Slope of the superconducting gap function in $Bi_2Sr_2CaCu_2O_{8+\delta}$ measured by vacuum tunneling spectroscopy

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Reproducible scanning tunneling microscope (STM) spectra of $Bi_2Sr_2CaCu_2O_{8+\delta}$ consistently exhibit asymmetric tunneling characteristics, with the higher peak conductance corresponding to a negatively biased sample. We consider various possible sources of this asymmetry that are not intrinsic to the superconducting state, including energy dependence of the normal-state densities of states of sample and/or tip, existence of bandwidth cutoffs, unequal work functions of tip and sample, and energy-dependent transmission probability. It is concluded that none of these effects can explain the sign and temperature dependence of the observed asymmetry. This indicates that the observed asymmetry reflects an intrinsic property of the superconducting state: an energy-dependent superconducting gap function with nonzero slope at the Fermi energy. It is pointed out that such a sloped gap function will also give rise to a thermoelectric effect in STM experiments, resulting in a *positive* thermopower. We discuss the feasibility of observing this thermoelectric effect with an STM and conclude that it is easily observable. Again, contributions to this thermoelectric effect may also arise from energy dependence of normal-state densities of states and from energy-dependent transmission probability. However, because each of these features manifests itself differently in the thermoelectric effect and in the tunneling characteristics, an analysis of thermoelectric currents and voltages together with the tunneling spectra as a function of temperature and tip-sample distance would allow for accurate determination of the slope of the gap function. It is suggested that it would be very worthwhile to perform these experiments, because the slope of the gap function reflects a fundamental property of the superconducting state. In particular, the theory of hole superconductivity has predicted the existence of such a slope, of universal sign, in all superconductors. It is furthermore argued that recent experimental results on vortex lattice imaging provide further strong evidence for the existence of the gap slope discussed here. [S0163-1829(99)04118-1]

I. INTRODUCTION

Tunneling spectroscopy is a powerful experimental tool that has the potential to provide fundamental information on the microscopic physics of high-temperature superconductivity. Unfortunately, due to the difficulty in preparation of well-characterized sample surfaces and tunnel junctions, often the spectra obtained have shown varying features that depend on the particular sample and/or the particular tunneling technique used. Consequently, it has been difficult to determine which of the features found in the large number of experimental studies that have been performed to date actually reflect intrinsic properties of high-temperature superconductors.

It is hence notable that several recent scanning tunneling microscopy (STM) studies of single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ cleaved in an ultrahigh vacuum have shown highly reproducible features.[1–3] These experiments are performed under carefully controlled conditions, and tunneling spectra are obtained at a large number of different positions on the sample and for a range of distances between tip and sample. The spectra do not show time dependence and generally exhibit either superconducting or semiconducting features, which has been suggested to depend on the oxygen stochiometry of the topmost Cu-O layer at the position of the tip.[1]

The spectra that exhibit superconducting features show structure in the tunneling conductance dI/dV that resembles the structure in the tunneling density of states of conventional BCS superconductors predicted by BCS theory: a decrease in the conductance for low voltages, where a gaplike structure develops, and a large enhancement of the conductance at the edges of this gaplike structure. A notable difference with the predictions of conventional BCS theory is that the spectrum is highly asymmetric: the peaks in dI/dV are symmetrically located around zero voltage but their height is substantially different, with the largest peak corresponding to a negatively biased sample. This asymmetry is found to persist to low temperatures. Another feature of the spectra, which is sometimes observed only for negative sample voltage,[1] and sometimes for both polarities,[3] is a dip in the conductance at voltages about twice the position of the conductance peaks.

In this paper we focus on the tunneling asymmetry in the peaks of dI/dV and consider various possible origins for it. First, it is clear that the tunneling geometry is asymmetric itself; an asymmetry may be expected to arise from different work functions of the tip and the sample. In fact, tunneling asymmetries are often seen in conventional tunnel junctions and have been explained assuming differences in work functions of the electrodes.[4] Second, as we will show, a tunneling asymmetry also arises from the fact that the tunneling probability is energy dependent, even in the absence of differences in work functions. Finally, a tunneling asymmetry could also arise from an energy-dependent normal-state density of states of either tip or sample, or from the existence of a bandwidth cutoff in either tip or sample. To focus the discussion we assume that the superconducting state in the sample is described by conventional BCS theory with an isotropic gap, so that the density of states in the superconducting state is

$$N[E(\epsilon)]=g_s(\epsilon)\frac{E}{\sqrt{E^2-\Delta^2}}, \tag{1}$$

with $g_s(\epsilon)$ the density of states of the sample in the normal state, and the quasiparticle energy E given in terms of the band energy ϵ by

$$E(\epsilon)=\sqrt{(\epsilon-\mu)^2+\Delta^2}, \tag{2}$$

with Δ the energy gap and μ the chemical potential. We will also allow for the possibility of broadening of the density of states arising from lifetime or other effects,[5] characterized by an inverse lifetime Γ.

We will show that none of the effects listed above are able to describe even qualitatively an asymmetry of the sign and temperature dependence seen experimentally. Briefly, asymmetries originating in asymmetric normal-state densities of states or bandwidth cutoffs tend to become smaller as the temperature is lowered, contrary to experimental observations. Asymmetry originating in energy dependence of the transmission probability persists down to low temperatures but is of the opposite sign to that observed experimentally. Asymmetry originating in differences in work functions can give the correct sign of the observed asymmetry at high temperature but the sign switches as the temperature is lowered, in contradiction with experiment.

We are thus led to the conclusion that the observed asymmetry originates in an intrinsic feature of the superconducting state. Such asymmetry will arise when the superconducting gap function Δ is itself energy dependent,[6] so that the quasiparticle energy Eq. (2) is

$$E(\epsilon)=\sqrt{(\epsilon-\mu)^2+\Delta(\epsilon)^2}. \tag{3}$$

The measured asymmetry reflects the slope of the gap function at the Fermi energy, $\Delta'(\mu)$. Hence, we argue that the experimental results are direct evidence for the existence of a slope in the superconducting gap function.

Another experimental consequence of a sloped gap function is that it should manifest itself in a thermoelectric effect in tunneling experiments.[7] It gives rise to a thermoelectric power of the tunnel junction given by $Q=\Delta(\mu)\Delta'(\mu)/eT$ for small temperature gradients. We argue that such a thermoelectric effect should be easily observable with an STM and that it should be experimentally searched for. Certainly, energy dependence of the normal-state densities of states and energy-dependent transmission could also contribute to a thermoelectric effect. However, we will show that analysis of the thermoelectric currents and voltages in conjunction with the tunneling characteristics dI/dV as a function of temperature and tip-sample distance should allow to clearly distinguish between the different sources of a thermoelectric effect, and in particular, to accurately determine the slope of the gap function. The sign of the observed tunneling asymmetry implies that the sign of the measured thermoelectric power will be *positive* (at least for small tip-sample distance), opposite to what one would predict from energy dependence of the electron transmission probability alone. In fact, thermoelectric experiments with STM with normal-metal electrodes have recently been reported,[8] with the sign of the measured themopower always negative.

The slope of the gap function at the Fermi energy is a fundamental property of the superconducting state, as it gives information on the underlying microscopic interactions that give rise to superconductivity in the system. For example, for an attractive Hubbard model[9] or for an electron-phonon model within a local approximation (Holstein model)[10] the superconducting gap function has zero slope. The detection of a gap slope would reveal the existence of an intrinsic electron-hole asymmetry of the superconductor and shed light on the theoretical understanding of superconductivity. In particular, the theory of hole superconductivity[11] has predicted the existence of such a slope, of universal sign, in all superconductors, with magnitude that scales with the critical temperature. However, a sloped superconducting gap function could also arise in other theoretical frameworks.

It is currently generally believed that high-T_c superconductors have an order parameter with complete or dominant d-wave symmetry.[12] The d-wave symmetry relates to the variation of the gap *on* the Fermi surface, while the slope of the gap function discussed here relates to variation of the gap in a direction *perpendicular* to the Fermi surface. Here we assume a gap that is constant on the Fermi surface for simplicity; however a simple generalization of our analysis could also be used to describe the variation of the amplitude of a d-wave gap, or more generally of an anisotropic gap, in a direction perpendicular to the Fermi surface, and we believe the conclusions would be similar concerning the existence of an *average* gap slope. Thus we argue that our analysis is relevant irrespective of whether the gap has s-wave or d-wave, or any other symmetry.

We note also that recent work[13] has argued that a gap with d-wave symmetry together with an assumption of directional tunneling can explain the experimentally observed tunneling asymmetries. We will discuss that work in the conclusions.

In the next section we discuss the tunneling formalism. Section III deals with barrier effects, and Sec. IV with density of states effects. In Sec. V we consider the effect of an energy-dependent gap function. Section VI discusses the expected thermoelectric effect in the presence of all these effects. We conclude in Sec. VII with a discussion.

II. FORMALISM

The tunneling probability across a barrier of thickness d for an electron of energy E above the Fermi level of the tip is given within the WKB approximation by

$$T(E,V)=e^{-2S(E,V)}, \tag{4a}$$

$$S(E,V)=\int_0^d\sqrt{\frac{2m}{\hbar^2}[V(x)-E]}\,dx, \tag{4b}$$

with m the electron mass and $V(x)$ the barrier potential. We assume a trapezoidal barrier, with work functions ϕ_t and ϕ_s for tip and sample, respectively, and a voltage V of the sample relative to the tip, as shown in Fig. 1, and obtain

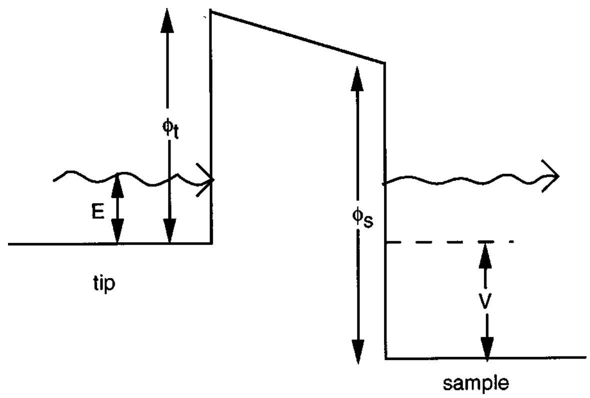

FIG. 1. Diagram of electron energy levels. V is the voltage of the sample relative to the tip, E is the electronic energy above the Fermi level of the tip. ϕ_t and ϕ_s are the work functions of tip and sample.

$$S(E,V)=\frac{\sqrt{2m}}{\hbar}\frac{2d}{3(\phi_t+V-\phi_s)}\times[(\phi_t-E)^{3/2}-(\phi_s-V-E)^{3/2}] \quad (5)$$

for the case where both the arguments raised to the 3/2 power are positive. When the voltage or energy are such that one of the arguments is negative, Eq. (5) still holds if that argument is set equal to zero. If the work functions ϕ_t and ϕ_s are similar and the voltage V is small, Eq. (5) reduces to

$$S(E,V)=\frac{\sqrt{2m}}{\hbar}d\left(\bar{\phi}-\frac{V}{2}-E\right)^{1/2}, \quad (6)$$

with $\bar{\phi}=(\phi_s+\phi_t)/2$ the average work function. The derivation assumes V, $V+E\ll\bar{\phi}$, and $|\phi_t-\phi_s|\ll\bar{\phi}$.

We assume that electrons tunneling into the superconductor go into a band of width D. Within a BCS formalism the tunneling current from sample to tip is given by[14]

$$I_{st}(V)=\int_{-D/2}^{D/2}d\epsilon[u^2(\epsilon)\{f_t[E(\epsilon)-eV]-f_s[E(\epsilon)]\}\times g_t[E(\epsilon)-eV]T(E(\epsilon)-eV,V)+v^2(\epsilon)\{f_s[E(\epsilon)]-f_t[E(\epsilon)+eV]\}g_t[-E(\epsilon)-eV]\times T(-E(\epsilon)-eV,V)]g_s(\epsilon). \quad (7)$$

Here, f_t and f_s denote Fermi functions for tip and sample, which may be at different temperatures, and g_t and g_s are normal-state densities of states for tip and sample. V is the voltage of the sample relative to the tip. The coherence factors are given by the usual form,

$$u^2(\epsilon)=\frac{1}{2}\left(1+\frac{\epsilon-\mu}{E(\epsilon)}\right), \quad (8a)$$

$$v^2(\epsilon)=\frac{1}{2}\left(1-\frac{\epsilon-\mu}{E(\epsilon)}\right), \quad (8b)$$

and we assume that the relation between quasiparticle energy E and band energy ϵ is given by Eq. (2). μ is the sample chemical potential.

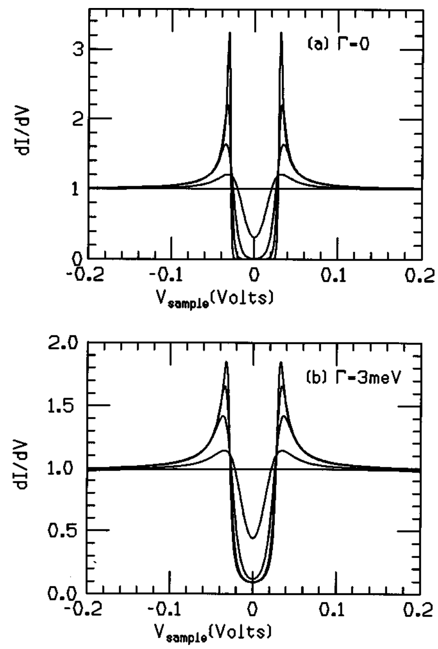

FIG. 2. Tunneling conductance for a superconductor with zero temperature gap $\Delta=30$ meV and $T_c=100$ K, for $T/T_c=0.1$, 0.25, 0.5, 0.85, and 1. Constant densities of states of sample and tip and energy-independent transmission are assumed. In the following figures, the same values for gap and temperatures are used. (a) $\Gamma=0$, (b) $\Gamma=3$ meV.

The tunneling current Eq. (7) can be written as

$$I_{st}(V)=\int_{-\infty}^{\infty}d\omega[f_t(\omega-eV)-f_s(\omega)]g_t(\omega-eV)\times T(\omega-eV,V)\rho_s(\omega), \quad (9)$$

with $\rho_s(\omega)$ the superconducting density of states,

$$\rho_s(\omega)=\int_{-D/2}^{D/2}d\epsilon\{u^2(\epsilon)\delta[\omega-E(\epsilon)]+v^2(\epsilon)\delta[\omega+E(\epsilon)]\}g_s(\epsilon). \quad (10)$$

Broadening effects[5] may be included by replacing the δ functions in Eq. (10) by

$$\delta(\omega)\rightarrow\frac{1}{\pi}\frac{\Gamma}{\omega^2+\Gamma^2}, \quad (11)$$

with Γ a phenomenological broadening parameter.

We consider for definiteness a band of width $D=0.5$ eV in the sample, a critical temperature $T_c=100$ K and zero temperature gap $\Delta(T=0)=30$ meV. The temperature dependence of the gap is assumed to follow the conventional weak-coupling BCS behavior. Figure 2 shows for reference the tunneling conductance obtained from Eq. (7) assuming unit transmission and energy-independent normal state densities of states of sample and tip, for broadening factors $\Gamma=0$ and $\Gamma=3$ meV. The amplitude of the peaks in dI/dV is proportional to u^2 and v^2 for positive and negative voltage, respectively. Because $u^2(\epsilon)=v^2(2\mu-\epsilon)$, tunneling characteristics are symmetric in the sign of V in the absence of other effects.

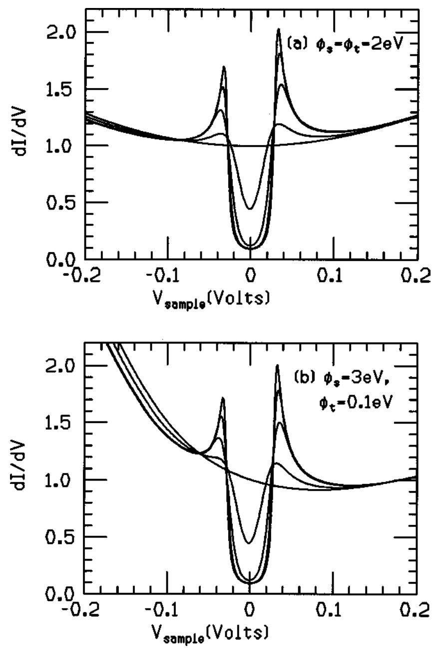

FIG. 3. Effect of finite barrier height. Barrier thickness is $d=20A$. In (a), work functions of tip and sample are equal. The conductance is symmetric in the voltage above T_c, and becomes asymmetric as the gap opens. In (b), the conductance above T_c is asymmetric due to unequal work functions; as the gap develops, the asymmetry switches sign. Broadening parameter is $\Gamma=3$ meV.

III. BARRIER EFFECTS

When we allow for energy-dependent transmission as given by Eq. (4), the tunneling spectra develop an asymmetry that increases as the temperature is lowered, as shown in Fig. 3(a). By decreasing the work functions of tip and sample or increasing the barrier thickness the size of the asymmetry is increased. Thus it would easily be possible to match the magnitude and temperature-dependence of the observed asymmetry. However, the sign of the asymmetry thus obtained, larger conductance for a positively biased sample, is opposite to what is seen experimentally.

At first sight the existence of this asymmetry may seem puzzling, since it occurs for equal work functions of tip and sample and for densities of states that are symmetric around the Fermi energy for both tip and sample. However, it is easily understood by inspection of the diagram in Fig. 4. Because the gap in the superconducting density of states of the sample blocks the transmission in that energy range, the transmission for positive sample involves electrons that are closer to the top of the barrier than that for a negative sample. As the temperature is raised and the gap in the density of states closes, this asymmetry will disappear.

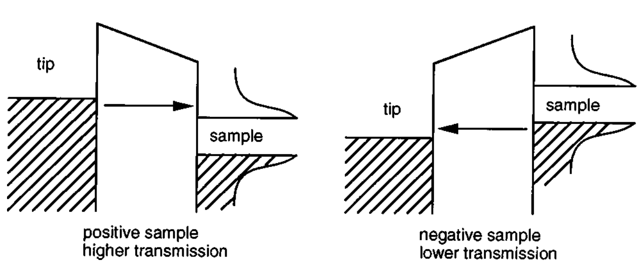

FIG. 4. Illustration of asymmetry in tunneling resulting from energy gap in the sample. For equal work functions, the transmission will be higher for positively biased sample because the tunneling electrons are closer to the top of the barrier.

Could energy-dependent transmission give rise to an asymmetry of the observed sign? It could happen two ways. First, if the transmission coefficient for an electron were to decrease as its energy increases. While this cannot happen in an STM, it could happen in a conventional tunnel junction if the Fermi level is in the band gap of the insulating barrier close to the valence band, as discussed by Gundlach.[15] Second, energy-dependent transmission would give rise to an asymmetry of the observed sign if the gap structure in the density of states shown in Fig. 4 occurred in the tip rather than in the sample. Clearly, neither of these scenarios appears to be relevant to the case under consideration.

Next we consider the effect of differences in the work function of tip and sample. This can give rise to asymmetry in tunneling, and, in fact, has been proposed to explain asymmetries observed in various conventional tunnel junctions.[4] If the work function of the tip is lower than that of the sample the tunneling conductance in the normal state will be larger for a negatively biased sample, in agreement with observations. Figure 3(b) shows results for a rather extreme case, with work functions $\phi_t=0.1$ eV, $\phi_s=3$ eV. While the asymmetry in the normal state is of the observed sign, as the temperature is lowered the peaks in the conductance exhibit the asymmetry of opposite sign, induced by the energy dependence of the transmission in the presence of the superconducting energy gap. It is found that no combination of work functions and barrier thicknesses can give rise to an asymmetry of the observed sign in the peaks of the tunneling conductance at low temperatures.

IV. DENSITY OF STATES EFFECTS

We next consider the effect of nonconstant density of states and of bandwidth cutoffs. Figure 5 shows the resulting conductance when the normal-state density of states of the sample and/or of the tip has a linear energy variation, reflected in the conductance for $T=T_c$ in Fig. 5. This is obtained for an electronic density of states in the tip that increases with energy, and/or one in the sample that decreases with energy. As the temperature is lowered it is seen that the magnitude of the asymmetry in the conductance peaks decreases, simply because the peaks arise from band energies increasingly closer to the Fermi energy. When we include broadening, as in Fig. 5(b), the asymmetry persists to lower temperatures, but there is still a clear progressive reduction in the magnitude of the asymmetry as the temperature decreases.

Furthermore, when we include the energy dependence of the transmission the asymmetry gets further reduced and may even change sign, as shown in Fig. 6. In (a) the normal-state asymmetry, of the sign seen experimentally, arises from a sloped density of states of either the tip or the sample. In (b), we assume that the band edge in the sample occurs very close to the Fermi energy, as shown by the dashed line, also giving rise to an asymmetry of the sign seen experimentally at high temperatures.[16] In the presence of energy-dependent transmission, the asymmetry changes sign as the temperature

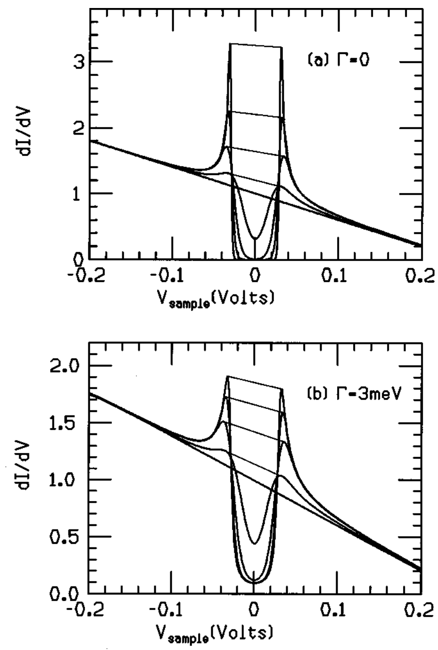

FIG. 5. Effect of nonconstant density of states. The energy variation of the normal-state density of states is linear, and given by the conductance curve for $T=T_c$. It corresponds to either a sample density of states that decreases with electronic energy, or a tip density of states that increases with energy, or a combination of the two. The asymmetry decreases as the temperature is lowered, both without (a) and with (b) broadening. The thin lines connecting the peaks are drawn to illustrate this effect.

is lowered. The work functions assumed here are not particularly small, and smaller work functions or larger tip-sample distances would make the obtained asymmetry deviate even further from what is observed.

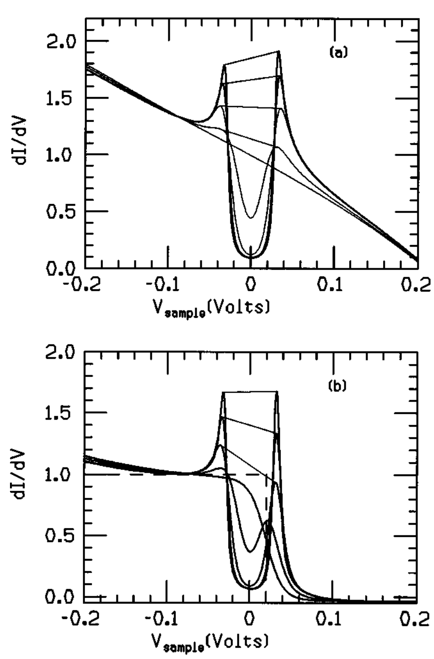

FIG. 6. Effect of sloped density of states (a) and of bandwidth cutoff (b) together with energy-dependent transmission. $\phi_s=\phi_t=1$ eV, barrier thickness $d=10A$. $\Gamma=3$ meV. The dashed line in (b) shows the density of states of the sample.

We conclude that asymmetries originating from nonconstant densities of states and bandwidth cutoffs cannot explain the experimentally observed asymmetry. The asymmetry originating from these effects invariably decreases rapidly as the temperature is lowered. The asymmetry originating in the energy dependence of transmission discussed in the previous section does persist to low temperatures but it is of the opposite sign to the one observed experimentally. Hence we conclude that the experimentally observed asymmetry must originate in an intrinsic asymmetry of the superconducting state itself.

V. ENERGY-DEPENDENT GAP FUNCTION

The simplest generalization of BCS theory with constant energy gap is to assume that the gap depends on wave vector k only through the band energy ϵ_k. Hence, the quasiparticle energy is

$$E_k(\epsilon_k)=\sqrt{(\epsilon_k-\mu)^2+\Delta^2(\epsilon_k)}. \tag{12}$$

For energies close to the Fermi energy,

$$\Delta(\epsilon)=\Delta(\mu)+\Delta'(\mu)(\epsilon-\mu). \tag{13}$$

The minimum quasiparticle energy will now occur not at $\epsilon_0=\mu$, but rather at

$$\epsilon_0=\mu-\Delta(\mu)\Delta'(\mu), \tag{14}$$

and is

$$\Delta_0=E(\epsilon_0)=\Delta(\mu)+O(\Delta'^2). \tag{15}$$

The BCS coherence factors Eq. (8) are no longer equal at the band energy corresponding to the minimum quasiparticle energy ϵ_0, rather,

$$u^2(\epsilon_0)=\frac{1}{2}(1-\Delta'), \tag{16a}$$

$$v^2(\epsilon_0)=\frac{1}{2}(1+\Delta'), \tag{16b}$$

with $\Delta'\equiv\Delta'(\mu)$. For electrons injected into (extracted from) the sample, i.e., positive (negative) sample voltage, the tunneling conductance is proportional to u^2 (v^2). Hence if the slope Δ' is positive, that is, if the gap function is an increasing function of electronic energy, the tunneling current and conductance will be larger for a negatively biased sample, as seen experimentally. We define

$$\left.\frac{dI}{dV}\right|_+=\text{height of }\frac{dI}{dV}\text{ peak for } V_{sample}>0 \tag{17}$$

and similarly for $V_{sample}<0$, and the tunneling asymmetry A as the difference in the height of the peaks divided by the average peak height

$$A=\frac{\left.\frac{dI}{dV}\right|_- - \left.\frac{dI}{dV}\right|_+}{\left[\left.\frac{dI}{dV}\right|_- + \left.\frac{dI}{dV}\right|_+\right]/2} \tag{18}$$

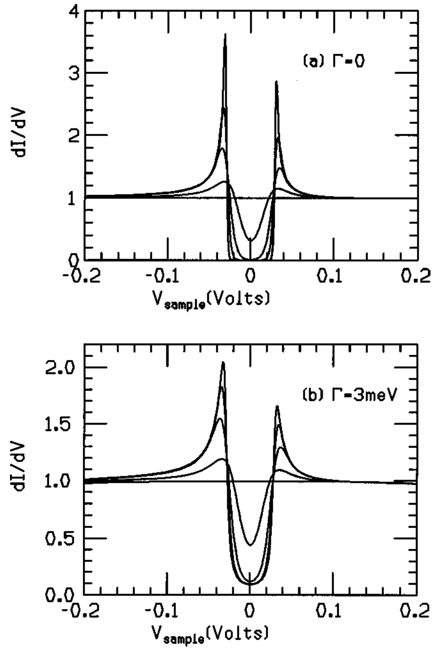

FIG. 7. Effect of intrinsic asymmetry, without (a) and with (b) broadening. Gap slope is $\Delta'=0.12$. Constant densities of states and energy-independent transmission are assumed.

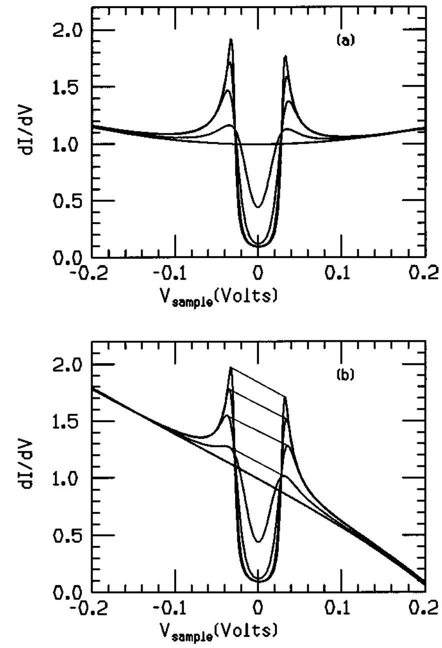

FIG. 8. Effect of intrinsic asymmetry and energy-dependent transmission. Gap slope is $\Delta'=0.12$, barrier parameters are $\phi_s=\phi_t=1$ eV, $d=10A$. (a) Constant density of states, (b) sloped density of states.

and we have at low temperatures[6]

$$A(T\rightarrow 0)=2\Delta'(\mu). \tag{19}$$

Figure 7 shows the tunneling conductance for gap slope $\Delta'=0.12$, with and without broadening. The asymmetry is clearly noticeable in both cases and approaches the theoretical limit Eq. (19) for the case of no broadening, while it remains somewhat smaller for $\Gamma\neq 0$.

However, the effect of the gap slope needs to be balanced with that of the energy-dependent transmission. Expanding the expression Eq. (5) for the transmission we obtain

$$T(E,V)=T_0[1+c(E+eV/2)], \tag{20a}$$

$$c(eV^{-1})=0.51\frac{d(A)}{\bar{\phi}(eV)^{1/2}}, \tag{20b}$$

with $\bar{\phi}$ the average of the work functions of tip and sample. We find for the tunneling asymmetry at low temperatures

$$A(T\rightarrow 0)=2\left(\Delta'(\mu)-\frac{c\Delta_0}{2}\right). \tag{21}$$

For a typical barrier $\bar{\phi}=1$ eV, $d=10A$ we have $c\sim 5$ eV^{-1}. Hence for a gap $\Delta_0=30$ meV the tunneling asymmetry will be of the observed sign if the gap slope is larger than about 0.08.

The slope of the gap function, Δ', may or may not depend on temperature. If Δ' is independent of T, then its effect will become increasingly important relative to the effect of energy-dependent transmission as the temperature is raised, according to Eq. (21), since Δ_0 decreases as T_c is approached. For example, for parameters where the intrinsic and barrier-induced asymmetry exactly cancel at low temperatures, the intrinsic asymmetry would dominate as T approaches T_c. Instead, if the gap slope scales with the gap itself, its effect on the asymmetry will have the same temperature dependence as that of energy-dependent transmission. We will assume that this is the case here, so that the gap slope approaches zero as $T\rightarrow T_c$. It should also be noted that this is the behavior predicted within the theory of hole superconductivity.[11]

Figure 8 shows the combined effects of intrinsic asymmetry and energy-dependent transmission, for a case where intrinsic asymmetry dominates: $\Delta'=0.12$, and barrier parameters $\phi_s=\phi_t=1$ eV, $d=10A$, leading to $A=0.044$ in Eq. (21). In the presence of also a nonconstant density of states, the asymmetry seen in the normal state persists strongly as the temperature is lowered, Fig. 8(b), unlike the cases where no intrinsic asymmetry exists (Figs. 5 and 6).

Equations (20) and (21) imply that the observed asymmetry should become weaker as the tip-sample distance increases and the effect of energy-dependent transmission increases. There is some evidence for this effect in the data shown in Fig. 13 of Ref. 2. The asymmetry for the smallest tip-sample distance (largest conductance) is approximately 12.5%, and it decreases to about 5–7 % as the tip-sample distance increases and the current decreases by about a factor of 10. The change in the tip-sample distance over that range will be approximately

$$\Delta d(A)=\frac{\ln 10}{1.02\sqrt{\bar{\phi}(eV)}}. \tag{22}$$

From Eqs. (20)–(22) we may deduce the values of the average work function and the change in the tip-sample distance in the experiment of Ref. 2. Assuming that the asymmetry decreased from 12.5% to 6% when the current decreased by a factor of 10 and a gap $\Delta_0=30$ meV yields

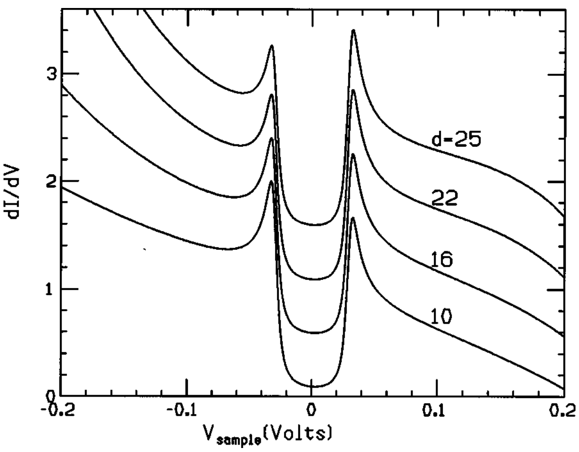

FIG. 9. Tunneling characteristics in the presence of nonconstant density of states, intrinsic gap slope, and energy-dependent transmission, at temperature $T/T_c=0.1$ for various tip-sample distances given in the figure (numbers next to the curves in A). The curves are offset increasingly by 0.5 for clarity. Broadening parameter is $\Gamma=3$ meV. Gap slope is $\Delta'=0.17$ and work functions are $\phi_s=\phi_t=0.53$ eV.

$$\overline{\phi}=0.53\ \text{eV}, \tag{23a}$$

$$\Delta d=3.1A \tag{23b}$$

which are not unreasonable. The gap slope implied by these data is

$$\Delta'=0.062+0.011d(A), \tag{24}$$

where d is the smallest tip-sample distance in the data in Fig. 13 of Ref. 2. For example, $\Delta'=0.17$ if $d=10A$. This analysis furthermore implies that the observed asymmetry will switch signs if the tip-sample distance were to be increased by another $3A$, corresponding to another factor of 10 drop in the conductance. In Fig. 9 we show the tunneling characteristics for these parameters at low temperatures for a range of tip-sample distances illustrating this effect. It would be of interest to obtain more detailed experimental information on the dependence of the tunneling asymmetry on tip-sample separation to compare with the theoretical expectation.

We conclude from this analysis that it is possible to account for the observed tunneling asymmetry in a simple way by assuming an energy-dependent gap function with a finite slope at the Fermi energy. The existence of energy-dependent transmission does not qualitatively modify the results, it only implies that the underlying gap slope giving rise to the observed asymmetry is larger than what one would have inferred assuming the transmission was energy-independent. To explain the energy dependence of the observed spectra above T_c it is furthermore necessary to assume that there is either energy dependence in the normal-state density of states and/or a large difference in the work functions of tip and sample. However, these factors would not be able to explain the persistence of the asymmetry to low temperatures. Accurate measurement of the dependence of the tunneling asymmetry on tip-sample distance at low temperatures should allow for a determination of the effect of energy-dependent transmission, and hence for an accurate determination of the gap slope. As we show in the next section, further information on the gap slope may be inferred by measurement of a thermoelectric effect.

VI. THERMOELECTRIC EFFECT IN STM

Further information on the existence of a slope in the gap function may be obtained from experiments where the tip and the sample are at different temperatures. To our knowledge such experiments at cryogenic temperatures have not yet been systematically performed,[18] but we are not aware of any fundamental reason that would not allow for thermally decoupling of tip and sample and keeping them at substantially different temperatures in vacuum. Assuming there is no heat conduction path between tip and sample, the only way to transport heat is through radiation. For example, assume the sample is at higher temperature than the tip. The power transfered from sample to tip per unit area is of order

$$P=\sigma(T_s^4-T_t^4), \tag{25a}$$

$$\sigma=5.6710^{-5}\ \text{erg sec}^{-1}\,\text{cm}^{-2}\,\text{K}^{-4}. \tag{25b}$$

To maintain a temperature difference between tip and sample the tip needs to conduct away the heat it absorbs sufficiently fast. A typical metallic conductivity is $\kappa\sim10^7$ erg $\text{sec}^{-1}\,\text{cm}^{-1}\,\text{K}^{-1}$, so the temperature gradient developed in the tip will be

$$\nabla T\sim\frac{\sigma}{\kappa}(T_s^4-T_t^4)\sim\frac{10^{-11}}{\text{cm K}^3}(T_s^4-T_t^4). \tag{26}$$

For example, for a tip of 1 cm length at $T=0$ at the end far from the sample, the end close to the sample would heat up to a negligible $T_t\sim10^{-3}$ K if the sample is at temperature $T_s=100$ K. A detailed analysis of heat transfer between tip and sample in an STM is given in Ref. 19.

There have been in fact several recent investigations of thermoelectric effects in scanning tunneling microscopy with normal-metal samples.[8,19–22] In these experiments, performed at room temperature, the tip is heated by laser power to a temperature approximately 10 K higher than the sample. It is noteworthy that in these experiments the thermopower measured is invariably negative, even when the samples are metals with positive bulk values for the thermopower. This is presumably due to the contribution to the thermopower from energy-dependent transmission, as discussed below.

If we assume that the gap slope Δ' is positive, as indicated by the tunneling asymmetry, a positive current will flow from the hotter to the colder electrode in our case in the absence of other effects, that is, the system will have positive thermopower. This is a consequence of the fact that the average quasiparticle charge in the superconductor, given by

$$q(\epsilon)=v^2(\epsilon)-u^2(\epsilon) \tag{27}$$

is positive on the average,[7] unlike the usual BCS case with a constant energy gap where quasiparticles are charge neutral on average.

For energy-independent transmission and constant densities of states the current from sample to tip originating from quasiparticles of lowest energy, i.e., with $E=\Delta_0$, is proportional to

$$J_{st}=(1-\Delta')[f_t(\Delta_0-eV)-f_s(\Delta_0)]+(1+\Delta')\times[f_s(\Delta_0)-f_t(\Delta_0+eV)]=f_t(\Delta_0-eV)-f_t(\Delta_0+eV)-\Delta'[f_t(\Delta_0-eV)+f_t(\Delta_0+eV)-2f_s(\Delta_0)] \quad (28)$$

and for small temperature gradient this yields for the zero current thermoelectric voltage of the sample

$$V_0=\frac{\Delta_0}{e}\Delta'\frac{T_s-T_t}{T_t}, \quad (29)$$

hence, the thermopower of the junction, Q, is directly proportional to the gap slope,

$$Q=\frac{\Delta_0}{eT_t}\Delta', \quad (30)$$

and is positive if the gap slope is positive. Note that the thermopower here increases as the temperature is lowered, unlike the usual situation in metals and semiconductors where it decreases linearly with temperature as $T\rightarrow 0$.

However, contributions to the thermopower will also arise from energy-dependent transmission as well as from nonconstant densities of states. If we consider the current arising only from quasiparticles of minimum energy, Δ_0, a nonconstant density of states of the sample will not contribute at low temperatures. Equation (28) generalizes to

$$J_{st}=(1-\Delta')(1+c\Delta_0)(1+c_t\Delta_0)[f_t(\Delta_0-eV)-f_s(\Delta_0)]+(1+\Delta')(1-c\Delta_0)(1-c_t\Delta_0)[f_s(\Delta_0)-f_t(\Delta_0+eV)], \quad (31)$$

assuming that the voltage $V\ll\Delta_0$. Here, c_t is the logarithmic derivative of the tip density of states at the Fermi energy,

$$c_t=g_t'(0)/g_t(0), \quad (32)$$

and we have approximately

$$J_{st}=f_t(\Delta_0-eV)-f_t(\Delta_0+eV)-[\Delta'-\Delta_0(c+c_t)]\times[f_t(\Delta_0-eV)+f_t(\Delta_0+eV)-2f_s(\Delta_0)], \quad (33)$$

which contributes to the zero voltage thermoelectric current proportionally to

$$J_{st}=\frac{\Delta_0}{e}[\Delta'-\Delta_0(c+c_t)][f_t(\Delta_0)-f_s(\Delta_0)]. \quad (34)$$

For the zero current thermoelectric voltage, we have for small temperature gradient

$$V_0=\frac{\Delta_0}{e}[\Delta'-\Delta_0(c+c_t)]\frac{T_s-T_t}{T_t}. \quad (35)$$

Hence, we conclude that the thermoelectric voltage may be of either sign depending on the magnitude of the parameters c and c_t arising from energy-dependent transmission and tip density of states, respectively.

In particular, note that the effect of energy-dependent transmission is twice as large here as it is in the tunneling asymmetry, Eq. (21). This is because the tunneling asymmetry occurs at finite voltage, $V=\Delta_0$, where the effect of energy-dependent transmission is smaller than at zero voltage. Hence it is possible to have a tunneling asymmetry of the observed sign, reflecting the intrinsic gap slope, together with a negative thermopower, dominated by energy-dependent transmission.

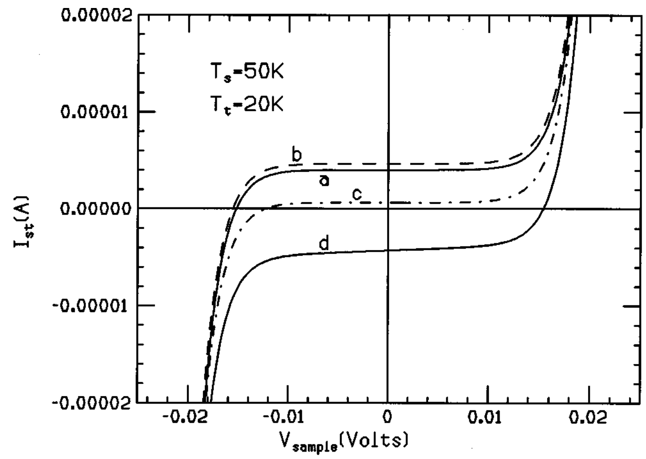

FIG. 10. Current versus voltage for sample and tip at different temperatures as given in the figure for various cases, with constant densities of states, zero gap slope, and energy-independent transmission except as indicated below. Curve labeled a (solid line): gap slope $\Delta'=0.12$; b (dashed line): tip density of states of slope $m_t/g_t(0)=-4$ eV^{-1}; c (dot-dashed line): sample density of states of slope $m_s/g_s(0)=4$ eV^{-1}; d (solid line): barrier parameters $\phi_s=\phi_t=0.5$ eV, $d=20A$, and gap slope $\Delta'=0.36$. No broadening is assumed.

It is also possible to obtain a negative thermopower just from the effect of tip density of states if c_t is positive, that could dominate over the effect of the intrinsic gap slope. However, note that this corresponds to a tip density of states that *increases* with electronic energy, and hence it would give rise to tunneling spectra with normal-state slope that is opposite to the slope that is observed experimentally.

Figures 10 and 11 illustrate some of the effects discussed above. Figure 10 shows current versus voltage in the presence of a large temperature difference between sample and tip, with the tip colder than the sample. The thermoelectric effect arising from intrinsic asymmetry and from a sloped tip density of states g_t is similar, as seen in Fig. 10 (curves labeled a and b), and it corresponds to positive thermoelectric power. However the dI/dV spectra [Figs. 11(a) and 11(b)] are very different, with the ones corresponding to the sloped g_t exhibiting an asymmetry that is opposite in sign to that observed experimentally and to that originating in the intrinsic effect, Fig. 11(a). Conversely, a sloped g_t that would give rise to an asymmetry of the sign seen experimentally would give rise to a thermopower opposite in sign to what is shown in Fig. 10, i.e., negative thermopower. Thus experimental results for tunneling spectra together with thermoelectric effect could clearly distinguish between competing hypothesis of intrinsic asymmetry and energy-dependent g_t.

An energy-dependent g_s of negative slope would give rise to an asymmetry in tunneling of the sign seen experimentally [Fig. 11(c)] and to a thermoelectric effect that has the same sign as that given by the intrinsic asymmetry, but the magnitude of the thermoelectric current is much smaller than in

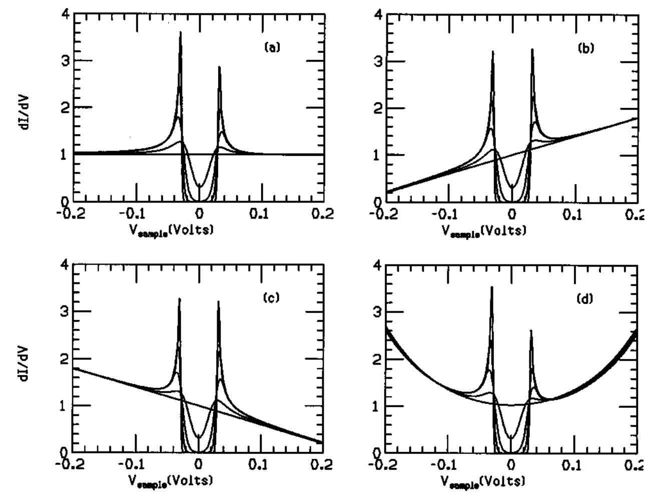

FIG. 11. Tunneling characteristics for the parameters of the four cases in Fig. 10. (a) sloped gap, (b) sloped tip density of states, (c) sloped sample density of states, (d) sloped gap and energy-dependent transmission. Note that the sloped densities of states of sample and tip give rise to tunneling conductance of opposite slope but to a thermoelectric effect of the same sign (Fig. 10). Note also that the tunneling asymmetry in (d) is of the sign expected from the gap slope, but the sign of the thermoelectric effect in Fig. 10 (curve labeled d) is opposite because the effect of energy-dependent transmission dominates there.

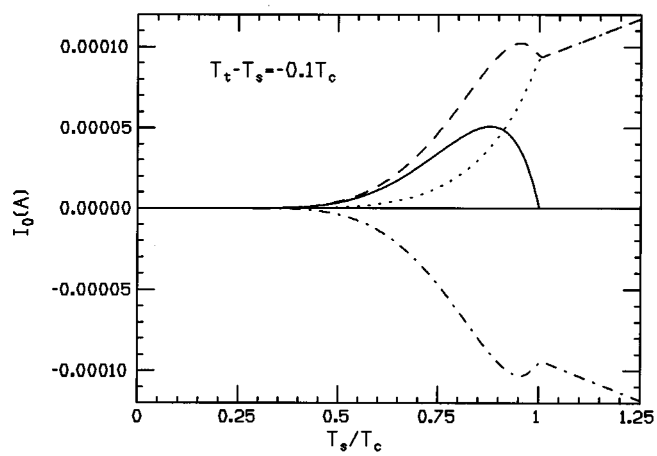

FIG. 12. Zero voltage thermoelectric current from sample to tip for the tip colder than the sample as function of sample temperature for fixed temperature difference between tip and sample. Constant densities of states, zero gap slope, and energy-independent transmission is assumed except as indicated below. Solid line: gap slope $\Delta'=0.12$; dashed line: tip density of state of slope $m_t/g_t(0)=-4\ \mathrm{eV}^{-1}$; dotted line: sample density of state of slope $m_s/g_s(0)=4\ \mathrm{eV}^{-1}$; dot-dashed line: barrier parameters $\phi_s=\phi_t=1\ \mathrm{eV}, d=7.84A$. $\Gamma=0$ in all cases.

the other cases, as seen in Fig. 10 (curve labeled c). The zero voltage thermoelectric current arising from all these effects is given by

$$I_{ts}=\frac{2}{eR}\int_{\Delta_0}^{\infty}dE\,\frac{E}{\sqrt{E^2-\Delta_0^2}}\left[\frac{\Delta'\Delta_0}{E}+(c+c_t)E+c_s\frac{E^2-\Delta_0^2}{E}\right]\times[f_t(E)-f_s(E)], \tag{36}$$

with $c_s=g_s'(0)/g_s(0)$. The effect of a nonconstant sample density of states is much smaller because the singularity at $E=\Delta_0$ is canceled for the term involving c_s. Thus it would be possible to discern the effect of intrinsic asymmetry versus sample density of states on the thermopower by calculating the expected magnitude of thermoelectric current and comparing with experiment.

The energy-dependent transmission gives rise to a tunneling asymmetry of opposite sign to that seen experimentally, as previously discussed, and to negative thermopower. However, because its effect on the thermopower is twice as large as on the tunneling asymmetry it is possible to choose parameters so that the tunneling asymmetry is of the sign observed experimentally, as determined by Δ', and yet the thermopower is negative, opposite to what the intrinsic asymmetry predicts. An example is shown in Fig. 10 (curve labeled d) and Fig. 11(d). However, this requires rather large values of both intrinsic asymmetry and energy-dependent transmission parameters.

Analysis of the thermoelectric effect as function of temperature would also allow us to clearly distinguish between an intrinsic origin and the other effects. Figure 12 shows the zero voltage thermoelectric current versus sample temperature T_s for a fixed temperature difference between sample and tip, $T_t-T_s=-0.1$, with the tip colder than the sample. In the presence of only the intrinsic asymmetry (solid line) the thermoelectric current would go to zero at T_c. Instead, energy-dependent density of states of either tip or sample (dashed and dotted lines) can give a thermoelectric current of the same sign, but it continues to increase as the temperature is raised above T_c. The same is true for the current arising from energy-dependent transmission, which in addition is of an opposite sign to that originating in intrinsic asymmetry (dash-dotted line). Results for these cases for temperature gradient of opposite sign, $T_t-T_s=0.1$, are the mirror image across the horizontal axis of these results.

In contrast, the magnitude of the zero current thermoelectric voltage V_0 is rather different for both signs of the temperature gradient, being larger when the tip is colder than the sample. At low temperatures we can derive an expression for V_0 that is not restricted to small temperature gradients,

$$V_0=[\Delta'-\Delta_0(c+c_t)]\frac{k_BT_t}{e}[e^{(\beta_t-\beta_s)\Delta_0}-1], \tag{37}$$

which shows that the thermoelectric voltage can become very large at low temperatures when the tip is colder than the sample. Again, the voltage goes to zero at T_c if it originates in intrinsic asymmetry and remains finite when it originates in the other factors. Results for the tip colder and warmer than the sample are illustrated in Fig. 13.

The effect of broadening on the thermoelectric effect is shown in Fig. 14. We assume only intrinsic asymmetry is present, but the effect is qualitatively the same when the thermoelectric effect originates in energy-dependent transmission or variations in the densities of states. The thermoelectric current at low temperatures increases as the broadening parameter increases. The thermoelectric voltage is reduced in the presence of broadening: the effect is most dramatic when the tip is colder than the sample, the large thermoelectric voltages obtained in the absence of broaden-

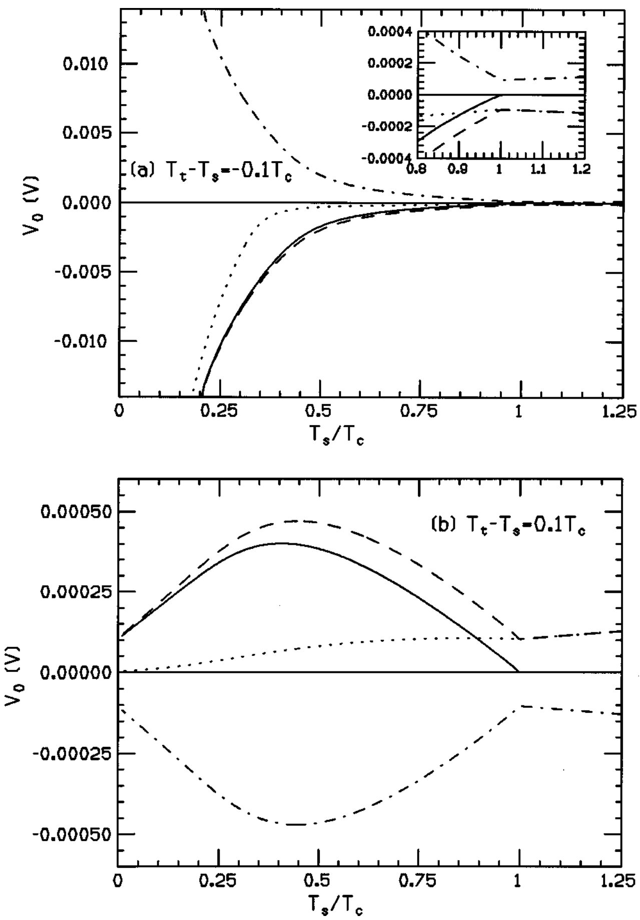

FIG. 13. Zero current thermoelectric voltage (of sample) for the four cases of Fig. 12. The line convention is the same as in Fig. 12. (a) Tip colder than the sample. Note that the voltage becomes very large at low temperatures. The inset shows the behavior of the voltage close to T_c. (b) Sample colder than tip. Note that the voltages are much smaller than in (a). Note also that both in (a) and (b) only in the case of intrinsic asymmetry alone (solid line) does the voltage go to zero at T_c.

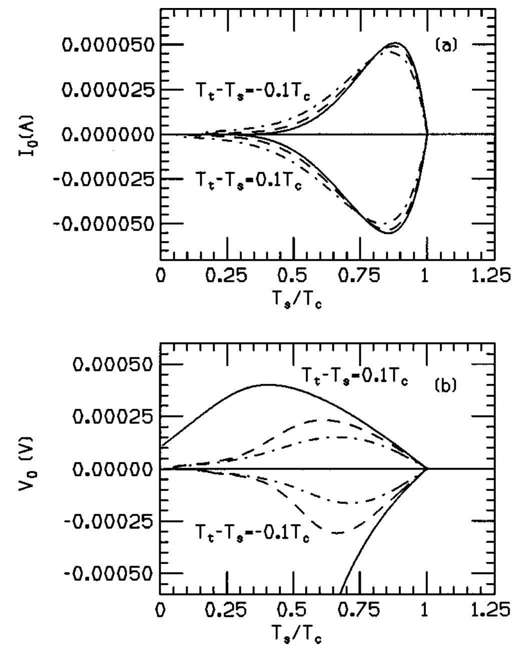

FIG. 14. Effect of broadening on thermoelectric current (a) and thermoelectric voltage (b). Gap slope is $\Delta'=0.12$, constant densities of states and energy-independent transmission is assumed. Solid lines are for $\Gamma=0$, dashed lines for $\Gamma=1$ meV, and dash-dotted lines for $\Gamma=3$ meV. The effect of broadening is to slightly increase the thermoelectric current. The thermoelectric voltage is somewhat decreased by broadening when the tip is hotter than the sample, and substantially so when the tip is colder than the sample.

ing at low temperatures are significantly reduced and become comparable to those obtained for opposite sign of the temperature gradient. This occurs because in the presence of broadening the thermoelectric current at low temperatures is dominated by low-energy electrons rather than electrons with energies above the energy gap.

Next we consider the situation where both intrinsic asymmetry and energy-dependent transmission exist. As the tip-sample distance is varied one or the other effect could dominate, as previously discussed. Results are shown in Figs. 15 and 16. For sufficiently small tip-sample distance, assuming tunneling remains ideal, the intrinsic asymmetry effect will dominate at low temperatures. Here we have assumed a gap slope $\Delta'=0.24$ and an average work function $\overline{\phi}=1$ eV. According to Eqs. (35) and (20b), the crossover between intrinsic-asymmetry–dominated and barrier-dominated thermoelectric effect at low temperatures should occur for tip-sample distance $d\sim 15A$. Indeed, intrinsic asymmetry is seen to dominate for $d=5A$ and $d=10A$. Note that the crossover in the thermoelectric voltage at low temperatures when the tip is colder than the sample [Fig. 16(a)] would be very sharp as function of tip-sample distance. Note also that in the tunneling conductance instead of the crossover between gap-slope–dominated asymmetry and barrier-dominated assymetry would instead occur only for $d\sim 30A$ for these parameters, i.e., the intrinsic asymmetry would dominate for a much larger range of tip-sample distances. Observation of

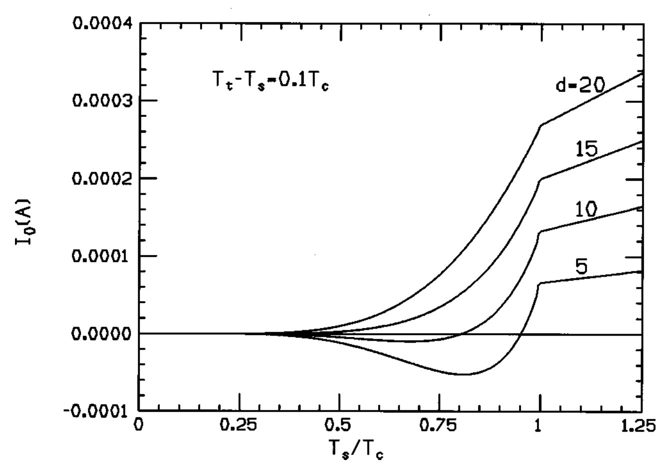

FIG. 15. Zero voltage thermoelectric current (from sample to tip) versus sample temperature in the presence of both finite gap slope, $\Delta'=0.24$, and energy-dependent transmission with barrier parameters $\phi_s=\phi_t=1$ eV and tip-sample distances given next to the curves. Since the sample is colder than the tip, positive thermopower corresponds to current going from tip to sample, which occurs when the barrier is thin and the intrinsic asymmetry dominates. For temperature gradient of opposite sign the curves would be the mirror images of these across the horizontal axis. $\Gamma=0$.

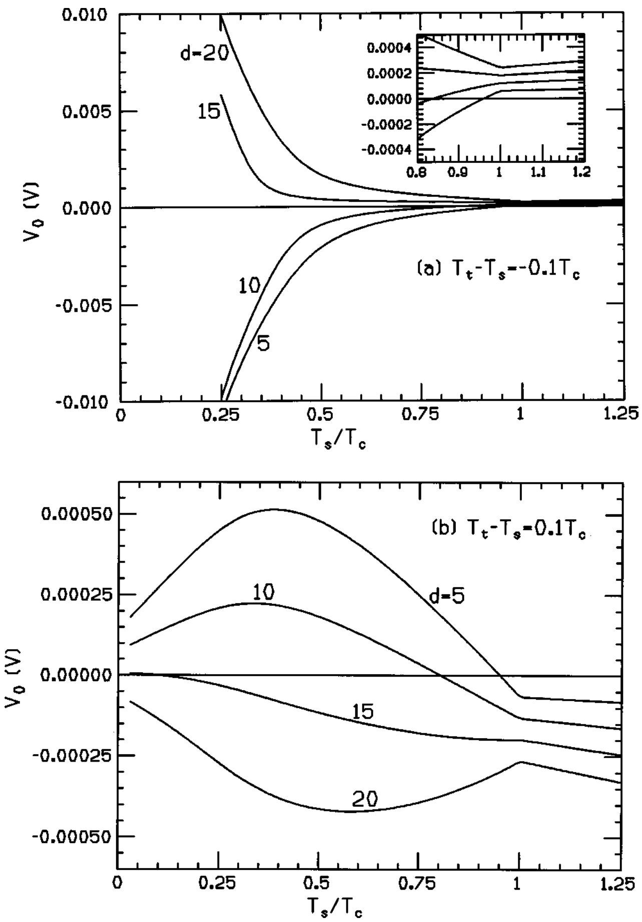

FIG. 16. Zero current thermoelectric voltage (of sample) for parameters as in Fig. 15 and (a) tip colder than sample and (b) tip hotter than sample. Thermoelectric power is positive for thin barriers and low temperatures. The inset in (a) shows the behavior close to T_c.

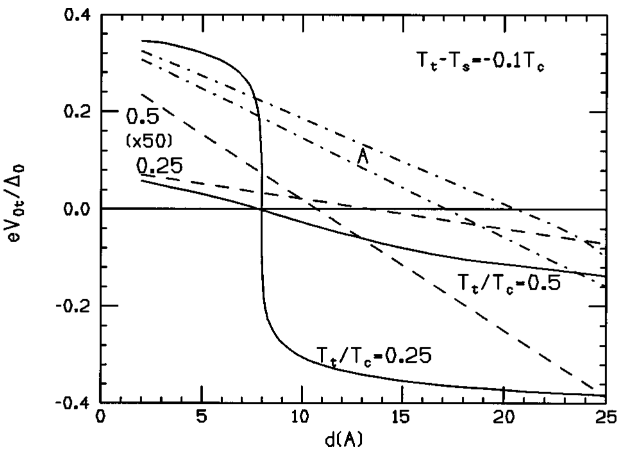

FIG. 17. Thermoelectric voltage (of tip) with $\Gamma=0$ (solid lines) and with $\Gamma=3$ meV (dashed lines) at two temperatures (labels next to the lines) for $T_t-T_s=-0.1T_c$ as function of tip-sample distance d. Sample density of states is the same as in Fig. 5. The data for $\Gamma=3$ meV are multiplied by 50. For small d intrinsic asymmetry dominates, for large d barrier-induced asymmetry dominates. Note that broadening causes the thermoelectric effect to be dominated by intrinsic asymmetry over a wider range of tip-sample distances. The tunneling asymmetry at temperature $T=0.1T_c$ is also shown for $\Gamma=0$ and $\Gamma=3$ meV (dash-dotted lines labeled A). Here again, the asymmetry remains dominated by the intrinsic effect for a larger d in the case where Γ is finite. Gap slope is $\Delta'=0.17$, work functions are $\phi_s=\phi_t=0.53$ eV. Note that the range of barrier thickness where intrinsic asymmetry dominates is substantially larger for the tunneling conductance asymmetry than for the thermoelectric effect.

the behavior discussed here would clearly evidence the competition between intrinsic and barrier-induced effects and allow for the extraction of the intrinsic gap slope.

Finally we compare in Fig. 17 the behavior of tunneling asymmetry and thermoelectric voltage as a function of tip-sample distance, in the presence of intrinsic asymmetry, barrier-induced asymmetry, and energy-dependent sample density of states. As discussed earlier, intrinsic asymmetry dominates in the tunneling asymmetry over a substantially larger range of tip-sample distances than in the thermoelectric effect. For both cases, broadening increases the range of tip-sample distances where intrinsic asymmetry dominates.

VII. DISCUSSION

We have discussed in this paper various factors that may give rise to asymmetry in STM tunneling conductance experiments. While the various effects discussed here can give rise to asymmetries of either sign, we have argued that the recent results on tunneling spectra of $Bi_2Sr_2CaCu_2O_{8+\delta}$ provide evidence for an asymmetry originating in an intrinsic property of the superconductor, an energy-dependent gap function. The slope of the gap function as a function of electronic energy implied by the experimental results is *positive*. We have suggested that further data on temperature and tip-sample dependence of the asymmetry may be able to further support this conclusion.

Furthermore, we have proposed that independent evidence for intrinsic asymmetry originating in a finite gap slope would be provided by thermoelectric experiments with STM, which should yield *positive* thermopower in certain parameter regimes. Such experiments, always yielding *negative* thermopower, have already been performed with normal metals, which suggests that they are quite feasible.

The combination of more extensive high-quality tunneling spectra and results for the thermoelectric effect should be able to determine unambiguously the sign and magnitude of the gap slope in $Bi_2Sr_2CaCu_2O_{8+\delta}$. The gap slope determines the average quasiparticle charge in the superconductor and is thus a quantity of fundamental interest. The theory of hole superconductivity[11] has predicted that the gap slope, and consequently also the average quasiparticle charge, is positive for all superconductors, and that its magnitude scales with the critical temperature. Temperature and carrier concentration dependence of the gap slope is also predicted by the theory.

Coffey and co-workers[13] have recently proposed that the observed asymmetry in tunneling conductance discussed here is evidence in favor of d-wave symmetry of the order parameter. Their analysis relies on an assumption of directional tunneling, and the parameters in their model are chosen in order to match the observed asymmetry. We note that for the case of a half-filled band and a band structure with only nearest neighbor coupling, that is an electron-hole symmetric system, their analysis predicts an asymmetry of the observed sign when the tunneling direction is parallel or close to one of the principal axes in the plane for both a *d*-wave and an *s*-wave gap. Even though the asymmetry predicted is larger for *d* wave, the essential elements giving rise to the asymmetry in this calculation appears to be the assumption of directional tunneling. While this assumption may have validity for the case of point contact tunneling it would appear not to be applicable to the STM experiments discussed here where the tip is mounted perpendicular to the Bi-O layers and tunneling is expected to yield an angular average over the *ab*-plane density of states.[1]

Recent STM experiments in the presence of a magnetic field[23] provide further strong evidence for the existence of an intrinsic asymmetry associated with the superconducting state. The data of Renner *et al.* clearly show that when the STM tip is moved from the vortex core to a region between vortices the peak for negative sample voltages grows faster than that for positive voltages, irrespective of what the sign of the asymmetry in the vortex core was. This strongly suggests that the asymmetry is directly associated with the superconducting state. As we have discussed, the opening of a gap in the absence of intrinsic asymmetry would lead to precisely the opposite effect, i.e., the positive voltage peak growing faster, due to the energy dependence of transmission. Further support for this point is provided by the fact that Renner *et al.* state that ''the sharpest contrast to map the vortex structure on BSCCO ($Bi_2Sr_2CaCu_2O_{3+\delta}$) is obtained using the conductivity at a negative sample voltage $V=-\Delta_p/e$'' (Δ_p is the peak position). In other words, the peak at negative voltages gives the strongest signal on whether the system is normal or superconducting. This is related to the prediction of the theory of hole superconductivity that photoemission should give a much clearer signal of the transition to the superconducting state than inverse photoemission.[24] Similarly one should find a change in the sign of the thermoelectric voltage as function of the tip position in measurements in the mixed state, with negative thermopower measured in the vortex core region and positive thermopower in regions far from the cores.

Future work will involve a calculation of tunneling spectra as function of position in the vortex lattice within a model with finite gap slope such as the model of hole superconductivity. Possible signatures of a finite gap slope in Josephson tunneling are being investigated. It would be of great interest to find other experimental signatures of a finite gap slope in superconductors.

ACKNOWLEDGMENTS

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