Scanning Tunneling Spectroscopy of a Vortex Core from the Clean to the Dirty Limit

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The local density of states of a superconducting vortex core has been measured as a function of disorder in the alloy system $Nb_{1-x}Ta_xSe_2$ using a low-temperature scanning tunneling microscope. The peak observed in the zero-bias conductance at a vortex center is found to be very sensitive to disorder. As the mean free path is decreased by substitutional alloying the peak gradually disappears and for $x=0.2$ the density of states in the vortex center is found to be equal to that in the normal state. The vortex-core spectra hence may provide a sensitive measure of the quasiparticle scattering time.

Recent low-temperature scanning tunneling microscope (STM) studies of the vortex lattice in $2H$-NbSe$_2$ [1] have revived theoretical [2-5] and experimental [6] interest in the electronic structure of flux lines in type-II superconductors. The STM permits a direct measurement of the *local* density of states with atomic resolution. Prior to these first STM measurements, the spatial variation of the density of states in the mixed state of a superconductor was inaccessible to experimental techniques. This quantity is obtained by measuring the local tunneling conductance versus bias voltage as a function of position. It is in fact this spectroscopic capability of the STM which allows one to differentiate between core regions and superconducting regions in the mixed state and, hence, to obtain an image of the vortex lattice in a superconductor. The first observation of the Abrikosov vortex lattice using an STM by Hess *et al.* [1] relied on the change in differential conductance at the energy gap of the superconductor to provide the contrast. These first measurements also revealed an unexpected peak in the differential conductance at zero bias at the center of a vortex. This peak has since been associated with low-lying quasiparticle bound states in the vortex core [2-4] and qualitative agreement between the experimental conductance curves and theory has been obtained. In addition, new radial structure in the vortex-core density of states has been predicted [2] and subsequently confirmed by experiment [7].

Although low-temperature STM holds promise as a powerful local probe of superconductivity, only pure $2H$-NbSe$_2$ has been investigated by this technique at present. NbSe$_2$ is a clean type-II superconductor with a transition temperature (T_c) of 7.2 K and a charge-density-wave (CDW) transition at 33 K which creates a gap in a fraction of the Fermi surface at low temperature. In this work we consider the effect of changing the superconductor's material properties in a systematic manner on the vortex-core spectra. In particular, we have investigated the evolution of the zero-bias conductance peak (ZBCP) observed in the center of a vortex

core as a function of disorder.

We have studied a range of compositions of the layered compound $2H$-Nb$_{1-x}$Ta$_x$Se$_2$ from $x=0$ to $x=0.2$. Previous work on this material system [8] shows that substitution of 3% of Ta suppresses the CDW transition found in pure NbSe$_2$ and further substitution decreases T_c, so that for $x=0.2$, $T_c=5$ K. Since Nb and Ta are isoelectronic and have similar atomic radii, large changes in the electronic properties are not expected with alloying; however, the mean free path is found to systematically decrease with increased Ta substitution [9]. Thus upon substitution one can continuously vary the properties from those of a clean superconductor to one approaching the dirty limit.

The ideal nature of the surface of these layered compounds plays an important role in the success of this experiment. Single crystals are grown by the usual iodine-assisted chemical transport reaction at 660-720 °C. The samples are cleaved in air just prior to installation in our low-temperature STM [9] and topographic images of the surface routinely show atomic resolution with large flat plateaus even up to 1×1-μm^2-size scales. The STM is situated in the bore of an 8.5-T superconducting magnet and is placed in 1.5 mTorr of He exchange gas in a vacuum can which separates it from the helium bath. The temperature of the STM can be lowered to 1.3 K and the magnetic field is always applied perpendicular to the basal plane of the crystals. For the following experiments chemically etched gold tips were used.

To image the vortex lattice we have used a scanning tunneling spectroscopy (STS) method which highlights the vortex-core peak structure directly. A small dither voltage of 400 μV is applied to the sample bias voltage, and the resulting ac current, which is proportional to the differential tunneling conductance, is detected with a lock-in amplifier. Images are acquired typically with a sample bias of 0.1 V and tunneling current of 0.5 nA. At each point in the STS image we freeze the tip and measure the differential conductance at zero bias and normalize this to the differential conductance at a voltage slight-
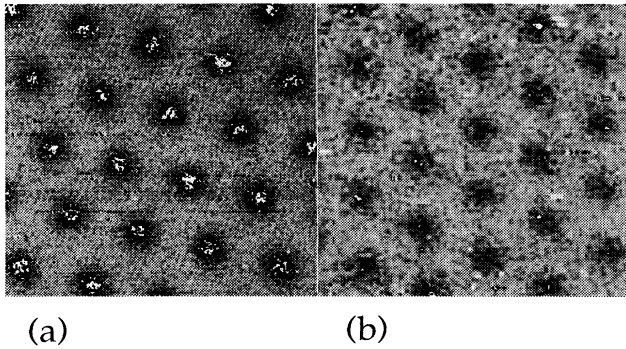
(a)   (b)

FIG. 1. 4000 Å×4000 Å STS images of the vortex lattice at 1.3 K and 0.3 T: (a) $x=0.00$ sample (pure NbSe$_2$); note that regions in which the conductance ratio is greater than 1 have been artificially saturated to white to highlight the spatial extent of the ZBCP in the vortex cores. (b) $x=0.20$; the ZBCP is absent in this case.

FIG. 2. Spectra acquired away from the flux lines at 1.3 K and 0.3 T for $x=0.00$ and $x=0.15$. A fit by a pair-breaking model of the density of states is indicated by the dashed lines. The fitting parameters are, for $x=0$, $\Delta=1.3$ meV, $\Gamma=0.51$ meV, and for $x=0.15$, $\Delta=1.0$ meV, $\Gamma=0.46$ meV. In both cases the temperature is taken to be $T=1.3$ K.

ly larger than the superconducting gap. Values of this ratio which are larger than 1 represent an increase in conductance at zero bias with respect to the background junction conductance. Figure 1 shows STS images acquired in this manner on two samples with (a) $x=0.00$ (pure NbSe$_2$) and (b) $x=0.20$ at a temperature of 1.3 K and magnetic field of 0.3 T. The images are 4000 Å ×4000 Å with white representing regions of low conductance ratio (0.5) and black (1.0) representing regions of higher conductance ratio (vortex-core regions). The vortex lattice is clearly observed with a spacing consistent with the applied magnetic field ($a_0=893$ Å). Note that in the vortex cores, regions with enhanced conductance relative to the normal state (conductance ratio greater than 1) are artificially colored white to highlight the spatial extent of the ZBCP. Note that these regions are absent in the $x=0.20$ sample. For the range of compositions studied the vortex lattice is found to be aligned with the atomic lattice as is found in pure NbSe$_2$ [1].

On the basis of these images the vortex cores are located and detailed conductance measurements are made. Figure 2 shows representative spectra outside the core regions in $x=0.00$ and $x=0.15$ Nb$_x$Ta$_{1-x}$Se$_2$ at $T=1.3$ K and a field of 0.3 T. A form typical of a normal-metal–superconductor tunnel junction is observed. Comparison with BCS theory, however, reveals that these spectra show an enhanced conductance at zero bias and the peaks at the gap edge are reduced relative to those expected. These spectra can be fitted using a pair-breaking model which phenomenologically incorporates the effects of inelastic scattering on the density of states [10]:

$$N(E,\Gamma)=\mathrm{Re}\frac{|E-i\Gamma|}{(E-i\Gamma)^2-\Delta^2},$$

where Δ is the superconducting gap and Γ is a pair-breaking parameter. The fit parameters are noted in the figure caption. The experimental temperature of 1.3 K is used to calculate the normalized conductance. There are
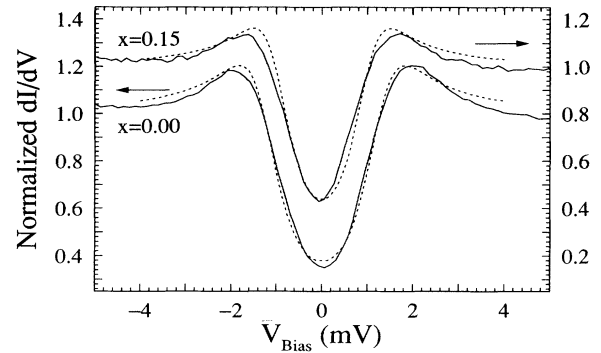
a number of possible origins to this nonideal tunneling behavior which is observed to the same extent for all the samples studied even in zero field. These include the large current densities inherent to STM which can cause pair breaking (although we have decreased the current by a factor of 10 with no noticeable change in the spectra). It is also possible that surface contamination plays a role. We do not believe that these relatively small deviations from BCS theory affect the overall conclusions of this work.

We now turn to the systematic behavior of the vortex-core spectra as a function of the Ta concentration. Figure 3 shows the spectra at the center of a vortex core for five samples with varying x. These tunneling characteristics were found reproducibly on different vortex cores and several samples with the same Ta concentration. All the spectra shown in Fig. 3 are taken at 0.3 T and 1.3 K after imaging the vortex lattice as described above. One notes the gradual disappearance of the peak at zero bias with increasing x. For $x=0.00$ the ratio of the conductance at zero bias to that at high bias is 1.2. This is less than the
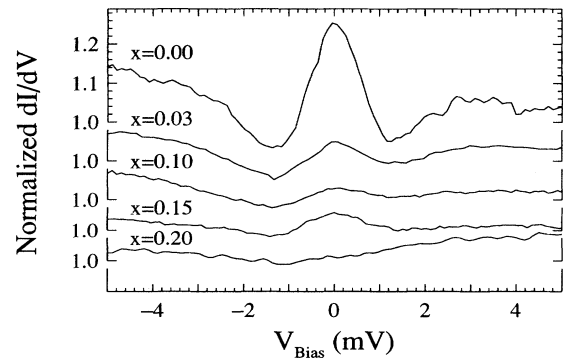

FIG. 3. Spectra taken at the center of a vortex core for various Ta substitutions at 1.3 K and 0.3 T. The spectra are normalized to the differential conductance at high bias.

value found by Hess, Robinson, and Waszczak [7] of 1.5 and is consistent with a difference in impurity concentration in the samples studied. [The residual resistivity ratio (RRR) of Hess's $NbSe_2$ sample was 31 compared to 22 for our $x=0$ sample.] We also note that the peak height we observe is a factor of 4 less than that predicted theoretically [2]. The dependence of the vortex-core spectra on x suggests a natural explanation of this discrepancy in terms of the importance of quasiparticle scattering even in pure $NbSe_2$.

Although a detailed theory for the reduction in the density of states at a vortex center as a function of impurity concentration does not exist, simple physical reasoning can explain this observation. The ZBCP has been shown to be due to the form of the low-energy quasiparticle wave functions [2] and reflects the higher probability of finding a quasiparticle at the Fermi energy at the vortex center. The wave functions depend on the nature of the confining pair potential the quasiparticles feel in a vortex. A rather rough analogy between the spatial variation of the pair potential and a potential well of depth Δ and radius $2\xi_\parallel$ for quasiparticles can be made [2]. Scattering on a scale smaller than the size of this "potential well" will cause a significant mixing of levels and ultimately destroy the ZBCP. One might expect this to occur when the coherence length $\xi_\parallel$ is of the order of the mean free path l.

Table I lists the material parameters for the samples used in this study. The Ginzburg-Landau coherence length parallel to the planes, $\xi_\parallel(T=0)$, has been determined from the upper-critical-field measurements of Dalrymple and Prober [9]. The mean free path was calculated from the low-temperature resistivity ρ_{LT} (determined just above T_c) measured using a four-point method on the same crystals used in the STM studies. As other authors [9,11] we have assumed a spherical Fermi surface [12] and an electron density of 1.55×10^{22} e/cm^3 (1 electron per molar volume). The mean free path is thus calculated as $l = \hbar (3\pi^2)^{1/3}/e^2 n^{2/3}\rho_{LT}$. We believe this provides a reasonable estimate of the mean free path in the basal plane. Although the Ginzburg-Landau coherence length is relatively constant for this alloy series, the BCS coherence length ξ_0 is expected to increase as the transition temperature decreases. We can estimate the BCS coherence length from the mean free path and the Ginzburg-Landau coherence length using the relation

$$\xi_\parallel(0) = 0.74\xi_0[\chi(0.88\xi_0/l)]^{1/2},$$

where χ is the Gor'kov function [13] which interpolates between the clean limit, $\xi_0 \ll l$, $\chi(0)=1$, and dirty limit, $\xi_0 \gg l$, $\chi(\infty)=1.33l/\xi_0$. One notes in Table I that ξ_0 indeed increases with increasing x such that for $x=0.10$, $\xi_0 > l$. Even for $x=0.15$, for which $\xi_0 \sim 3l$, a small indication of the ZBCP is still present. This may reflect the fact that the tightly bound quasiparticle states have a spatial extent less than $\xi_\parallel$.
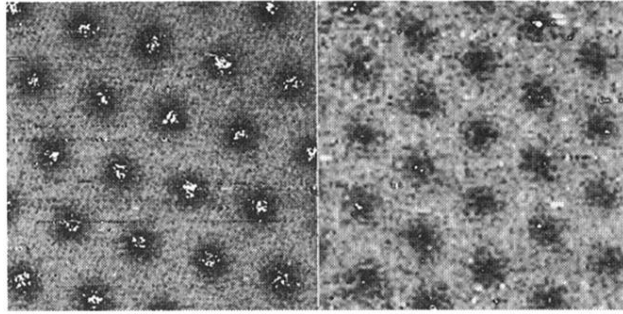
TABLE I. Material parameters for $Nb_{1-x}Ta_xSe_2$ samples studied.

x	T_c (K)	ρ_{LT} ($\mu\Omega$ cm)	RRR	$\xi_\parallel(0)$ (Å)	l (Å)	ξ_0 (Å)
0.00	7.2	6.4	22.0	84	320	130
0.03	6.8	11.0	12.7	73	186	122
0.10	5.9	16.9	6.2	80	121	156
0.15	5.4	26.9	5.1	82	76	199
0.20	5.1	29.3	4.7	86	70	224

In summary, we have shown the result of systematically decreasing the mean free path (while keeping the Ginzburg-Landau coherence length relatively constant) on the density of states in a vortex core. Our results show that the conductance peak is a property of superconductors in the clean limit. These results also confirm theoretical work by Watts-Tobin, Kramer, and Pesch [14] showing that in the dirty limit the local density of states at the center of a vortex core is exactly the same as that in the normal state. The evolution of the zero-bias peak with disorder suggests that the vortex-core density of states should provide a sensitive measure of the quasiparticle scattering time.

[1] H. F. Hess, R. B. Robinson, R. C. Dynes, J. M. Valles, Jr., and J. V. Waszczak, Phys. Rev. Lett. **62**, 214 (1989).
[2] J. D. Shore, M. Huang, A. T. Dorsey, and J. P. Sethna, Phys. Rev. Lett. **62**, 3089 (1989).
[3] F. Gygi and M. Schluter, Phys. Rev. B **41**, 822 (1990); **43**, 7609 (1991).
[4] U. Klein, Phys. Rev. B **40**, 6601 (1989).
[5] A. W. Overhauser and L. L. Daemen, Phys. Rev. Lett. **62**, 1691 (1989).
[6] R. Berthe, U. Hartmann, and C. Heiden, Appl. Phys. Lett. **57**, 2351 (1990).
[7] H. F. Hess, R. B. Robinson, and J. V. Waszczak, Phys. Rev. Lett. **64**, 2711 (1990).
[8] Ch. Renner, Ph. Niedermann, A. D. Kent, and Ø. Fischer, J. Vac. Sci. Technol. A **8**, 330 (1990).
[9] B. J. Dalrymple and D. E. Prober, J. Low Temp. Phys. **56**, 545 (1984).
[10] R. C. Dynes, V. Narayanamurti, and J. P. Garno, Phys. Rev. Lett. **41**, 1509 (1978).
[11] D. E. Prober, R. E. Schwall, and M. R. Beasley, Phys. Rev. B **21**, 2717 (1980).
[12] Admittedly an approximation for these anisotropic materials—assuming a strictly two-dimensional free-electron Fermi surface results in a 30% increase in the mean free path listed in Table I.
[13] See, for example, N. R. Werthamer, in *Superconductivity*, edited by R. Parks (Marcel Dekker, New York, 1969), Chap. 6, p. 338.
[14] R. Watts-Tobin, L. Kramer, and W. Pesch, J. Low Temp. Phys. **17**, 71 (1974).

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