Influence of temperature on the nodal properties of the longitudinal thermal conductivity of YBa$_2$Cu$_3$O$_{7-x}$

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The angle dependence at different temperatures of the longitudinal thermal conductivity $\kappa_{xx}(\theta)$ in the presence of a planar magnetic field is presented. In order to study the influence of the gap symmetry on the thermal transport, angular scans were measured up to a few Kelvin below the critical temperature $T_c$. We found that the four-fold oscillation of $\kappa_{xx}(\theta)$ vanishes at $T > 20$ K and transforms into a one-fold oscillation with maximum conductivity for a field of 8 T applied parallel to the heat current. Nevertheless, the results indicate that the $d$-wave pairing symmetry remains the main pairing symmetry of the order parameter up to $T_c$. Numerical results of the thermal conductivity using an Andreev reflection model for the scattering of quasiparticles by supercurrents under the assumption of $d$-wave symmetry provide a semiquantitative description of the overall results.

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I. INTRODUCTION

The order parameter in high-temperature superconductors (HTS) has been proved to have mainly a $d_{x^2-y^2}$ pairing symmetry. In particular, tri-crystal phase-sensitive measurements have determined the presence of a predominant $d_{x^2-y^2}$-gap symmetry up to the superconducting critical temperature $T_c$ over, if any, other minor components smaller than 5% of that with the $d_{x^2-y^2}$-symmetry. Thermal transport measurements have also shown the predominance of this symmetry for the gap function. In particular, when a magnetic field is rotated parallel to the CuO$_2$ planes, the longitudinal thermal conductivity shows a fourfold oscillation which can be explained in terms of both Andreev scattering of quasiparticles by vortices (AS) and Doppler shift (DS) in the energy spectrum of the quasiparticles if one takes a $d_{x^2-y^2}$-gap into account. Nevertheless, the fourfold oscillation in the thermal conductivity has been resolved up to $\sim 15$ K. Above this temperature, no direct evidence for this type of symmetry from this kind of measurements has been published. Furthermore, deviations from the expected angular pattern within a pure $d_{x^2-y^2}$-symmetry have been attributed to the effect of pinning of vortices.

The variation of the thermal conductivity as a function of angle $\theta$ between the heat current and the magnetic field applied parallel to the CuO$_2$ planes depends mainly on the heat transport by quasiparticles, their interaction with the supercurrents (vortices) and the symmetry of the order parameter. An angular pattern showing properties of the order parameter symmetry can only be achieved when the temperature is low enough so that the quasiparticle momentum is close to the nodal directions of the gap. Otherwise, thermal activation would also induce quasiparticles at different orientations from those of the nodes and hence, the sensitivity of the probe to measure gap characteristics will be reduced. The first question we address in this paper is related to the temperature range at which the nodal characteristics of the gap are directly observable by thermal conductivity. The second question we would like to clarify in this paper is whether the thermal activation of quasiparticles with increasing temperature, treated phenomenologically within a Fermi liquid approximation for thermal transport including the $d_{x^2-y^2}$-gap function, can explain the experimental results in the whole temperature range, in particular the change of symmetry of $\kappa_{xx}(\theta)$ as a function of temperature. In this paper, we calculate numerically the thermal conductivity at different angles and temperatures at fixed magnetic field, assuming an Andreev reflection model for the scattering of quasiparticles by supercurrents, originally proposed by Yu et al. within the two-dimensional BRT expression for the thermal conductivity, and compare it to the experimental data.

Angle scans in a magnetic field applied parallel to the CuO$_2$ planes were performed in order to measure the angular variation of the longitudinal thermal conductivity $\kappa_{xx}(\theta)$ in two single crystals of YBa$_2$Cu$_3$O$_{7-x}$ high-temperature superconductor. The overall results agree with the theoretical model and confirm the predominance of the $d_{x^2-y^2}$-gap up to $T_c$. The measurements provide also new results that improve our knowledge of the thermal transport at temperatures at which the nodal properties of the gap are not directly observable.

Following the experimental and sample details of the next section, we present in Sec. III the main experimental results. In Sec. IV we describe the used model and compare it with the experimental data. A brief summary is given in Sec. V.

II. EXPERIMENTAL AND SAMPLE DETAILS

In order to rule out effects concerning shape and structure characteristics of the crystal we have used two different samples of YBa$_2$Cu$_3$O$_{7-x}$ (YBCO): a twinned single crystal with dimensions (length × width × thickness) 0.83 × 0.6 × 0.045 mm$^3$ and critical temperature
FIG. 1: Top view of the sample arrangement and the definition of the angle $\theta$.

$T_c = 93.4$ K previously studied in Refs.4,9,10,11 and an untwinned single crystal with dimensions $2.02 \times 0.68 \times 0.14$ mm$^3$ and $T_c = 88$ K. For the measurement of the thermal conductivity, a heat current $J$ was applied along the longest axis of the crystal studied. In the untwinned sample, $J$ was parallel to the $a$-axis and in the twinned crystal was parallel to the $a/b$-axes (twin planes oriented along (110)). In both cases the position of the lattice axis with respect to the crystal axis was determined using polarized light microscopy and X-ray diffraction.

The longitudinal temperature gradient ($\nabla_x T$) was measured using previously calibrated chromel-constantan (type E) thermocouples and a dc picovoltmeter. Special efforts were made in order to minimize the misalignment of the plane of rotation of the magnetic field applied perpendicular to the CuO$_2$ planes of the sample. This misalignment was minimized step by step, measuring the angle dependence of $\kappa_{xx}(\theta)$ until a satisfactory symmetrical curve was obtained. In this way, we estimate a misalignment smaller than $0.5^\circ$. An in-situ rotation system enabled measurement of the thermal conductivity as a function of the angle $\theta$ defined between the applied field and the heat flow direction along $+\hat{x}$, see Fig. 1. For more details on the experimental arrangement see Ref.4.

As pointed out by Aubin et al.4 and observed in Refs.4,9, the effect of the pinning of vortices plays an important role in determining the correct angle pattern in this kind of measurement. In fact, when the angle of the magnetic field is changed, a non uniform vortex distribution due to pinning forces may appear. As argued in Ref.4, the pinning of the Josephson-like vortices parallel to the planes is strongly affected by vortices perpendicular to the planes which may appear due to the misalignment of the crystal axes with respect to the applied magnetic field. In this situation, even hysteresis in the angular patterns of the thermal conductivity can be measured. We note that pinning of vortices is influenced by the distribution of the oxygen vacancies in the sample as well as by defects and impurity centers. Thus, in order to rule out the influence of this effect in the measurements, a field-cooled procedure have to be used. This procedure consists of two steps. In the first step the sample is driven into the normal state by heating to a few Kelvin above $T_c$ and the angle is changed. Secondly, it is cooled down to the desired temperature at constant field.

In Fig. 2 we show the longitudinal thermal conductivity as a function of the temperature in both crystals at zero magnetic field. As argued by many authors (see for example Refs.13,14,15), the observed behavior in Fig. 2 provides qualitative information about the quality of the sample. The height of the peak observed in the temperature dependence of the thermal conductivity is related to the relative contributions between the density of impurity scattering centers and the strength of the inelastic electron-electron scattering. Crystals showing large peaks may have a small amount of impurity scattering centers15 and/or larger quasiparticle-related inelastic contribution. The origin of the peak is explained in terms of a competition between the decrease of the in-
III. EXPERIMENTAL RESULTS AND DISCUSSION

The angular patterns of the longitudinal thermal conductivity $\kappa_{xx}$ at different temperatures for the twinned and untwinned crystals are shown in Figs. 3 and 4 respectively. At low enough temperatures ($T < 20K$), a magnetic field parallel to the CuO$_2$ planes of the sample produces a fourfold oscillation in the longitudinal thermal conductivity as the field is rotated from $\theta = 90^\circ$ to $\theta = -90^\circ$. We note that at both $\theta = 90^\circ$ and $\theta = -90^\circ$ the magnetic field is perpendicular to the heat flow. At $\theta = 0^\circ$ the field is parallel to the heat flow (see Fig. 1). As noted in Refs. 2 and the angular patterns are not free of the phononic contribution to the total thermal conductivity. However, this contribution seems to be constant under variations of the magnetic field orientations for two reasons. 1) For fields parallel to the CuO$_2$ planes, vortices are unlikely to have a normal core (Josephson vortices), making a change of the phonon attenuation with field and angle unlikely. 2) There is no experimental evidence that the phononic contribution does change substantially with field. The similarity between the temperature dependencies of $\kappa_{xx}$ and $\kappa_{xy}$ below $T_c$ where a large change in the quasiparticle density occurs indicates phonon-electron scattering. In fact, no remarkable phonon-electron scattering have been addressed in those experiments where the measured magnetic field dependencies are larger than the oscillation amplitudes measured in this paper. Therefore, measurable contributions of the phonons via quasiparticle-electron scattering are more unlikely to occur in the angular patterns. This characteristic makes the angular profiles of the thermal conductivity suitable to be compared with electronic models by using the quantity $\kappa_{xx}(\theta) - \kappa_{xx}(90^\circ)$ where the phononic contribution is subtracted.

As pointed out in Ref. 17, the variation of the thermal conductivity in Figs. 3 and 4 can be explained with a model involving a Doppler shift (DS) in the energy spectrum of the quasiparticles along with an accurate inclusion of the impurity scattering and/or assuming Andreev scattering (AS) of quasiparticles by vortices. Briefly, in the mixed state the quasiparticles are in the presence of a phase gradient produced by the superfluid flow of the vortices. Thus, as viewed from the laboratory frame they experience a Doppler shift in their energy spectrum given by the scalar product of the momentum and superfluid velocity, $\mathbf{p} \cdot \mathbf{v}_s$. When a quasiparticle of momentum $\mathbf{p}$ is moving parallel to the magnetic field, the product is zero and hence, no DS occurs. Thus, the angular characteristic of this effect is to produce an excess of quasiparticles in the direction perpendicular to the field and thereby, reducing the “local” thermal resistance at this orientation. When the field is placed parallel to the heat current, the Doppler shift is the same for both nodal directions at $\theta = 45^\circ$ and $\theta = -45^\circ$. Therefore and since thermal resistances must be added in parallel, a simple picture in which there are only quasipar-
particles at the nodes would produce a fourfold oscillation in the thermal conductivity with opposite sign to that observed in the measurements. However, as pointed out in Refs.\textsuperscript{14,15,24}, the DS affects both the carrier density as well as the scattering rate of quasiparticles, and since at high enough temperatures the latter dominates, the quasiparticles move more easily parallel to the magnetic field. Therefore, the sign of the fourfold oscillation observed in the experiments is also recovered within this picture.

In the AS mechanism a phase gradient, namely, the superfluid flow surrounding the vortices may induce Andreev reflection of the quasiparticles\textsuperscript{25}. Thus, the DS in the energy spectrum $p \cdot v_s$ is implicitly taken into account in the AS picture. As viewed from the laboratory frame, when the quasiparticle energy equals the value of the gap, then the quasiparticle is transformed into a quasihole reversing its velocity and hence, decreasing its contribution to the thermal conductivity. Thus, a quasiparticle with momentum $p$ parallel to the magnetic field does not experience a DS and hence, no Andreev reflection can take place. The field acts as a filter for the quasiparticle that contributes to reduce the total temperature gradient. Note that the AS picture gives rise to a fourfold oscillation in the thermal conductivity as the magnetic field is rotated parallel to the CuO$_2$ planes without more considerations.

Then, qualitatively both AS and DS can explain in principle the angle profiles observed at low temperatures. However as pointed out in Ref.\textsuperscript{24}, neither AS nor DS alone can explain the magnetic field dependence of the oscillation amplitude in the whole field range $0 \, T \leq B \leq 9 \, T$. Therefore, a more realistic picture of the thermal transport should take both into account. As argued in Ref.\textsuperscript{25}, this scenario produces different regimes influenced by the predominance of either the DS or the AS in the magnetic field dependence. Thus, at constant temperature the strength of the magnetic field becomes the parameter that changes the regime. At low magnetic fields the DS dominates and the AS dominates at high fields\textsuperscript{14,27}. In both cases, an increase of the oscillation amplitudes with increasing magnetic field strength is predicted. In Fig.\textsuperscript{3} the oscillation amplitudes of the thermal conductivity at $3 \, T$ and $8 \, T$ for different temperatures in the twinned crystal are shown.

As the temperature increases the fourfold oscillation is no longer observable, see Figs.\textsuperscript{3} and \textsuperscript{4}. This fact can be understood if one takes into account the thermal activation of the quasiparticles at different orientations from those of the nodes of the order parameter. In other words, if we suppose that the same processes that govern the thermal transport at low temperatures (AS and DS) are responsible for the high temperature behavior as well, we have to conclude that an increasing number of carriers appears in the direction of the heat current as the temperature is raised. As we shall see below from the numerical results using the AS model, this is, in fact, what takes place as the temperature is increased. However, although the observed angle profiles can be well described by the numerical analysis, the amplitude of the oscillations are smaller than the simulation results at $T > 30 \, K$. As argued for the peak of the curves in Fig.\textsuperscript{2} and shown in the following section, this discrepancy can be solved by the inclusion of an inelastic scattering into the calculations\textsuperscript{14,15}.

In Fig.\textsuperscript{4} we show the angle dependence of the untwinned crystal at $8 \, T$. Similar angle profiles as for the twinned sample have been found, but the amplitude of the oscillation is considerably smaller. This can be explained in terms of a larger concentration of impurity scatterers in the untwinned sample, as discussed above and in Ref.\textsuperscript{24}. In fact, if the impurity scattering rate is sufficiently large, the relative weight of the directionality of the AS and DS mechanisms to $\kappa_{xx}(\theta)$ weakens. The influence of the impurity scattering has been considered in the original models by inclusion of an impurity rate\textsuperscript{2,4,23,24}.

We note also that the symmetry of the $\kappa_{xx}(\theta)$—curves differs slightly for the twinned and untwinned samples at the same absolute or reduced temperature. The effect of impurity scatterers can be accounted for by an impurity

![FIG. 4: Angle dependence patterns of the longitudinal thermal conductivity at different temperatures and 8 T for the untwinned sample. The results are normalized to the value of the thermal conductivity when the field was applied perpendicular to the heat current, at $\theta = 90^\circ$.](image-url)
dependent gap parameter as done in Ref. 23. In general, however, it can be viewed as an effect related to the ratio of the modulus of the gap and the density of states of the quasiparticles at a temperature T and momentum p.

Therefore, since our twinned sample has a larger critical temperature $T_c$ than our unwinned sample, the oxygen deficiency in the latter could be also responsible for a reduced density of states, and therefore, for a different angle patterns respect to the sample with a higher $T_c$.

Figure 5 shows the complete angle patterns measured in the twinned crystal at 8 T. At $T > 78$ K no clear change in the thermal conductivity when the field is rotated parallel to the CuO$_2$ planes is observed. This can be explained by taking into account the temperature dependence of the gap, which vanishes at $T_c$, and the relative increase of the inelastic scattering rate.

IV. COMPARISON WITH THE TWO-DIMENSIONAL THERMAL TRANSPORT THEORY

To compare the experimental results with theory we take recently published results of the longitudinal and transverse thermal conductivity into account that indicate that at high fields ($B > 2$ T), applied parallel to the planes, the main scattering mechanism for the quasiparticles appears to be the Andreev reflection by vortex supercurrents. Therefore, we use the formulation proposed by Yu et al. using a two dimensional model of the BRT expression for the thermal conductivity, which has been useful in the interpretation and discussion of previous results. Under this model the longitudinal thermal conductivity can be written as

$$\kappa_{xx}(\theta) = \frac{1}{2\pi^2 c k_B T^2 h^2} \int_{p_F}^{\infty} d^2 p \frac{v_{gx} v_{gz} E_p^2}{\Gamma(B, p, T)} \text{sech}^2 \left( \frac{E_p}{2k_B T} \right),$$

where $v_{gx}$ is the x-axis component of the group velocity, and $E_p$ is the quasiparticle energy. For this energy we use a free quasiparticle model $E_p^2 = (p^2/2m_{eff} - \mu)^2 + \Delta^2(p, T)$ where $m_{eff}$ is the effective mass of the quasiparticle. (B, p, T) may be taken as a relaxation rate given by the sum of the following scattering mechanisms acting in series: scattering of QP by impurities $\Gamma_{imp}(p)$, by phonons $\Gamma_{ph}(B, p, T)$, by quasiparticles $\Gamma_{qp}(B, p, T)$, and AS by vortex supercurrents $\Gamma_v(B, p, T)$. The expression for this scattering rate according to the model for the AS, proposed by Yu et al., is given by

$$\Gamma_v(B, p, T) = \Gamma_0 \exp \left\{ \frac{-m_{eff}^2 a_v^2 [E_p - |\Delta(p, T)|]^2}{p_F^2 h^2 \ln(a_v/a_0) \sin^2 \psi(p)} \right\},$$

where $a_v$ is the intervortex spacing given in this model by

$$a_v^2 = \frac{\nu_F^2 h^2}{\pi \Delta_0^2 \gamma m_{eff}^2} \frac{B_{ab}}{B}.$$  

We use the BCS-like parameterization for the temperature dependence of the gap amplitude

$$\Delta(T) = \Delta_0 \tanh \left( 2.2 \sqrt{\frac{T_c}{T} - 1} \right),$$

and the $d_{x^2-y^2}$-pairing symmetry in the resulting gap $\Delta(p, T)$ enters as follows

$$\Delta(p, T) = \Delta(T) \left[ \frac{\cos(p_x a/h) - \cos(p_y a/h)}{1 - \cos(p_F a/h)} \right].$$

We showed recently that at low temperatures and high fields, a quantitative agreement between this model and the oscillation amplitudes of the thermal conductivity tensor is achieved only if the intervortex spacing is
increased by about five times the value defined in Eq. (3) if we use the parameters from Refs. 13, 18, 19, 28, 29. Ginzburg-Landau parameter $\kappa = 100$, anisotropy $\gamma = 4$, $B_\text{c2}^0 = 650$ T. Furthermore, from those fits, for the twinned crystal we obtained a momentum independent impurity scattering $\Gamma_\text{imp} \simeq 0.12$ ps and $(\Gamma_0^{-1})_B (B = B_\text{c2}) \simeq (3/2) \Gamma_\text{imp}^{-1}$. Assuming a total scattering rate given by the sum of the AS and impurity rates, i.e. $\Gamma(B, p, T) = \Gamma_v(B, p, T) + \Gamma_\text{imp}$, the model given by Eqs. (1) to (5) reproduces remarkably well the symmetry of the curves in Fig. 5 in the whole measured temperature range. Because in this calculation we do not take explicitly into account the inelastic scattering rate $\Gamma_\text{in}$, given by phonons $\Gamma_\text{ph}$ and by quasiparticles $\Gamma_\text{qp}$, the calculated oscillation amplitude above 20 K increases up to $\simeq 10$ times that observed in the experiment.

In Fig. 6 we show the results of the numerical simulation. Each of the curves in this figure has been multiplied by a factor $f(T)$ in order to fit the experimental oscillation amplitude (Fig. 5). We note that the symmetry of the results can be explained quite satisfactorily by this model. This result, on one hand, supports the predominance of $d_{x^2−y^2}$-pairing symmetry of the order parameter up to temperatures close to $T_c$ and, on the other hand, confirms the idea of a competition between the thermal activation of quasiparticles in the direction of the thermal current and the gap structure. We note that the $d_{x^2−y^2}$-gap symmetry is the only gap function that can explain the whole angular patterns up to $T_c$. Of course, a $s$-gap also gives a onefold oscillation similar to the experiments above $\sim 50$ K, but it does produce neither the fourfold oscillation below $\sim 15$ K nor the curves between 15 K and 50 K. Thus, the set of curves in Fig. 6 can be only explained if one uses a main $d_{x^2−y^2}$-gap into the calculations. Furthermore, the anisotropy of $d$-wave gap proposed for BSCCO does not seem to occur in YBCO since in the latter the nodes are observed at both field orientations $\theta = 45^\circ$ and $\theta = -45^\circ$ (see also Refs. 13, 18, 19, 28, 29).

The factor $f(T)$ is related to the inelastic scattering rate $\Gamma_\text{in}$, which was not taken explicitly into account in the previous calculations. However, an approximation can be carried out to get roughly the temperature dependence of the total scattering rate, which could be in part associated to the temperature dependence of $\Gamma_\text{in}$. Since the AS mechanism is weakly temperature dependent below $T/T_c < 0.8$, we may approximate the total scattering rate as

$$\Gamma_\text{imp} + \Gamma_\text{v} + \Gamma_\text{in} \sim (\Gamma_\text{imp} + \Gamma_\text{v}) f^{-1}(T).$$  \hspace{1cm} (6)

The temperature dependence of the inelastic scattering rate is given then by the factor $f(T)$ in this approximation as

$$\Gamma_\text{in} \propto f^{-1}(T) - 1.$$  \hspace{1cm} (7)

In Fig. 7 we plot $f^{-1}(T) - 1$. As expected, we recover the overall behavior of the inelastic scattering rate already described by many authors. 13, 18, 19, 28, 29. Below $T_c$.
it decreases rapidly and becomes negligible in comparison with the impurity and AS scattering rates below $\sim 25$ K. Although the approximation given by (6) is too rough to obtain the true temperature dependence of the inelastic scattering rate, it is instructive to compare the obtained power dependence with results from literature. Thermal Hall angle measurements performed in the same YBCO twinned crystal show that $\cot(\theta_H) = n_H/\tau_H \propto T^4$ down to $\sim 20$ K ($n_H$ is the effective mass and $\tau_H$ is the Hall scattering time of the quasiparticles responsible for the Hall signal)\textsuperscript{19} The power law dependence obtained for $\tau_H^{-1} \propto T^4$ is similar to that obtained for the longitudinal scattering rate assuming a $d$–wave pairing.\textsuperscript{20} Since quasiparticle-quasiparticle scattering mechanism should be the dominant temperature dependent inelastic scattering below $T_c$, we expect a rate proportional to the density of quasiparticles. Interestingly, within the simple two-fluid model we expect a density of quasiparticles proportional to $(T/T_c)^4$. On the other hand, a $T^3$ dependence is expected within the spin-fluctuation scattering picture.\textsuperscript{29}

V. SUMMARY

In summary, we have measured the longitudinal thermal conductivity $\kappa_{xx}$ in two single crystals of YBCO in the presence of a planar magnetic field which was rotated parallel to the CuO$_2$ planes, from $T \sim 10$ K up to a few Kelvins below $T_c$. Fourfold oscillations were recorded below $\sim 20$ K. Above this temperature the angle dependence of the longitudinal thermal conductivity changes; from the minimum at $\theta = 0^\circ$ (field parallel to the heat current) a maximum develops at high $T$. The observed behavior in the whole temperature range can be very well reproduced by a model involving Andreev scattering of quasiparticles by vortices and the two-dimensional BRT expression for the thermal conductivity assuming a $d$–wave pairing. The overall results agree with the $d_{x^2-y^2}$-pairing symmetry of the order parameter. This agreement suggests that the mechanisms that influence the behavior of the quasiparticles below $T_c$ and above $\sim 10$ K are well described by the Fermi liquid theory at nearly optimal doping. The small differences found in the angle patterns between the twinned and untwinned samples can be explained in terms of the different impurity concentration as well as oxygen deficiency and are accounted for by the phenomenology exposed in this work.

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