Fluctuation Dominated Josephson Tunneling with a Scanning Tunneling Microscope

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We demonstrate Josephson tunneling in vacuum tunnel junctions formed between a superconducting scanning tunneling microscope tip and a Pb film, for junction resistances in the range 50–300 kΩ. We show that the superconducting phase dynamics is dominated by thermal fluctuations, and that the Josephson current appears as a peak centered at small finite voltage. In the presence of microwave fields ($f = 15.0$ GHz) the peak decreases in magnitude and shifts to higher voltages with increasing rf power, in agreement with theory.

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Scanning tunneling microscopy (STM) has been extensively used in the study of high-T_c superconductors (HTSC), providing a spectroscopic tool with unparalleled energy and spatial resolution [1]. Yet, while superconducting tips have been demonstrated in the past [2], all STM studies so far have been performed using normal-metal tips, thus probing only the single-particle excitation spectrum, the gap structure which is a consequence of superconductivity, but not the superconducting (SC) ground state itself. Results from STM measurements of HTSC show excitation gaps in situations where superconductivity is believed to be absent (pseudogap), such as in vortex cores [3,4] and above T_c in underdoped samples [5], as well as inhomogeneities in the gap structure in reportedly high quality BiSrCaCuO crystals [6] and thin films [7]. These results, due to the nature of the measurements, do not remove the ambiguity with respect to the existence of a finite SC pair amplitude in the situations studied. In light of this outstanding problem, it seems desirable to have a way to directly probe the SC pair amplitude with high spatial resolutions on the order of ξ, the coherence length. This can be achieved by performing STM experiments with SC tips [8], measuring the contribution from Josephson pair tunneling to the total tunneling current.

In this Letter we report on measurements of STM vacuum tunnel junctions formed between a SC tip and a conventional SC Pb film at $T \sim 2$ K. We find an enhancement of the tunneling current at low bias voltages for junction resistances of 50–300 kΩ, and demonstrate that this is due to Cooper pair tunneling by considering both the dc and ac Josephson effects in the presence of strong thermal fluctuations. We show that the information obtained from this new type of experiment is sufficient to deduce the local Josephson I_cR product and, therefore, the local amplitude of the SC condensate.

We have recently developed a method for the reproducible fabrication of SC STM tips [9]. Tips made by deposition of Ag(30 Å)/Pb(5000 Å) proximity bilayer onto conventional $Pt_{0.8}Ir_{0.2}$ STM tips exhibit a well-developed BCS gap at low temperatures. From $\Delta(T)$ measurements, Δ_0 and T_c were estimated to be 1.33 meV and 6.8 K, respectively [9]. Here we use these tips to form superconductor/insulator/superconductor (*S/I/S*) vacuum tunnel junctions against a Pb film (Fig. 1) grown *in situ* on a graphite substrate. Because of the high mobility of Pb atoms on the graphite surface during the room temperature deposition, the Pb film is not uniform. We therefore use the STM to first scan over the sample surface and identify SC Pb clusters. Thus we can selectively position the tip over the area of interest for this work, with spatial resolutions as reported in Ref. [9].

For this configuration we observe typical quasiparticle tunneling conductance spectra for *S/I/S* junctions with normal state resistance $R_N = 100$ MΩ, as shown in Fig. 2. Note the vanishingly small conductance for $|eV| < 2\Delta$ at low temperatures, which confirms that tunneling is the dominant charge transfer mechanism. At higher temperatures, thermal excitations of quasiparticles across the gap give rise to a conductance peak at $\pm(\Delta_{Pb} - \Delta_{tip}) \approx 0$ meV. At high junction resistances, one can expect the SC phases of the two superconductors comprising the junction to be completely decoupled, as their coupling, described by the Josephson binding energy [10,11],

$$E_J = \frac{\pi\hbar}{4e^2}\frac{\Delta(T)}{R_N}\tanh\frac{\Delta(T)}{2k_BT}, \tag{1}$$

is vanishingly small compared to k_BT. For example, for a typical junction of 100 MΩ, $E_J/k_B \sim 0.5$ mK. Here T is the temperature, Δ is the SC gap, R_N is the junction resistance, and all other symbols have their usual meaning.

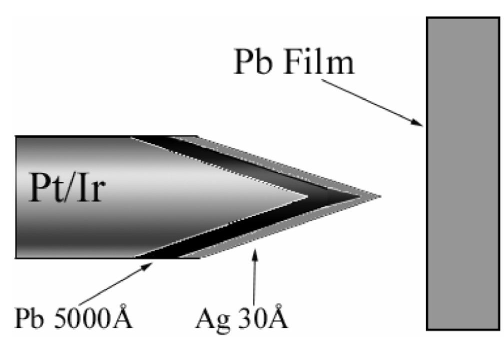

FIG. 1. Schematic depiction of the experimental setup.

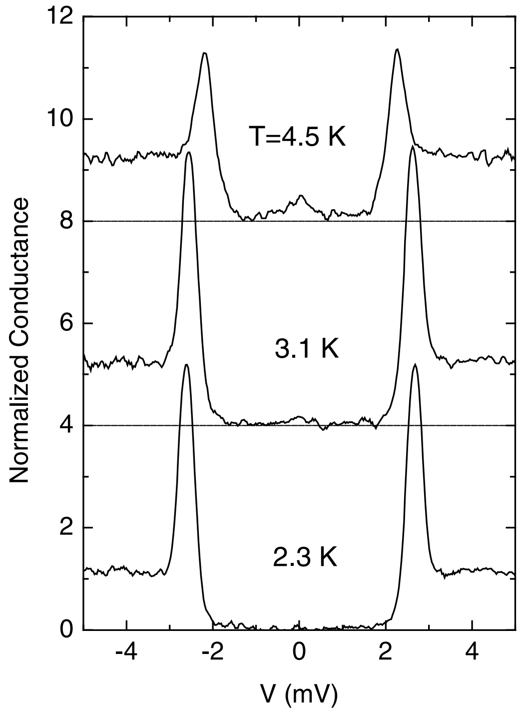

FIG. 2. Normalized dI/dV curves for a high resistance ($R_N = 100\ \mathrm{M}\Omega$) *S/I/S* junction at various temperatures. The curves are offset for clarity, and the horizontal lines correspond to zero conductance of each curve.

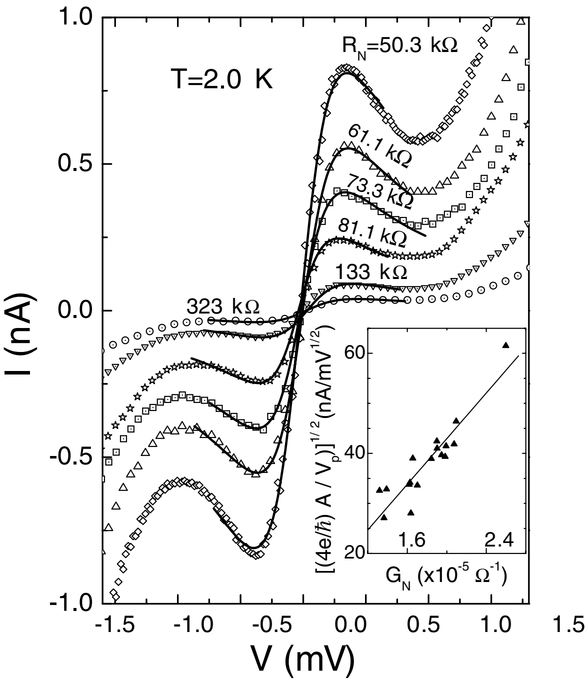

FIG. 3. Low bias current-voltage characteristics for various junction resistances at $T = 2.0$ K. The data (points skipped for clarity) are represented by symbols, and the lines represent two-parameter fits to theory. Inset: $I_{c,0} \times \sqrt{e/k_BT}$ vs G_N (see text).

As the junction resistance is decreased, E_J increases. When E_J becomes larger than k_BT, a supercurrent of magnitude $I = I_{c,0} \sin\phi$ can flow across the junction at zero voltage [12]; here $I_{c,0} = 2eE_J/\hbar$ and ϕ is the relative phase. When a finite voltage V is applied across the junction, ϕ oscillates according to $\dot{\phi} = 2eV/\hbar$. These are the well-known Josephson equations and the Josephson effect in low resistance junctions with macroscopically sized electrodes is well understood and well studied [12].

The present situation is different—we note that the tip electrode area is at most $1-2\ \mathrm{nm}^2$, thus the very small junction capacitance C leads to large charging energies of the junction, $E_C = e^2/2C$. In addition, the junction resistance cannot be made arbitrarily small and there are two factors limiting the range of this parameter. First, because of the small junction area, the current densities are high and a rough estimate suggests that junction resistances below few tens of kΩ will result in current densities sufficient to destroy the superconductivity in the tip. A second limit on the resistance is the transition from vacuum tunneling to a point contact regime which was observed to occur at about $R_N \sim 10$ kΩ [13]. Experimentally, we were able to achieve resistances as low as 40 kΩ.

Figure 3 shows typical data from measurements of the current-voltage characteristics at our base temperature 2.0 K for various junction resistances. Note that the data is drawn for voltages below the sum of the energy gaps on a magnified current scale. The full I-V curves show a well-defined gap feature at $\pm(\Delta_{\mathrm{tip}} + \Delta_{\mathrm{Pb}}) = 2.6$ meV. The location of the gap feature does not change with decreasing resistance, and the high bias part of the curves falls on the same line when the curves are scaled by R_N. The low bias part of these curves, however, does not reduce to the same line upon scaling. We observe a current peak centered at finite voltage near zero bias, the height of which grows when R_N is decreased. Since the SC gap is temperature dependent, the fact that the gap-edge feature appears at the same voltage regardless of the junction resistance is an indication that there is no self-heating of the junction (see also Ref. [14]). The current peak observed in Fig. 3 cannot, therefore, be attributed to enhanced quasiparticle thermal excitations across the gap, and must be a signature of pair tunneling.

One can model the dynamics of the phase ϕ in such a loosely coupled Josephson junction by a point particle moving in a periodic washboardlike potential landscape $U(\phi) = E_J(1 - \cos\phi)$ [15] and being subject to a stochastic force due to thermal noise [16,17]. For very high junction resistances (very shallow potential) the motion of the phase is completely randomized by thermal fluctuations as $k_BT \gg E_J$. But as the resistance decreases the phase spends on average more and more time near the minima of $U(\phi)$. We note that even at the lowest resistance achieved in this work $E_J/k_B \sim 1$ K, thus E_J is comparable to, but still smaller than, the thermal energy in the system. In this case the phase motion can be viewed as diffusive, and the

I-V characteristics of such a junction have been calculated using this approach by Ivanchenko and Zil'berman [17] and Harada *et al.* [18] (and also by Ingold *et al.* [19] using a perturbative approach) to have the form

$$I(V) = \frac{I_{c,0}^2 Z_{\rm env}}{2} \frac{V}{V^2 + V_p^2}, \qquad (2)$$

where $V_p = (2e/\hbar) Z_{\rm env} k_B T_n$, considering the thermal fluctuations as Johnson noise generated by a resistor $Z_{\rm env}$ at temperature T_n. The solid lines in Fig. 3 represent fits of our data to Eq. (2), with V_p and $A \equiv I_{c,0}^2 Z_{\rm env}/2$ the only fitting parameters. Using Eqs. (2) and (1), a plot of $\sqrt{(4e/\hbar)A/V_p}$ vs $G_N = 1/R_N$ (Fig. 3 inset) is expected to be linear with zero intercept and slope $\pi\Delta/2e\sqrt{k_B T_n}$. We obtain from a linear fit $T_n = 5.8 \pm 0.6$ K and $Z_{\rm env} = 1.5 \pm 0.2$ kΩ. The value of $Z_{\rm env}$ is consistent with our experimental setup. The value of the noise temperature T_n is higher than the actual temperature of the junction, which is not surprising since the isolation of the junction from room temperature circuitry is not perfect [20]. It is also consistent with values reported from similar measurements on ultrasmall, high resistance planar junctions [18,21].

We have repeated the measurements described here by probing different regions of the Pb film, and by using several tips and several samples. We find that the values we get for T_n and $Z_{\rm env}$ are reproducible within error bars, from which we conclude that these parameters are sample independent, and are determined only by the details of the experimental setup. We stress that knowledge of these values allows for the determination of $I_{c,0}$ in any sample from the I-V curves.

Since our data agrees very well with theory, and it is extremely difficult to explain our results using a quasiparticle-only picture, especially once self-heating is ruled out [14], we are led to the conclusion that this is a fluctuation dominated dc Josephson effect. This effect was previously observed in ultrasmall planar junctions and is well documented [18,22,23]. Furthermore, we confirm that this effect stems from Josephson tunneling by measuring the response of the junction in the presence of microwave fields with frequency $f = 15.0$ GHz, fed into a cylindrical cavity containing the STM by a semirigid coaxial cable antenna [24].

In the absence of fluctuations, when $E_J \gg k_B T, E_C$, a voltage source driven Josephson junction exhibits phase locked Shapiro current spikes at voltages corresponding to integral multiples of $\hbar\omega/2e$, where $\omega = 2\pi f$ is the angular frequency of the rf field [12,24], and the height of the kth order spike I_k at voltage V_k is proportional to the Bessel function of order k of the reduced ac voltage induced on the junction. When strong thermal fluctuations dominate the phase dynamics, and especially in our case where the typical frequency of phase-slip events $f_T \sim k_B T_n/h \sim 1.2 \times 10^{11}$ Hz is much greater than the frequency of the applied microwave field $f = 1.5 \times 10^{10}$ Hz, phase locking cannot be achieved and the I-V characteristics will exhibit a broad peak instead of discrete spikes. This has been formally calculated by Falci *et al.* [25] who obtain the following expression for the pair tunneling current:

$$I(V) = \sum_{k=-\infty}^{\infty} J_k^2\left(\frac{2eV_{\rm ac}}{\hbar\omega}\right) \times I_{\rm dc}\left(V - \frac{k\hbar\omega}{2e}\right), \qquad (3)$$

where J_k is the kth order Bessel function, $V_{\rm ac}$ is the induced ac voltage on the junction, and $I_{\rm dc}$ is the dc characteristics of the junction. Numerical evaluation of Eq. (3) shows a broad current peak that diminishes in magnitude and moves away from zero bias as the intensity of the radiation is increased, and the center of mass of the Shapiro spikes shifts to higher voltages. In Figure 4 we present (a) a calculation of the differential conductance dI/dV, due to pair tunneling according to Eq. (3), and (b) our numerically differentiated data, which is in very good agreement with the calculation. Note that the calculation does not account for the quasiparticle tunneling current background. The observed conductance peaks shift linearly to higher bias voltages with increased ac voltage as predicted by Eq. (3). At higher bias voltages (not shown) we observe the expected quasiparticle tunneling current with a gap edge that smears, due to photon assisted tunneling, with increasing microwave power.

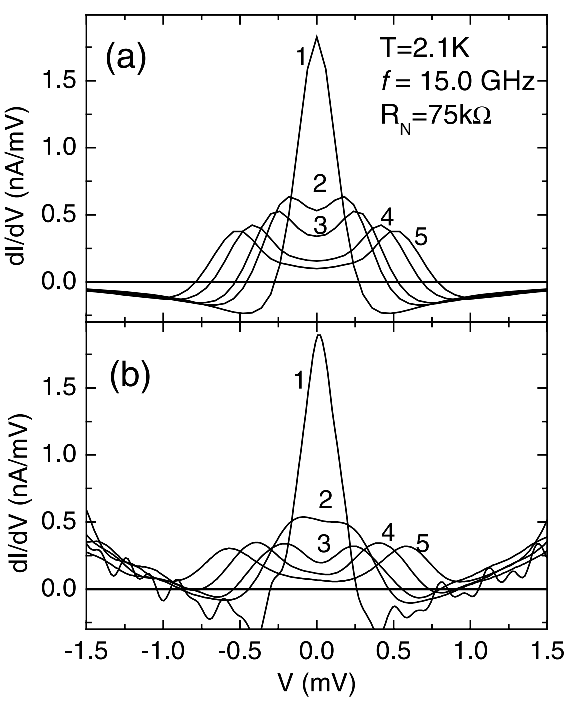

FIG. 4. (a) Calculation of the conductance at low bias voltages due to Josephson tunneling in the presence of microwave fields ($f = 15.0$ GHz) for various induced ac voltages on the junction. (b) Numerically differentiated current-voltage data for a junction with $R_N = 75$ kΩ at $T = 2.1$ K. Curves 1–5 correspond to $V_{\rm ac} = 0$, 0.31, 0.39, 0.54, and 0.65 mV, respectively.

To summarize, we have demonstrated a STM-based Josephson probe. We have shown that the tunneling characteristics obtained in this experiment are in good agreement with the model of fluctuation dominated Josephson tunneling. While the fluctuation dominated dc Josephson effect has been observed previously in ultrasmall planar junctions [18,22,23], this is to our knowledge the first observation of the corresponding ac effect in the presence of microwave fields. We have shown that the Josephson I_cR product can be determined locally from the junctions I-V curves, and we therefore expect that a Josephson STM will prove useful as a probe of the SC pair amplitude on length scales of the order of the coherence length in high-T_c materials. In addition, the ease with which the junction resistance in a STM configuration can be controlled, makes this system favorable for studying the effects of thermal fluctuations and the Coulomb blockade (at low enough temperatures) [26] on the Josephson phase dynamics.

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[1] See, for example, S. H. Pan, E. W. Hudson, K. M. Lang, H. Eisaki, S. Uchida, and J. C. Davis, Nature (London) **403**, 746 (2000).
[2] S. H. Pan, E. W. Hudson, and J. C. Davis, Appl. Phys. Lett. **73**, 2992 (1998).
[3] S. H. Pan, E. W. Hudson, A. K. Gupta, K.-W. Ng, H. Eisaki, S. Uchida, and J. C. Davis, Phys. Rev. Lett. **85**, 1536 (2000).
[4] Ch. Renner, B. Revaz, K. Kadowaki, I. Maggio-Aprile, and Ø. Fischer, Phys. Rev. Lett. **80**, 3606 (1998).
[5] Ch. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and Ø. Fischer, Phys. Rev. Lett. **80**, 149 (1998).
[6] C. Howland, P. Fournier, and A. Kapitulnik, cond-mat/0101251.
[7] T. Cren *et al.,* Phys. Rev. Lett. **84**, 147 (2000); T. Cren, D. Roditchev, W. Sacks, and J. Klein, Europhys. Lett. **54**, 84 (2001).
[8] J. Šmakov, I. Martin and A. V. Balatsky, cond-mat/0009310.
[9] O. Naaman, W. Teizer, and R. C. Dynes, Rev. Sci. Instrum. **72**, 1688 (2001).
[10] V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963).
[11] In Pb junctions, E_J is 78.8% of this value due to strong coupling effects.
[12] L. Solymar, *Superconductive Tunneling and Applications* (Wiley, New York, 1972).
[13] J. K. Gimzewski and R. Möller, Phys. Rev. B **36**, 1284 (1987).
[14] $S/I/N$ dI/dV curves taken with the same tip in the same run over a patch of bare graphite for a wide range of junction resistances are essentially identical. Since the power dissipated in the tip due to quasiparticle relaxation is inversely proportional to the resistance, we can rule out the importance of self-heating effects.
[15] K. K. Likharev, *Dynamics of Josephson Junctions and Circuits* (Gordon and Breach, New York, 1986).
[16] V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. **22**, 1364 (1969).
[17] M. Ivanchenko and L. A. Zil'berman, Zh. Eksp. Teor. Fiz. **55**, 2395 (1968) [Sov. Phys. JETP **28**, 1272 (1969)].
[18] Y. Harada, H. Takayanagi, and A. A. Odintsov, Phys. Rev. B **54**, 6608 (1996).
[19] G.-L. Ingold, H. Grabert, and U. Eberhardt, Phys. Rev. B **50**, 395 (1994).
[20] One would note that the present model of Eq. (2) neglects the effects of shot noise in the junction, as well as any correlations in the thermal noise. Both effects may additionally contribute to the seemingly elevated noise temperature. The more complex case of a nonwhite noise spectrum due to a general electromagnetic environment is treated by Ingold *et al.* in Ref. [19].
[21] W. C. Danchi, J. Bindslev Hansen, M. Octavio, F. Habbal, and M. Tinkham, Phys. Rev. B **30**, 2503 (1984).
[22] S. A. Buckner, J. T. Chen, and D. N. Langenberg, Phys. Rev. Lett. **25**, 738 (1970).
[23] C. J. Muller and R. de Bruyn Ouboter, Physica (Amsterdam) **194B–196B**, 1043 (1994).
[24] R. Kleiner *et al.,* Phys. Rev. Lett. **76**, 2161 (1996).
[25] G. Falci, V. Bubanja, and G. Schön, Z. Phys. B **85**, 451 (1991).
[26] K. K. Likharev and A. B. Zorin, J. Low Temp. Phys. **59**, 347 (1985).