Vacuum tunneling of superconducting quasiparticles from atomically sharp scanning tunneling microscope tips

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We report on the study of atomically sharp superconducting tips for scanning tunneling microscopy and spectroscopy. The results clearly show vacuum tunneling of superconducting quasiparticles from atomically sharp tips. Observed deviations of the energy gap of the superconducting tip from its bulk value are attributed to the proximity effect. We show that a combination of a superconducting tip and an atomic resolution scanning tunneling microscope provides a means of achieving very high resolution local spectroscopy. We also discuss how this combination paves the way for a number of important applications. © *1998 American Institute of Physics.* [S0003-6951(98)04446-5]

Since early in its history, scanning tunneling microscopy (STM) with nonsuperconducting tips has been used to successfully study the local properties of superconducting samples.[1–4] Interestingly, although it has long been proposed,[5] use of superconducting tips for STM has not been achieved. We note that a number of important experiments have used scanning tunneling microscopes to create adjustable microscopic junctions with superconducting electrodes.[6–8] We distinguish these techniques from the use of a sharp superconducting tip (having one or a few atoms at the end), with vacuum tunneling, for atomically resolved microscopy and spectroscopy. One might naturally ask whether such a sharp tip could even exhibit a superconducting density of states (DOS) at the end. Recently, Yazdani *et al.* noted the existence of a BCS single electron excitation spectrum above a normal metal adatom on a superconducting Nb substrate.[9] This implies that atomically sharp superconducting STM tips might be achievable. In this letter we report the successful realization of such tips, and describe their application to the study of both normal and superconducting samples.

For this work, we use a very-low-temperature STM,[10] which has spectroscopic resolution better than 30 μV at its base temperature of 250 mK. The superconducting tips are made from 0.2 mm diam Nb wire by mechanical sharpening. The tip installation is carried out with the STM open to air at room temperature. The STM chamber is then sealed, evacuated to 10^{-6} Torr, and cooled to 4.2 K. The natural oxide of Nb at the apex of the tip is removed, and the tip is further sharpened, by field emission against a Au target in cryogenic ultrahigh vacuum at 4.2 K. After this process, the sharpness of the tip is verified by identification of atomic scale features on the Au surface.

Next, the relationship between tunneling current I and tip displacement from the surface z is recorded to confirm the characteristic exponential behavior of vacuum tunneling [$I \propto \exp(-z/z_0)$ where z_0 is a decay constant]. From these data, the effective barrier height, which represents the convoluted work function of the tip and the sample, can be extracted.[11] As an example, the results of a I vs z measurement for a Nb tip is plotted in log/linear format in Fig. 1(a). The curve is linear over three decades of current, which confirms the vacuum nature of the tunneling, and a work function of 2.9 eV is deduced from its slope.

To evaluate the superconducting properties of this Nb tip, the differential conductance $G(V) = dI/dV$ of tunneling between tip and sample is measured as a function of bias voltage V.[12] This function is a measure of the convoluted local DOS of the sample and the tip.[13] We use the Au field

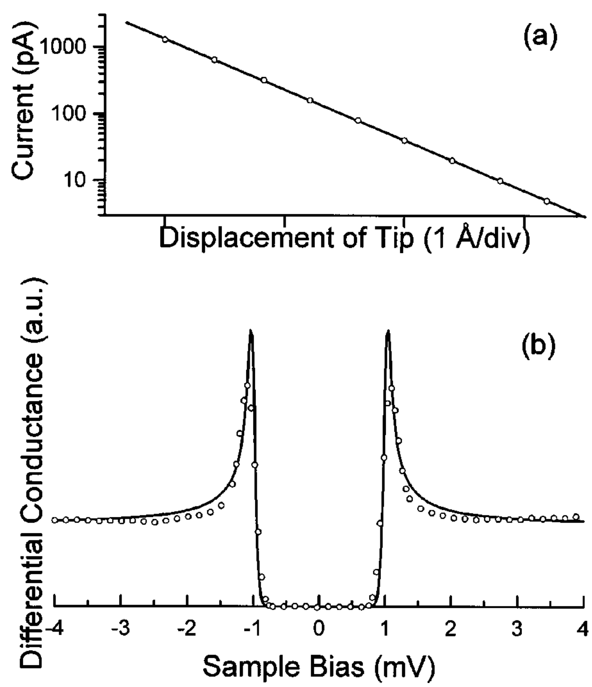

FIG. 1. (a) A log/linear plot of the tunneling current vs displacement of a Nb tip from a Au surface. The circles are the measured data, and the solid line is the linear fit. The exponential relationship over three decades of current indicates the vacuum nature of the tunneling. (b) Tunneling conductance vs sample bias for the same Nb tip at 335 mK. The solid line is the calculated conductance using a BCS DOS and an energy gap $\Delta_{tip}(0) = 1.0$ meV. The circles are the measured data (only a fraction of the measured data points are shown to allow clear display of the underlying calculated curve). The spectrum indicates a superconducting quasiparticle DOS at the tip end.

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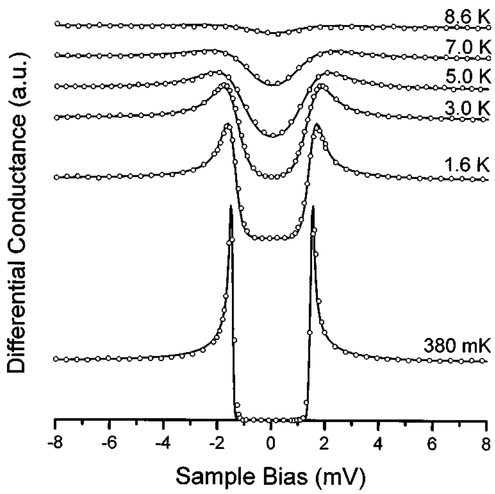

FIG. 2. Measured differential conductance spectra (circles) for tunneling between a Nb tip and a Au surface at several temperatures. The spectra are displaced vertically from each other for clarity. The solid lines are the calculated conductance using a BCS DOS with $\Delta_{tip}(0)=1.47$ meV and the temperatures shown. The excellent agreement of the data and the calculations confirms that the temperature dependence of the tip DOS is very similar to that of bulk Nb.

emission target as the sample. Figure 1(b) shows the resulting differential conductance spectrum taken at 335 mK. It clearly demonstrates a superconducting quasiparticle DOS, as evidenced by the zero of conductance below the gap voltage and the high peaks at the gap edge. To clarify that the superconducting features in this spectrum are due to the tip, and not due to possible deposition of Nb onto the surface during field emission, this measurement is performed at several different locations separated by more than 100 nm from the location where the field emission was performed. The absence of significant variations between the spectra at different locations indicates that the energy gap in the DOS observed is that of the tip.

It is interesting to note that the measured superconducting energy gap value of the tip Δ_{tip} varies from tip to tip after the field emission process, ranging from a few tenths of an meV to near the bulk gap value of Nb [1.53 meV at $T=0$ K (Ref. 14)]. The origin of this deviation is not fully understood. One possible explanation is that the superconductivity observed at the end of the tip has propagated from the bulk due to the proximity effect.[15] Thus the measured gap will depend on the dimensions, structure, and composition of the apex of the tip, which can be modified during field emission.

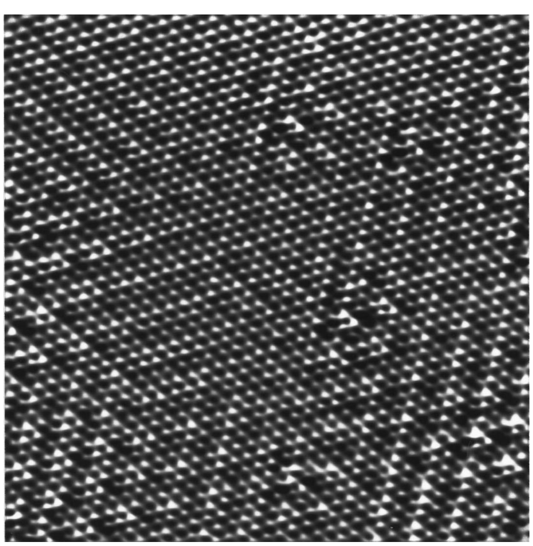

FIG. 3. Topographic image (150 Å square) of a $NbSe_2$ surface acquired at 4.2 K in constant current mode with the superconducting tip whose tunneling spectrum is shown in Fig. 2. A charge density wave pattern is clearly visible. The atomic resolution of the image indicates that this superconducting tip is atomically sharp.

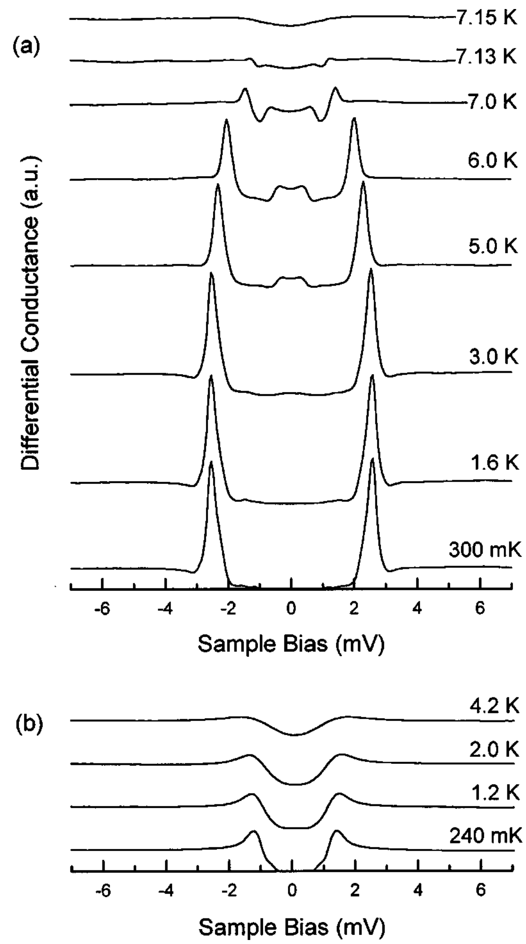

FIG. 4. (a) Measured differential conductance spectra for tunneling between a superconducting Nb tip and a $NbSe_2$ surface. The characteristic SS tunneling peaks $\Delta_{tip}+\Delta_{NbSe}$ and $\Delta_{tip}-\Delta_{NbSe}$ vary in voltage and change in magnitude, with temperature, as expected. (b) Measured differential conductance spectra for tunneling (NS) between a PtIr tip and a $NbSe_2$ surface. Comparison between the SS and NS tunneling spectra, plotted in the same scale, shows the dramatic enhancement in sharpness of the features obtained with a superconducting tip.

In Fig. 2, differential conductance spectra from another Nb tip (prepared and evaluated for its work function and sharpness as described above) are shown from 8.6 K to 380 mK. Each trace is displaced vertically for clarity. One can clearly see the superconducting gap in the DOS developing at the tip with falling temperature. The solid lines in Fig. 2 are the expected spectra for each temperature, calculated from a BCS DOS using a value of the energy gap for the tip $\Delta_{tip}(0)=1.47$ meV. Note that the agreement between the measured data and calculations is much better than in Fig. 1(b). In general, the smaller the deviation of the measured gap from its bulk value, the better the agreement with the BCS DOS, especially near the edge of the gap. This is an expected consequence of the proximity effect.[16]

To study superconducting–tip scanning tunneling microscopy on a superconductor, we now use this tip to perform measurements on a sample of $NbSe_2$. This is a well-studied layered material, which shows a charge density wave, and has a superconducting phase transition at $T_c = 7.2$ K. The superconducting energy gap, as measured by STM, is $\Delta_{NbSe}(0) = 1.11$ meV.[3] A $NbSe_2$ single crystal sample is cleaved *in situ* at 4.2 K and exchanged for the Au sample. Figure 3 shows an image of the $NbSe_2$ surface at $T = 4.2$ K, taken with the tip whose tunneling conductance spectra are shown in Fig. 2. Both the atomic lattice on the surface and the charge density wave are clearly resolved. This confirms that the tip is atomically sharp. Again, conductance spectroscopy is carried out on this $NbSe_2$ surface as a function of temperature and the results are displayed in Fig. 4(a).

The two pairs of peaks in Fig. 4(a) are characteristic of the superconducting–superconducting (SS) nature of the tunneling, and clearly distinguish it from normal–superconducting (NS) tunneling. The experimental values of $V = \pm(\Delta_{tip} \pm \Delta_{NbSe})/e$ at which they occur are in excellent agreement with the values calculated from the BCS theory[17] at all temperatures measured. This confirms not only the existence of a superconducting DOS at the end of the tip, but also that tunneling in SS-STM is physically similar to that in planar junctions, albeit in a junction of far smaller area.

The use of superconducting tips in low-temperature scanning tunneling microscopes has a number of significant advantages. Most important of these is the improvement in spectroscopic resolution when compared to normal–tip STM. This is due to the sudden rise in conductance at the gap edge and the associated high peak in the DOS of the superconducting tip.[18] An example of this can be seen by comparing the SS tunneling of Fig. 4(a) with the NS tunneling of Fig. 4(b), where we show the conductance spectra acquired on $NbSe_2$ with a nonsuperconducting PtIr tip. There is a dramatic enhancement of sharpness of the features in the spectrum obtained with a superconducting tip, which could prove significant in many scanning tunneling spectroscopy (STS) measurements, since it exists even at the commonly available temperature of 4.2 K.

Of even greater significance is the fact that superconducting-tip STM opens the door for many important future applications. One example is spin polarized STM and STS. Here, the Zeeman splitting of the quasiparticle DOS for the two spin orientations in the superconducting tip can provide a spin polarized electron source[5] capable of being scanned, with atomic resolution, over the surface to be studied. Another example is Josephson tunneling in SS-STM. This would be significant in its own right, since it would provide a new type of tunable Josephson junction. It could also become a useful instrument to study, for example, spatial variations of the order parameter in exotic superconductors.[19]

In conclusion, reliable operation of atomically sharp superconducting Nb tips, with vacuum tunneling, in low-temperature STM and STS has been achieved. The measured energy gap at the ends of these tips varies, and we attribute deviations from the bulk value to the proximity effect. SS tunneling between a Nb tip and a $NbSe_2$ surface is studied from 250 mK to 9 K, and compared to NS tunneling with a PtIr tip under similar conditions. This mode of STM not only provides a much improved spectroscopic resolution, but also opens the door for many future applications, including scanning spin-polarized electron tunneling, and scanning Josephson tunneling.

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