Dynamics of Tunneling Centers in Metallic Systems

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Dynamics of tunneling centers (TC) in metallic systems is studied, using the technique of bosonization. The interaction of the TC with the conduction electrons of the metal involves two processes, namely, the screening of the TC by electrons, and the so-called electron assisted tunneling. The presence of the latter process leads to a different form of the renormalized tunneling frequency of the TC, and the tunneling motion is damped with a temperature dependent relaxation rate. As the temperature is lowered, the relaxation rate per temperature shows a steep rise as opposed to that in the absence of electron assisted process. It is expected that this behavior should be observed at very low temperatures in a careful experiment. The present work thus tries to go beyond the existing work on the interaction of electron assisted process. It is expected that this behavior should be observed at very low temperatures in a careful experiment. The present work thus tries to go beyond the existing work on the dynamics of a two-level system in metals, by treating the electron assisted process.

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I. INTRODUCTION

The existence of double-well structures in metals has now been known for a long time. They arise in diverse physical situations, the most well-known of which is metallic glasses. The existence of tunneling states in glasses was proposed long back [1]. It is believed that because the atoms in a glass are 'quenched' in a random configuration, there is a possibility of an atom, or group of atoms to see a complicated potential landscape in its neighbourhood, which may have at least two neighbouring local minima. In such a situation, the atom, or the group of atoms can spontaneously tunnel from one well to the other. In this, and similar situations, the entity is referred to as a tunneling center (TC). Another well known, and well studied example is that of hydrogen trapped in Nb(OH)$_2$. At very low temperatures ($\sim 70K$), the hydrogen, which can normally diffuse quantum-mechanically, is localized in some trap-sites formed by oxygen atoms [2]. The geometry of the system is such that the hydrogen can stay in two close-by, energetically equivalent sites around the trapping atom. This results in the formation of a TC. Incoherent neutron scattering reveals the tunneling of the hydrogen, which is influenced by the conduction electrons of the metal. These experiments have been theoretically analyzed using the model of a particle in a double-well, in contact with free electrons [3,4]. Lately, the conductance behavior of narrow metal constrictions has been explained within the framework of scattering of conduction electrons from a TC [5]. The behavior of certain heavily doped conducting polymers has also been attributed to the existence of TCs [6]. Universal conductance fluctuations have also been used to study the dynamics of a single two-level defect in a metal [7]. In short, tunneling centers in metals have proved to be of considerable experimental and theoretical importance.

Theoretical scenario is the following. The TC can be classified to be either slow or fast, depending on its detailed behavior. When the TC is slow, the effect of the conduction electrons is to provide some kind of friction to the tunneling motion of the particle. What actually happens is that the particle being charged, has a screening conduction electron cloud around it. As it tunnels from one site to the other, the electrons have to follow its motion adiabatically. Now it turns out that the electrons cannot actually follow the motion of the particle instantaneously - the adiabatic approximation breaks down. The tunneling of the atom is a local perturbation which can give rise to an infinite number of electron-hole pairs in the electron gas, with infinitesimal energy, and thus to the so-called infra-red divergence. The result is that the tunneling of the particle is governed by the relaxation of the conduction electron cloud around it. The tunneling is thus heavily suppressed and at zero temperature the particle tends to localize in a particular well. This small energy behavior of the electron gas can be described in terms of charge density excitations of the electron gas, which are approximately bosonic in nature. In addition, if the low-temperature dynamics of the particle is confined to the two-dimensional Hilbert space spanned by the two ground states of the isolated wells, one can map the problem onto the model of dissipative two-level system (TLS) studied extensively by Leggett et al [8]. One has to choose what is called the Ohmic dissipation in order to correctly describe the behavior of electronic excitations.

In a slow TC, the atom remains stationary during an electron scattering. On the other hand, if the TC is fast, the process where there is a scattering of an electron simultaneously with the tunneling of the particle, has to be taken into account. This process was originally proposed by Kondo [9] and further work along this direction has been done by Zawadowsky and collaborators [10-15]. Here the electrons assist the tunneling motion of the particle, and hence the particle cannot be localized in one of the two wells, even at very low temperatures. The particle, tunneling from one well to the other, scatters...
the electrons between different orbital angular momentum states. If we describe the TLS, formed by the particle in a double-well, by a pseudo-spin 1/2, the situation is quite similar to the Kondo problem where the impurity spin-flip causes a spin-flip of the electrons. Here the role of the electron-spin in the Kondo problem is played like two independent channels of electrons to which the TC couples. It has been shown earlier that this problem can be mapped on to that of the two-channel Kondo model, originally proposed by Nozières and Blandin [12], which has come under a lot of attention because of its non-Fermi-liquid low temperature behavior.

In an actual TC in metals, both the processes should be present - infact, the amplitude of screening by conduction electrons is much larger than that for electron assisted tunneling (because the tunneling particle is heavy). The interplay between these two processes may yield interesting dynamics at low temperatures. The dynamics of a TLS, with electron screening, has been studied in detail, and the high as well as low temperature behavior is well understood. At very low temperatures, the TLS undergoes weakly-damped coherent oscillations, with a tunnel frequency which is strongly renormalized by the coupling to the electrons. The damping coefficient increases roughly linearly with temperature, whereas the effective tunnel frequency follows a weak power-law. As the temperature is increased, the thermally excited electron-hole pairs destroy the coherence of the particle. As a result, tunneling gets incoherent, and resembles stochastic jump diffusion. The jump rate, from one site to the other, however, has a very non-trivial temperature dependence. It follows a power-law, the jump rate decreasing with increasing $T$.

The dynamics of a slow TLS, in all temperature regimes, has been studied within the so-called dilute boson gas approximation (DBGA), to the underlying functional integral expression for the dynamics of the TLS [13]. For weak coupling of the TLS to the electrons, some calculations have gone beyond the DBGA, to study the low-temperature dynamics [13]. These two approaches do not treat the electron assisted tunneling process. Very recently conformal field theory has been used to study the very low temperature scaling behavior of the two-channel Kondo model [13]. The results obtained are relevant for the anomalous resistivity of the metal arising out of the scattering of electrons off the TLS. This approach has been highly successful. We are, however, interested in the actual dynamics of a TLS and not on its effect on the metallic system. Such interest stems from the fact that while experimentally probing the dynamics of a TLS, one usually deals with things like the tunnel frequency and the relaxation rate of the tunneling atom.

In this paper, we calculate the Laplace transform of the time correlation function of the TLS, which, in the context of neutron scattering from the tunneling particle, is related to the incoherent structure factor. The correlation function will give the necessary information regarding the interplay between the tunneling dynamics and the relaxation behavior, so commonly seen in all quantum dissipative systems.

II. THE MODEL

In the following we assume that the TC consists of a single particle in an asymmetric double well potential. The temperature is low enough for the dynamics of the particle to be effectively described by the Hilbert space spanned by the two lowest states of the isolated wells. In this approximation, the TC becomes a TLS. Now, the interaction of the TLS with the conduction electrons can be described by the following Hamiltonian (see e.g., [12])

$$H' = \frac{1}{2} \hbar \epsilon \sigma_z + \frac{1}{2} \hbar \Delta_0 \sigma_x$$
$$+ \frac{1}{2} \sqrt{2K} \rho \sigma_z \sum_{p,k,k'} (c_{pkl}^\dagger c_{pkl} - c_{pkl}^\dagger c_{pkl}^-)$$
$$+ \frac{\hbar \Delta_1}{2 \rho \epsilon_F} \sum_{p,k,k'} (\sigma_+ c_{pkl}^\dagger c_{pkl}^- + \sigma_- c_{pkl}^\dagger c_{pkl}^+)$$
$$+ \sum_{p,kl} \epsilon_p(k) c_{pkl}^\dagger c_{pkl}$$

In (1), TLS is described by the pseudo-spin $\sigma$. The two eigenstates of $\sigma_z, | \pm >$ correspond to the tunneling particle being in the left and the right wells, respectively. The electrons are described by a spherical wave representation, due to the presence of a point scatterer in the Fermi sea. The first two terms in (1) describe the bare TLS, $\epsilon$ being the asymmetry between the two wells and $\Delta_0$ the bare tunneling frequency. The third term represents the screening of the TC by the conduction electrons. The fourth term in (1) describes electron assisted tunneling, $\Delta_1$ being the strength of the process. The last term is the Hamiltonian of the electrons in the partial wave representation. The indices $k$ and $k'$ represent the wavevectors of the electrons; $l$ is the index for the partial wave and in the present model runs over two values, + and −; $p$ represents the spin of the electrons and runs over two values $\pm 1/2$; $\epsilon_p(k)$ is the energy of the electrons with wave-vector $k$ and spin $p$; $\rho$ is the density of states at the Fermi level of the electron gas; $K$ is a dimensionless coupling constant which describes the screening interaction of the TLS with the electrons, whose value is restricted to be $0 \leq K \leq 0.5$ [19]. Operator $c_{pkl}^\dagger (c_{pkl})$ creates (annihilates) an electron in a state with spin $p$, a wavevector $k$, energy $\epsilon_p(k)$ and partial wave state +. The energy of the excitations is assumed to have an upper cutoff, equal to the Fermi energy of the electron gas, $\epsilon_F$. The Hamiltonian in (1) can, in fact, be thought of as a highly anisotropic two-channel Kondo Hamiltonian with
two external magnetic fields $\epsilon$ and $\Delta_0$, pointing along $z$- and $x$-axis respectively. It also differs crucially from the Kondo problem due the fact that the value of the so-called $J_{fr}$ in the present case is very different. Due to this, the present model shows coherent oscillation of the pseudo-spin, which is absent in in the conventional model.

We have shown earlier that the dynamics of the single channel Kondo problem can be efficiently studied by bosonizing the Hamiltonian \[20\]. The success of this method in treating the effect of the low-energy excitations on the impurity spin motivates us to also apply it to the present problem. The two spin channels of the electrons being completely independent, the bosonization of one does not affect that of the other. Consequently, the bosonized form for \(1\) can be written down in a manner similar to that of the single channel Kondo Hamiltonian (for a detailed account of bosonization the reader is referred to \[21\]). The resulting Hamiltonian then assumes the following form

\[
H' = \frac{1}{2} \hbar \epsilon \sigma_z + \frac{1}{2} \hbar \Delta_0 \sigma_x \\
+ \frac{1}{2} \hbar \sqrt{2 \pi K v_F / L \sigma_z} \sum_{k,p} \sqrt{\omega_k \omega_p} e^{i\omega_k/2\omega_c}(b_{pk}^\dagger + b_{pk}) \\
+ \frac{1}{2} \hbar \Delta_1 \sum_p [\sigma_+ e^{-\xi_p} + \sigma_- e^{\xi_p}] + \sum_{kp} \hbar \omega_k b_{pk}^\dagger b_{pk} \tag{2}
\]

where $\xi_p = \frac{1}{2} \sum_p (\frac{\pi v_F}{L \omega_k})^{1/2} \exp(-\omega_k/L \omega_c)(b_{pk}^\dagger - b_{pk})$. In \[2\], $p$ is the channel index, $\omega_k = v_F k$, $v_F$ being the Fermi velocity and $\omega_c$ is a high frequency cutoff related to $\epsilon_F$. $L$ is the length of the normalization box and the limit $L \to \infty$ is taken such that $(2\pi/L) \sum_k \to \int dk$. The boson operator $b_{pk}^\dagger$ creates a particle in the state $k$ of the $p$th channel, which is an excitation of the electron gas. The charge density excitations drop out of the problem.

If $\Delta_1$ is zero, the Hamiltonian given by \[2\], without $\Delta_0$, can be diagonalized exactly. The strategy then, is to treat the terms proportional to $\Delta_0$ and $\Delta_1$ as a perturbation. To this end we perform a unitary transformation $U H' U^{-1}$ on the Hamiltonian where $U = \exp[\sigma_z \sqrt{K/2} \sum_p \xi_p]$. The transformed Hamiltonian has the following form

\[
H = \frac{1}{2} \hbar \epsilon \sigma_z + \sum_{k,p} \hbar \omega_k b_{pk}^\dagger b_{pk} \\
+ \frac{1}{2} \hbar \Delta_0 [\sigma_+ e^{\sqrt{2} \xi} + \sigma_- e^{-\sqrt{2} \xi}] + \frac{1}{2} \hbar \Delta_1 \\
\times \sum_p [\sigma_+ e^{\sqrt{2} \xi + \xi_p} + \sigma_- e^{-\sqrt{2} \xi + \xi_p}] \tag{3}
\]

One can see that for $\Delta_0$, $\Delta_1 = 0$, the Hamiltonian in \[3\] is diagonal in the usual representation. In the following we denote the first and the second terms of \[3\] by $H_S$ and $H_B$, respectively, and the other two terms by $H_I$. A suitable perturbation theory can now be done on $H$ by treating $H_I$ perturbatively, which amounts to treating $K$ exactly and doing an expansion in $\Delta_0$ and $\Delta_1$.

The quantity of interest here is the Laplace transform of the time correlation function of the pseudo-spin, $C(t) = \langle \sigma_x(0) \sigma_x(t) \rangle$, given by

\[
\hat{C}(z) = \int_0^\infty e^{-zt} < \sigma_x(0) \sigma_x(t) > dt. \tag{4}
\]

Here, the angular brackets denote canonical ensemble average, and the Heisenberg time evolution of $\sigma_x$ is dictated by $H$. In the following analysis, the Greek indices $\mu, \nu$ denote the impurity spin states, and the “states” of the Liouvillean operators are denoted by $| \nu \rangle$. We shall now focus our attention on $\hat{C}(z)$ which can be put in the following form:

\[
C(z) = \sum_{\nu, \nu'} \langle \nu | \rho_S | \nu' \rangle C(0) < \nu' | e^{z U(z)} | \nu > \tag{5}
\]

In writing \[5\] we have made use of the Liouvillean operator formalism to introduce a “bath-averaged time evolution operator” $[U(t)]_{av}$, where $U(t) = e^{iL_{fr}t}$, $L$ being the Liouvillian associated with $H$. In addition, we have factorized the canonical density matrix $\rho$, as $\rho \approx \rho_S \cdot \rho_B$, where $\rho_S$ is the density operator associated with $H_S$ and $\rho_B$ is the density operator for a bath of noninteracting bosons given by $H_B$. The bath average $[\cdot]_{av}$ implies a multiplication by $\rho_B$ and a trace over the bath states. The bath-averaged time evolution operator contains all the information regarding the influence of the electronic environment on the TLS. We calculate $[U(z)]_{av}$ using the resolvent expansion formalism where the resolvent is treated perturbatively to yield a “self-energy” which is second order in $H_I$, resulting in $[U(z)]_{av} \approx \frac{1}{z - i L_S + \{ L_I (z - i L_S - i L_B)^{-1} L_I \}_{av}^{-1}}$, where $L_S, L_I$ and $L_B$ are Liouvillean operators associated with $H_S$, $H_I$ and $H_B$ respectively \[3\]. The calculation proceeds along the lines of Ref. \[2\] which treats the dynamics of a TLS in contact with a bosonic bath.

We first calculate the matrix for the self-energy $[L_I (z - i L_S - i L_B)^{-1} L_I ]_{av}$ and it turns out that in the second order resolvent expansion, it can be represented in terms of certain correlation functions of a gas of free bosons. In fact, one needs quantities like $< e^{+ \xi(t)} e^{-\xi(t)} >$ where the subscript $B$ indicates that the correlation function is calculated using only a free-boson Hamiltonian \[3\]. These correlation functions can be calculated using certain well known properties of a set of harmonic oscillators using the approximation $T \ll \hbar \omega_c / k_B$ and $t \ll 1/\omega_c$. The self-energy, being related to the Liouvillean operators, is a $4 \times 4$ matrix within the space of the states of the pseudo-spin. But this matrix turns out to be block diagonal, and the $2 \times 2$ blocks can be handled with ease. Consequently, the block-diagonal matrix for the self-energy is combined with the diagonal matrix for $(z - i L_S)$ and inverted to yield the averaged time-evolution operator $[U(z)]_{av}$. 
III. RESULT

Once the bath-averaged time-evolution operator is known, we are all set to calculate the Laplace transformed correlation function, the quantity of central importance to us. We plug in the form of $[\hat{U}(z)]_{\alpha\nu}$ in (9) to yield:

$$
\hat{C}(z) = \frac{z + \tanh \left( \frac{\hbar \epsilon z}{2} \right) \left[ \Phi_-(z + i\epsilon) - \Phi_-(z - i\epsilon) \right]}{z \left[ z + \Phi_+(z + i\epsilon) + \Phi_-(z - i\epsilon) \right]}, \quad (6)
$$

where $\Phi_\pm(z) = F(z) \pm F'(z)$, and

$$
F(z) = \frac{1}{4} \Delta_0^2(\Theta/\omega_c) \Gamma(1 - 2K) \frac{\Gamma(1 + K + z/\Theta)}{(z + \Theta K) \Gamma(1 - K + z/\Theta)} e^{i\pi K} 
- \frac{\Delta_0^2}{2 \omega_c^2} \frac{\Gamma(2\eta - 1) \Gamma(1 - \eta + z/\Theta)}{(z + \eta \Theta) \Gamma(1 + \eta + z/\Theta)} e^{-i\pi \eta}
+ \frac{\Delta_0^2}{32\pi^2} \frac{\Gamma(1 + 2\eta) \Gamma(1 - \delta + z/\Theta)}{(z - \delta \Theta) \Gamma(1 + \delta + z/\Theta)} e^{i\pi \delta}, \quad (7)
$$

In (6), $\Theta = 2\pi k_B T/\hbar$, $\eta = \sqrt{2K} - K$, $\delta = \sqrt{2K} - 2K$ and $F'(z)$ is the same as $F(z)$ except that the exponential factors are replaced by their complex conjugates. Some comments at this stage. The expression for the Laplace transformed correlation function, given by (6), describes the dynamics of a TC in a metal. The dynamics is governed by the screening of the TC by the conduction electrons, as well as the electron assisted process. The first term represents the effect of screening of the TC by the conduction electrons, while the second term accounts for the electron assisted process. The third and the fourth terms in (6) are much smaller in magnitude compared to the first two, because of the presence of the extra factors $\Theta/\omega_c$.

IV. DISCUSSION AND CONCLUSION

The second order treatment of $\Delta_0$ and $\Delta_1$ is equivalent to some kind of a DBGA if one were to employ the functional integral formalism, pioneered by Leggett et al [3]. Let us, for the time being, neglect electron assisted tunneling, by putting $\Delta_1 = 0$, which reduces the expression in (6) to the following:

$$
\hat{C}(z) = \frac{z + \tanh \left( \frac{\hbar \epsilon z}{2} \right) \sin \pi K [G(z + i\epsilon) - G(z - i\epsilon)]}{z \left[ z + \cos \pi K \{G(z + i\epsilon) + G(z - i\epsilon)\} \right]}, \quad (8)
$$

where

$$
G(z) = \frac{1}{2} \Delta_0^2(\Theta/\omega_c) \frac{\Gamma(1 - 2K) \Gamma(1 + K + z/\Theta)}{(z + \Theta K) \Gamma(1 - K + z/\Theta)}, \quad (9)
$$

This is just the DBGA result for a dissipative asymmetric TLS, for ohmic dissipation, derived by Leggett et al [9] using the path-integral formalism, and by Dattagupta et al [3] using the resolvent expansion formalism. By including the $\Delta_1$-dependent terms in the expression for the correlation function, we extend the DBGA result to the case where the electron assisted process plays an important role, in addition to the screening effect.

In order to make explicit analysis simpler we look at the particular case of a symmetric double-well where $\epsilon = 0$. The real part of $\hat{C}(z)$, which may describe the neutron scattering structure factor, is in general not a Lorentzian. In order to force a Lorentzian form, we further assume that $K \ll 1$. With this simplification, the expression (6) can be approximated by

$$
\hat{C}(z) \approx \frac{1}{z + \Delta_0^2/(z + \gamma_0) + 2\Delta_1^2/(z + \gamma_1)}, \quad (10)
$$

where $\Delta_0 = \Delta_0(\Theta/\omega_c)^K \Gamma(1 - 2K) \cos(\pi K)]^{1/2}$, $\Delta_1 = (\Delta_1/\omega_c)(\Theta/\omega_c)^{\gamma_0} \Gamma[2\eta\cos(\pi \eta)/(1 - 2\eta)]^{1/2}$, $\gamma_0 = K\Theta$ and $\gamma_1 = \eta\Theta$. In writing (10) we have neglected the last two terms in (6) (recall the approximation $\omega_c \gg \Theta$). The real part of the Laplace transformed correlation function given by (10) describes two Lorentzian lines of width $\gamma_L = \gamma_0 + 2\Delta_1^2\gamma_1/(1 + 2\Delta_1^2)$, centered at $\omega = \pm[\Delta_0^2/(1 + 2\Delta_1^2) - \gamma_0 + 2\Delta_1^2\gamma_1/(1 + 2\Delta_1^2)]$, centered at $\omega = \pm[\Delta_0^2/(1 + 2\Delta_1^2) - \gamma_0 + 2\Delta_1^2\gamma_1/(1 + 2\Delta_1^2)]$. One can see that the tunnel frequency of the particle is renormalized due to the effect of metallic electrons. The renormalization is not only due to the overlap of electron states corresponding to the two positions of the particle (which leads to a reduction factor of the form $(k_B T/\hbar \omega_c)^K$) but also due to the effect of electron assisted process. This will show up in the effective tunnel splitting of a particle, at very low temperatures where it is in the coherent tunneling regime. The width of the peaks, because of the $\Delta_1$ dependent term, also deviates from the usual “Korringa” form at very low temperatures.

The DBGA is known to break down at very low temperatures for small frequencies. In order to circumvent this difficulty we shall concentrate on the real part of $\hat{C}(z)$ near the resonances. Without any interaction with electrons the resonances for the TLS are at $z = \pm i\Delta_0$. So, we replace $F(z)$ in the expression (6) for $\hat{C}(z)$, by $F(i\Delta_0)$. Thus, the relaxation rate is given by $\gamma = Re[C(i\Delta_0)]$, and the tunnel-splitting is given by $\omega = -Im[C(i\Delta_0)]$. Figures 3 and 4 show plots of the two against temperature. In the plots, everything is scaled with $\Delta_0$, to yield dimensionless variables. One can see that for low enough temperature, the electron assisted process has a dramatic effect on the relaxation rate. At very low temperature, such a system is known to go into a Kondo-like correlated state. This may be the reason for this increased contribution to relaxation. This feature possibly can be experimentally observed. The tunnel-splitting, however, does not show a significant qualitative change in its temperature dependence. The plots indicate that at high temperatures the electron-assisted
process does not have a significant effect. But as the temperature is lowered, it is this process which dominates the dynamics of the TLS.

In conclusion, we have studied the dynamics of a particle in a double-well potential, in the presence of conduction electrons where the effect of electron assisted tunneling is taken into account in addition to conventional damping effects. In the appropriate limit the relevant expression reduces to the DBGA result for the dynamics of an Ohmic dissipative two-level system. In the coherent tunneling regime, the tunnel frequency is modified by the effect of electrons. The relaxation rate shows an unexpected rise as the temperature is lowered, as opposed to the case where the electron assisted process is absent. We believe that this feature should be observable in a careful experimental study.

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FIG. 1. For plotting the relaxation rate and tunnel splitting against $\Theta$, we scale all quantities with $\Delta_0$, so that $\Gamma = \gamma/\Delta_0$, $\tau = \Theta/\Delta_0$, $\Delta = \Delta_1/\Delta_0$ and $D = \omega_c/\Delta_0$. The plot shows dimensionless relaxation rate per temperature $\Gamma/\tau$ against the temperature $\tau$, for $D = 100.0$, $K = 0.05$, $\Delta = 1.0$ (solid line), and $\Delta = 0.0$ (dashed line). At low temperatures the behavior of the solid line indicates the increased relaxation due to the electron-assisted process.

FIG. 2. $\Omega = -Im[\hat{C}(i\Delta_0)/\Delta_0]$ plotted against temperature, for $D = 100.0$, $K = 0.05$, $\Delta = 1.0$ (solid line), and $\Delta = 0.0$ (dashed line).