Dissipative Quantum Tunneling of a Single Microscopic Defect in a Mesoscopic Metal

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The transition rates of individual two-state systems in a mesoscopic disordered metal (Bi) are studied via time-dependent conductance fluctuations. A striking increase in the transition rates is observed as the temperature T is decreased below 1 K for defects with energy splitting $\varepsilon < k_B T$. In contrast, the transition rates decrease monotonically as T is lowered if $\varepsilon > k_B T$. We show that these phenomena are a manifestation of dissipative quantum tunneling and estimate, for one particular defect, the defect-conduction-electron coupling parameter, $\alpha \approx 0.24$.

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Understanding the dynamics of a two-state system is crucial for describing light particle diffusion in metals [1], glasses at low temperature [2], chemical reaction rate theory [3], low-frequency ($1/f$) noise in metals [4,5], electromigration [6], and collective macroscopic tunneling [7]. We report the first detailed experimental study of the tunneling dynamics of an *individual*, microscopic two-state defect in a mesoscopic metal as a function of temperature and defect energy. Transitions between low-lying states of the system can be induced by thermally activated (over-the-barrier) processes, but our measurements are done at temperatures low enough ($T \lesssim 1$ K) compared to the barrier height that quantum-mechanical (through-the-barrier) tunneling is the dominant process. Our samples are small enough that at these temperatures the interactions of the defect are exclusively with the thermal reservoir of conduction electrons and phonons; i.e., defect-defect interactions can be neglected.

Our experimental probe is the low-frequency conductance. The motion of a single defect can be resolved experimentally because in mesoscopic metals (particularly of reduced dimensionality), the spatial fluctuation of a single-scattering defect over a distance $\sim k_F^{-1}$ is reflected in a greatly enhanced universal conductance fluctuation (UCF) $\lesssim e^2/h$ due to multiple electron interference [5,8,9]. We exploit this sensitivity of electronic properties to individual atomic coordinates to study the properties of the scattering centers themselves. The ability to examine the transition rates as a function of defect energy splitting is facilitated by the recent discovery of magnetic-field tuning of a two-state system in a mesoscopic metal [10].

Our most striking result is the observation of a tunneling rate that increases as the temperature decreases below 1 K. Furthermore, application of a moderate magnetic field can, in specific cases, suppress this behavior. We show that these effects are fully consistent with a model of incoherent tunneling of a particle between two positional eigenstates. By comparison to theories of dissipative tunneling for Ohmically damped two-state systems we extract, for one particular defect, an estimate of the defect-conduction-electron coupling parameter $\alpha \approx 0.24$.

The experiments were carried out on 30-nm evaporated polycrystalline films of Bi fabricated into five-terminal bridges with lateral dimensions between 60 and 300 nm. We measure the conductance change δG as a function of time; typical magnitudes are $\delta G \lesssim 10^{-1} e^2/h$ at 0.5 K. Our receiver bandwidth is 10^{-3}–20 Hz (see Ref. [10] for more details). In the data described here, δG was always bistable, with only two discrete values separated by an amount greater than all other noise sources. Thus it was always possible to assign the conductance to one of two states of a microscopic defect in one arm of the bridge. We obtained the mean times in the upper and lower states by recording several hundred transitions per datum and fitting the resulting histogram of lifetimes $P(t)$ with an exponential decay function, $P(t) \propto e^{-\gamma_i t}$. We denote the reciprocal lifetimes in the excited (fast) or ground (slow) states by γ_f and γ_s, respectively.

Figure 1 shows how the transition rates for the same two-state defect vary with temperature. The different behavior in the two panels demonstrates the dramatic role of an external magnetic field, applied perpendicular to the plane of the substrate. At low field for $T > 1$ K, both $d\gamma_f/dT$ and $d\gamma_s/dT$ are positive, which we interpret as arising from thermal activation. At $H = 0.5$ T, both the fast and slow rates pass through a minimum at 1 K, and then *increase substantially as T decreases further.* At $H = 4.4$ T, both $d\gamma_f/dT$ and $d\gamma_s/dT$ are positive for all T; i.e., there is no anomalous speeding up. At 0.5 K γ_s is nearly an order of magnitude smaller than γ_f. We have also measured how the rates depend on magnetic field at the fixed temperature $T = 0.46$ K. Between 0 and 9 T the fast rate varies by less than a factor of 2; over this same field range E changes by over an order of magnitude [10].

We first emphasize that a thermally activated process leads to rates γ_f and γ_s which both decrease exponentially as T decreases. Therefore, our results show that a tunneling process must control the rates in the $T < 1$ K region. To explain the difference between the results shown

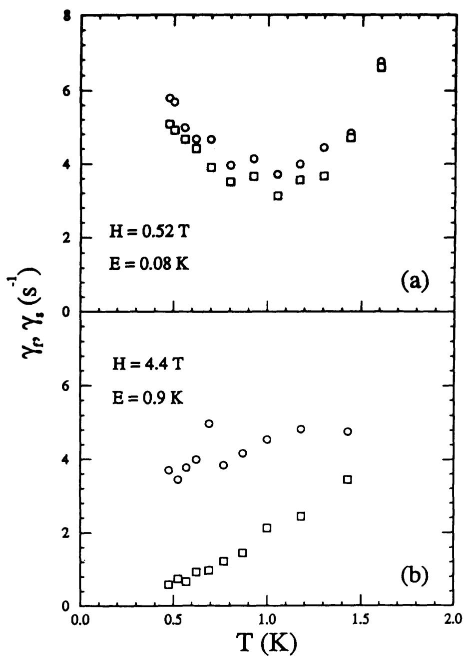

FIG. 1. Transition rates γ_i of a single bistable microscopic defect in Bi as a function of temperature at two different magnetic fields. The fast rate γ_f and the slow rate γ_s of transitions of a two-state system are determined by thermally activated processes above 1 K. Below 1 K, quantum-mechanical tunneling determines the rates. (a) For $H=0.5$ T, corresponding to an energy splitting of 0.08 K, the rates show an anomalous increase below 1 K. (b) When the defect energy is tuned to 0.9 K by a 4.4-T magnetic field, $d\gamma_i/dT>0$ for both rates at all temperatures. This indicates that the tunneling process is sensitive to the defect energy.

in Figs. 1(a) and 1(b), we note that the magnetic field changes the energy splitting of the defect, E. Since to make a transition from the lower to upper state the defect must obtain energy E from the bath, when $E/k_BT\lesssim 1$ it is reasonable that lowering the temperature suppresses stimulated transitions to the excited state. Detailed balance for a two-state system relates E to the ratio of the transition rates by $e^{E/k_BT}=\gamma_f/\gamma_s$. Therefore, measurement of γ_i vs T in various magnetic fields allows one to obtain $E(H)$ [10]. The data in Fig. 1 yield splittings of 0.08 ± 0.02 and 0.9 ± 0.05 K at fields of 0.52 and 4.37 T, respectively.

Within the simplest two-state model, the temperature dependence of the transition rate γ_f from the excited state to the ground state is determined by the sum of spontaneous and stimulated decay. The former is independent of T, and the latter decreases as T is lowered. Thus, within this model $d\gamma_f/dT>0$ at all T. Though Fig. 1(b) is consistent with this picture, it cannot account for the anomalous upturn observed in Fig. 1(a).

We now show that our results are all consistent with the theory of quantum tunneling of a single two-state system coupled to conduction electrons [1,7]. As is standard, we assume that a charge-density coupling exists between conduction electrons and the two-state system [8,9,11] so that each of the two conductance values corresponds to the particle being localized in a particular well [1,7]. The Hamiltonian of the system (neglecting explicit phonon couplings) is

$$H=\frac{1}{2}\varepsilon\sigma_z-\frac{1}{2}\hbar\Delta\sigma_x+\sigma_z\sum_{kk'\eta}V_{kk'}c^{\dagger}_{k\eta}c_{k'\eta}+H_e\,, \qquad (1)$$

where ε is the asymmetry energy, Δ is the tunneling matrix element, and the σ_i are Pauli matrices. The third term describes the interaction between the electron bath and the defect [12]; $V_{kk'}$ describes the scattering potential, and $c^{\dagger}_{k\eta}$ creates a fermion of wave vector k, energy ξ_k, and spin η. The bath itself is described by H_e; for noninteracting fermions, $H_e=\sum_{k\eta}\xi_k c^{\dagger}_{k\eta}c_{k\eta}$. The coupling of the defect to the bath is characterized quite generally in terms of a parameter α which describes the coupling strength as well as the spectral density of bath excitations [1,7,13]. For an s-wave potential ($V_{kk'}=V$) and a bath of free fermions, α is determined solely by the scattering phase shift for electrons at the Fermi surface [14,15]; in the limit of weak scattering, $\alpha=\frac{1}{2}(n_0V)^2$, where n_0 is the density of electron states at the Fermi level [1,14,15]. For $0<\alpha<\frac{1}{2}$, the region relevant for metals [14,15], the model exhibits two general regimes, depending on the relative magnitudes of T, ε, and Δ [1,7,16]. The relevant parameter is the renormalized tunneling matrix element Δ_r, related to Δ by $\Delta_r=\Delta(\Delta/\omega_c)^{\alpha/(1-\alpha)}$, where ω_c is the bath cutoff frequency [17]. If $\hbar\Delta_r$ is greater than both ε and αT, the effects of electron coupling are relatively weak and can be treated as a small perturbation on the dynamics of the defect [18]. In this regime, coherent oscillations in the time evolution of the particle are expected and transitions can be induced between energy eigenstates of the tunneling system [19,20]. If, on the other hand, either ε or αT is much greater than $\hbar\Delta_r$, then the effective coupling of defect and electrons is so strong that it is appropriate to treat the tunneling matrix element as a small perturbation. Here, tunneling is incoherent since the rapid fluctuations of the bath act to dephase the tunneling particle. The fast transition rate has the form [21]

$$\gamma_f=\frac{1}{2}\Delta_r^{2(1-\alpha)}\left[\frac{2\pi k_BT}{\hbar}\right]^{2\alpha-1}\frac{e^{\varepsilon/2k_BT}}{\Gamma(2\alpha)}\left|\Gamma(\alpha+i\varepsilon/2\pi k_BT)\right|^2, \qquad (2)$$

where Γ is the (complex) gamma function. When $k_BT/\varepsilon\gg1$, $\gamma_f\sim T^{2\alpha-1}$, so that the transition rates increase as T decreases (since $\alpha<\frac{1}{2}$). When $k_BT/\varepsilon\ll1$, γ_f decreases as T decreases.

We now examine the experimental results in Fig. 1(a) in the context of this model. For the small ε case in Fig. 1(a), the rate minima occur at 1 K, for which $k_BT/\varepsilon\approx10$. Although the temperature range is limited, we can nonetheless estimate the power-law exponent $\gamma_f \sim T^{-\zeta}$ with $\zeta \approx 0.5$, so that $\alpha \approx 0.25$. Evaluating Eq. (2) with $\alpha = 0.25$, we find $\hbar\Delta_r \sim 4\times10^{-7}$ K. Thus the parameters are completely consistent with the asymmetric (biased) dissipative tunneling model, i.e., $\hbar\Delta_r \ll \varepsilon, \alpha T$ and $0 < \alpha < \frac{1}{2}$. In addition, the observed exponential distribution of two-state occupancy times is consistent with the picture of incoherent tunneling, where rapid fluctuations of the bath interrupt the particle's oscillatory wave function and destroy its phase coherence, so that the probability of a transition is independent of the system's previous history.

The theory can be used to understand the effects of changing either the magnetic field or the temperature. Inspection of Eq. (2) shows that $T^{1-2\alpha}\gamma_f$ depends on the ratio ε/k_BT but not on ε and k_BT separately. If one assumes that as H is varied $\varepsilon(H)$ changes but α and Δ do not, then one can define $y \equiv k_BT/\varepsilon$ and write $T^{1-2\alpha}\gamma_f = f_{\alpha,\Delta}(y)$, where f is a scaling function defined by Eq. (2). In Fig. 2, we plot $T^{1-2\alpha}\gamma_f$ vs k_BT/ε for five data sets obtained with one defect, two of which are shown in Fig. 1. There is one magnetic-field sweep at the fixed temperature $T = 0.46$ K, and four temperature sweeps at different values of the magnetic field. The solid line is $f(y)$ for $\alpha = 0.24$ and $\hbar\Delta_r = 4.4\times10^{-7}$ K. One sees that Eq. (2) is fully consistent with the decay rates of all the data sets over a substantial region of y, $0.5 < y < 10$.

The form of the scaling function explains the differences between Figs. 1(a) and 1(b). The data of Fig. 1(a) correspond to large values of $y = k_BT/\varepsilon$, where the scaling function tends to a constant and $\gamma_f \propto T^{-(1-2\alpha)}$. In contrast, the data of Fig. 1(b) have $y \lesssim 1$, where the scaling function increases sharply enough as a function of y that $d\gamma_f/dT > 0$.

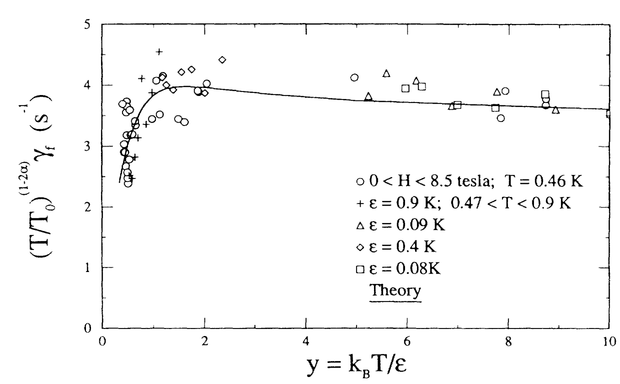

FIG. 2. Plot of the scaled fast transition rate $(T/T_0)^{1-2\alpha}\gamma_f$ vs $y \equiv k_BT/\varepsilon$, where $T_0 = 1$ K. The solid line is the prediction of the dissipative tunneling model with the conduction-electron–defect coupling parameter $\alpha = 0.24$. The circles correspond to a field sweep at $T = 0.46$ K; the other data points correspond to temperature sweeps for four different energy splittings of the same defect; plusses, 0.9 K; diamonds, 0.4 K; triangles, 0.09 K; squares, 0.08 K.

There are some caveats that should be considered in discussing Fig. 2. The theoretical scaling function depends very weakly on y at $y \gtrsim 2$ and the scatter in the data is large enough that we cannot claim to verify its detailed form. The calculation (solid line) in Fig. 2 is not a fit to the data but represents our best estimate, with uncertainty in α of about ± 0.05. The uncertainties in Δ_r and α are correlated; for a given α, the uncertainty in Δ_r is $\sim 10\%$.

Theoretically, we have assumed that varying the magnetic field H changes only ε. The parameter α which describes the coupling between the defect and the electrons depends on the spectral density of the bath and the local charge density in the vicinity of the defect, both of which could be affected by a magnetic field. If α were to depend on H, it would change the renormalized matrix element Δ_r, even if the bare tunneling matrix element Δ is H independent. Previous observations of random H-dependent defect energies [10] were consistent with charge-density fluctuations at the defect arising from electron interference in the mesoscopic sample [11]. Thus one might expect changes in the effective scattering strength as H varies. However, it is reasonable that the apparent changes in α are small because bath excitations of all energy scales up to the upper cutoff $\hbar\omega_c$ contribute to the effects on tunneling whereas the magnetic field affects low-energy processes only. Figure 2 indeed indicates that, within the uncertainties, α (and Δ_r) are H independent.

We do not suggest that the same value of α applies to all defects. In fact, application of Eq. (2) to other defects shows that quite different values of α and Δ_r are necessary to describe the data. This is not surprising since the scattering potential $V_{kk'}$ and bare tunneling matrix element Δ both depend on the specific tunneling configuration.

It is useful to place our results in the context of previous experiments on dissipative tunneling. Earlier work using different methods and systems has clearly observed enhanced diffusion rates at relatively low temperatures for light particles such as positive muons in the metals Cu, Al, and Sc [22–24]. In Nb with H interstitials trapped at O impurities, neutron diffraction has been able to measure both tunnel splittings [25] and inelastic linewidths [26], with the latter used to infer enhanced hopping rates. In addition, macroscopic quantum tunneling in SQUIDs has been observed in a strongly dissipative regime [27].

We have argued that our present experimental technique maps the conductance onto positional eigenstates of a bistable tunneling defect. The transitions are measured in real time at extremely low frequencies. Thus this method is similar to the time-domain method employed in superconducting macroscopic tunneling studies [27]. However, the fluctuating entity here is a *microscopic* object, detected as a result of multiple electron interference in a normal metal. In contrast with studies of light interstitials, we have not intentionally introduced tunneling impurities into our samples. We believe, although cannot prove, that the tunneling particles have mass $\sim m_{Bi}$. One argument to support this view is that the number of active defects appearing in our sample volume and bandwidth is consistent with the number of intrinsic tunneling systems expected in a highly disordered, or glassy, metal [2]. The number of defects in our samples is also in general agreement with the amplitude of previously measured $1/f$ noise in the UCF regime for evaporated Bi samples [28]. Finally, several individual defects have been studied in different evaporations [10]; all show similar behavior.

In summary, we have measured the time evolution of a single two-state system in a mesoscopic metal. The tunneling rates either decrease or increase as the temperature T is lowered, depending on the relative sizes of the energy asymmetry ε and $k_B T$, in agreement with dissipative tunneling theory. Changing the magnetic field H changes ε but not, within the uncertainties, the coupling between the defect and the electron bath or the tunneling matrix element. Our ability to tune the energy ε has therefore allowed a detailed test of the dissipative tunneling model for a microscopic system.

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