Chapter 2

Atomic Tunneling from a STM/AFM Tip: Dissipative Quantum Effects from Phonons

2.1 Introduction

We study the effects of phonons on the tunneling of an atom between two surfaces[1]. In contrast to an atom tunneling between two positions in the bulk material, the phonons couple very strongly to the atom, qualitatively changing the tunneling behavior. This is the first example of dissipative *ohmic* coupling from phonons for a two-state system. We propose an experiment in which an atom tunnels from the tip of an STM, and show how its behavior would be similar to the Macroscopic Quantum Coherence behavior predicted for SQUIDS. An ability to tune and calculate many parameters would lead to detailed tests of the standard theories[1].

[1] A general introduction for non-specialist can be found on the web at: http://www.lassp.cornell.edu/ ardlouis/dissipative/QAtunneling.html.

The fascinating experiments carried out by Eigler and colleagues[2, 3, 4, 5] have demonstrated 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, the collapse of the wavefunction has been attributed to the interaction of a quantum system with a macroscopic environment[6]. Numerous authors have shown how interactions with an external environment can wipe out measurable effects of off-diagonal elements of the density matrix and as such give a plausible mechanism for the wave-function collapse process. In its modern form this idea is called decoherence[7] and is currently receiving renewed interest in its applications to the study of quantum computing. However, this in no way solves the measurement problem as many authors assert. Even if one were to solve the quantum mechanics of a sub-system interacting with a measuring device (for example a two-state system with two states, up and down), this leaves unanswered the question: why did measurement A produce an up-state and measurement B a down-state; what broke the symmetry? This in contrast to the spontaneous breaking of symmetry in a classical system (for example an Ising model with no external field) which could always (in principle) be traced back to a thermal fluctuation. With this caveat in mind, it is clear that understanding how an environment "collapses" a wave-function is still of great importance to understanding the measurement problem – but it doesn't solve it, it only pushes back the boundary of ignorance. As recently emphasized by Leggett[8], 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 to which it is coupled? 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"[9]. In a metal, the conduction electrons must be rearranged when a quantum subsystem changes state: the initial and final electron ground–state wavefunctions 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 at all. In this chapter, we show that phonons, an environment even more ubiquitous than conduction electrons, can also cause an overlap catastrophe. In particular, we demonstrate 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 [10] 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).

2.2 An Atom Between a Tip and a Surface feels a Double Well Potential

Eigler and collaborators report on an "atomic switch", in which a Xe atom is reversibly transfered 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[3, 4, 11]. Several theories have been put forth to explain the atomic switch experiment[12, 13, 14, 15, 16]. The potentials calculated for the Xe-tip-surface system have a double well shape as depicted in figure 2.1, 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 like Xe[17]. Of course the exponential dependence of the tunneling amplitude Δ on system parameters and the precise shape of the potential makes it very difficult to accurately predict its magnitude from first principles. The point is more that because the atom can be brought arbitrarily close to where it sticks to the surface, the tunneling rate can be made arbitrarily large.

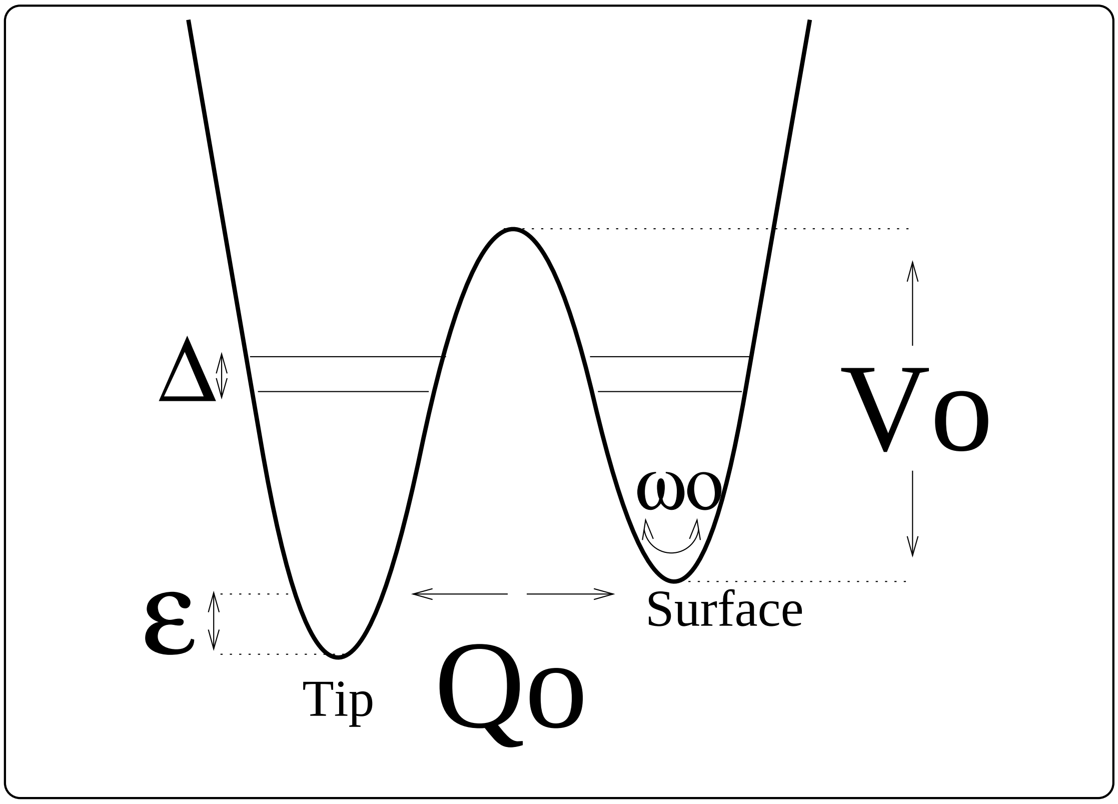

Figure 2.1: **Double-Well potential for the Tunneling Atom**
The atom tunnels from the STM/AFM tip to the substrate surface and back under the influence of a potential like the one shown above. For temperatures: $k_B T << \hbar\omega_0/2 < V_0$, the tunneling is between the lowest harmonic oscillator type states in each well. The tunneling amplitude Δ gives the energy splitting of the symmetric and anti-symmetric 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.

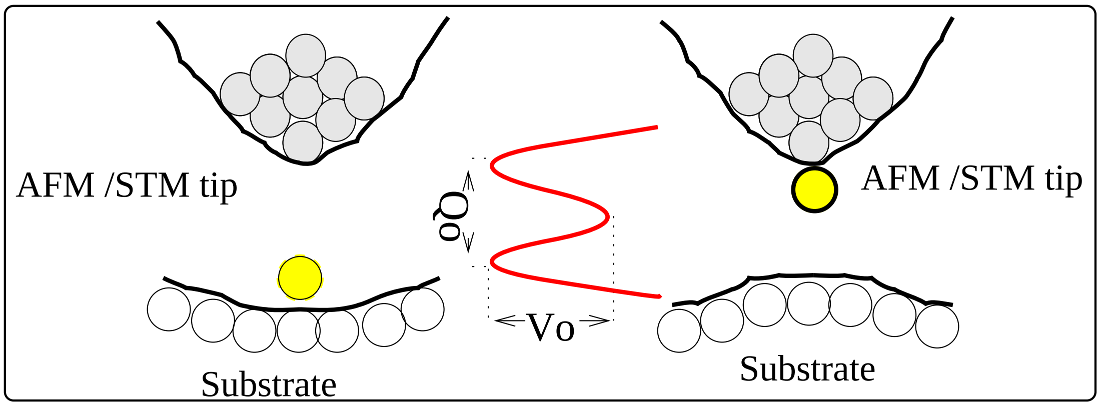

Figure 2.2: **Schematic drawing of the STM/AFM + atom system** The tunneling of the atom couples to phonons via the response of the substrate to the external force $\pm\Delta\mathbf{F}/2$ that depends on the position of the atom. The coupling to an environment drastically alters the tunneling behavior. 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.

2.3 The Effect of the Phonon Environment: Ohmic Dissipation

We now proceed to the main part of this chapter: the effect of the phonon environment on our quantum tunneling system, and consider temperatures such that $k_B T << \hbar\omega_0/2 < V_0$, allowing us to truncate the Hilbert space to two states, one for each well[18].

As depicted in figure (2.2), the AFM/STM + atom system will exert a different force $\pm\Delta\mathbf{F}/2$[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 a Hamiltonian for the two-state system

[2] The use of the word force is an unfortunate misnomer: force is a purely classical concept. Instead this should be the gradient of the potential. However, in keeping with common conventions in the literature, we continue to use the word force.

with the displaced phonons:

$$H = \frac{\hbar\Delta}{2}\sigma_{\mathbf{x}} + \frac{\epsilon}{2}\sigma_{\mathbf{z}} + \sum_{\mathbf{k}\sigma}\left[\frac{1}{2}m_{\mathbf{k}\sigma}x_{\mathbf{k}\sigma}^2\mathbf{1} + \frac{1}{2}m_{\mathbf{k}\sigma}\omega_{\mathbf{k}\sigma}^2\left(x_{\mathbf{k}\sigma}\mathbf{1} + q_{\mathbf{k}\sigma}\sigma_{\mathbf{x}}\right)^2\right]. \tag{2.1}$$

The $\{x_{\mathbf{k}\sigma}\}$ are the normal coordinates for a given polarization σ and wavevector $\mathbf{k}$. The masses of the normal coordinate particles are denoted by $m_{\mathbf{k}\sigma}$, and the phonon angular frequency by $\omega_{\mathbf{k}\sigma}$. 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 $P(t) = \sin^2(\Delta t)$. For a potential of the form:

$$V(q) = V_0\left(1 - (\frac{2q}{Q_0})^2\right)^2, \tag{2.2}$$

the bare tunneling amplitude Δ can be found in WKB approximation or by an instanton analysis; namely:

$$\Delta = 2\sqrt{3}\omega_0(\hbar S_I/\pi)^{1/2}\exp[-\frac{S_I}{\hbar}] \tag{2.3}$$

$$S_I = \int_{-\frac{q_0}{2}}^{\frac{q_0}{2}}(2mV(q))^{1/2}dq, \tag{2.4}$$

where S_I is the instanton action. The prefactor comes from Gaussian fluctuations in the instanton approach so that for our potential:

$$S_I = \frac{2}{3}(2mV_0q_0^2)^{1/2} = \frac{8\sqrt{2}}{3}\left(\frac{V_o}{\omega_0}\right) \tag{2.5}$$

$$\Delta \approx 3.80(\hbar\omega_0V_0)^{1/2}\exp\left[-3.77\left(\frac{V_0}{\hbar\omega_0}\right)\right] \tag{2.6}$$

The tunneling amplitude Δ is roughly the geometric mean of the zero point energy and the barrier height times the exponential of their ratio.

What is now 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 (plus phonons) with the atom in state two (plus phonons). The new renormalized 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. If one assumes that the phonons are all in their ground states (Gaussians) then their effect on Δ can easily be calculated to be:

$$\begin{aligned} \Delta_r &= \Delta e^{-W} \\ W &= \sum_k \frac{\lambda_k^2 Q_0^2}{4\hbar m \omega_k^3} \end{aligned} \tag{2.7}$$

where λ_k is the coupling parameter given by:

$$\lambda_k = \left(\frac{m_k \omega_k^2 q_k}{Q_0} \right) \tag{2.8}$$

(the polarization index σ has been suppressed). For atoms tunneling in the bulk, this approach gives good qualitative results[19, 20]. 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 (or more precisely, how the atom couples to the phonon environment through the gradient of the potential). While the defect tunneling in a solid only exerts a *dipole* force, resulting in $q_k \sim k^{-1}$[19], the external tip plus atom system exerts a *monopole* "force" on the surface, resulting in $q_k \sim k^{-2}$[21]. 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. This "overlap catastrophe" is not the real physical picture though, and Leggett and co-workers have worked out a more sophisticated scheme that splits up the

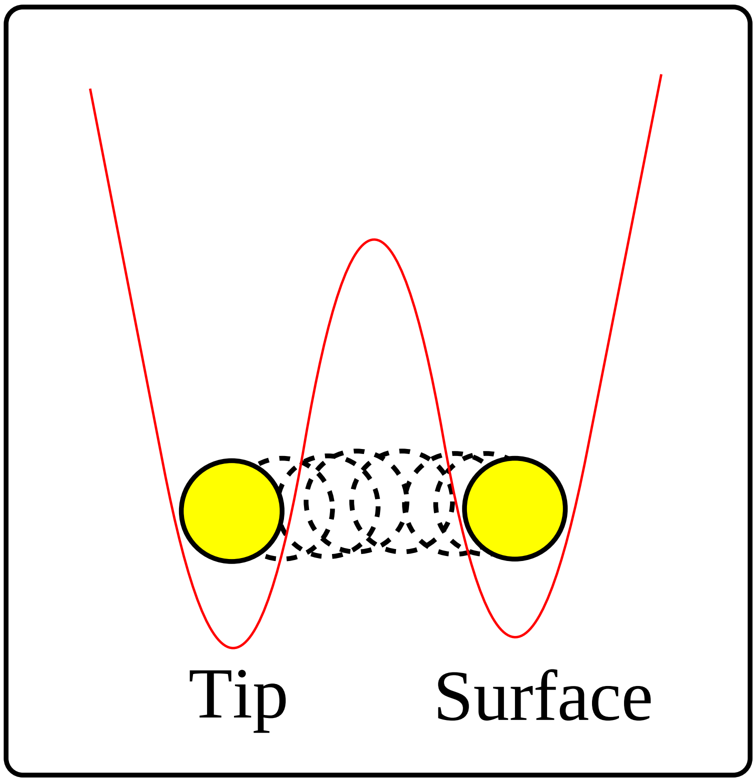

Figure 2.3: **Effect of long-frequency phonons on tunneling**
High frequency phonons simply renormalize the tunneling frequency Δ. However, the adiabatic approximation implicit in such a Frank-Condon factor approach breaks down for long-wavelength phonons with frequencies $\omega_k \leq \Delta$. In fact these phonons, which cause an infrared divergence in a simplistic Frank-Condon overlap integral, can safely be ignored.

effects of high and low frequency oscillators(phonons)[10]. They replace Δ by a "renormalized" tunneling element:

$$\Delta \to \Delta_r = \Delta \exp\left(\frac{-q_0^2}{2\pi\hbar}\int_{\Delta_r}^{\infty}\frac{J(\omega)}{\omega^2}d\omega\right), \tag{2.9}$$

where

$$J(\omega) = \frac{\pi}{2}\sum_k \frac{\lambda_k^2}{\omega_k m_k}\delta(\omega-\omega_k) \tag{2.10}$$

is the "spectral density". The lower-limit frequency cutoff is explained in figure (2.3). Eq(2.9) is then iterated to self-consistency. Leggett *et al.* find that the "renormalized" tunnel splitting becomes:

$$\Delta_r = \Delta \left(\frac{\Delta}{\hbar\omega_c} \right)^{\alpha/(1-\alpha)}, \tag{2.11}$$

where the ω_c is a cutoff frequency $\Delta << \omega_c << \omega_0$ used to separate high and low frequency oscillators and drops out of most relevant calculations. The effect of the environment can now be characterized by a dimensionless coupling parameter α defined as:

$$\alpha = \frac{\eta Q_0^2}{2\pi\hbar}, \tag{2.12}$$

with η equivalent to the friction coefficient in the macroscopic limit[22]. 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 "superohmic" variety[10]. The tunneling element is renormalized to Δ_r for $\alpha < 1$, and is zero for $\alpha > 1$. More sophisticated renormalization group calculations confirm this simple picture[23]. 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 analogue of Anderson's "overlap catastrophe " in an electron gas[9]. We emphasize that this would be the first case in which an overlap catastrophe is caused by phonons[24].

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

$$\alpha = \frac{1}{64\pi^2\hbar\rho c_t^3} G(\sigma)(\Delta F)^2, \tag{2.13}$$

where ρ is the density in $kg\ m^{-3}$, 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[25][3] The critical $\Delta\mathbf{F}$ for which $\alpha = 1$ ranges from about 3 nanoNewtons(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 needed for atomic tunneling, the typical force of a tip on a surface is of the order of a nN per Å separation[26], putting us right into the interesting regime[27]!

[3] For a more detailed discussion of Rayleigh modes, and why they produce *ohmic* coupling, see Appendix **B**.

2.4 Proposed Experimental Measuring Schemes

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[28, 29], and there is only incoherent tunneling with a rate[10]:

$$\tau^{-1} = \Delta_r \frac{\Gamma(\alpha)}{\Gamma(\alpha + 1/2)} \frac{(\pi)^{2\alpha+1/2}}{2} \left(\frac{k_B T}{\hbar\Delta_r}\right)^{2\alpha-1}, \tag{2.14}$$

from which the parameter α could be extracted and compared to our predictions[30, 31]. 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 of the rate from *decreasing* with increasing temperature to *increasing* with increasing temperature when α crosses $1/2$ from below[32] (See figure 2.4 for a quick summary of the regimes described above). An STM with sufficient control to manipulate atoms that can also vary temperature from $7K$ to $300K$ has already been built[33]!

Second, a more ambitious experiment might test the coherence predictions: observing the tunneling turn off as α is increased. Coherent oscillations, one of the goals of the MQC community[10], will occur if $T < \hbar\Delta/k_B\pi\alpha$, and $\alpha < 1/2$[34]. For experimentally accessible temperatures (say $1K$) with $\alpha = 0.1$ this means a tunneling rate of at least $10GHz$. Recent developments in high-speed STM have achieved a time resolution of picoseconds[35], and we envisage a direct measurement of the

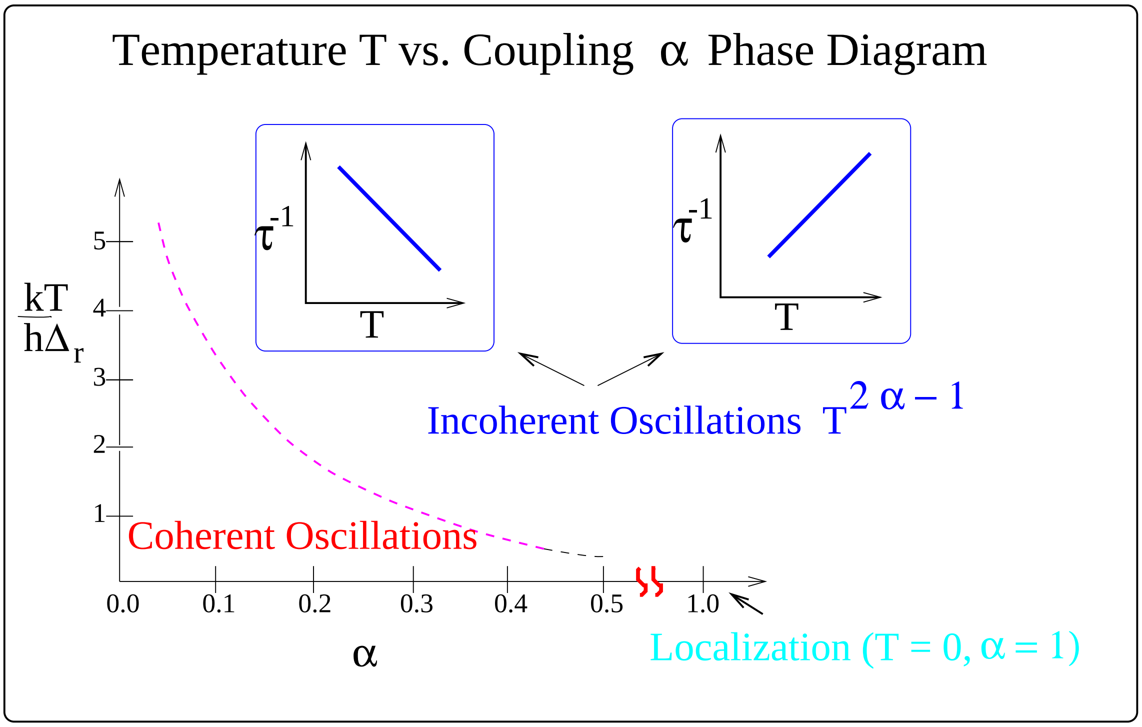

Figure 2.4: **Temperature T v.s. α Phase diagram**
The two parameters of interest are the temperature T (normalized by the renormalized tunneling frequency Δ_r) and the coupling constant α. At zero temperature the coherent oscillations are expected to disappear for $\alpha \geq 0.5$ and localization sets in for $\alpha \geq 1$. The coherent oscillations initially persist, as the temperature is raised, slowly becoming more and more damped until they cross over to incoherent behavior. What's particularly interesting is that the oscillation frequency has a *qualitatively* different power-law dependence on T when α crosses $\frac{1}{2}$. At even higher temperatures we anticipate Arrhenius behavior[4].

correlations by sending in pairs of voltage pulses[36]. This could be accomplished by a low-temperature STM coupled to picosecond lasers that generate the voltage pulses(as was done in Refs.[35]). By comparing with the temporal Bell Inequalities described in Appendix **A**, these correlations could be tested for quantum coherence. Varying the intra-pair 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 $\epsilon = \Delta$. The inter-pair time must be much greater than the intra-pair time, and the pairs 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[37].

2.5 Conclusions for Atomic Tunneling with Dissipation

In summary, we have shown how the phonons couple in a fundamentally different way to an external particle at a surface than to an interstitial particle 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/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 all 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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[17] A number of authors have calculated well parameters. If we take for example some parameters from reference [16] for Xe on Ni(110) ($V_0 = 7meV, Q_0 = 0.6\text{Å}$), we can obtain tunneling elements up to $\hbar\Delta/k_B \sim 0.5K$! In this paper, we use the example of Xe simply because a lot is know about that system. Of course 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 reference [13], so that by varying the tip position and the bias, both Δ and ϵ can be varied independently.

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[20] A similar effect is at work in the small-polaron problem, where the phonon-overlap integral lowers the hopping rate of an itinerant electron.

[21] This can be seen by the following argument: $q_{\mathbf{k}i} \sim D(k)^{-1}_{ij} \sum_l \Delta F_j(\mathbf{r}_l) e^{-i\mathbf{k}\cdot\mathbf{r}_l}$, where $D(k)_{ij}$ is the dynamical matrix which goes as $\sim k^2$ for small k. Thus $q_k \sim k^{-1}$ for a dipole force, and $q_k \sim k^{-2}$ for a monopole force.

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[27] Note that we have ignored the phonons in the tip. Although they are part of the environment, their effect is expected to be small. On the one hand, if we approximate the tip as another flat surface, the total coupling will just be sum of the α's from each surface. As the tip is usually made of tungsten or some other stiff material, it will typically have an α an order of magnitude smaller than the α from the substrate surface. On the other hand, if we approximate the tip as a $1-d$ vertical line of atoms, the coupling will be *sub-ohmic*, and the tunneling will be quenched at all coupling strengths. We have also ignored possible coupling to electron hole pairs. Of course for an insulator they won't matter, and for example in the case of Xe on Ni(110), the electron- hole pair dissipation is typically a factor 100 smaller than the phonon dissipation[12]. When it is not negligible, it will give an additive contribution to α. An estimate of the force difference ΔF can be obtained from *ab-initio* total energy pseudopotential calculations quite accurately with standard techniques (Thomás Arias, private communication).

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[37] Coherent oscillations with ohmic coupling have been observed by H. Wipf *et al* Europhys. Lett., **4** 1379 (1987) for Hydrogen tunneling in Niobium. In this case, the measurement is in frequency space on a large number of trapped H atoms interacting with conduction electrons. Recent calculations (Daniel S. Fisher and Aris L. Moustakas, "On the Localization of Heavy Particles in Metals", Harvard preprint HU-AM-94-001 (1994)) have shown that simultaneous tunneling of the atom and electrons will inhibit localization caused by the conduction electron bath that gives ohmic dissipation in the aforementioned experiments[31, 37] and thus a phonon bath may be the only experimentally accessible route to this effect.