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Dissipative quantum tunneling of a single defect in Bi

Kookjin Chun and Norman O. Birge
Department of Physics and Astronomy and Center for Fundamental Materials Research, Michigan State University, East Lansing, Michigan 48824
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We have studied the dynamics of single bistable defects in submicrometer Bi wires at temperatures 0.1–2 K. The defect motions cause measurable changes in the sample resistance via universal conductance fluctuations. The temperature dependence of the transition rates agrees quantitatively with the predictions of dissipative quantum tunneling theory, which describes tunneling of a defect in a double-well potential with strong dissipation from the electron bath. We report detailed measurements of a single defect as a function of magnetic field, and discuss the field dependence of the defect-potential asymmetry ε, the defect-bath coupling constant α, and the renormalized tunneling matrix element Δ_r.

The quantum-mechanical problem of a particle tunneling in a double-well potential is of great theoretical[1] and experimental interest.[2] Interactions of the tunneling system with a dissipative environment can have a striking effect on the tunneling dynamics. The most interesting case is that of Ohmic dissipation, which occurs when an atom tunnels in a metal in the presence of conduction electrons. In this case the tunneling rate can *increase* with decreasing temperature, due to the nonadiabatic nature of the low-energy electron-hole excitations.[3]

Dissipative quantum tunneling has been observed in a variety of physical systems. A particularly striking example is the observation of a *single* defect tunneling between metastable states in a disordered metal, reported recently by Golding, Zimmerman, and Coppersmith.[4] These authors studied the defect motion by measuring discrete conductance jumps of a submicrometer bismuth wire. Because the sample dimensions were comparable to the phase-breaking length for quantum transport at low temperatures, the sample's conductance was highly sensitive to the positions of the scattering centers.[5] Golding, Zimmerman, and Coppersmith found that the defect dynamics were consistent with predictions of dissipative quantum tunneling theory.[6] In particular, the tunneling rates increase with decreasing temperature when $kT>\varepsilon$, where ε is the energy asymmetry of the defect double-well potential. When $kT<\varepsilon$, the tunneling rates follow a simple picture of stimulated absorption and spontaneous emission; the faster rate is roughly temperature independent and the slower rate decreases as $e^{-\varepsilon/kT}$. They also observed that ε for a single defect varies with magnetic field,[7] which had been predicted[8] based on local fluctuations in the electron density that occur in disordered metals.

The work described above left several open questions. First, the data presented for fixed magnetic field were either in the regime $kT<\varepsilon$ or $kT>\varepsilon$; there were no data showing the temperature crossover from one regime to the other.[4] Second, the magnetic-field dependence of ε was not completely understood; in addition to the random variation with field predicted by theory,[8] there was an overall trend of increasing ε with field for the defects studied.[7] Third, data taken at different temperatures and magnetic fields were consistent with the hypothesis that the defect-electron bath coupling constant α and the renormalized tunneling matrix element Δ_r are independent of field.[4] Although there are theoretical grounds for this hypothesis, it was not tested quantitatively in the experiments.

We have also studied the dynamics of individual defects in submicrometer Bi wires. Bi was chosen because of its high resistivity and correspondingly large resistance fluctuations due to quantum interference effects.[9] We report extensive measurements of a single defect as a function of temperature (0.1–2 K) and magnetic field (0–7 T). The advantage we have over the earlier work[4] is our extension of the lower limit of the temperature range from about 0.5 to 0.1 K, which allows us to make a more thorough comparison between experiment and theory and address the questions listed above. We find that the temperature dependence of the defect tunneling rates agrees quantitatively with the predictions of dissipative quantum tunneling theory over the entire temperature range from $kT \ll \varepsilon$ to $kT \gg \varepsilon$. We also observe a variation of ε with magnetic field, but the minimum value of ε for this particular defect occurs for $B\neq 0$. The question of whether α and Δ_r vary with field is more difficult to address quantitatively. Although the theoretical fits to our data are excellent, they depend only weakly on α over the experimental temperature range. A scaling plot shows that our data are consistent with the hypothesis that α and Δ_r are independent of B for this defect.

Our sample is a thermally evaporated Bi wire 20 nm thick, patterned using electron beam lithography into a five-terminal device for use in a Wheatstone bridge. Each arm of the sample has a linewidth of 0.1 μm, a length of 0.5 μm, and a resistance of about 1 kΩ. At 4.2 K and below, the resistance of a Bi wire such as this often exhibits spontaneous switching between two distinct values, due to the motion of a bistable defect in one arm of the sample.[10] The magnitude of the jumps is a random function of applied magnetic field,[7] as expected from universal conductance fluctuation theory.[11] In this wire, the largest jumps were of order $0.2e^2/h$ at 1 K and $1e^2/h$ at 0.1

K, the increase at lower temperature due to the increasing length scales for quantum transport.[9] The data presented here were obtained by adjusting the magnetic field to achieve the maximum signal, then measuring the transition rates of the defect as a function of temperature. At each temperature, several hundred transitions were recorded and analyzed.

The experimental temperature range was determined by two factors. Above about 2 K several defects were active, so it was difficult to determine unambiguously the tunneling rates of a particular defect. Below 0.1 K the signal-to-noise ratio was very small. Although the resistance change due to the motion of a single defect increases as the temperature is lowered, the drive current must be decreased even faster to avoid sample heating. We used a drive current of 0.7 nA at 0.1 K.

Raw data of resistance versus time were analyzed using two independent methods. First, the individual jumps are found using a comparator with hysteresis (Schmitt trigger). The dwell times in each state are histogrammed and fit to a single exponential to find the mean dwell time, τ, and the transition rate to the other state, $\gamma=\tau^{-1}$. We call the fast and slow transition rates γ_f and γ_s. The histograms show that the transitions are independent random events characterized by a single rate for each state. The second analysis method is based on fitting a Debye-Lorentzian line shape to the power spectrum,[12] which determines the sum of the two rates, $\gamma=\gamma_f+\gamma_s$. The ratio of the rates is then obtained by fitting a histogram of the raw conductance values to the sum of two Gaussians and comparing the areas. The two analysis methods yield consistent values of γ_f and γ_s.

Figure 1 shows γ_f and γ_s versus temperature for four different values of the magnetic field. We first observe that the rates obey the principle of detailed balance, i.e., $\gamma_f/\gamma_s=e^{\varepsilon/kT}$, for each value of the field. Figure 2 shows $\ln(\gamma_f/\gamma_s)$ versus $1/T$ for the four data sets, along with linear fits through the origin. The slopes of these lines give values for ε/k of 402, 213, 90, and 40 mK, respectively, with a standard deviation of ± 10 mK. Since we will later show that these four data sets come from the same defect, Fig. 2 illustrates the random variation of ε with B observed by Zimmerman, Golding, and Haemmerle.[7] We emphasize that the value $\varepsilon/k=213$ mK near zero field is greater than the values of 40 and 92 mK at $B=2.274$ and 2.286 T; thus the systematic increase of ε at low fields observed for the three defects in Ref. 7 is not universal. We also note the rapid variation of ε with B, illustrated by its change from 40 to 90 mK when the field changed from 2.274 to 2.286 T.

Next we discuss the temperature dependence of the rates. Figure 1(b), with $\varepsilon/k=213$ mK, most clearly illustrates three distinct temperature regimes. When $kT<\varepsilon$, γ_f is roughly temperature independent, while γ_s decreases rapidly with decreasing temperature. As the temperature is raised, the rates cross over to a qualitatively different regime. The ratio of the two rates still obeys detailed balance, but both rates *decrease* with increasing temperature, following a power law,[3] $T^{2\alpha-1}$. This behavior, shown more vividly in Figs. 1(c) and 1(d), is an essential feature of dissipative tunneling in metals. Above 1.2 K, the rates increase rapidly, due first to phonon-assisted tunneling, and eventually to thermal activation over the barrier.

We limit quantitative analysis to the temperature range below 1.2 K, i.e., below the onset of phonon-assisted tunneling. The temperature dependence of the total tunneling rate, $\gamma=\gamma_f+\gamma_s$, has been calculated in terms of the tunneling matrix element Δ_r, the asymmetry ε, and the coupling constant α. For the case of incoherent tunnel-

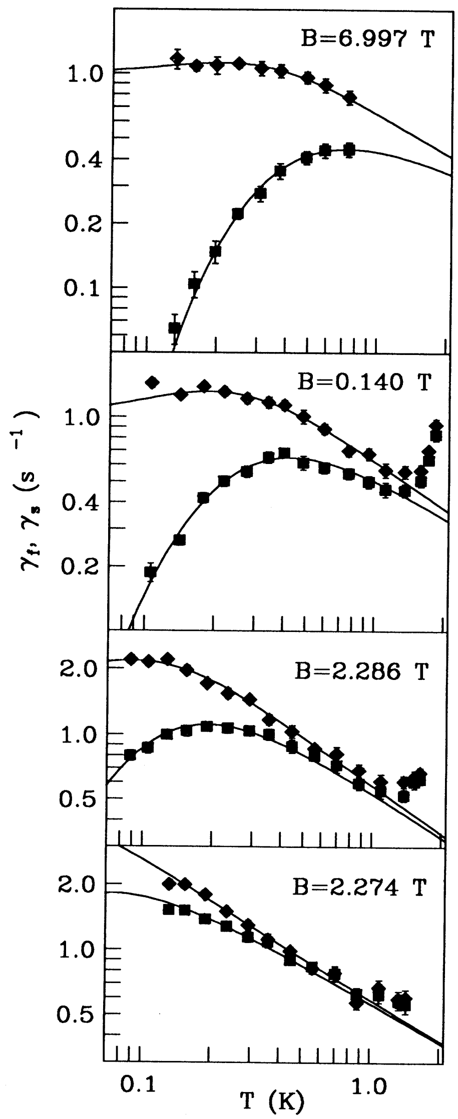

FIG. 1. Fast and slow transition rates vs temperature, for four values of the applied magnetic field. Solid lines are fits of Eq. (1) to the data for $T<1.2$ K. The values of ε/k (from Fig. 2) are 402, 213, 92, and 40 mK, from top to bottom. The values of α obtained from the fits are 0.22, 0.16, 0.14, and 0.21, respectively, with corresponding values of Δ_r of 8.3×10^3, 2.5×10^4, 3.4×10^4, and 1.0×10^4 s^{-1}. Uncertainties are discussed in the text.

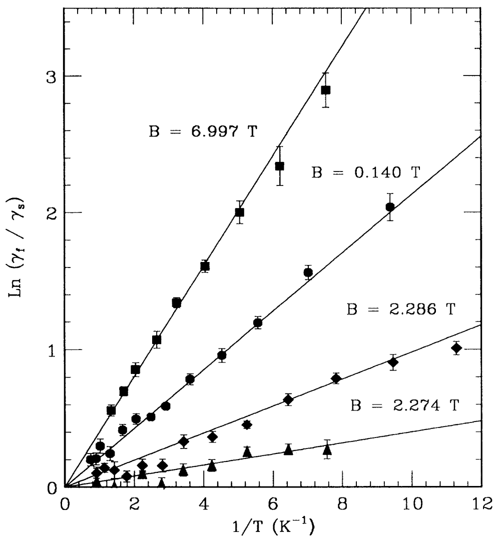

FIG. 2. $\ln(\gamma_f/\gamma_s)$ vs $1/T$, for the four data sets of Fig. 1. The slopes of the lines are 402, 213, 92, and 40 mK, respectively, with an uncertainty of ± 10 mK.

ing ($\Delta_r \ll kT$), the result is[13]

$$\gamma = \frac{\Delta_r}{2}\left[\frac{2\pi kT}{\hbar\Delta_r}\right]^{2\alpha-1}\frac{\cosh(\varepsilon/2kT)}{\Gamma(2\alpha)}\left|\Gamma\left[\alpha + i\frac{\varepsilon}{2\pi kT}\right]\right|^2 . \tag{1}$$

Equation (1) and the detailed balance relation together determine both γ_f and γ_s. For an atom tunneling in a metal, α is constrained to lie between 0 and $\frac{1}{2}$.[14] The tunneling matrix element Δ_r has been renormalized by interactions with phonons and with high-energy (adiabatic) electron-hole excitations. The solid lines in Fig. 1 are two-parameter least-squares fits of Eq. (1) to the data. (The asymmetry ε is fixed by the fits in Fig. 2.) The agreement between theory and experiment is excellent for all $T<1.2$ K. (In the future, with more high-temperature data points, we will include phonon-assisted tunneling in our analysis.[15]) The values of α and Δ_r determined from the fits are given in the figure caption.

Given quantitative agreement between theory and experiment at each value of the magnetic field, we can ask whether the coupling constant α varies with magnetic field for this defect. Since the variation of ε with B is believed to be due to fluctuations in the local charge density near the defect,[8] and since α also depends on this charge density,[14] it is plausible that α also varies with B. Golding, Zimmerman, and Coppersmith[4] argued, however, that α should be relatively insensitive to field because it depends on electron-hole excitations with energies up to a cutoff equal to the defect vibrational frequency.[16] We will present two analyses of our data aimed at answering this question. The first analysis, which assumes that the variation in α with B obtained from the fits is significant, leads to an inconsistency in the interpretation. The second analysis will show that the data are consistent with a single value of α, independent of B.

The values of α and Δ_r determined from a single data set are highly correlated; nevertheless one can estimate their joint uncertainties from contour plots of χ^2. The uncertainty in α obtained this way is about 0.02. Since α varies over the range 0.14–0.22 for the four data sets, this analysis suggests that the variation of α with B is significant. The correlation between α and Δ_r also extends to the family of fits in Fig. 1—in fact the values of α and Δ_r determined from the four fits fall on a straight line when plotted as $\log(\Delta_r)$ versus $\alpha/(1-\alpha)$. This correlation is highly suggestive, because the relation between Δ_r and the bare (unrenormalized) tunneling matrix element Δ is[6]

$$\Delta_r = \Delta(\Delta/\omega)^{\alpha/(1-\alpha)} , \tag{2}$$

where ω is a characteristic vibrational frequency of the defect in one of its two wells.[17] If α varies with magnetic field, but Δ and ω do not, then Eq. (2) predicts a linear dependence of $\log\Delta_r$ on $\alpha/(1-\alpha)$, with an intercept and slope equal to $\log\Delta$ and $\log(\Delta/\omega)$, respectively. With this interpretation, our values of α and Δ_r give the results $\hbar\Delta/k = 1.8$ μK and $\hbar\omega/k = 0.23$ K. While the first of these values is reasonable, the second is not. Not only is 0.23 K extremely low for a vibrational frequency (even for a defect that tunnels at low temperature), but such a low value violates the condition $kT \ll \hbar\omega$ upon which Eq. (1) is based.[6,13]

What is wrong with the above analysis? The correlation between α and Δ_r obtained from the four data sets lies along the same direction in parameter space as the correlation deduced from the χ^2 contours of a single fit. Moreover, the χ^2 analysis may underestimate the uncertainty in α, because it assumes that experimental errors are normally distributed.[18] These observations suggest that all four data sets can be fit with unique values of α and Δ_r. In fact, such a global fit is quite satisfactory, as can be seen in a scaling plot of all the data. This plot was

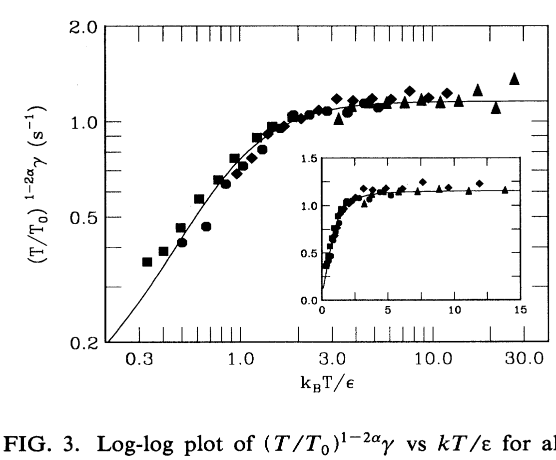

FIG. 3. Log-log plot of $(T/T_0)^{1-2\alpha}\gamma$ vs kT/ε for all four data sets, with $\alpha=0.195$ and $T_0=1$ K. The solid line is a fit of Eq. (1) to all the data, with $\Delta_r = 1.3\times 10^4$ s^{-1}. The four data sets are $B=6.997$ (■), 0.140 (●), 2.286 (♦), and 2.274 T (▲). Inset: Same quantities plotted on linear axes. (This plot is similar to Fig. 2 of Ref. 4, except that we use γ rather than γ_f on the vertical axis.)

introduced by Golding, Zimmerman, and Coppersmith,[4] who pointed out that the quantity $\gamma T^{1-2\alpha}$ depends on temperature only through the ratio kT/ε. (Golding, Zimmerman, and Coppersmith plot the quantity $\gamma_f T^{1-2\alpha}$, rather than $\gamma T^{1-2\alpha}$, leading to a slightly different shape of the curve.) Figure 3 shows a plot of $\gamma(T/T_0)^{1-2\alpha}$ versus kT/ε, with $T_0=1$ K and $\alpha=0.195$ for all the data. The data are clearly consistent with the interpretation that α and Δ_r are independent of magnetic field for this defect.

To determine more accurately the dependence of α with field, we would need either a larger experimental temperature range, or smaller experimental uncertainties. We have attempted to minimize obvious sources of systematic error. We were very careful to limit Joule heating of the sample from the measurement current. (The tunneling rates provide an excellent measure of the defect temperature, through the detailed-balance relation.[19]) The effect of external interference was minimized by rf filters on all electrical leads to the sample.[20] It is possible that other defects in the sample may influence the dynamics of the defect we are studying.[21] Even small changes in the measured tunneling rates can lead to significant uncertainty in the evaluation of α, especially when $kT \leq \varepsilon$. In the future, we will pursue further measurements in the power-law regime $kT > \varepsilon$, where we can most accurately determine α.

In summary, we have measured the tunneling rates of a single defect in a metal over a broad range of temperature and magnetic field. The rates agree quantitatively with predictions of dissipative quantum tunneling theory. Our data confirm the observation that the defect energy asymmetry ε varies randomly with field, and they support the hypothesis that the defect-electron bath coupling constant α is nearly independent of field. These results illustrate the power of conductance measurements of mesoscopic samples to obtain microscopic information about individual defects in disordered materials. While the experiments to date have been restricted to polycrystalline Bi samples, we expect to observe similar behavior in other disordered metals.

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[18] See W. Press *et al.*, *Numerical Recipes* (Cambridge University, Cambridge, 1986), p. 534; an analysis of uncertainties based on χ^2 can be unduly influenced by a stray data point, e.g., the second lowest point at $B=0.14$ T in Fig. 1.

[19] We have studied the role of Joule heating on the electron and defect temperatures in this sample, K. Chun and N. O. Birge (unpublished).

[20] The success of the rf filtering was confirmed by the reproducible behavior of one defect over a two-month period.

[21] There is strong evidence for defect-defect interactions at much higher temperatures; see K. Ralls and R. A. Buhrman in Ref. 10.