Josephson Effect in Pb/I/NbSe$_2$ Scanning Tunneling Microscope Junctions

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We have developed a method for the reproducible fabrication of superconducting scanning tunneling microscope (STM) tips. We use these tips to form superconductor/insulator/superconductor tunnel junctions with the STM tip as one of the electrodes. We show that such junctions exhibit fluctuation dominated Josephson effects, and describe how the Josephson product $I_R N$ can be inferred from the junctions’ tunneling characteristics in this regime. This is first demonstrated for tunneling into Pb films, and then applied in studies of single crystals of NbSe$_2$. We find that in NbSe$_2$, $I_R N$ is lower than expected, which could be attributed to the interplay between superconductivity and the coexisting charge density wave in this material.

I. INTRODUCTION

Scanning tunneling microscopy (STM) has been proven to be an invaluable tool in the study of superconductors by serving as a probe of the density of states with very high energy resolution and sub-nanometer spatial resolution. STM has also been successfully used to create tunnel junctions on materials that do not readily form high quality planar junctions (e.g., in MgB$_2$), or in cases where a particular small region of the sample is of interest.

In recent years it has been realized that the use of superconducting (SC) STM tips may provide a complementary technique to conventional STM studies, since in SC STM both quasiparticle and Josephson tunneling can be measured locally with a scannable superconductor/insulator/superconductor (S/I/S) junction. Here we report on experiments we performed using SC STM tips. We measure the tunneling characteristics of Pb and NbSe$_2$ samples at $T$~2 K for various values of the junctions’ normal state resistances. For junction resistances below 100 kΩ we observe fluctuation dominated Josephson effects, and estimate the Josephson $I_R N$ product, a physically important quantity, for tunneling into NbSe$_2$.

II. EXPERIMENTAL TECHNIQUE

Superconducting tips are prepared according to Ref. 8 by deposition of Pb(5000 Å)/Ag(30 Å) proximity bilayers on conventional PtIr tips. Fig. I(a) shows a typical $dI/dV$ curve for tunneling at 2.1 K between a SC tip and a Pb(5000 Å)/Ag(30 Å) sample deposited on a graphite substrate at the same time as the tips were made. Sharp peaks are seen at voltages $eV = \pm 2 \Delta_{Pb}$, and the Pb phonon structure is clearly observed, confirming the high quality of the junction and that single-step tunneling is the dominant conduction mechanism. At these thicknesses (30 Å) the Ag contributes very little to the junction characteristics. The method described above for SC tip fabrication is robust and reproducible, yielding over 80% working tips, with virtually indistinguishable SC properties.

The measurement of the Josephson effect in STM junctions is challenging due to their small size and the associated high junction resistance. The energy scale for the phase coupling of a Josephson junction is the Josephson binding energy, $E_J = \Delta R_Q/2R_N$, where $R_Q = \hbar/4e^2 \sim 6.45$ kΩ, $\Delta$ is the SC gap, and $R_N$ is the normal state resistance of the junction. In junctions for which $k_B T \geq E_J$, phase locking is destroyed by thermal fluctuations, and no dc supercurrent can be observed. Indeed, for Pb junctions with resistances $\sim 100$ MΩ, the Joseph-
son binding energy is \( E_J/k_B \sim 0.5 \text{ mK} \), and we see no trace of the dc Josephson effect at our base temperature of 2.1 K (Fig. 1(a)).

Several authors have shown \(^{10,11}\) that when \( E_J \) is smaller than, but comparable to \( k_BT \), the phase dynamics in a small Josephson junction is dominated by thermal fluctuations. In this regime pair tunneling would be observed, but the pair current will be dissipative, with the voltage drop proportional to the rate of change of the relative phase across the junction. Ivanchenko and Zil’berman \(^{12}\) found that the current-voltage \( (I - V) \) relation for small \( E_J/k_BT \) due to this incoherent pair tunneling has the form:

\[
I(V) = \frac{I_c^2 Z_{env}}{2} \frac{V}{\sqrt{V^2 + V_p^2}},
\]

where \( I_c = 2eE_J/h \), \( Z_{env} \) is the effective impedance of the junction’s environment, and \( V_p = (2e/h)Z_{env}k_BT_n \) with \( T_n \) defined as the effective noise temperature.

Fig. 1(b) shows representative \( I - V \) curves taken on a Pb/Ag film at 2.1 K for various junction resistances \(^{13}\) below 120 kΩ. A current peak in the \( I - V \) characteristics emerges from the quasiparticle background as the junction resistance is lowered. The \( I - V \) curves can be fitted with good agreement to Eq. 1 for \( |V| < 1.0 \text{ mV} \), with \( A \equiv I_c^2 Z_{env}/2 \) and \( V_p \) as the only fitting parameters. The dependence of the quantity \( \sqrt{(4e/h)A/V_p} \) on \( G_N = 1/R_N \) should be linear with a slope equal to \( I_c R_N/\sqrt{k_BT_n} \). Fig. 2 shows such a plot for several Pb/Ag samples and a NbSe\(_2\) sample. Knowing the noise temperature \( T_n \), one can infer the Josephson product \( I_c R_N \).

Fluctuation dominated dc and ac Josephson effects were observed and carefully analyzed in Ref. \(^{13}\), confirming that the features we observe in the tunneling characteristics of these STM junctions are indeed signatures of pair tunneling. We have shown \(^{13}\) that the noise parameters \( T_n \) and \( Z_{env} \) depend only on the experimental setup and not on the sample studied or the tip used. Once these parameters are measured by determining a sample with a known \( I_c R_N \) (e.g. Pb), this quantity can be inferred for any sample measured under the same experimental conditions. \(^{14}\)

III. RESULTS AND DISCUSSION – NbSe\(_2\)

We apply the method described above to the measurement of the Josephson product for tunneling between a SC STM tip and a 2H–NbSe\(_2\) crystal. 2H–NbSe\(_2\) is a layered material that undergoes a charge density wave (CDW) transition at 33 K and a superconducting transition at 7.2 K. Niobium diselenide has been studied extensively in the past \(^{15,16,17}\), but to our knowledge, no reliable Josephson tunneling data exist for this system. While NbSe\(_2\) appears to be a conventional BCS superconductor, it shows anisotropy of the SC order parameter, transport and optical properties \(^{15}\), as well as coexisting CDW and SC states. These make NbSe\(_2\) an interesting system, especially since the high-T\(_c\) cuprates are believed to share some of these properties. \(^{16}\)

\( I - V \) curves were measured at 2.1 K on a 2H–NbSe\(_2\) single crystal \(^{18}\) cleaved at room temperature in vacuum (5 \times 10\(^{-7}\) torr). For junction resistances larger than 40 kΩ, the scaled tunneling current \( I(V) \times R_N \) is independent of \( R_N \). We use these curves to represent the background quasiparticle current, with no Josephson contribution, which we then subtract from the \( I - V \) curves measured at lower \( R_N \) to find the contribution of pair tunneling to the total current. This is done because junctions with NbSe\(_2\) as one of the electrodes have considerable quasiparticle weight below the gap even at this temperature of 2.1 K.

Fig. 2 shows a plot constructed from the fits to Eq. 1 for several Pb/Ag films and the NbSe\(_2\) data shown in the inset. From linear fits to the data we obtain the slopes 0.75 \( \pm 0.03 \text{ mV}^{1/2} \) and 0.46 \( \pm 0.03 \text{ mV}^{1/2} \) for Pb/Ag and NbSe\(_2\) respectively. The ratio of \( I_c R_N(\text{Pb}/\text{I}/\text{NbSe}_2) \) and \( I_c R_N(\text{Pb}/\text{I}/\text{Pb}) \) is thus 0.61 \( \pm 0.05 \). Since \( I_c R_N(\text{Pb}/\text{I}/\text{Pb}) = 1.671 \text{ mV} \) is known \(^{19}\) from the Ambegaokar-Baratoff (AB) formula \(^{19}\), we get \( I_c R_N(\text{Pb}/\text{I}/\text{NbSe}_2) = 1.02 \pm 0.08 \text{ mV} \).

The SC gap in NbSe\(_2\) can be described quite well by

\[
\begin{align*}
\Delta &= \frac{\hbar \bar{\Delta}}{2e} \quad \text{for} \quad |V| \ll 10 \text{ mV}, \\
\Delta &= \Delta_0 \quad \text{for} \quad |V| \gtrsim 10 \text{ mV},
\end{align*}
\]

where \( \Delta \) and \( \bar{\Delta} \) are the tunneling and superconducting gap values, respectively. The linear fits give \( \bar{\Delta} = 14.5 \text{ mV} \), and the data can be fitted on a linear scale above 50 mV.

\[\text{FIG. 2: Plot of } \sqrt{(4e/h)A/V_p} \text{ vs. } G_N \text{ for three Pb/Ag samples (Pb1, Pb2, Pb3) and a NbSe}_2 \text{ sample (diamond symbols). Solid lines are linear fits to the data. The error in } G_N \text{ is estimated at } 5\%. \text{ Inset: pair current vs. voltage for the NbSe}_2 \text{ sample, and junction resistances between 33 and 14.5 kΩ.}\]
an anisotropic s-wave gap function, with $\Delta(0)$ ranging from 0.7 to 1.4 meV over the Fermi surface (FS). Estimates using the AB formula for Pb/I/NbSe$_2$ junctions yield $I_c^{AB}R_N \sim 1.25$ mV, significantly higher than the one we observe, even when using the smallest gap parameter in NbSe$_2$. This suggests that the gap anisotropy alone cannot explain our result and we must look for additional possible explanations.

A further reduction in $I_cR_N$ may result from the existence of a CDW and its effect on the SC state. Gabovich et al. calculated the Josephson product for CDW and SDW superconductors and found that it decreases from the AB limit by an amount that depends on the ratio between the CDW and the SC energy gaps, and the degree to which the FS is affected by the formation of a CDW gap. In light of our results it appears that explanations along these lines should be sought, especially since the effects of a CDW may be important in the interpretation of Josephson data in high-$T_c$ cuprates.

In summary, we measured the Josephson effect in junctions formed between SC STM tips and Pb and NbSe$_2$ samples. In NbSe$_2$ we find the Josephson product $I_cR_N = 1.02 \pm 0.08$ mV. This result is smaller than expected from the AB formula, but may be described within the theory of Gabovich et al. for Josephson tunneling in CDW superconductors. These results suggest that the interplay between the SC state and the CDW may have an important role in Josephson tunneling.

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1. H. F. Hess et al., J. Vac. Sci. Technol. A 8, 450 (1990).
2. Ali Yazdani et al., Science 275, 1767 (1997).
3. S. H. Pan et al., Nature (London) 403, 746 (2000).
4. J. E. Hoffman et al., Science 295, 466 (2002), and references therein.
5. G. Rubio-Bollinger, H. Suderow, and S. Vieira, Phys. Rev. Lett. 86, 5582 (2001).
6. A. D. Truscott, R. C. Dynes, and L. F. Schneemeyer, Phys. Rev. Lett. 83, 1014 (1999).
7. S. H. Pan, E. W. Hudson, and J. C. Davis, Appl. Phys. Lett. 73, 2992 (1998).
8. O. Naaman, W. Teizer, and R. C. Dynes, Rev. Sci. Instrum. 72, 1688 (2001).
9. J. Šmakov, I. Martin, and A. V. Balatsky, Phys. Rev. B 64, 212506 (2001).
10. M. Ivanchenko and L. A. Zil’berman, Zh. Eksp. Teor. Fiz. 55, 2395 (1968) [Sov. Phys. JETP 28, 1272 (1969)].
11. V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. 22, 1364 (1969); G.-L. Ingold et al., Phys. Rev. B 50, 395 (1994); Y. Harada et al., Phys. Rev. B 54, 6608 (1996).
12. The junction normal resistances are determined from the slope of the $I - V$ curves for voltages above the sum of the gaps.
13. O. Naaman, W. Teizer, and R. C. Dynes, Phys. Rev. Lett. 87, 097004 (2001).
14. The noise temperature in this experiment is estimated to be $T_n = 57 \pm 5$ K. This is much higher than the bath temperature, and can be traced to noise sources at room temperature, and leakage of radiation into our cryostat.
15. T. Yokoya et al., Science 294, 2518 (2001).
16. S. V. Dordevic, D. N. Basov, R. C. Dynes, and E. Bucher, Phys. Rev. B 64, 161103 (2001).
17. E. W. Hudson, Ph.D. Thesis, University of California, Berkeley, 1999.
18. C. S. Oglesby, E. Bucher, C. Kloc, and H. Hohl, J. Cryst. Growth 137, 289 (1994).
19. V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. 10 486 (1963).
20. A. M. Gabovich, D. P. Moiseev, A. S. Shpigel, and A. I. Voitenko, Phys. Stat. Sol. 161, 293 (1990).
21. We use $\Delta_{Pb}=1.35$ meV, and include a factor of 0.788 due to strong coupling effects.