^{i\vec{k}\cdot\vec{R}_i}C_{\vec{k},\mu}$ annihilates a conduction electron with momentum $\vec{k}$ and spin μ in the Wannier state around that ion.

III. THE FORMALISM

We begin introducing another basis formed by two sets of *s*-wave operators, that are sited around the magnetic probe and at the ICF ion. Let

$$C_{\varepsilon\mu}=\frac{1}{\sqrt{\rho}}\sum_{\vec{k}}\,e^{i\vec{k}\cdot\vec{R}_f}\delta(\varepsilon-\varepsilon_{\vec{k}})C_{\vec{k}\mu} \tag{3}$$

be an operator that annihilates a *s*-wave electron around the ICF ion sited at $\vec{R}_f$, with energy $\varepsilon=\varepsilon_{\vec{k}}$ and spin μ, obeying the usual anticommutation relation $\{C_{\varepsilon\mu},C^{\dagger}_{\varepsilon'\mu'}\}=\delta(\varepsilon-\varepsilon')\delta_{\mu\mu'}$, and

$$d_{\varepsilon\mu}=\frac{1}{\sqrt{\rho}}\sum_{\vec{k}}\,e^{i\vec{k}\cdot\vec{R}_i}\delta(\varepsilon-\varepsilon_{\vec{k}})C_{\vec{k}\mu} \tag{4}$$

an operator that annihilates an electron in a *s*-wave state at the magnetic probe ion, with energy $\varepsilon=\varepsilon_{\vec{k}}$ and spin μ, with the anticommutation relation $\{d_{\varepsilon\mu},d^{\dagger}_{\varepsilon'\mu'}\}=\delta(\varepsilon-\varepsilon')\delta_{\mu\mu'}$, where ρ is the one-electron density of states per spin. In terms of the operator $d_{\varepsilon\mu}$, the field operator for the conduction electrons around the magnetic probe ion is $\Psi_{\mu}(\vec{R}_i)=\int d\varepsilon\sqrt{\rho}d_{\varepsilon\mu}$ and, similarly, the field operator for the conduction electrons around the ICF ion is $\Psi_{\mu}(\vec{R}_f)=\int d\varepsilon\sqrt{\rho}C_{\varepsilon\mu}$. The basis formed by the operators $C_{\varepsilon\mu}$ and $d_{\varepsilon\mu}$ is not an orthogonal one. Then, using the Gram-Schmidt orthogonality process, we can obtain another operator

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$$\overline{C}_{\varepsilon\mu}=\frac{1}{\sqrt{1-\dfrac{\sin^2(k_FR)}{k_F^2R^2}}}\left[d_{\varepsilon\mu}-\frac{\sin(k_FR)}{k_FR}C_{\varepsilon\mu}\right], \quad (5)$$

which obeys the anticommutation relations $\{\overline{C}_{\varepsilon\mu},\overline{C}^{\dagger}_{\varepsilon'\mu'}\}=\delta(\varepsilon-\varepsilon')\delta_{\mu\mu'}$ and $\{\overline{C}_{\varepsilon\mu},C^{\dagger}_{\varepsilon'\mu'}\}=0$, with $R=|\vec{R}_f-\vec{R}_i|$ being the distance between the magnetic probe and the ICF ions. In this basis the Hamiltonians H and H_x can be written as

$$H=H_0+H_{vf}, \quad (6)$$

$$H_0=D\int_{-1}^{+1}\overline{C}^{\dagger}_{\varepsilon\mu}\overline{C}_{\varepsilon\mu}d\varepsilon, \quad (7)$$

$$H_{vf}=D\left[\int_{-1}^{+1}\varepsilon C^{\dagger}_{\varepsilon\mu}C_{\varepsilon\mu}d\varepsilon+\frac{\varepsilon_f}{D}C^{\dagger}_{f\mu}C_{f\mu}+\sqrt{\frac{2\Gamma}{\pi D}}(f^{\dagger}_{0\mu}C_{f\mu}+\text{H.c.})\right], \quad (8)$$

and

$$H_x=[\Theta_1 g^{\dagger}_{0\uparrow}g_{0\downarrow}+\Theta_2(g^{\dagger}_{0\uparrow}f_{0\downarrow}+f^{\dagger}_{0\uparrow}g_{0\downarrow})+\Theta_3 f^{\dagger}_{0\uparrow}f_{0\downarrow}]I_-+\text{H.c.}, \quad (9)$$

where

$$f_{0\mu}=\frac{1}{\sqrt{2}}\int_{-1}^{+1}d\varepsilon C_{\varepsilon\mu}, \quad (10)$$

$$g_{0\mu}=\frac{1}{\sqrt{2}}\int_{-1}^{+1}d\varepsilon \overline{C}_{\varepsilon\mu}, \quad (11)$$

$$\Theta_1=-2A\left[1-\frac{\sin^2(k_FR)}{k_F^2R^2}\right], \quad (12)$$

$$\Theta_2=-2A\frac{\sin(k_FR)}{k_FR}\left[1-\frac{\sin^2(k_FR)}{k_F^2R^2}\right]^{1/2}, \quad (13)$$

$$\Theta_3=-2A\frac{\sin^2(k_FR)}{k_F^2R^2}, \quad (14)$$

$\Gamma=\pi\rho V^2$, k_F is the moment at the Fermi level and the energy ε is measured in relation to the Fermi level in units of the half-width D of the conduction band.

According to Refs. 8–10, in the Wilson numerical renormalization group method the conduction bands that appear in Eqs. (7) and (8) are divided into logarithmic intervals $\pm D\Lambda^{-j-z}$ ($j=1,2,\ldots$), where z is a continuous parameter and $\Lambda>1$, and are written in a ''hopping'' form, suitable to be numerically diagonalized, in terms of an orthonormal set of fermion operators $\{f_{n\mu}\}$ and $\{g_{n\mu}\}$ around the ICF and the magnetic probe site, respectively,

$$H_0^N=\Lambda^{(N-1)/2}\left\{\sum_{n=0}^{N-1}\varepsilon_n(g^{\dagger}_{n\mu}g_{(n+1)\mu}+g^{\dagger}_{(n+1)\mu}g_{n\mu})\right\}, \quad (15)$$

$$H_{vf}^N=\Lambda^{(N-1)/2}\left\{\sum_{n=0}^{N-1}\varepsilon_n(f^{\dagger}_{n\mu}f_{(n+1)\mu}+f^{\dagger}_{(n+1)\mu}f_{n\mu})+\tilde{\varepsilon}_f C^{\dagger}_{f\mu}C_{f\mu}+\tilde{\Gamma}^{1/2}(f^{\dagger}_{0\mu}C_{f\mu}+\text{H.c.})\right\}, \quad (16)$$

where

$$\tilde{\Gamma}=\left(\frac{2}{1+\Lambda^{-1}}\right)^2\frac{2\Gamma}{\pi D}, \quad (17)$$

$$\tilde{\varepsilon}_f=\left(\frac{2}{1+\Lambda^{-1}}\right)\frac{\varepsilon_f}{D}, \quad (18)$$

ε_n is function of Λ and z and is obtained numerically, N is an integer, and the scale factor $\Lambda^{(N-1)/2}$ was introduced in order to make the lowest energy scale in H_{vf}^N and H_0^N of order 1. For $z=1$ can be written as[8]

$$\varepsilon_n=\Lambda^{-n/2}(1-\Lambda^{-n-1})(1-\Lambda^{-2n-1})^{-1/2}(1-\Lambda^{-2n-3})^{-1/2}.$$

The diagonalization of the Hamiltonians given by Eqs. (15) and (16) is performed iteratively in subspaces of the same charge and spin (Q,S,S_z), using the renormalization group transformations

$$H_{vf}^{N+1}=\Lambda^{1/2}H_{vf}^N+\varepsilon_N(f^{\dagger}_{N\mu}f_{(N+1)\mu}+f^{\dagger}_{(N+1)\mu}f_{N\mu}) \quad (19)$$

and

$$H_0^{N+1}=\Lambda^{1/2}H_0^N+\varepsilon_N(g^{\dagger}_{N\mu}g_{(N+1)\mu}+g^{\dagger}_{(N+1)\mu}g_{N\mu}), \quad (20)$$

respectively.

In this new basis, according to Eq. (9), the magnetic probe ion interacts with the conduction electrons only via the first orbitals $g_{0\mu}$ ($f_{0\mu}$) of the chain Hamiltonians [Eqs. (15) and (16)].

IV. THE RELAXATION RATE

The relaxation rate, which is the central point of the present work, is calculated using the Fermi golden rule[7]

$$\frac{1}{T_1}=\frac{4\pi}{h}\sum_{I,F}P_I|\langle I|H_x|F\rangle|^2\delta(E_I-E_F), \quad (21)$$

where T_1 is the relaxation time, P_I is the statistical Boltzmann weight [$P_I=\exp(-E/k_BT)/\Sigma_i\exp(-E_i/k_BT)$], E_I and E_F are the energies of the initial ($|I\rangle$) and the final ($|F\rangle$) many-particle states, respectively, T is the temperature and k_B is Boltzmann's constant. According to Eq. (9) for H_x, the term $\langle F|H_x|I\rangle$ is written as

$$|\langle F|H_x|I\rangle|^2=\Theta_1^2|\langle F|g^{\dagger}_{0\uparrow}g_{0\downarrow}|I\rangle|^2+\Theta_2^2|\langle F|g^{\dagger}_{0\uparrow}f_{0\downarrow}|I\rangle|^2+\Theta_2^2|\langle F|f^{\dagger}_{0\uparrow}g_{0\downarrow}|I\rangle|^2+\Theta_3^2|\langle F|f^{\dagger}_{0\uparrow}f_{0\downarrow}|I\rangle|^2 \quad (22)$$

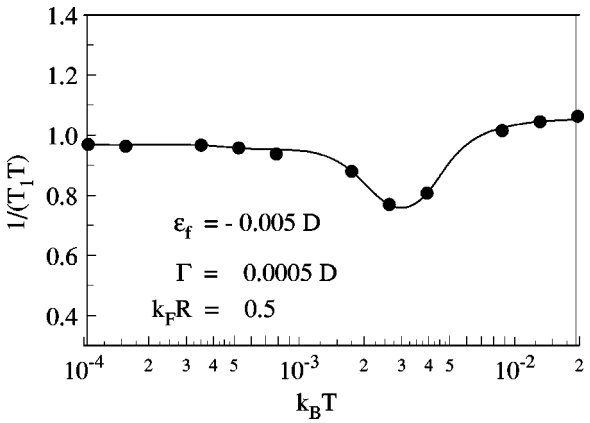

FIG. 1. The relaxation rate as a function of temperature for $\varepsilon_f = -0.005D$, $\Gamma = 0.0005D$, and $k_F R = 0.5$. For $k_B T \approx \varepsilon_f$, the rate $1/(T_1 T)$ presents a minimum around ε_f due to the resonant coupling between the ICF orbital and the conduction band. For low temperature $k_B T \ll \varepsilon_f$, Γ the system shows a typical Korringa relaxation rate.

and the eigenstates $|I\rangle$ and $|F\rangle$ are direct products of the eigenstates of the Hamiltonians H_0^N and H_{vf}^N.

The sum in Eq. (21) was perfomed according to Ref. 10. Figure 1 shows the numerical result for $1/(T_1 T)$ as a function of the temperature, where we take the coupling constant $A = \sqrt{h/(4\pi k_B \rho^2)}$. For $k_B T \ll \varepsilon_f, \Gamma$ the system presents a typical Korringa relaxation rate. For $k_B T \approx \varepsilon_f$, the rate $1/(T_1 T)$ decreases due to the resonant coupling between the ICF ion and the conduction band. For low temperature an analytical expression for $1/T_1$ is obtained, which is useful to verify the accuracy of the numerical renormalization group calculation.

In order to calculate the relaxation rate in the zero temperature limit we diagonalize the Hamiltonians H_0^N and H_{vf}^N and write the many-particle eigenstates in terms of many single particle states. For large odd N the diagonalization of H_0^N and H_{vf}^N into a set of single-particle levels gives[8]

$$H_0^N = \sum_{j=-(N+1)/2}^{j=(N+1)/2} \overline{\eta}_j \overline{a}_{j\mu}^{\dagger} \overline{a}_{j\mu}, \tag{23}$$

$$H_{vf}^N = \sum_{j=-(N+1)/2}^{j=(N+1)/2} \hat{\eta}_j \hat{a}_{j\mu}^{\dagger} \hat{a}_{j\mu}, \tag{24}$$

where $\overline{\eta}_j = \Lambda^{j-1}$ $(j \gg 1)$, $\overline{\eta}_{-j} = -\overline{\eta}_j$ $(\overline{\eta}_0 \equiv 0)$,

$$\hat{\eta}_j = \Lambda^{j-1+\gamma}, \tag{25}$$

$$\hat{\eta}_{-j} = -\hat{\eta}_j, \tag{26}$$

$$\cot(\pi\gamma) = \frac{\hat{\eta}_j}{|\hat{\eta}_j|} \frac{\Lambda^{(N-1)/2} \ln \Lambda}{\pi \alpha_0^2 \tilde{\Gamma}} [\hat{\eta}_j - \tilde{\varepsilon}_f \Lambda^{(N-1)/2}], \tag{27}$$

$\hat{a}_{j\mu}$ are written in terms of the operators $a_{j\mu}$ and the elements of an orthogonal matrix U_{ij},

$$\hat{a}_{j\mu} = \sum_i U_{ij} a_{i\mu}, \tag{28}$$

$$U_{ij} = \Lambda^{(n-1)/4} \tilde{\Gamma}^{1/2} \alpha_{0i} \frac{U_{0j}}{\hat{\eta}_j - \eta_i} \quad \text{for} \quad i \neq 0, \tag{29}$$

$$U_{0j} = \left(\frac{|\hat{\eta}_j| (\ln \Lambda)^2}{|\hat{\eta}_j| (\ln \Lambda)^2 + \frac{\pi^2 (1 - \Lambda^{-1})}{2} \tilde{\Gamma} \Lambda^{(N-1)/2} [1 + \cot^2(\pi\gamma)]} \right)^{1/2}. \tag{30}$$

The operators $g_{0\mu}$ and $f_{0\mu}$ that appears in the Hamiltonian H_x given by Eq. (9), can be written in terms of $\overline{a}_{j\mu}$ and $\hat{a}_{j\mu}$ as

$$g_{0\mu} = \Lambda^{-(N-1)/4} \left[\sum_{j=-(N+1)/2}^{j=(N+1)/2} \alpha_{0j} \overline{a}_{j\mu} \right], \tag{31}$$

$$f_{0\mu} = \Lambda^{-(N-1)/4} \sum_{j=-(N+1)/2}^{(N+1)/2} \sum_{l=-(N+1)/2}^{(N+1)/2} \alpha_{0l} U_{lj} \hat{a}_{j\mu}, \tag{32}$$

with $\alpha_{0j} = ((1 - \Lambda^{-1})/2)^{1/2} \Lambda^{(j-1)/2}$ and $\alpha_{00} \equiv 0$.

For low temperature, Eq. (21) can be written as[11]

$$\frac{1}{T_1} = \lim_{\Lambda \to 1} \lim_{N \to \infty} k_B T \rho^2 \left(\frac{2}{1 + \Lambda^{-1}} \right)^2 \Lambda^{(N-1)} \sum_{l,m} W(l,m),$$

$$W(l,m) = |\langle F_m^l | H_x | I \rangle|^2 \delta(E_l - E_m) \delta(E_m - E_0), \tag{33}$$

where $\rho = 1/D$ is the density of states, $|I\rangle$ is taken as the many-particle ground state, $|F_m^l\rangle$ is the final many-particle excited states associated to a hole and an electron in the single-particle energy levels E_m and E_l, respectively, and E_0 is the Fermi energy level ε_o multiplied by the factor $2\Lambda^{(N-1)/2}/(1 + \Lambda^{-1})$. Using $g_{0\mu}$ and $f_{0\mu}$, given by Eqs.

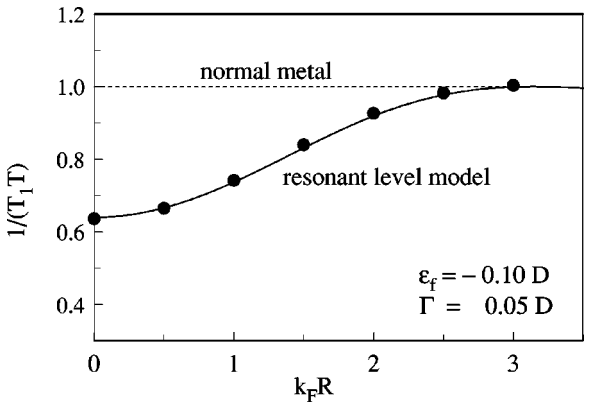

FIG. 2. The low temperature behavior of the relaxation rate as a function of $k_F R$. The numerical renormalization group results, which are represented by circles, are in good agreement with the analytical results (full line) given by Eq. (34).

(31) and (32), and the Sommerfeld-Watson[12] transformation to evaluate the sums over m and l in Eq. (33), we obtain the relaxation time

$$\frac{1}{T_1} = \left[1 - \frac{\Gamma^2}{[\varepsilon_f^2 + \Gamma^2]} \frac{\sin^2(k_F R)}{(k_F R)^2} \right]^2 T. \qquad (34)$$

Figure 2 presents the numerical renormalization group results for $1/(T_1 T)$ as a function of $k_F R$ for low temperature, obtained from Eq. (21) (circles), which are in good concordance with the analytical results given by Eq. (34) (full line). The accuracy of the numerical renormalization group method encourage us to study the model that takes into account the Coulomb interaction $U C_{f\uparrow}^{\dagger} C_{f\uparrow} C_{f\downarrow}^{\dagger} C_{f\downarrow}$ between the electrons in the ICF orbital, which is presently in progress.

We have shown that the renormalization group method is useful to calculate the relaxation time of a magnetic probe ion embedded in an ICF host, which is represented by the resonance model. The method is appropriate to investigate the effect of the screening and the Coulomb interaction in the fluctuating valence orbital, a complex many-body problem. As the ICF orbital is coupled to the chain Hamiltonian of the conduction band only via the $f_{0\mu}$ orbital, the numerical error is minimized in the iterative process which takes place in the renormalization group calculation.

ACKNOWLEDGMENT

The authors would like to thank the CNPq/Brazil for the financial support.

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