Field-induced nodal order parameter in the tunneling spectrum of $YBa_2Cu_3O_{7-x}$ superconductor

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We report planar tunneling measurements on thin films of $YBa_2Cu_3O_{7-x}$ at various doping levels under magnetic fields. By choosing a special setup configuration, we have probed a field-induced energy scale that dominates in the vicinity of a node of the *d*-wave superconducting order parameter. We found a high doping sensitivity for this energy scale. At optimum doping, this energy scale is in agreement with an induced id_{xy} order parameter. We found that it can be followed down to low fields at optimum doping but not away from it.

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I. INTRODUCTION

It is by now well established that cuprate superconductors have an order parameter with a dominant *d*-wave symmetry, characterized by node lines along the [110] and equivalent directions.[1] Nodal quasiparticles then become the dominant low energy excitations.[2] Interesting phenomena have been predicted to occur when the *d*-wave superconductor is subjected to a magnetic field perpendicular to the superconducting planes: an energy gap should develop at the nodes, which increases with the square root of the applied field. This has been explained by Laughlin[3] who assumed an additional imaginary id_{xy} component, which increases with the magnetic field. Discrete Landau energy levels were also predicted to develop in the nodal regions by Gor'kov and Schrieffer[4] and Anderson.[5] However, the observation of nodal finite energy levels has encountered theoretical and experimental difficulties.

It has been argued that in the mixed state, superfluid screening currents result in a Doppler shift of the Landau levels larger than the level spacing, rendering their observation impossible.[4,6–8] This Doppler shift will also obscure a possible id_{xy} component.[9] Tunneling experiments performed along a nodal direction, which should, in principle, be an ideal method to probe nodal states, are dominated by low energy Andreev–Saint-James surface states due to the *d*-wave symmetry, resulting in a characteristic zero bias conductance peak (ZBCP). The degeneracy of the Andreev–Saint-James states is lifted by screening currents splitting the ZBCP, an effect that can be confused with that of nodal finite energy levels. Spontaneous time-reversal symmetry breaking effects are also sometimes observed.[10–14] In addition, it is not trivial to distinguish between the predictions of the Landau states and of the minority order-parameter theories. This is because the energy of the first Landau level is equal to the amplitude of the id_{xy} component predicted by Laughlin.

To address these difficulties, we have used field cycles that enable us to distinguish between the Doppler shift and other possible spectral contributions. We have concluded that data taken in decreasing fields are essentially free of Doppler-shift effects and can be used to identify finite energy nodal states. We confirmed that this energy follows the predicted square root of field behavior at optimum doping. By extending our measurements to 20 mK, we were able to follow the evolution of these states down to fields of the order of a few 1000 G, where the Landau level interpretation is excluded due to the long length of the corresponding trajectories. Finally, we report that the doping level has a strong influence on the splitting behavior of the ZBCP. The square root behavior is not obeyed at low fields when deviating from optimum doping. Such deviations were reported before;[14,15] however, the data were taken in increasing fields and were an admixture of the studied phenomena and Doppler shift. In the new data presented and studied in this paper, the Doppler-shift contribution is essentially eliminated, and the doping effect appears far more clearly. In summary, our results favor the existence of an additional id_{xy} order-parameter component predicted by Laughlin rather than that of the formation of nodal energy levels predicted by Gor'kov and Schrieffer[4] and Anderson.[5]

This paper will be organized as follows. We begin with a theoretical background of the Doppler-shift effect. In Sec. III, we present our experimental setup enabling us to distinguish between the two contributions. Our tunneling results at optimum doping will be shown together with the low temperature measurements at 20 mK (Sec. IV). We then compare our results to theory in Sec. V and finish with our conclusions and findings.

II. THEORETICAL BACKGROUND

We wish to discuss the differences between two theoretical approaches regarding the development of finite nodal energy states under applied magnetic fields.

In the first approach, by Laughlin,[3] the free energy of a *d*-wave superconductor subjected to a magnetic field perpendicular to the superconducting planes can be minimized by the inclusion of an additional id_{xy} component to the main $d_{x^2-y^2}$ component (illustrated in Fig. 1 by the id_{xy} order parameter marked in purple). The id_{xy} component breaks the symmetry in such a way that opposite currents will flow on opposite faces of the sample, creating a magnetic moment parallel to the applied field. If the moment and the applied field are parallel to each other, the free energy will be minimized.

In the second approach, following Anderson,[5] we consider the motion of a quasiparticle in a nodal region (Fig. 1). Under an applied magnetic field **B**, it acquires a velocity component parallel to the Fermi surface. In the absence of a superconducting order parameter, it will be in one of

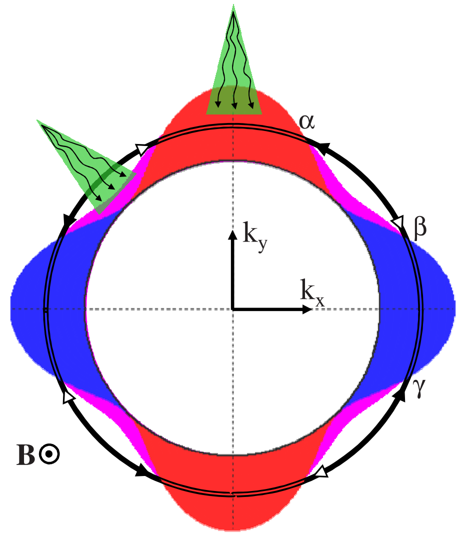

FIG. 1. (Color online) Schematic illustration of the electronic momentum space in a *d*-wave superconductor under an applied magnetic field. The *x* and *y* axes are parallel to the [100] and [010] crystallographic directions, respectively. For simplicity, we assume a cylindrical Fermi surface, a $d_{x^2-y^2}$-wave superconducting gap, nodes at $\pm 45°$ from the principal axes, and a magnetic field **B** parallel to the *z* axis. The quasiparticle cycle of multiple Andreev–Saint-James reflections forming the nodal energy level is marked by the solid line. As an alternative theoretical description for the energy scale, an induced id_{xy} order parameter (marked in purple) is predicted to develop mainly in the vicinity of the nodes. The collimation of the injected electrons in a planar tunneling configuration of the experiment is marked by the green triangles for two different junction orientations. The field-induced nodal energy scale can only be probed for tunneling along the node direction (left triangle).

the Landau levels, determined by the cyclotron frequency $\omega_c = \frac{eB}{m^* c}$, where *e* is the quasiparticle charge, *c* is the speed of light, and m^* is the effective electron mass. However, when $\hbar\omega_c < \Delta$, where Δ is the amplitude of the superconducting gap, the usual cyclotron motion is not possible. Instead, as schematically described in Fig. 1, a series of Andreev–Saint-James reflections in momentum space will occur.[13] This process can only occur at certain energy levels for which the total phase change during a Saint-James cycle is a multiple of 2π. These energy levels correspond to values of the current that flows around the Fermi surface.

Interestingly, the amplitude of the minority component is the same as the energy of the first Landau level.[4,5] The two approaches lead to the same energy exactly:

$$\varepsilon(B) = \pm 2\sqrt{\hbar\omega_c \Delta}. \tag{1}$$

Nonetheless, there are substantial differences between these two approaches. While the Gorko'v–Schrieffer–Anderson theory assumes that the order parameter is not altered by the magnetic field, its modification is a key prediction in Laughlin's theory. The latter also predicts a transition temperature above which the *d*-wave symmetry is recovered. Finally, Gor'kov and Schrieffer and Anderson predict a series of energy levels while Laughlin only predicts a finite gap value.

Tunneling along a nodal direction of a *d*-wave superconductor is done through a surface where zero-energy bound states are present due to the interference of quasiparticles that undergo Andreev–Saint-James reflections from lobes of the order parameter having phases that differ by π.[13,16] These bound states should appear as a conductance peak at zero bias in an in-plane tunneling spectrum.[17] As shown by Fogelström *et al.*, this zero bias peak splits into two spectral peaks due to a Doppler shift from superfluid currents flowing parallel to the surface.[15] The peaks bias are proportional to $v_s \cdot p_F$, where v_s is the superfluid velocity and p_F the Fermi momentum of the probed states. For example, when a magnetic field *H* is applied parallel to the surface, Meissner current Doppler shift will produce spectral peaks which are linear with *H* up to a field of the order of the thermodynamical critical field where saturation is reached [about 1 T in the case of $YBa_2Cu_3O_{7-x}$ (YBCO)].

Since a nodal energy scale and a Doppler shift will both split the zero bias conductance peak under an applied field, as has been observed,[10–12,14] one must find a method to distinguish between both mechanisms and determine the difference in the predicted field dependences. An obvious difference between them is that field-induced nodal energy scales are best observed in the absence of superfluid currents, while a Doppler-shift effect exists only in their presence.

A method that we have already used[18] consists in performing magnetic field cycles. Meissner currents are quite different in increasing and decreasing fields because the Bean–Livingston barrier which can retard the penetration of vortices through strong surface currents, up to a field of the order of the thermodynamical critical field, is only effective against flux penetration (increasing fields) and not against flux exit (decreasing fields). As a result, strong surface Meissner currents on the scale of the London penetration depth exist only in increasing fields.[19–21] Other types of current that can produce a Doppler shift are screening currents around vortices[22] and Bean's critical state currents.[23] The latter reverse sign with field reversal and, in high fields, extends into the entire thickness of the sample. They are typically weaker than the Meissner currents in the Bean–Livingston regime.

In planar tunneling experiments, electrons are injected across a dielectric barrier into the superconductor. The transmission probability decays exponentially with the increasing angle between the electron's momentum and the normal to the interface, resulting in a collimated current. In a typical junction, the momentum divergence has an angle of 10°–20° known as the tunneling cone. The width of this cone, which can vary slightly from one junction to another, will influence the Doppler shift of the zero-energy surface bound states. However, it will not modify the energy of nodal states nor that of an induced id_{xy} order-parameter component.

In summary, a zero bias conductance peak is expected to appear in a *d*-wave tunneling along the node direction and to split into two spectral peaks when a magnetic field is applied perpendicular to the CuO_2 planes due to a field-induced nodal energy scale or via a Doppler-shift effect. The two effects behave differently in a magnetic field. In the next section, we will show a way to minimize the Doppler effect which allowed us to probe the field-induced nodal energy scale alone.

TABLE I. Sample characterization. As explained in the text, from the resistivity temperature dependence measurement, $R(T)$, we can estimate the sample doping. The zero field spectral peak bias value is noted as δ_0. T_c is the temperature at zero resistivity. The transition temperature width is determined by a Gaussian fit to $\frac{dR}{dT}$ at the transition.

Name	Figs.	Orientation	Thickness (Å)	Doping regime	T_c (K)	T_c width (K)	δ_0 (mV)	T (K)
S1	2,5,7	(110)	1600	Optimal	88.1	1.4	0	0.3
S2	3	(110)	1600	Optimal	90	1	0	4
S3	3	(100)	1000	Under	84	1.5	0	4.2
S4	4,7	(110)	1200	Optimal	88.2	1.3	0	0.02
S5	6	(110)	1200	Over	87.3	1.5	1.4	4.2
S6	7	(110)	600	Under	87	1	0	4.2
S7	7	(110)	1200	Over	87.7	0.3	1.3	4.2
S8	7	(110)	1200	Over	85.7	1.1	1.6	4.2
S9	7	(110)	1200	Over	88.3	0.8	1.8	1.3
S10	7	(110)	1200	Over	86.7	0.7	1.8	1.3
S11	7	(110)	1200	Over	87.7	0.8	1.9	1.3
S12	7	(110)	1200	Over	88.3	0.6	1.5	1.3
S13	7	(110)	1200	Over	88.0	1	1.35	1.3
S14	7	(110)	1200	Over	87.9	0.3	2.25	1.3

III. EXPERIMENT

Thin YBCO films were grown using dc off-axis sputtering deposition. In order to minimize (103) oriented grains, a buffer layer of $PrBa_2Cu_3O_{7-x}$ was first deposited using rf off-axis sputtering on top of the substrate.[24] We used $SrTiO_3$ and $LaSrGaO_4$ substrates for the (110) and (100) oriented films, respectively. These films have a well defined [001] direction parallel to the surface of the film. θ-2θ x-ray diffraction patterns showed the relevant peaks for the desired orientation. Scanning electron microscopy and atomic force microscopy showed a well defined crystallographic growth and surface roughness of a few nanometers. Resistivity measurements showed the expected in-plane anisotropy. In addition, the temperature dependence of the *ab* plane resistivity allowed us to estimate the doping level in our films. It changes from a positive curvature for overdoped, linear with temperature for optimally doped, and negative curvature for underdoped films.[25–28] *I-V* characteristics were measured using a current source and a digital voltmeter. The tunneling conductance spectra were calculated by differentiating the $I(V)$ curves. Table I shows the characterization values for the 15 representative junctions used in the figures of this paper.

In our experiments, the tunneling junction is created by placing an indium electrode on top of the surface of freshly prepared thin YBCO films. At the metal-superconductor interface, a thin insulating indium oxide layer is then formed, which is stable over weeks and many thermal cycles. We can verify the quality of the tunneling junction by several methods. First, by lowering the temperature below the indium critical temperature, we can measure the well-known indium tunneling spectrum which dominates the low energy spectra. Also, the indium spectrum disappears as we increase the magnetic field or heat up the sample above the indium's critical field and temperature, respectively. Second, we ensure that the high bias conductance is insensitive to the magnetic field and temperature below the YBCO critical temperatures, as expected for tunneling spectroscopy.

Our sample and the field configuration are favorable for several reasons. First, the magnetic field is applied parallel to the surface (which avoids threading the tunnel junction with vortices) and at the same time perpendicular to the CuO_2 planes, as required for the observation of finite energy nodal states. Second, the undesired Doppler shift due to superfluid currents is minimized because in our geometry, $v_s \cdot p_F$ is at a minimum since the dominant currents flow parallel to the interface. Third, by comparing data taken in increasing and decreasing fields, it is possible to identify the effect of the Doppler shift due to strong surface Meissner currents that exist only in increasing fields.[19–21] Therefore, measurements in decreasing fields are less affected by the Doppler effect. Also, the intensity of the Meissner currents themselves can be minimized by using films whose thickness is smaller than the London penetration depth.[18,29] Finally, we have performed experiments both on (110) and (100) oriented films. While for the [110] direction we expect to probe the field-induced energy scale, for the [100] direction, it should not be observed. We emphasize that in our method due to the planar geometry of the junction and the resulting tunneling cone, we probe a specific direction in k space rather than the **k** averaged local density of states, as probed by scanning tunneling microscopy.

IV. RESULTS

A. Probing the field-induced nodal energy scale

In Fig. 2, we show tunneling spectra for an optimally doped (110) oriented film as a function of magnetic field

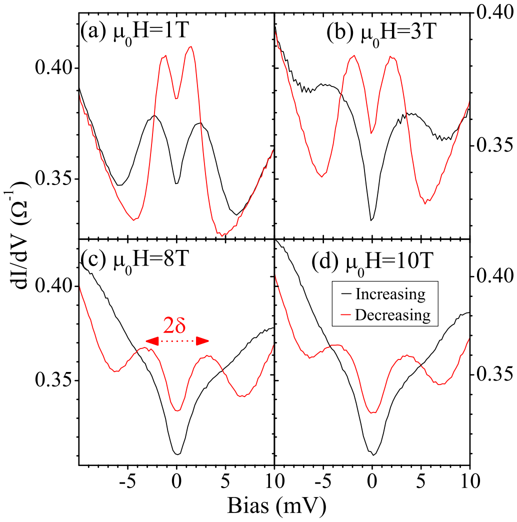

FIG. 2. (Color online) Tunneling spectra, $dI/dV(V)$, for various magnetic fields (sample S1, at 0.3 K). Increasing from zero magnetic field (black) and decreasing from 16 T (red), they present different tunneling spectra in a given magnetic field. We define 2δ as the distance between the two maxima in the spectrum. For a given magnetic field, $\delta(H)$ is always larger in increasing fields than in decreasing one. This is due to the Doppler-shift effect resulting in shifting and smearing of the field-induced nodal energy scale peaks to higher biases in increasing magnetic fields. In fields higher than 4 T, the peaks can only be identified in decreasing magnetic fields.

applied perpendicular to the CuO_2 planes. We notice that the spectral peak bias values, $\delta(H)$, are always larger in increasing rather than in decreasing fields. In addition, the peak seen in decreasing fields is well defined for all fields, while that seen in increasing fields becomes very broad and is too broad to be identified (Fig. 2), while in decreasing fields, it remains well defined up to more than 22 T (see, for example, Fig. 7).

We demonstrate in Figs. 3 and 4 that the spectral peak bias value does not increase linearly at low fields and does not saturate at high field. This is in contradiction with the Doppler shift theory of zero-energy surface bound states. A substantial difference between the behavior of (100) and (110) oriented films is observed. While for thin (110) films the spectral peaks are clearly seen even at low fields, no such peaks can be detected in thin (100) films. Since in (100) films the only splitting mechanism is the Doppler-shift effect, the absence of the spectral peaks in such films is indicative of the insignificance of that effect in films thinner than the London penetration depth. Upon increasing the (100) oriented film thickness, the contribution of the Doppler shift is also increased and the spectral peaks are recovered.[12]

The third qualitative evidence supporting the argument that we probe a field-induced nodal energy scale rather than a Doppler-shifted zero bias conductance peak comes from the shape of the tunneling conductance at zero bias. While the Doppler-shifted ZBCP results in a V-shape conductance,[15,22] one expects a U-shaped conductance at zero bias for the field-induced nodal energy scale.[30] Because it is difficult to distinguish between these two shapes due to thermal population effects, one should perform measurements at very low temperatures. In Fig. 4, we show tunneling

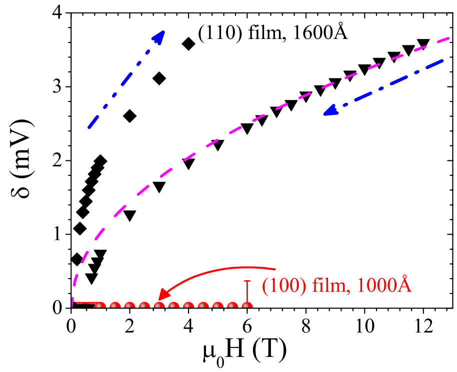

FIG. 3. (Color online) Spectral peak bias value, δ, as a function of magnetic field at 4.2 K. Black triangles (diamonds) represent the peak positions in decreasing (increasing) magnetic fields for a (110) oriented film having a thickness of 1600 Å (sample S2). While a Doppler shift in increasing fields prevents observation of the field-induced nodal energy scale, the measurements in decreasing fields are free of the Doppler-shift effect and allow unambiguous identification of the field-induced nodal energy scale. The dashed purple line is a fit to $\delta = AH^{1/2}$, with $A = 1.02 \pm 0.05$ meV/$T^{1/2}$, in excellent agreement with Eq. (1). The red circles represent (100) oriented film with a thickness of 1000 Å (sample S3). The field-induced nodal energy scale is not observed, as expected.

spectra taken at 20 mK under high magnetic fields. The observed U-shaped conductance is in agreement with the field-induced nodal energy scale scenario and contrasts the Doppler-shift model.

We have therefore demonstrated that a Doppler shift due to Meissner screening currents does not play an important role in our tunneling measurements of thin films in decreasing fields. However, the effect of a nearby vortex[22] or Bean's critical state currents[23] could still take place in a decreasing magnetic field. These currents reverse their polarity when decreasing the magnetic field from the maximum value reached. If they dominate, the total current should be zero at some field. In this case, a zero bias peak should reappear at a finite magnetic field. Such a behavior was never observed, ruling out that the spectral peaks in decreasing fields arise from the effect of a nearby vortex or strong Bean critical state currents (see, for example, a field cycle in Fig. 8).

Finally, we note that the peak position measured in decreasing fields is extremely reproducible for a variety of samples and junctions. This is in contrast with the expected behavior of the Bean critical currents and the Doppler-shift effect that are strongly dependent on film thickness, tunneling cone, and surface barrier formation.

In contrast with the difficulties encountered in trying to explain the data shown in Figs. 2 and 3 by a Doppler shift of zero-energy surface bound states, a field-induced nodal energy scale provides a reasonable explanation. As shown in Fig. 4, the data taken in decreasing fields fit the predicted square root dependence on the magnetic field. The field hysteresis shown in Figs. 2 and 3 can be understood as due to a Doppler shift by Meissner currents in increasing fields; the peaks are shifted to higher bias and are broadened until they cannot be identified anymore in very high fields.

To summarize, we have shown that for an optimally doped sample, tunneling measurements on a (110) oriented

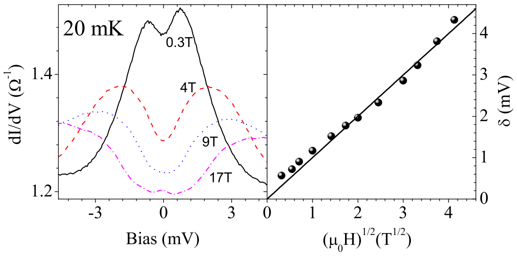

FIG. 4. (Color online) (a) Tunneling spectra taken at 20 mK in decreasing magnetic fields (sample S4). Note the constant conductance in the vicinity of zero bias supporting the existence of an energy scale $\varepsilon = \pm\delta(H)$. (b) Spectral peak value in decreasing fields versus square root of the applied magnetic field measured at 20 mK. The line is a fit to high fields having a slope of 1.01 $\mathrm{meV/T^{1/2}}$.

film reveal an energy scale that can be understood either as an additional complex order parameter in the form of id_{xy} or as the first Landau state in the vicinity of the d-wave node.

B. Implausibility of nodal Landau levels

After ruling out the Doppler-shift effect, we now concentrate on distinguishing between the nodal Landau-state approach of Gor'kov and Schrieffer[4] and Anderson[5] and the nodal order-parameter component predicted by Laughlin.[3] Although the first Landau level and the nodal order parameter have identical field dependences, the following evidences favor the latter.

An important feature is observed in Fig. 4: only two peaks at $\pm\delta$ are visible. This is incompatible with the theories in Refs. 4 and 5, which predict a series of energy levels that should manifest themselves as peaks in the tunneling spectrum. These peaks have never been observed. One could argue that the higher energy levels are hidden due to scattering. The visibility of the first peak at relatively high temperatures ($T > 4.2$ K) under small magnetic fields ($H \simeq 0.3T$) suggests that the scattering processes are not strong enough to obscure the higher peaks at 20 mK and 9 T; yet, these peaks are not observed in Fig. 4.

Further evidence comes from estimating the trajectory length at the node area. If we define θ as the angle from the antinodal direction at which the particles meet the superconducting gap Δ and perform an Andreev–Saint-James reflection, and $\pm\alpha$ as the trajectory angle measured from the node, we get $\alpha = 2(45-\theta)$. Using Eq. (1), we calculate α for small angles to be $\alpha = \frac{\varepsilon}{\Delta} = 2\sqrt{\frac{\hbar eB}{mc\Delta}}$. In the node area, the trajectory can be treated classically using the cyclotron frequency ω_c. In the node vicinity, the trajectory time $t = \frac{\alpha}{\omega_c}$ gives the trajectory length x:

$$x = tV_F = 2\sqrt{\frac{\hbar mc}{eB\Delta}}V_F, \qquad (2)$$

where V_F is the Fermi velocity. Using Eq. (2), we calculate that for $B = 1$ T, $\Delta = 20$ meV, and $V_F = 10^6$ m/s, the trajectory length is 8600 Å, which is several times larger than the film's thickness. Scattering from the surface will not allow the formation of Landau states unless the applied field is of the order of 100 T. However, in fact, the nodal scale is observed at low temperatures (20 mK) down to a fraction of a tesla (Fig. 4).

Two additional indications favoring Laughlin's theory over that Gor'kov and Schrieffer and Anderson will be discussed in the following sections: the effects of doping in Sec. IV C and a first-order phase transition of the field-induced nodal energy scale measured by Elhalel *et al.*[29] in Sec. V.

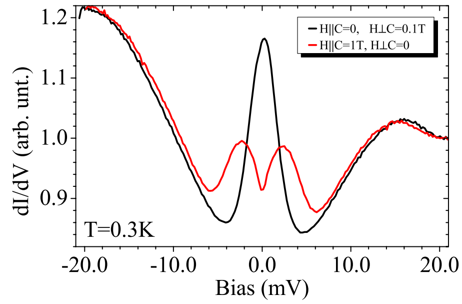

FIG. 5. (Color online) Tunneling conductance for a (110) film taken at 0.3 K (sample S1). The black line is measured in a magnetic field of 0.1 T applied parallel to the CuO_2 planes where the indium counterelectrode is in its normal state without field-inducing spectral peaks. The red curve is for 1 T in increasing fields applied perpendicular to the CuO_2 planes, where field-induced nodal energy scale peaks are observed.

C. Effect of doping on field-induced nodal energy scale

1. Underdoped case

For underdoped samples at zero field, only a single peak is observed at zero bias (the ZBCP). We shall now demonstrate that this is not due to the thermal smearing of two peaks at finite energy. To do that, we reduced the temperature to 0.3 K while applying a small magnetic field perpendicular to the c axis (and parallel to the sample's surface). This field quenches superconductivity in the indium counterelectrode but has no effect on the ZBCP, as has been demonstrated by Krupke and Deutscher.[12] The results are shown in Fig. 5. The sample is slightly underdoped at $T_c = 88.1$ K. The black solid line shows a sharp zero bias peak without any observable splitting. An upper limit for the bias of possible spontaneous peaks is extracted from this measurement to be $2k_BT = 0.06$ meV. We note that even at 16 T, we do not observe any zero bias peak splitting under this configuration, which ensures the sample's orientation. By contrast, when the field is applied parallel to the sample's c axis, the two spectral peaks at ± 2.2 meV are clearly seen.

In Fig. 6, we show a typical field dependence of the nodal energy scale. At low fields up to 1 T, a single peak is observed at zero bias. Upon increasing the magnetic field, a field-induced nodal energy scale appears at lower energies when compared to the case of optimum doping, until it reaches a crossover field (marked by H^* in Fig. 6) at which it recovers the optimally doped $\sqrt{H}$ behavior. In underdoped samples, there appears to be a well defined field below which the ZBCP does not split. This field increases rapidly with underdoping.

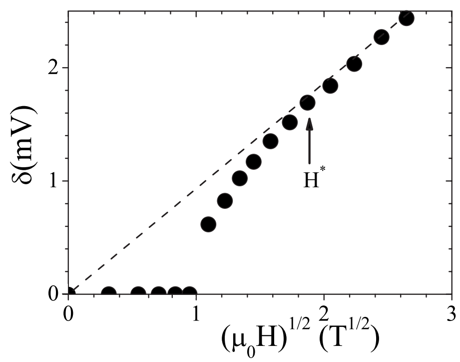

FIG. 6. Spectral peak value, δ, as a function of the square root applied magnetic field (sample S6). The line is a linear fit to the high magnetic field data. It has a slope of 0.9 $meV/T^{1/2}$.

2. Overdoped case

All overdoped samples exhibit spontaneous zero field spectral peaks. Dagan and Deutscher[14] have shown a correlation between the spontaneous spectral peak bias values, δ_0, and the rate at which this bias increases with field. They concluded that the spontaneous peaks are due to a modification in the order-parameter symmetry near the surface in the vicinity of optimum doping from pure d wave for underdoped samples to $d \pm id_{xy}$ or $d \pm is$ in overdoped ones.

In this section, we present a study of overdoped samples with different oxygen doping levels in high *decreasing* magnetic fields starting from fields as high as 32.4 T. The results are summarized in Fig. 7. For slightly overdoped films (see all data points above the dashed line), we find zero field spectral peaks, with a minute shift at low magnetic fields. The zero and low magnetic field data are qualitatively in agreement with the measurements of Dagan and Deutscher.[14] However, new behavior is observed at high magnetic fields. At these high fields, all data points collapse to a single line having a slope of 1 $meV/H^{1/2}$ (dashed line). This is the same slope as found at high fields for optimally doped and underdoped samples.

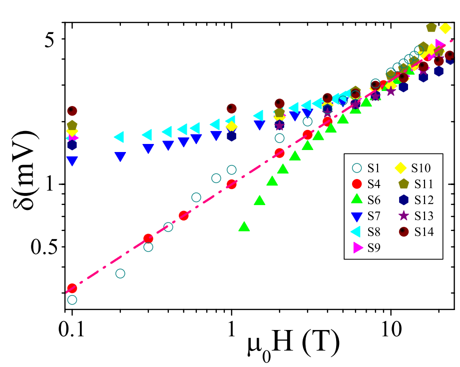

FIG. 7. (Color online) Spectral peak positions, δ, as a function of magnetic field for various doping levels films at log-log scale. All data shown here were taken in decreasing magnetic field. In overdoped films, the tunneling spectrum exhibits peaks at zero magnetic field, where, for underdoped ones, peaks are missing at low magnetic fields. All δ values coincide at high magnetic fields and follow the dashed line having a slope of 1 $meV/H^{1/2}$.

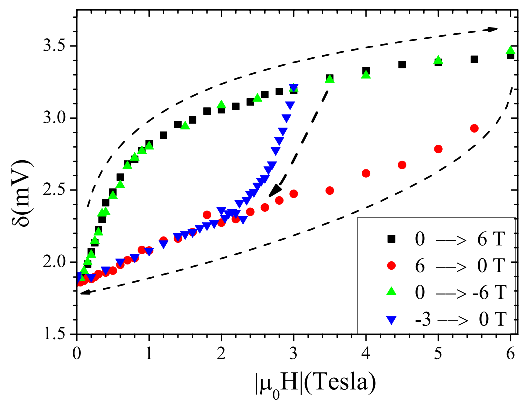

FIG. 8. (Color online) Spectral peak position, δ, as a function of the absolute magnetic field in both polarities (sample S5). We found no evidence that the polarity of the magnetic field influences the peak position for a given field.

It has been suggested that finite energy peaks can result from trapped vortices and their associated Doppler-shifting supercurrent at the surface.[22] Another explanation could be a minority imaginary component of the superconducting order parameter.[15,31] Both cases break time-reversal symmetry, as the spontaneous currents flow in a specific direction. Applying additional currents should then result in either increasing or decreasing of the net current, assuming that the time-reversal symmetry is broken macroscopically. However, we find no differences in the tunneling spectra for both polarities. This is shown in Fig. 8. When the magnetic field is increased for a zero field cooled sample, the spontaneous peak value should reduce for about one-half of the samples, or when the induced current is opposite to the spontaneous one. However, in over 100 junctions measured in this study, we never observed that the spontaneous peak's bias decreased with increasing magnetic field.

Theoretical studies by Asano *et al.*[32,33] and Kalenkov *et al.*[34] show that surface impurities could also result in spontaneous spectral peaks. These models predict that scattering of impurities cause bound states that dominate the low bias spectrum. However, the Andreev–Saint-James bound states will still be present, and a three peaked structure at zero bias should be present. We did not observe such behavior in any of the junctions presented here. Moreover, according to these models, the "impurity" spectral peak bias value should be proportional to the amount of impurities at the surface. In the case of a clean junction, a zero bias peak should be present. However, when a magnetic field is present, a splitting via a Aharonov–Bohm-like phase shift will occur.[32] This means that at high bias, the zero field spectral peaks will be shifted by an external magnetic field and, for a clean interface, no magnetic field shifted peaks should be observed. However, an earlier study by Dagan and Deutscher[14] (see also Fig. 7) shows the opposite trend. At low magnetic fields, junctions showing a spontaneous spectral peak shift to a lesser degree than those showing a zero bias conductance peak. This rules out impurities as a possible explanation for spontaneous peaks.

V. DISCUSSION

As mentioned before, both theories–Laughlin's[3] id_{xy} theory and Landau states by Gor'kov and Schrieffer[4] and Anderson,[5] result in exactly the same field dependence for the induced nodal scale. However, there are two main differences between these approaches that can be checked experimentally. First, Laughlin predicts a weak first-order phase transition to the id_{xy} state which is not predicted by the Landau-state theorem. This phase transition was, in fact, demonstrated recently by Elhalel *et al.*[29] Second, the Gor'kov–Schrieffer–Anderson theory predicts a series of states while in Laughlin's theory, only one energy scale appears. The second peak is not observed even down to 20 mK. Additionally, we showed that the trajectories between two successive Andreev–Saint-James reflections are much longer than the film thickness. It is therefore unlikely that such states exist in the thin films used in our measurements.

Laughlin's theory, however, has no doping dependence, which is a key feature observed in our measurements. Following Laughlin, Deutscher *et al.*[35] suggested a doping dependence correction to the free energy in the form

$$F = a\delta^2 + b\delta^3 - c\delta B. \tag{3}$$

Here, b and c are calculated by Laughlin. a is a doping dependent term, $a = a_0(x - x_c)$, where a_0 is a negative constant and x_c is the optimal carrier concentration.

Using Eq. (3), we calculate a minimum F for $\delta \neq 0$ at zero field only in the overdoped regime where $x > x_c$, while at higher fields, the square root behavior of δ is recovered for both underdoped and overdoped regimes. We can therefore conclude that our data are better described by the modified Laughlin theory[3,35] with an additional order-parameter component.

We have shown that time-reversal symmetry is not broken macroscopically even when spontaneous spectral peaks appear in the tunneling measurements. An experiment similar in concept was conducted by Tsuei *et al.* where they measured the spontaneous half-flux-quantum vortex in a tricrystal experiment and found no difference between spontaneous vortices at opposite polarities.[36] The tricrystal experiment claimed to rule out a minority component to the superconductor order parameter.

Because our measurements, as well as the tricrystal ones, are macroscopic, domain regions with alternating spontaneous current directions can reconcile both experimental results. The origin of such regions could be alternating minority order parameters having $\pm id_{xy}$ or $\pm is$ symmetries as both plus and minus states are degenerate. In fact, such configurations could be energetically favorable. In such a case, at the domain wall region, the spontaneous current should be zero. Our technique has the advantage that it can probe the zero magnetic field state and the microscopic time-reversal symmetry breaking regardless of the spontaneous current direction at the microscopic scale.

Since the order parameter changes over a length scale set by the superconductor coherence length, one should be able to find nanometer scale regions (the domain wall region) where the minority component order parameter is zero, while, in other regions (inside the domains), it should be finite. Only a microscopic study, for example, with a scanning tunneling microscope, may be able to probe such domains. Furthermore, in the domain wall region, the order-parameter symmetry should be purely d wave. Therefore, measurements aiming at detecting node-line excitations, such as thermal conductivity, may be dominated by the domain wall regions.

VI. SUMMARY

In conclusion, tunneling experiments revealed that the spectrum of quasiparticle states in nodal regions of a d-wave superconductor is profoundly modified by applying a magnetic field perpendicular to the CuO_2 planes. Doppler shift of the field-induced nodal energy scale has been identified and, as expected, it is large enough to prevent observation of a field-induced nodal energy scale; however, it has been minimized by choosing an appropriate geometry. We showed that the zero field spectral peaks cannot be explained by either inelastic scattering or trapped vortices at the surface but rather by a domainlike structure of minority order-parameter components with alternating signs. We studied the interplay between the spontaneous spectral peaks with the formation of the field-induced nodal energy scale and film doping. Although the low energy states are in agreement with theories based either on the explicit description of Andreev–Saint-James reflections by the order parameter away from the nodes[4,5] or on a free-energy expression that takes into account a gain in energy due to the interaction between the applied field and the magnetic moment created by an id_{xy} component,[3] the absence of higher energy level peaks and the unreasonably long length of the trajectories that would be necessary to observe Landau levels are in contradiction with the former, while the doping dependence and the first-order phase transition observed by Elhalel *et al.*[29] are in favor of the existence of a field-induced id_{xy} order parameter.

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