Dynamics of the Biased Two-Level System in Metals

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We study the influence of conduction electrons on spectral and dynamical properties of the biased two-level system using path-integral methods. The structure factor for inelastic neutron scattering is calculated in the case of weak coupling ($K \ll 1$) and for a special value of the coupling strength ($K = \frac{1}{2}$). The effect of interbounce interactions is systematically taken into account at low temperatures, thus removing shortcomings of the dilute-bounce-gas approximation. The method and results are relevant also for the "macroscopic quantum coherence" problem.

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At very low temperatures the quantum dynamics of light interstitials in metals are strongly influenced by the nonadiabatic interaction with conduction electrons.[1] Specifically, the anomalous increase with decreasing temperature of the muon hopping rate in aluminum and copper below 10 K has been attributed to nonadiabatic screening effects.[2] The important influence of conduction electrons on defect tunneling has been seen even more clearly in neutron-spectroscopy experiments on hydrogen trapped by oxygen in niobium.[3] The proton in the $Nb(OH)_x$ samples seems to constitute a two-level system which is ideally suited for studying the influence of dissipation on quantum dynamics. In this system the dimensionless parameter K describing the coupling to the normal-state conduction electrons is very small, namely, near $K=0.05$. A theoretical analysis of the dissipative two-state system based upon the path-integral method has been presented by Leggett *et al.*,[4] and the theoretical aspects related to the fermionic effects have been surveyed by Kondo.[5] Most of the explicit results for the system dynamics were derived within the so-called dilute-bounce-gas approximation (DBGA) of the underlying functional-integral expression.[4,6] For a *symmetric* system the effect of the interbounce interactions is of order K^2, whereas the terms that are kept give nontrivial effects of order K. Thus the DBGA is a systematic weak-coupling approximation for a symmetric system down to $T=0$. Previously, the DBGA was employed to calculate the structure factor for a symmetric two-state system coupled to a Fermi bath.[7] The theoretical predictions were found to be in good agreement with recent experiments on hydrogen trapped by oxygen in normal-state niobium.[3]

For *asymmetric* two-state systems the effect of the interbounce interactions contributes to the order K at very low temperatures. Thus the DBGA breaks down in the presence of a bias, which shows up in a number of shortcomings[4,6] (see below).

In this Letter we present the corresponding systematic treatment of the dynamics for $K \ll 1$. We also give the exact solution for the special value $K=\frac{1}{2}$. A major motivation for this work is the recent observation of significant energetic shifts between the two interstitial sites of a given H atom in Nb due to static lattice strains from surrounding defects.[8] The shifts were shown to increase with increasing defect concentration. In view of current neutron-scattering experiments for such systems our attention here is concentrated on the calculation of the structure factor.[9] The method is relevant also for relaxation phenomena in metallic glasses[10] and for the "macroscopic quantum coherence" problem.[4,6]

Let us consider the dynamics of a defect tunneling between two trap sites with a bias energy $\hbar\epsilon$ between the ground states in the two traps and with a tunnel matrix element Δ_0. In the regime $\hbar\Delta_0, \hbar\epsilon, k_BT \ll \hbar\omega_0$ (but $k_BT/\hbar\Delta_0$ and $k_BT/\hbar\epsilon$ arbitrary), where $\hbar\omega_0$ is the energy of excitation in a single well, the original double-well potential problem can be truncated to an effective dissipative two-level system.[4,11] The dynamics of the isolated system is simply described by the pseudospin Hamiltonian $H_0 = -(\Delta_0\sigma_x + \epsilon\sigma_z)\hbar/2$. If we assume that the system starts out at time $t=0$ from one of the two states, say from $\sigma_z = +1$, then the evolution is described by $P(t) = \langle\sigma_z(t)\rangle$. In the absence of dissipation $P(t)$ shows oscillatory behavior, $P(t) = \epsilon^2/\Delta_b^2 + (\Delta_0^2/\Delta_b^2)\cos(\Delta_b t)$, where $\Delta_b = (\Delta_0^2+\epsilon^2)^{1/2}$. It has been shown by Kondo[1,5] that the screening cloud of the normal-state conduction electrons causes long-range interactions between tunneling transitions of the form $Q(t) = S(t) \pm i\pi K\,\mathrm{sgn}(t)$, where

$$S(t) = 2K \ln\left[\frac{\beta\hbar D}{\pi}\sinh\left(\frac{\pi t}{\beta\hbar}\right)\right]. \tag{1}$$

Here D is of the order of ω_0.[5] We note that an Ohmic heat bath set up by bosons leads to the same form of interaction as given in (1). The formally exact solution of

$P(t)$ in the form of a power series in Δ_0 is given by[4,6]

$$P(t)=\tfrac{1}{2}\sum_{n=0}^{\infty}(-\Delta_0^2)^n\left(\frac{\cos(\pi K)}{2}\right)^{n-1}\int_0^t dt_{2n}\int_0^{t_{2n}}dt_{2n-1}\cdots\int_0^{t_2}dt_1\sum_{\{\zeta_j=\pm 1\}}\cos\left(\zeta_1\pi K-\epsilon\sum_{j=1}^{n}\zeta_j(t_{2j}-t_{2j-1})\right)$$
$$\times\exp\left(-\sum_{j=1}^{n}S_{2j,2j-1}-\sum_{\substack{j,k=1\\ j>k}}^{n}\zeta_j\zeta_k\Lambda_{jk}\right). \quad (2)$$

Here we have arranged the tunneling transitions in pairs, usually called bounces. Each bounce may be viewed as a neutral pair of charges $q_\pm=\pm 1$, where the label $\zeta_j=\pm 1$ indicates whether the charge q_+ is preceded or succeeded by the charge q_-. The pair potential $S_{2j,2j-1}=S(t_{2j}-t_{2j-1})$ denotes the self-interaction of the bounce j and Λ_{jk} represents the set of interactions between the bounces j and k,

$$\Lambda_{jk}=S_{2j,2k-1}+S_{2j-1,2k}-S_{2j,2k}-S_{2j-1,2k-1}. \quad (3)$$

Finally, the sum over the label ζ_j in (2) runs over the 2^n possibilities of arranging n bounces of two different types.

For later convenience we decompose $P(t)$ as $P(t)=P_+(t)+P_-(t)$, where $P_+(t)$ and $P_-(t)$ denote the symmetric and antisymmetric contributions with respect to the bias $\hbar\epsilon$, respectively. In the limit $t\to\infty$, $P_+(t)$ drops to zero while $P_-(t)$ approaches the equilibrium value P_∞. We note that $P(t)$ is the relevant quantity in experiments on macroscopic quantum coherence.[4,6] The other dynamical quantity of interest is the symmetrized equilibrium correlation function $C(t)=[\langle\sigma_z(t)\sigma_z(0)\rangle+\langle\sigma_z(0)\sigma_z(t)\rangle]/2$, which is relevant in the neutron-scattering characteristics of the system. By comparing (2) with the corresponding exact path-integral expression for $C(t)$, we find the relation

$$C(t)=P_+(t)+P_\infty P_-(t). \quad (4)$$

Here we have neglected spin-bath correlations between the positive- and negative-time parts of the path (see Ref. 4, p. 35). While these correlations are relevant for the algebraic long-time tails of $C(t)$ for $T=0$, their effect on the neutron-scattering characteristics is negligibly small.[9] The inelastic part of the neutron-scattering function for a system tunneling between positions $\mathbf{d}/2$ and $-\mathbf{d}/2$ is related to the Laplace transform $\tilde{C}(\rho)$ of $C(t)$ by[6,12] $S(\mathbf{k},\omega)=\sin^2(\mathbf{k}\cdot\mathbf{d}/2)J(\omega)/\pi$, where $J(\omega)=2\,\mathrm{Re}\tilde{C}(\rho=i\omega)/[1+\exp(\beta\hbar\omega)]$ is the structure factor.

The true frequency scale of the problem is set by the effective tunnel splitting of a symmetric two-state system at zero temperature,[4]

$$\Delta=[\cos(\pi K)\Gamma(1-2K)]^{1/(2-2K)}\Delta_0(\Delta_0/D)^{K/(1-K)}. \quad (5)$$

It is convenient to introduce dimensionless frequencies $\lambda=\rho/\Delta$, $\nu=\omega/\Delta$, $\sigma=\epsilon/\Delta$, and temperature $\tau=k_BT/\hbar\Delta$. By extracting a thermal detailed-balance factor the dimensionless structure factor $j(\nu)$ is given by

$$J(\nu\Delta)=(2/\Delta)j(\nu)/(1+e^{\nu/\tau}). \quad (6)$$

Clearly, it is impossible to evaluate the complete series (2) for arbitrary K. We shall be able, however, to do so for weak damping $K\ll 1$ and also for $K=\frac{1}{2}$.

Let us first briefly consider the DBGA which is formally obtained by disregarding the interactions Λ_{jk} in (2). Then, the series (2) is in the form of a convolution and the Laplace transform $\tilde{p}(\lambda)=\Delta\tilde{P}(\rho=\lambda\Delta)$ can be summed to

$$\tilde{p}(\lambda)=\frac{1-\tan(\pi K)[g(\lambda_+)-g(\lambda_-)]/2i\lambda}{\lambda+[g(\lambda_+)+g(\lambda_-)]/2}, \quad (7)$$

$$g(\lambda)=(2\pi\tau)^{2K-1}\frac{\Gamma(K+\lambda/2\pi\tau)}{\Gamma(1-K+\lambda/2\pi\tau)}, \quad (8)$$

where $\lambda_\pm=\lambda\pm i\sigma$ and where $\Gamma(z)$ is Euler's gamma function. In the corresponding expression for $\tilde{c}(\lambda)=\Delta\tilde{C}(\rho=\lambda\Delta)$ the asymmetric contribution in σ of (7) is multiplied by P_∞, where P_∞ is the limiting value of $\lambda\tilde{p}(\lambda)$ as $\lambda\to 0$. Further, the structure factor $j(\nu)$ is given by[13]

$$j(\nu)=\mathrm{Re}\tilde{c}(\lambda=i\nu). \quad (9)$$

It follows from (7) and (8) that $P(t)$ approaches the equilibrium value $P_\infty=\tanh(\sigma/2\tau)$, regardless of the parameter K. Hence the DBGA predicts a symmetry breaking as $\tau\to 0$ for arbitrarily small K and arbitrarily small bias. This result is in marked contrast with the expectation based upon a standard quantum-mechanical analysis[14] that for $K\ll 1$

$$P_\infty=(\sigma/\sigma_b)\tanh(\sigma_b/2\tau), \quad (10)$$

where $\sigma_b=(1+\sigma^2)^{1/2}$. From this we may conclude that for a biased system with $K\ll 1$ the bounces actually form a dilute gas only for temperatures where the above results for P_∞ concur with $P_\infty=\sigma/2\tau$. This is the case in the region $\tau\gtrsim\tau_0$ where $\tau_0=\sigma_b$. Another indication for the breakdown of the DBGA below τ_0 is that $j(\nu)$ becomes negative within the DBGA around $\nu=0$ for $\tau<\tau_c$ where $\tau_c\simeq\tau_0$. This clearly is an unphysical result.

We now examine the self-consistency of the DBGA for $K\ll 1$. It is obvious that for fairly high temperatures (1) may be approximated by

$$S(t)=2K\Delta\pi\tau\ln(D/2\pi\tau\Delta)t. \quad (11)$$

With this choice the interbounce interactions Λ_{jk} defined in (3) cancel out, and $\tilde{p}(\lambda)$ is found from (1) exactly in the form (7) with

$$g(\lambda)=\delta^2/(\lambda+2\pi\tau K)\,, \tag{12}$$

where $\delta^2=(2\pi\tau)^{2K}/\Gamma(1-2K)$. On comparing (12) with (8) we see that the condition for the self-consistency of the DBGA is $2\pi\tau\gg|\lambda_\pm|$, which in the relevant frequency range indeed corresponds to the temperature restriction $\tau\gtrsim\tau_0$. With use of (4), (7), and (12) we then find in the parameter region $K\ll 1$, $\tau\gtrsim\tau_0$, and arbitrary bias, the solution

$$\tilde{c}(\lambda)=\frac{1}{\lambda}\left[1-\frac{\delta^2\lambda+(1-\sigma^2/4\tau^2)w}{\lambda^3+u\lambda^2+v\lambda+w}\right], \tag{13}$$

where $u=4\pi\tau K$, $v=u^2/4+\sigma^2+\delta^2$, and $w=2\pi\tau\delta^2K$. Decomposing (9) with (13) into partial fractions the spectral function $j(\nu)$ is obtained in the form

$$j(\nu)=\frac{1}{2}\frac{A_1\gamma+(\nu_0-\nu)A_2}{(\nu-\nu_0)^2+\gamma^2}+\frac{1}{2}\frac{A_1\gamma+(\nu_0+\nu)A_2}{(\nu+\nu_0)^2+\gamma^2}+\frac{A_3\gamma_r}{\nu^2+\gamma_r^2}\,, \tag{14}$$

where $\lambda=-\gamma_r$ and $\lambda=-\gamma\pm i\nu_0$ are the real and complex roots of the cubic pole condition in (13), respectively. For $\tau_0\lesssim\tau\lesssim\tau_1$, where $\tau_1=1/2\pi K$, we find from the cubic equation the approximate solutions

$$\begin{aligned}&\nu_0^2=\sigma^2+\delta^2-(\pi K\delta\tau)^2(\delta^4+4\sigma^2)/(\sigma^2+\delta^2)^2\,,\\&\gamma=\pi K\tau(2\sigma^2+\delta^2)/(\sigma^2+\delta^2)\,,\\&\gamma_r=2\pi K\tau\delta^2/(\sigma^2+\delta^2)\,.\end{aligned} \tag{15}$$

Further, the amplitude factors in (14) are given by

$$\begin{aligned}&A_1=1-\sigma^2/\nu_0^2,\quad A_2=A_1(\gamma-\gamma_r)/\nu_0\,,\\&A_3=\sigma^2/\nu_0^2-P_\infty^2\,,\end{aligned} \tag{16}$$

where $P_\infty=\sigma/2\tau$. Thus we find that the structure factor has two narrow resonances of width γ around $\nu=\pm\nu_0$ describing inelastic scattering and a third resonance of width γ_r around $\nu=0$ describing quasielastic scattering. The relative intensity of the inelastic peaks of $J(\omega)$ is governed by the detailed-balance factor in (6). Corresponding, $P(t)$ and $C(t)$ show damped oscillations with frequency ν_0 and damping rate γ. In addition, there is a contribution describing relaxation with a rate γ_r into the equilibrium values $P(t\to\infty)=P_\infty$ and $C(t\to\infty)=P_\infty^2$, respectively. As the temperature is increased from $\tau\simeq\tau_0$ to $\tau\simeq\tau_1$, ν_0 initially increases and then drops down again, while γ and γ_r increase linearly with temperature. Near the temperature τ_1 the three peaks of $j(\nu)$ merge into a single broad quasielastic peak. Now, it is essential to insert into (14), instead of (15), the exact solutions of the cubic pole condition. So it is more convenient to compute $j(\nu)$ from the original expression (9) using (13). Only for high temperatures $\tau\gg\tau_1$ does this expression simplify to $j(\nu)=\gamma_r/(\nu^2+\gamma_r^2)$, where γ_r is the tunneling rate

$$\gamma_r=K(2\pi\tau)^{2K+1}/[(2\pi\tau K)^2+\sigma^2]\,.$$

As the temperature is increased further the quasielastic peak narrows according to the power law τ^{2K-1}. This anomalous behavior has been observed by neutron spectroscopy for hydrogen jump rates in $Nb(OH)_x$.[8]

In the low-temperature region $\tau<\tau_0$ the approximation (11) for $S(t)$ is not valid. We should expect, therefore, that it is inconsistent to drop the interactions Λ_{jk}, and this is indeed what we find for a biased system. Now, for weak coupling the systematic treatment is to keep the interactions Λ_{jk} of linear order in K in the self-energy of the system. We then find from (2)

$$\tilde{p}(\lambda)=\frac{1+\pi K(\sigma/\lambda)/(\lambda^2+\sigma^2)}{\lambda+\lambda/(\lambda^2+\sigma^2)+k(\lambda)}\,, \tag{17}$$

where $k(\lambda)=k_0(\lambda)+k_1(\lambda)$ is the self-energy correction of linear order in K. Here, $k_0(\lambda)$ is the contribution kept in the DBGA,

$$k_0(\lambda)=-\Delta\int_0^\infty dt\,e^{-\lambda\Delta t}\cos(\epsilon t)S(t)\,, \tag{18}$$

while $k_1(\lambda)$ describes the sum over all irreducible multibounce clusters. These are configurations in which the first bounce interacts with the last one. After performing the ζ_j summation in (2) we find

$$k_1(\lambda)=-\sum_{n=2}^{\infty}(-1)^n\Delta^{2n-1}\prod_{j=1}^{n}\int_0^\infty ds_j\,e^{-\lambda\Delta s_j}\prod_{i=1}^{n-1}\int_0^\infty dr_i\,e^{-\lambda\Delta r_i}\Lambda_{n1}\sin(\epsilon s_n)\sin(\epsilon s_1)\prod_{k=2}^{n-1}\cos(\epsilon s_k)\,, \tag{19}$$

where $s_j=t_{2j}-t_{2j-1}$ is the length of the bounce j and where $r_j=t_{2j+1}-t_{2j}$ is the interval between the bounces j and $j+1$. Note that $k_1(\lambda)$ vanishes for a symmetrical system, as already mentioned before, while it becomes relevant for a biased system in the region $\tau\lesssim\tau_0$. Use of the dilute-gas approximation for these clusters gives the result (17) with (18) and (19). Now, it is consistent to make this approximation, since the effect of the neglected interactions is of order K^2 only. Next, it is important that the expressions $k_0(\lambda)$ and $k_1(\lambda)$ can be evaluated exactly for the pair potential (1). With the use of a spectral decomposition of (1) and with changes of the orders of intergration we finally obtain

$$\begin{aligned}&k(\lambda)=\frac{K\{2\lambda^3 s(\lambda)+\sigma^2\bar{\lambda}_+\bar{\lambda}_-[\bar{\lambda}_+ s(\bar{\lambda}_-)+\bar{\lambda}_- s(\bar{\lambda}_+)]\}}{(\lambda+\lambda-\sigma_b)^2}\,,\\&s(z)=\psi(1+z/2\pi\tau)-\pi\tau/z+\ln(2\pi\tau)\,,\end{aligned} \tag{20}$$

where $\lambda_\pm = \lambda \pm i\sigma$ and $\bar{\lambda}_\pm = \lambda \pm i\sigma_b$, and where $\psi(z)$ is the digamma function. The formula (17) with (20) gives the complete solution to the dynamics of the biased system in the weak-coupling case at low temperatures. We note that (17) yields the correct expression (10) for P_∞. Further, $\tilde{p}(\lambda)$ possesses simple poles at $\lambda = -\gamma_r$ and $\lambda = -\gamma \pm i\nu_0$, where

$$\nu_0^2 = \sigma_b^2 + 2K[\mathrm{Re}\psi(1 + i\sigma_b/2\pi\tau) + \ln(2\pi\tau)],$$
$$\gamma_r = \pi K \coth(\sigma_b/2\tau)/\sigma_b, \qquad (21)$$
$$\gamma = \gamma_r/2 + 2\pi K\sigma^2\tau/\sigma_b^2.$$

These formulas give the correct solution for $K \ll 1$ in the region $\tau \lesssim \tau_0$. The structure factor is found again in the form (14) with amplitude factors $A_1 = 1/\sigma_b^2$, $A_2 = A_1(\gamma - \gamma_r)/\sigma_b$, and $A_3 = [\sigma/\sigma_b \cosh(\sigma_b/2\tau)]^2$. Note that (21), and also $j(\nu)$, smoothly matches with the above DBGA results near $\tau = \tau_0$. Now, in contrast to the DBGA result, the amplitude of the quasielastic peak is positive for all τ, and it vanishes for $\tau = 0$.

For the special value $K = \frac{1}{2}$ the spectral properties of a biased two-state system can be determined exactly for all temperatures. The crucial point now is that for $K \to \frac{1}{2}$ all $\cos(\pi K)$ factors in (2) need to be compensated by divergent factors $\Gamma(1-2K)$ arising from the short-distance behavior of the attractive interactions in (2). It is found that in the expression for $p_+(\lambda)$ the bounce lengths shrink to zero in the limit $K \to \frac{1}{2}$, so that the interactions Λ_{jk} cancel out exactly, yielding $p_+(\lambda) = 1/(1+\lambda)$.[4] The expression for $p_-(\lambda)$ has one $\cos(\pi K)$ factor less. The rigorous analysis shows that now the first bounce is substituted by an irreducible multibounce cluster, which again can be evaluated exactly. In the end we find that exact result

$$\tilde{p}(\lambda) = \frac{1}{1+\lambda}\left\{1 + \frac{1}{i\pi\lambda}[h(\lambda_+) - h(\lambda_-)]\right\}, \qquad (22)$$

$$h(z) = \psi(1/2 + 1/4\pi\tau + z/2\pi\tau), \qquad (23)$$

which enables us to compute $P(t)$ and especially $P_\infty = (2/\pi)\mathrm{Im}h(i\sigma)$. We remark that the DBGA also gives $\tilde{p}(\lambda)$ in the form (22) where, however, the term $1/4\pi\tau$ in the argument of $h(z)$ is omitted. Readily the structure factor is obtained as $j(\nu) = \mathrm{Re}\{\tilde{p}_+(\lambda = i\nu) + P_\infty \tilde{p}_-(\lambda = i\nu)\}$. This formula describes a broad non-Lorentzian line shape. Again one finds that $j(\nu)$ is positive for all temperatures while the corresponding DBGA result becomes negative around $\nu = 0$ for sufficiently low temperatures.

In conclusion, a major result of this work is that the structure factor shows a quasielastic peak in addition to the inelastic peaks in the region of weak coupling, nonzero bias, and low temperatures $\tau \ll \tau_1$.

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[13]The DBGA formula for $j(\nu)$ was also found by S. Dattagupta, H. Grabert, and R. Jung (to be published) using quantum relaxation theory and calculating the self-energy in second order in the tunnel splitting.

[14]We note that P_∞ is exactly related to the partition function Z through $P_\infty = 2\tau\partial \ln Z/\partial\sigma$. By substituting $Z = \cosh(\sigma_b/2\tau)$, we find the formula (10).