

Accomplishments

(1)

We have developed a superconducting scanning tunneling microscope (S-STM)^{1,2} which is a direct and local probe of the pair wave function of superconducting materials via the Josephson effect and quasiparticle spectra via scanning tunneling spectroscopy (STS). The novel feature of this device is a superconducting tip (Pb with an Ag capping layer) in close proximity to a superconducting sample to form a superconductor-insulator-superconductor (SIS) tunnel junction. The operation of this S-STM has been verified in the observation of Josephson tunneling between the tip and different sample systems including Pb films and NbSe₂. At the experimental base temperature ($T=2.1\text{K}$) and large junction normal resistance ($R_N > 10^4 \Omega$) the phase is not locked in the washboard potential, but is excited thermally to be diffusive. The observed Josephson current is therefore dependent on the bias voltage near zero bias due to the phase motion. The experimental data for a Pb film sample shown in Fig. 1 follow phase diffusion model proposed by Ivanchenko-Zil'berman, $I(V)=AV/(V^2+V_P^2)$ where $A=I_C^2 Z_{ENV}/2$ and $V_P=(2e/\hbar)Z_{ENV}k_B T_N$. The phase diffusion branch in the IV data is characterized by an effective noise temperature, T_N and the environmental impedance, Z_{ENV} . A plot of $[(4e^2/\hbar)A/V_P]^{1/2}$ vs $G_N (=1/R_N)$ gives us $I_C R_N/(k_B T_N)^{1/2}$ from the slope, and using the known value of $I_C R_N(\text{Pb})=1.671\text{mV}$, we can find T_N and Z_{ENV} for our STM Josephson junctions. Current values of these parameters measured in Berkeley are 15.1K and 294 Ω , respectively (Inset of Figure 1). Once we know these two parameters intrinsic to the STM Josephson junctions, we can derive the $I_C R_N$ product of the Josephson Effect into any material. In our study of the anisotropic layered material NbSe₂ as a precursor to Bi₂Sr₂CaCu₂O_{8+ δ} (BSCCO)³, the $I_C R_N$ product was in good agreement with the predicted value by Ambegaokar-Baratoff formula after some efforts to lower the noise temperature. In our previous proposal, the next material for study was chosen to be BSCCO.

(2) High T_C superconductors and spatial inhomogeneities

Recent conventional STM/STS studies⁴⁻⁷ of the high T_C superconductors reveal that there is a wealth of both spatial and spectral structure. It is clear that nanometer-scale inhomogeneities appear in the electronic spectra and, most likely, the superconducting energy gap⁴. Extensive experimental and theoretical studies of these materials support the concept of spatial inhomogeneity, spontaneous decomposition, ordering, stripes, etc., however, an important aspect remains unexplored. Specifically, does the superconducting order parameter also exhibit these spatial variations or will measurements show only an electronic spatial variation? Does the pseudogap signal a fluctuating superconductor? These questions motivated us to develop our S-STM in order to study the superconducting order parameter of BSCCO.

For the work on the S-STM, the major accomplishments in the current grant period are observations of c-axis Josephson tunneling between a conventional superconductor (Pb) and variously doped BSCCO samples. Our samples included an optimally doped one with $T_C=92\text{K}$

(OP92) and three overdoped samples with $T_c=79\text{K}$, 81K and 76K (OV79, OV81 and OV76, respectively). In all these measurements, we clearly saw the Josephson coupling. Samples were provided through our collaboration with Professor Yoichi Ando at Osaka University and Dr Shimpei Ono at the Central Research Institute of Electric Power Industry in Japan. Initial work investigated samples OP92 and OV79 that were provided several years ago. They have recently grown underdoped samples with $T_c=64\text{K}$ (UD64), OP94, OV81 and OV76 samples (6 to 12 single crystals for each doping).

BSCCO samples were cleaved in ultrahigh vacuum (5×10^{-8} Torr or below) at room temperature and cooled down *in situ* to the measurement temperature 2.1K . We obtained atomic resolution images of OP92 at 2.1K scanned by the superconducting tip. Figure 1(a) is a set of IV characteristics with the junction normal resistance, R_N , ranging from $4\text{M}\Omega$ to $5\text{k}\Omega$. The inset shows the large bias range measurement of the superconducting gap of OP92 taken at the same location before observing the Josephson currents. By defining 2Δ as the difference between the quasiparticle (QP) coherence peaks we derive $\Delta=53\text{meV}$. The IVs of higher R_N in the main frame clearly show the Pb gap at 1.38mV , confirming that we do have vacuum tunneling between the tip and the sample. As the R_N is gradually decreased, the phase diffusion branch near zero bias appeared, indicating fluctuating Josephson coupling. Figure 1(b) magnifies the IVs of Figure 2(a) in the region near zero bias voltage for the three lowest R_N values. To extract the pair tunneling current, we subtracted the QP background below the Pb gap using the average of several scaled IVs taken with slightly higher R_N (inset of Figure 1(b)). This subtraction scheme is used for all the data sets shown in this report. Figure 1(c) shows the good agreement of the phase diffusion model fits (symbols) to the fluctuated Josephson current measurements (lines). The slope of the plot in Figure 1(d) yields the $I_c R_N$ product; an important quantity measuring the strength of the Josephson coupling. We constrain the fit to the origin on physical grounds: at zero conductance, there should be no current. This fit yields an $I_c R_N$ of $33\mu\text{V}$. We do not observe any Josephson coupling when Δ is more than 70meV , but we still do not understand this

Higher spatial resolution spectroscopic measurements were also performed on a length scale of a few tens of angstroms. Results are in Figure 2(a). The inset shows the local density of states (LDOS) taken at the point of the surface before and after the low R_N (in other words high current) measurements. It clearly shows typical dI/dV (black curve) taken on the BiO plane before the low R_N measurements, but after the high current density measurements, the superconductivity of the sample at that point appears to have changed (red curve). Again we observe good phase diffusion model fits to the observed thermally fluctuated Josephson currents in Figure 2(c). This result gives us confidence that we are in fact observing the pair tunneling between Pb and OV-BSCCO. Moreover, in the $I_c R_N$ plot of Figure 2(d), the measured $I_c R_N$ of $334\mu\text{V}$ is much higher than the OP sample result. Figure 3 shows the data taken at the different spatial point in the same experimental run for that shown in Figure 2. Note that the LDOS (the inset of Figure 3(a)) has well defined gap even after the low R_N measurements, but the lowest R_N is larger than that used for the measurements in Figure 2. Our preliminary results also indicate that the superconducting gap of BSCCO (difference between the coherence peaks) becomes bigger as R_N is decreased. The LDOS finally becomes a pseudogap-like spectrum with no coherence peak. Moreover, we often observe that the Josephson currents will disappear when R_N is decreased further and then reappears when it is again increased. This result is unexpected. In figure 4 we show the $I_c R_N$ plot of sample OV76 measured at 2.1K . Each Josephson current is

labeled in the chronological order in which it was taken. The Josephson coupling increases as we decrease the tunnel resistance (expected) but then unexpectedly disappears at a lower R_N !

Increasing R_N (decreasing G_N) results in the Josephson coupling returning. This behavior is all unexpected and requires further work.

The $I_C R_N$ products over the surface for OP and OV samples are plotted as a function of the superconducting gap of BSCCO, Δ in Figure 4. The $I_C R_N$ product becomes maximum near the BSCCO gap, $\Delta \sim 45\text{meV}$ (Δ_{AVE} , the average gap observed by other groups for similar doping samples) and then decreases as the measured Δ increases further, indicating an inverse relation. The spatial studies of both $I_C R_N$ products and Δ for OV79 are in Figure 5. The spectra in Figure 7(c) are measured along the arrow shown in Figure 5(a). We can see the spatial variations and the inverse relation on the length scale of the superconducting coherence length, ξ .

Summarizing our observations:

- (1) **C-axis Josephson couplings** between Pb and both OP and OV-BSCCO. This is surprising if BSCCO is strictly a d-wave superconductor
- (2) $I_C R_N$ of the OP sample seemed to be much smaller than those of OV samples
- (3) **$I_C R_N$ inhomogeneity** is correlated with the gap inhomogeneity on the length scale of ξ in the OV samples
- (4) **Inverse correlation** between $I_C R_N$ and Δ in OV samples
- (5) Degradations of the superconductivity of BSCCO by high current density

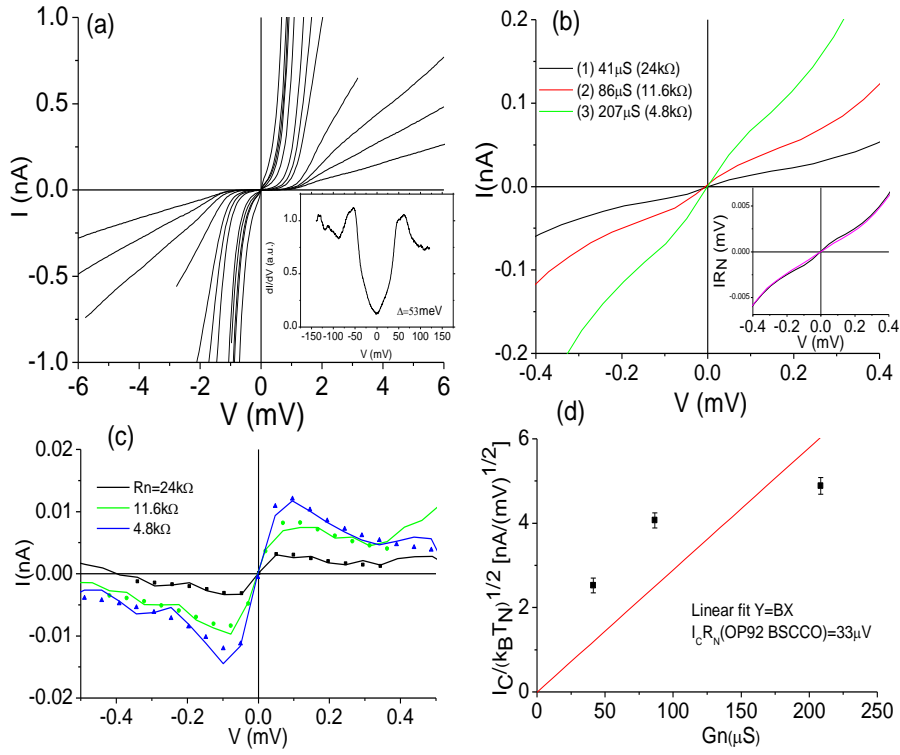


Figure 1: **a**, SIS IVs measured in **Pb/I/OP92** STM Josephson junctions at 2.1K. Inset: dI/dV taken at the same point. $\Delta=53\text{meV}$. **b**, IVs before QP background subtraction, magnified in the region near zero bias voltage. Inset shows an averaged scaled IV for QP background (pink curve). **c**, Thermally fluctuated Josephson currents (solid lines) between **Pb** and **OP92** sample measured at three different R_N at 2.1K with QP background subtracted. Dots represent the phase diffusion model fits. **d**, $I_C R_N$ plot for **Pb/I/OP92** STM Josephson junctions with $I_C R_N$ product $33\mu\text{V}$ derived from a linear fit $Y=BX$ (see the text)

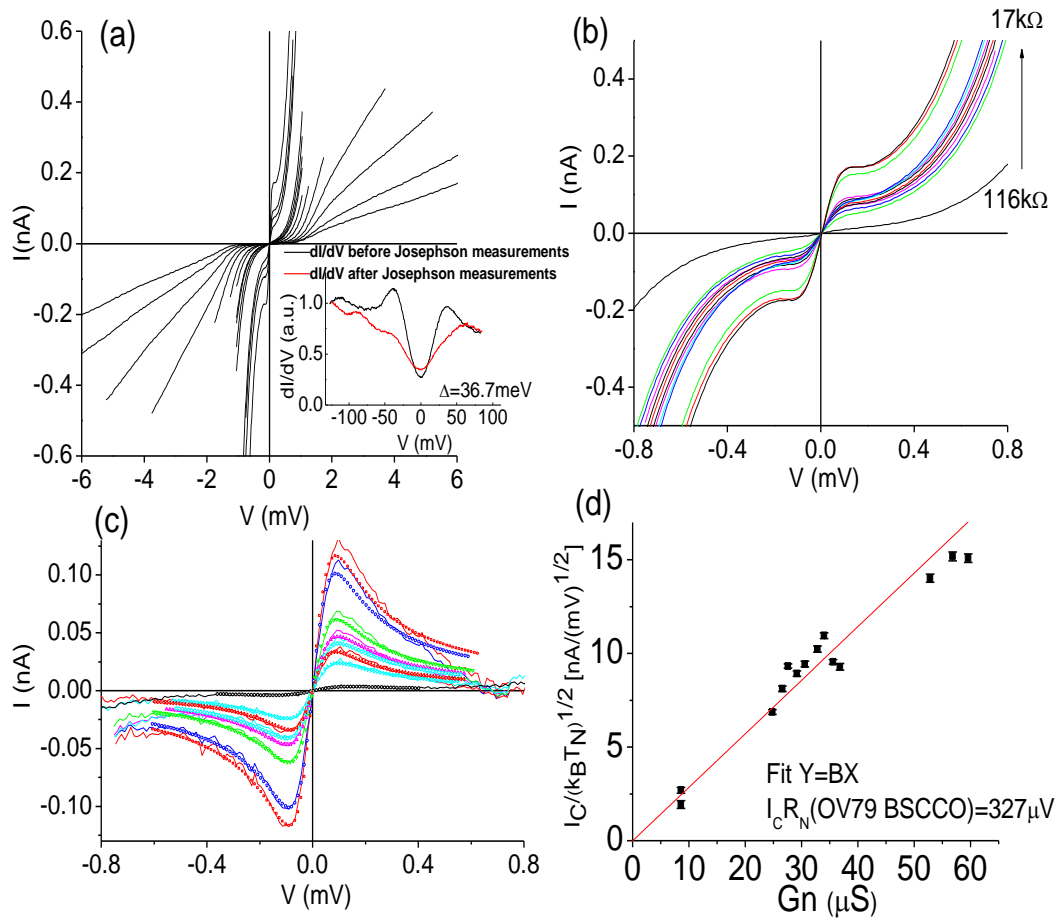


Figure 2: **a**, SIS IVs of **Pb/I/OV79** STM Josephson junctions measured at 2.1K with varying R_N . Inset: dI/dV taken before (black) and after (red) these Josephson measurements. Note that LDOS changed drastically after Josephson measurements. **b**, IVs with the phase diffusion branches near zero bias voltage. **c**, Thermally fluctuated Josephson currents (solid lines) between **Pb** and **Ov79** (QP background subtracted) and the phase diffusion model fits (dots). **d**, $I_C R_N$ plot for **Pb/I/OV79** STM Josephson junctions.

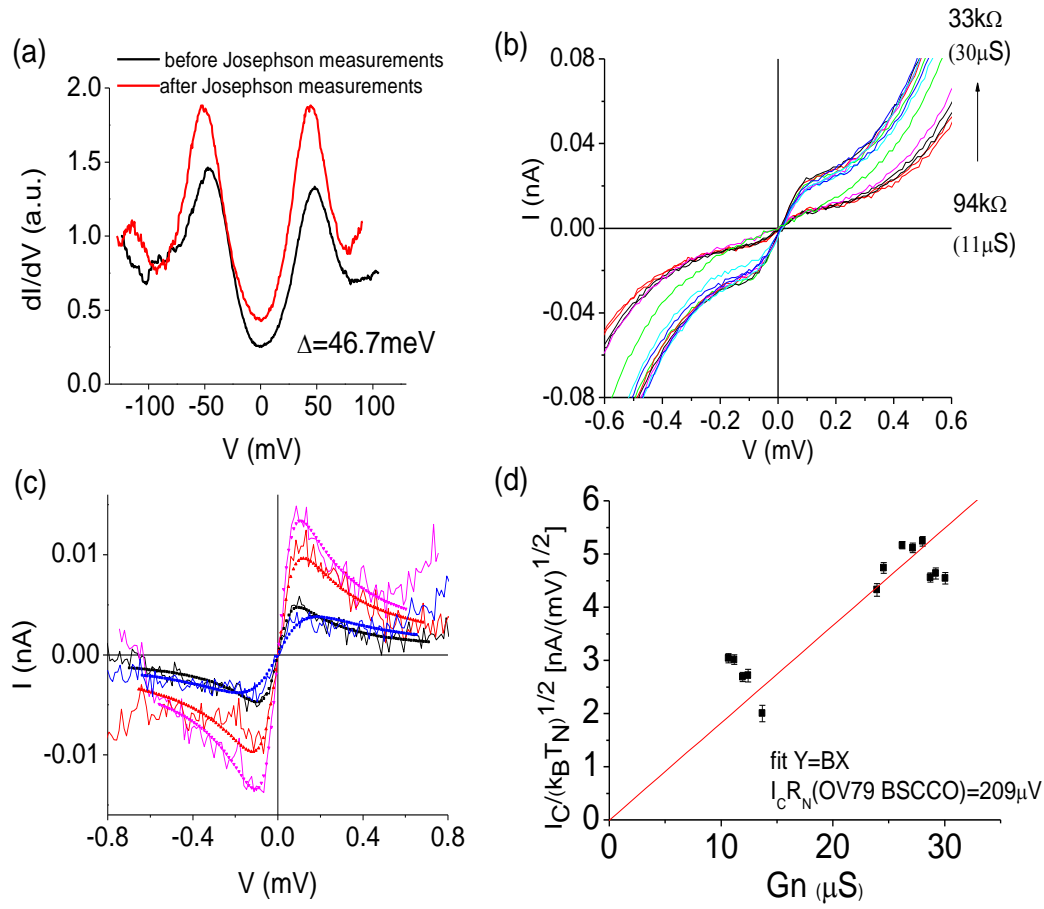


Figure 3: **a**, dI/dV of **OV79 BSCCO** taken before (black) and after (red) Josephson measurements taken at a different surface point in the same experimental run as the data shown in Figure 3. **b**, IVs of **Pb/I/OV79** STM junctions near zero bias voltage. **c**, Thermally fluctuated Josephson currents (solid lines) and the phase diffusion model fits (dots). **d**, $I_C R_N$ plot. Note that the highest G_N (lowest junction resistance R_N) is smaller (larger) than that in Figure 3.

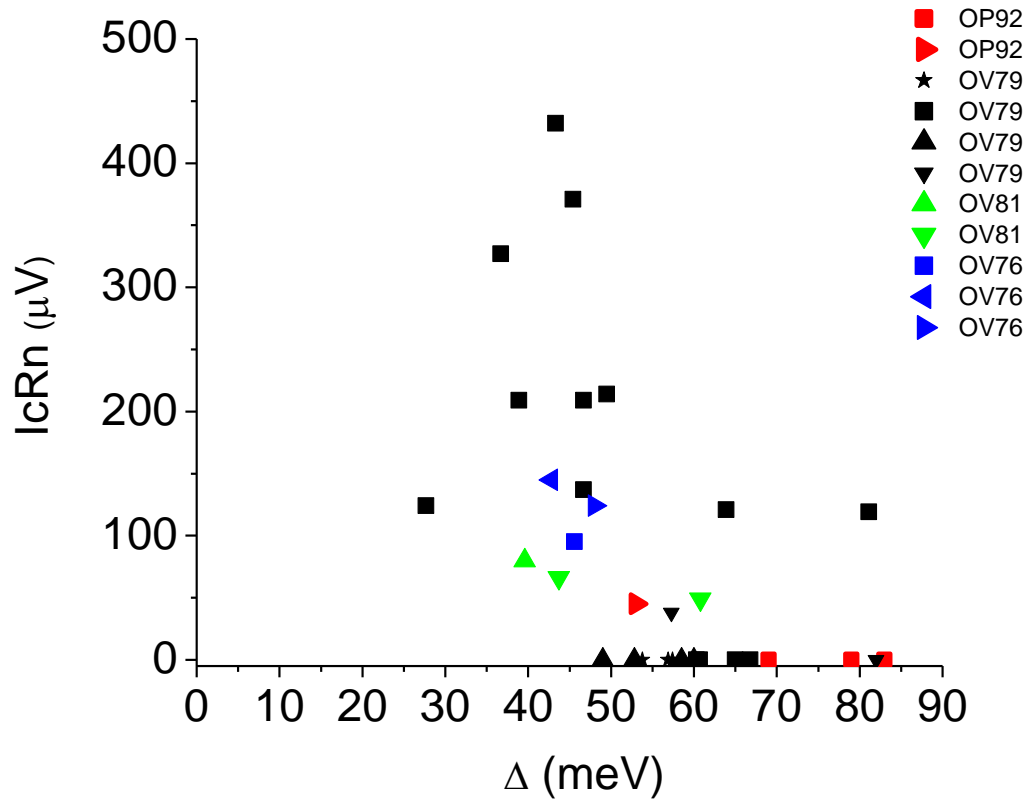


Figure 4: **a**, All the data sets of the superconducting gap of BSCCO, Δ and $I_c R_n$ derived from linear fit $Y=BX$ for OP and OV samples, indicating the inverse relation (see text).

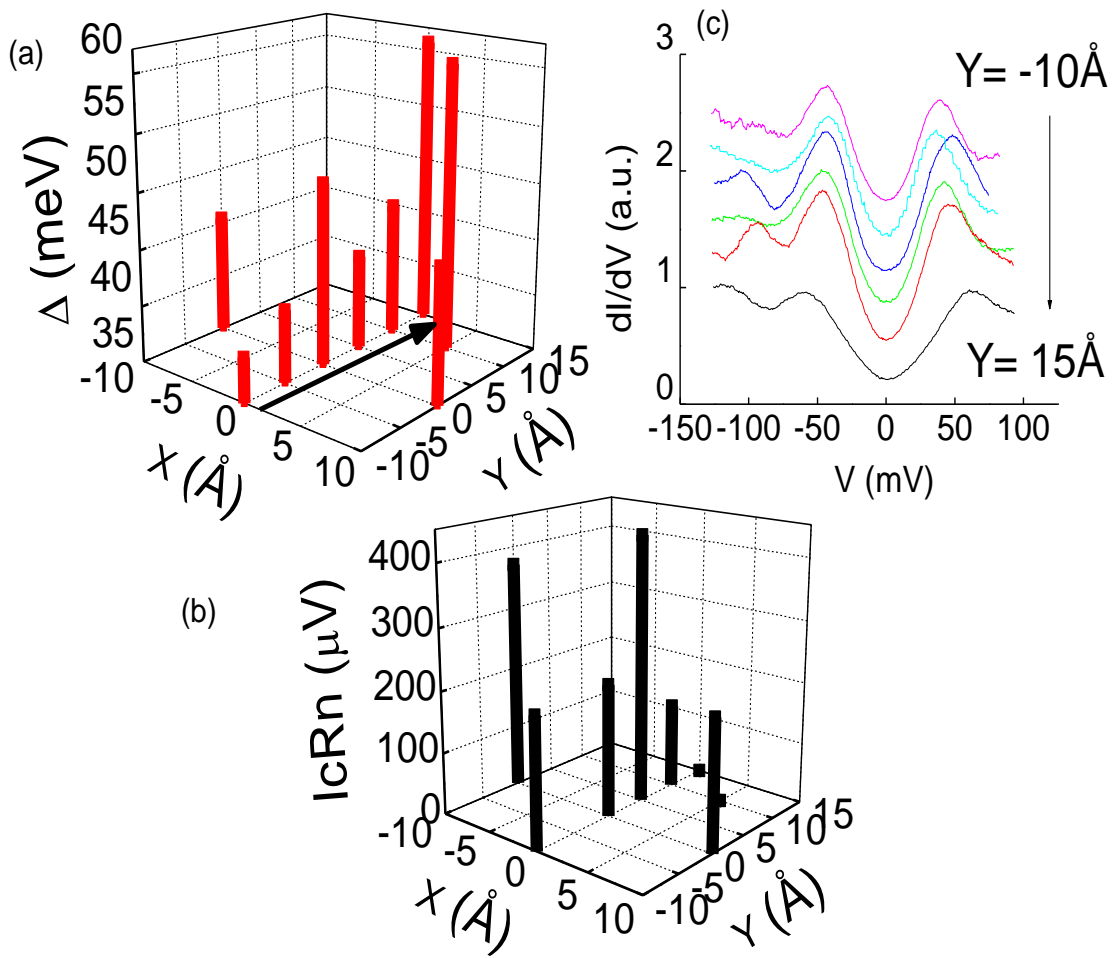


Figure 5: **a**, Spatial dependence of Δ of the **OV79** sample **b**, spatial dependence of the corresponding $I_c R_n$ products. $I_c R_n$ products were derived from linear fit $Y=BX$. Note that zero Josephson couplings where $\Delta \sim 60\text{meV}$. **c**, dI/dV spectra measured along the arrow shown in **a**.

Publications

1) Scanning Josephson tunneling microscopy of single-crystal $\text{Bi(2)Sr(2)CaCu(2)O(8+\delta)}$ with a conventional superconducting tip

By: Kimura, Hikari; Barber, R. P., Jr.; Ono, S.; et al.

PHYSICAL REVIEW LETTERS Volume: 101 Issue: 3 Article Number: 037002

Published: JUL 18 2008

2) Hikari Kimura, R. P. Barber, Jr., S. Ono, Yoichi Ando, and R. C. Dynes, “*Josephson scanning*

tunneling microscopy: A local and direct probe of the superconducting order parameter”

Physical Review, B 80, 144506 _2009_

3) S. A. Cybart, S. M. Anton, S. M. Wu, J. Clarke, and R. C. Dynes, “*Very large scale integration of nanopatterned $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ Josephson junctions in a two-dimensional array*”, **Nano Letters**, nl901785j, 2009.

4) S. M. Wu, S. A. Cybart, P. Yu, M. D. Rossel, J. X. Zhang, R. Ramesh and R. C. Dynes, “*Reversible control of exchange bias in a multiferroic field effect device*”, **Nature Materials**, nmat2803, 2010.

5) S. A. Cybart, T. Dalilouch, , S. M. Wu, S. M. Anton, J. A. Drisko, J. M. Parker and R. C. Dynes, “*Measurements and simulations of two dimensional arrays of SQUIDs from ion damage Josephson junctions*”, accepted **J. Appl. Phys.** **August 2012**

6) C. Hwang, T. G. Rappaport, P. Yu, M. B. Silva Neto, S. A. Cybart, S. J. Shin, A. V. Fedorov, E. E.

Haller, R. C. Dynes, R. Ramesh, D. H. Lee, and A. Lanzara, “*Complex Magnetic Ordering in Graphene Compounds*”, submitted, **August 2012**

7) S. A. Cybart, P. Roediger, T. J. Wong, E. Ulin-Avila, and R. C. Dynes, “*Angled reactive ion etching of high-aspect-ratio nanostructures*”, submitted, **Appl Phys Lett.** **August 2012**

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