Atomic Tunneling from a Scanning-Tunneling or Atomic-Force Microscope Tip: Dissipative Quantum Effects from Phonons

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We study the effects of phonons on the tunneling of an atom between two surfaces. In contrast to an atom tunneling in the bulk, the phonons couple very strongly and qualitatively change the tunneling behavior. This is the first example of *Ohmic* coupling from phonons for a two-state system. We propose an experiment in which an atom tunnels from the tip of an scanning-tunneling microscope, and show how its behavior would be similar to the macroscopic quantum coherence behavior predicted for SQUIDs. The ability to tune and calculate many parameters would lead to detailed tests of the standard theories.

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The fascinating experiments carried out by Eigler and co-workers [1–4] have shown that scanning-tunneling microscopy can be used not only for imaging, but also to directly demonstrate fundamental aspects of quantum mechanics. Inspired by their success, we propose an experiment to test the effect of a dissipative environment on a quantum mechanical system. While often ignored in many applications of quantum mechanics, the environment can have important effects on a simple quantum system. In fact, from the dawn of quantum mechanics, people have been attributing the collapse of the wave function to the interaction of a quantum system with a macroscopic environment [5]. As recently emphasized by Leggett [6], condensed-matter physics provides a natural laboratory for studying this: How does the surrounding macroscopic crystalline environment change the behavior of an embedded quantum system? In particular, what can realistic models of the environment tell us?

A tantalizing view of the importance of the environment is given by the "overlap catastrophe" [7]. In a metal, the conduction electrons must be rearranged when the quantum subsystem changes state: The initial and final electron ground-state wave functions are so different that the bath of conduction electrons can force the subsystem to dissipate energy during each transition, or even keep the transition from occurring. In this Letter, we show that phonons, an environment even more ubiquitous than conduction electrons, can also have an overlap catastrophe. In particular, we show that quantum tunneling behavior between two surfaces can be dramatically altered by the coupling to phonons. We show how detailed calculations of the macroscopic quantum coherence (MQC) community [8] could be tested experimentally in an admittedly microscopic system using the tunneling of atoms onto and off of the tip of a scanning-tunneling microscope (STM) or atomic-force microscope (AFM).

Eigler and co-workers report on an "atomic switch," in which a Xe atom is reversibly transferred from a Ni(110) surface to the W tip, as well as on "transfer near contact" processes in which an STM tip is brought so close to a surface that the absorbed Xe atom spontaneously hops to it [2,3,9]. Several theories have been put forth to explain the atomic switch experiment [10–14]. The potentials calculated for the Xe-tip-surface system have a double-well shape, as depicted in Fig. 1, and bringing the tip just slightly closer to the surface than the switch operating conditions can give significant tunneling amplitudes, even for a heavy atom such as Xe. If we take, for example, some calculated parameters from Ref. [14] for Xe on Ni(110) ($V_0 = 7$ MeV, $Q_0 = 0.6$ Å), we can obtain tunneling elements up to $\hbar\Delta/k_B \sim 0.5$ K. Here we use the example of Xe simply because a lot is known about that system. Lighter elements such as He would have even larger tunneling amplitudes. By adding an electric field, the wells can be biased in either direction quite sensitively, as was shown in Ref. [11], so that by varying the tip position and the bias both Δ and ϵ can be varied independently. Of course the exponential dependence of the tunneling

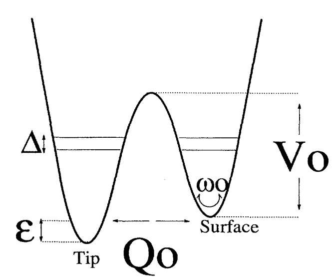

FIG. 1. Double-well potential for the tunneling atom. The atom tunnels from the STM or AFM tip to the substrate surface and back under the influence of a potential such as the one shown above. For temperatures $k_BT \ll \hbar\omega_0/2 < V_0$, the tunneling is between the lowest harmonic oscillator type states in each well. The tunneling amplitude $\hbar\Delta$ gives the energy splitting of the symmetric and antisymmetric superpositions of tip + surface states, and is determined by the barrier height V_0, the effective distance Q_0, the mass of the particle, as well as the possible asymmetry energy ϵ. The small oscillation frequency ω_0 is determined by the same parameters.

amplitude $\hbar\Delta$ on system parameters and the precise shape of the potential makes it very difficult to accurately predict its magnitude. The point is more so because the atom can be brought arbitrarily close to where it sticks to the surface, the tunneling rate can be made arbitrarily large.

We now proceed to the main part of this Letter: the effect of the phonon environment on our quantum tunneling system, and consider temperatures such that $k_B T \ll \hbar\omega_0/2 < V_0$, allowing us to truncate the Hilbert space to two states, one for each well [15].

As depicted in Fig. 2, the (AFM or STM) + atom system will exert a different force $\pm\Delta\mathbf{F}/2$ on the surface depending on where the atom is. The phonons will relax, switching their equilibrium positions in response to the atom being on the tip or the surface. This can be modeled by the Hamiltonian of the two-state system with the displaced phonons

$$H = \frac{\hbar\Delta}{2}\boldsymbol{\sigma}_\mathbf{x} + \frac{\epsilon}{2}\boldsymbol{\sigma}_\mathbf{z} + \sum_{\mathbf{k}\sigma}\left[\frac{1}{2}m_{\mathbf{k}\sigma}x_{\mathbf{k}\sigma}^2\mathbf{1} + \frac{1}{2}m_{\mathbf{k}\sigma}\omega_{\mathbf{k}\sigma}^2(x_{\mathbf{k}\sigma}\mathbf{1} + q_{\mathbf{k}\sigma}\boldsymbol{\sigma}_\mathbf{x})^2\right]. \tag{1}$$

The $\{x_{\mathbf{k}\sigma}\}$ are the normal coordinates for a given polarization σ and wave vector $\mathbf{k}$. The masses of the normal coordinate particles are denoted by $m_{\mathbf{k}\sigma}$, and the phonon angular frequency is $\omega_{\mathbf{k}\sigma}$. The $\{q_{\mathbf{k}\sigma}\}$ are the new equilibrium positions in the presence of the force $\pm\Delta\mathbf{F}/2$.

In the absence of phonons, the probability of finding the atom at the tip at time t, given that it was started there at time $t = 0$, oscillates back and forth with the usual form $P(t) = \sin^2(\Delta t)$. What now is the effect of the phonons (environment) on the behavior of the atom (embedded quantum system)? Is there a *qualitative* change of behavior? For atoms tunneling between two states in the bulk, an approach often taken to account for the environment is calculating the overlap integral of the atom in state one + relaxed phonons with the atom in state two + relaxed phonons. The new renormalized

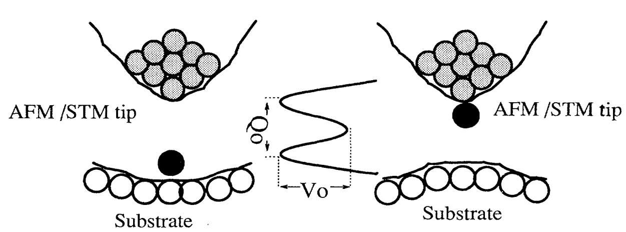

FIG. 2. Schematic drawing of the (STM or AFM) + atom system. The tunneling of the atom couples to phonons via the response of the substrate to the external force $\pm\Delta\mathbf{F}/2$ that depends on the position of the atom. The coupling to an environment drastically alters the tunneling behavior. For small coupling and low temperature, the tunneling is coherent with a renormalized tunneling frequency. For larger coupling and zero temperature, there is no tunneling at all, while at finite temperature there is incoherent tunneling with a power law dependence on temperature.

tunneling amplitude is just the bare amplitude multiplied by a Frank-Condon phonon-overlap factor. The atom still oscillates back and forth, but now with a reduced rate. For atoms tunneling in the bulk, this approach gives good qualitative results [16,17]. However, for an atom tunneling between surfaces, the situation is quite different, as can be seen by considering the force exerted by the atom on its environment. While the defect tunneling in a solid only exerts a *dipole* force, resulting in $q_k \sim k^{-1}$ [16], the external tip + atom system exerts a *monopole* force on the surface, resulting in $q_k \sim k^{-2}$ [18,19]. The external case has therefore a stronger coupling at low frequencies, and a naive calculation of the Frank-Condon factor gives an infrared divergence, a tell-tale sign that the adiabatic approximation breaks down. In fact, the case of tunneling between surfaces corresponds to "Ohmic" dissipation in the language of the macroscopic quantum tunneling (MQT) literature, as opposed to the bulk case, where the dissipation is of the "super-Ohmic" variety [8]. The effect of the environment can now be characterized by a dimensionless coupling parameter α defined as

$$\alpha = \frac{\eta Q_0^2}{2\pi\hbar}, \tag{2}$$

with η equivalent to the friction coefficient in the macroscopic limit [20]. The tunneling element is renormalized to $\Delta_r = \Delta(\Delta/\omega_c)^{\alpha/(1-\alpha)}$ for $\alpha < 1$, and is zero for $\alpha > 1$ [21]. In other words, for small coupling parameter α, the effect of the phonons is *quantitative* only, reducing the tunneling frequency, while if α crosses 1, the effect is *qualitative*: there is a transition to no tunneling at all. This fascinating effect is the phonon analog of Anderson's overlap catastrophe in an electron gas [7]. We emphasize that this would be the first case in which an overlap catastrophe is caused by phonons [22].

Having shown that an atom tunneling from an STM or AFM tip to a surface undergoes Ohmic dissipation from the phonon environment, we come to the question: Is the dimensionless coupling parameter α in an interesting regime? Assuming a point force $\Delta F/2$ on a semi-infinite isotropic medium with a linear (Debye) dispersion we find

$$\alpha = \frac{1}{64\pi^2\hbar\rho c_t^3}G(\sigma)\,(\Delta F)^2, \tag{3}$$

where ρ is the density in $\mathrm{kg\,m^{-3}}$, c_t is the transverse sound velocity, and $G(\sigma)$ is a tabulated function of Poisson's ratio $\sigma = (3\lambda + \mu)[6(\lambda + \mu)]$, with roughly equal contributions from the acoustic bulk and Rayleigh modes [23]. The critical $\Delta\mathbf{F}$ for which $\alpha = 1$ goes from about 3 nN for W to 0.3 nN for Pb. Another order of magnitude reduction can be obtained for some organic materials. At the short distances we need for atomic tunneling, the typical force of a tip on a surface is of the order of a nN per Å separation [24], putting us right into the interesting regime [25].

We envisage a number of realizations of the atom-surface tunneling experiments. First, it has been calculated that for $k_B T \geq \hbar\Delta/\pi\alpha$ the coherent oscillations are destroyed [26,27], and there is only incoherent tunneling with a rate [8]

$$\tau^{-1} = \Delta_r \frac{\Gamma(\alpha)}{\Gamma(\alpha + 1/2)} \frac{(\pi)^{2\alpha+1/2}}{2} \left(\frac{k_B T}{\hbar \Delta_r}\right)^{2\alpha-1}, \quad (4)$$

from which the parameter α could be extracted and compared to our predictions. (For example, an AFM operating in the noncontact mode can measure the oscillations by the force modulation of $\pm\Delta\mathbf{F}/2$. The tunneling current of an STM is significantly larger when the atom is on the tip rather than when it is on the surface [2].) Beautiful experiments [28] on individual two-state tunneling defects coupled to conduction electrons in mesoscopic metals and on SQUIDs [29] have confirmed this power law behavior of the rate with temperature. However, in contrast to our system, these experiments do not allow one to vary α, or to calculate it from first principles. We believe this would be the first quantitative test of the α parameter, giving valuable insight into the validity of the linear coupling model of a dissipative environment. By varying the distance between the tip and the surface, or by varying the position within the surface unit cell, the force $\Delta\mathbf{F}$, and thus the coupling parameter α, could be tuned. Of course, simultaneously, this would heavily affect the tunneling frequency Δ by changing the potential barrier. However, one could still see the qualitative change in temperature dependence of the rate. Of particular interest would be the crossover from *decreasing* with increasing temperature to *increasing* with increasing temperature of the rate when α crosses 1/2 from below [30].

Second, a more ambitious experiment might test the coherence predictions: watching the tunneling turn off as α is increased. Coherent oscillations, one of the goals of the MQC community [8], will occur if $T < \hbar\Delta/k_B\pi\alpha$, and $\alpha < 1/2$ [31]. There has been extensive discussion of the problems of measuring coherent oscillations in the MQC literature [32]. For example, Peres [33] has claimed that noninvasive measurements are impossible in this case: a measurement introduces an energy of order $\hbar\omega_0$. This has been disputed by Leggett and Garg [34], and, in fact, we find that the uncertainty in momentum P caused by measuring the position to better than $Q_0/2$ is $(\Delta P)^2/2 \geq \hbar\omega_0(\hbar\omega_0/64V_0)$, which gives a sizable window for the direct measurement we propose. This expression can be easily derived from the position-momentum uncertainty principle. Peres makes too crude an approximation for ΔP, and thus overestimates the effect of localizing the object to be measured (flux in his case) on its conjugate variable [19]. More specifically, for the case of Xe on Ni(110) theories for the switch experiment estimate that for a bias voltage greater than $\hbar\omega_0$, about one in 2000 electrons will inelastically scatter and excite the atom into a higher vibrational state [11]. For a current of ~0.1 nA the atom would be excited on average about once every μs. Thus an STM in imaging mode can only measure oscillations if $1/\Delta \ll 1$ μs, a rate that is easily reached for close enough tip-surface separation. If the bias is less than $\hbar\omega_0$, excitations can only occur through multiple electron transitions, so the excitation rate will be much smaller and lower oscillation rates can be measured.

For experimentally accessible temperatures (say 1 K) with $\alpha = 0.1$, this means a tunneling rate of at least 10 GHz. Recent developments in high-speed STM have achieved a time resolution of picoseconds [35], and we envisage a direct measurement of the correlations by sending in pairs of voltage pulses [32]. This could be accomplished by a low-temperature STM coupled to picosecond lasers that generate the voltage pulses (as was done in Ref. [35]). Varying the intrapair time by $\pi/2\Delta$ will give an oscillation of the integrated current that reaches a maximum of 40% of the current difference between the atom on the tip and the surface for a slightly biased well with $\epsilon = \Delta$ [19]. The interpair time must be much greater than the intrapair time, and the pulses must be long compared to the electron tunneling time (which is on the order of fs). These would be the first measurements of the coherent oscillations of a single entity with Ohmic dissipation, and, while the atom is not "macroscopic," our proposed experiment should be a fertile testing ground of new measurement schemes, and may shed light on the even more difficult problem of observing MQC in SQUIDs [36].

In summary, we have shown how the phonons couple in a fundamentally different way to particles at a surface than to particles in a bulk, and can cause an overlap catastrophe, just as electrons can. More precisely, the coupling of an atom to phonons at the surface produces Ohmic dissipation, and *qualitatively* changes the tunneling behavior, in contrast to the well-known case of tunneling in the bulk where the effect of phonons is only *quantitative*. We have proposed an experiment with an atom tunneling between an STM or AFM tip and a surface, and show how this could test theories of MQC, albeit in a microscopic setting. Because the parameters Δ, ϵ, and α can be varied by changing the tip-surface position and the bias, many different regimes of the theories of two-state systems with Ohmic dissipation can be put to the test.

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