Thermal conductivity of layered organic superconductor β-$(BDA\text{-}TTP)_2SbF_6$ in a parallel magnetic field: Anomalous effect of coreless vortices

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(Received 9 June 2004; revised manuscript received 27 September 2004; published 31 January 2005)

Thermal conductivity κ of the organic superconductor β-$(BDA\text{-}TTP)_2SbF_6$ was studied down to 0.3 K in magnetic fields H of varying orientation with respect to the superconducting plane. Anomalous plateau shape of the field dependence, κ vs H, is found for orientation of magnetic fields precisely parallel to the plane, in contrast to usual behavior observed in the perpendicular fields. We show that the lack of magnetic-field effect on the heat conduction results from coreless structure of vortices, causing both negligible scattering of phonons and constant in field electronic conduction up to the fields close to the upper critical field H_{c2}. Usual behavior is recovered on approaching H_{c2} and on slight field inclination from parallel direction, when normal cores are restored. This behavior points to the lack of bulk quasiparticle excitations induced by magnetic field, consistent with the conventional superconducting state.

DOI: 10.1103/PhysRevB.71.024531 PACS number(s): 74.25.Fy, 74.70.Kn

I. INTRODUCTION

In a commonly accepted picture of heat conduction in clean conventional superconductors, thermal conductivity κ at low temperatures $(T \ll T_c)$ is dominated by phonons.[1–3] The phonon contribution κ^g decreases rapidly on application of a magnetic field H, exceeding the lower critical field H_{c1}, so that thermal resistivity, $W=1/\kappa^g$, increases linearly with H due to scattering of phonons on the lattice of Abrikosov vortices.[4] The electronic part of thermal conductivity κ^e is determined by the intervortex quasiparticle tunneling, so it is negligibly small at the fields of the order of H_{c1} and shows an activated increase on H approaching the upper critical field H_{c2}. Since experimentally measured conductivity includes contributions of both electrons and phonons, $\kappa=\kappa^e+\kappa^g$, it shows a field dependence with a minimum at a field H_{min}, $H_{c1}<H_{min}<H_{c2}$, as shown schematically in Fig. 1. The value of H_{min} and the actual shape of the $\kappa(H)$ curve are both determined by the ratio of κ^e and κ^g.

In unconventional superconductors, variation of the thermal conductivity with magnetic field is more complicated.[5,6] Since quasiparticles in unconventional superconductors spread from the vortex cores into the bulk[7] and are delocalized, the electronic contribution is generally more important here.[8] In the $T=0$ limit, κ^e rapidly increases with field,[9–13] while at higher T the thermal conductivity remains constant in field, entering the so-called plateau regime.[14] This regime was experimentally identified in the cuprates in the field range $H \ll H_{c2}$,[9,14] and is predicted to extend close to H_{c2}.[15] The plateau was explained as caused by the electronic contribution, when an increase of the quasiparticle (QP) density is compensated by the decrease of their mean-free-path[15,16] (this implicitly assumes negligible phonon conductivity as compared to electronic contribution, $\kappa^g \ll \kappa^e$). When the phonon contribution is not negligible, the phonon scattering in the mixed state correlates well with the density of quasiparticles in the bulk, as represented by the electronic specific heat.[17]

Notable deviations from both of these pictures were found recently in layered superconductors in the case when the magnetic field was aligned precisely parallel to the plane.[18,19] In organic superconductors λ-$(BETS)_2GaCl_4$,[18] and κ-$(BEDT\text{-}TTF)_2Cu(NCS)_2$,[19] the thermal conductivity at the lowest temperatures ($\sim$0.3 K) remained almost independent of magnetic field, in the former case up to the fields of the order of H_{c2}, in stark contrast to the usual behavior observed in the perpendicular field. Since configuration of thermal conductivity measurements with magnetic fields parallel to the plane is used for determination of the symmetry of the superconducting gap,[20–24] understanding of this unusual field dependence is of notable importance.

It can be envisaged that the difference between the behavior in parallel and perpendicular fields may come from the difference in the structure of vortices in two-dimensional (2D) superconductors. Penetration of a magnetic field into a

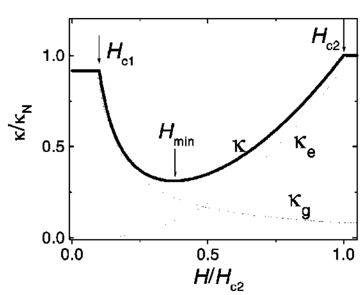

FIG. 1. Schematic presentation of the field dependence of thermal conductivity in isotropic conventional superconductors and in layered superconductors in magnetic field perpendicular to the superconducting plane.

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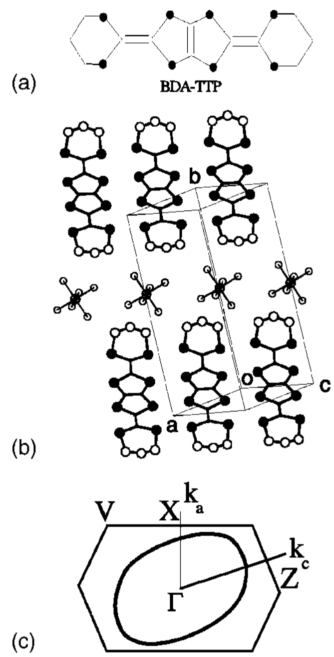

FIG. 2. Molecule of BDA-TTP (a). Solid dots indicate positions of sulfur atoms. In the crystal lattice of β-(BDA-TTP)$_2$SbF$_6$ (b), layers of donor molecules in the crystallographic *ac* plane alternate along *b* axis with layers of SbF$_6$ anions, leading to two-dimensional electronic structure with simple cylindrical Fermi surface, shown in panel (c) (Ref. 28).

superconductor induces a flow of a supercurrent, tending to diverge at the flux line. At a distance $\sim\xi/2$ (where ξ is coherence length) from the center, the supercurrent exceeds a critical value and the normal core is formed (Abrikosov vortex). In layered superconductors this scheme works when field is perpendicular to the plane. However, when field is parallel to the layer, the flow of supercurrent is restricted by the interplane tunneling, so that in case of weak coupling it never exceeds the critical value, resulting in no normal core (Josephson vortex) (see Ref. 25 for a review). For inclined fields, the two vortex systems form crossed vortex lattices.[26] Since the properties of the vortices in parallel field are notably different from those in bulk superconductors studied until recently, it is of interest to follow systematically the effect of the vortex structure on heat conduction.

Most efficiently this study can be done on materials of intermediate anisotropy. Here, the amplitude of the order parameter inside the core, despite being suppressed as compared to the bulk, remains finite.[27] In this case, with increase of temperature and field, the core of vortex can change to normal, and thus the effect of this transformation on thermal conductivity can be studied *in situ*.

In this paper we study the thermal conductivity of the organic superconductor β-(BDA-TTP)$_2$SbF$_6$, possessing relatively high transition temperature T_c of above 6 K.[28] As usual for two-dimensional molecular conductors,[29] the donor molecules of BDA-TTP [Fig. 2(a)] are packed in layers alternating with the layers of hexafluoroantimonate anions [Fig. 2(b)]. This structure brings about notable anisotropy of the electronic structure: simple extended Hückel model calculation predicts two-dimensional cylindrical Fermi surface, Fig. 2(c), occupying half of the first Brillouin zone. Experimental study of β-(BDA-TTP)$_2$SbF$_6$ indeed found the Fermi surface in reasonable correspondence with this prediction[30] and determined a rather large effective mass of 12.4 m_0. It also revealed sizable warping of the Fermi surface perpendicular to the plane, as indicated by observation of a rather broad coherence magnetoresistance peak, with the ratio of the Fermi energy in the plane ϵ_F to interplane transfer integral $t_\perp$ of 250–350 (Ref. 30) [approximately one order of magnitude smaller than the anisotropy of $\sim$3700 found in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$,[31] and comparable to anisotropy (1/280) of β-(BEDT-TTF)$_2$IBr$_2$ (Ref. 32)]. The superconducting state of the material was described as anisotropic three dimensional (3D).[33,34] It is essential for our study that the upper critical field for orientation parallel to the plane, $H_{c2\parallel}$, is about 12 T,[33] so that the whole *H*-*T* domain of superconductivity can be covered with measurements in a superconducting solenoid.

As a result of this study we found quite unusual field dependence of thermal conductivity in a parallel field, showing the plateau regime up to the fields as high as 0.6 H_{c2} and an anomalous minimum close to H_{c2}. This was in stark contrast to the usual dependence, observed on the same crystals in identical conditions in the perpendicular field. By studying transformation of the $\kappa(H)$ dependence with inclination of the field from parallel orientation and on variation of temperature, we show that the anomalous dependence can be understood in the usual scheme for conventional superconductors, adjusted for vortices without normal core. The lack of quasiparticles in the core brings simultaneously two effects, namely, negligible scattering of phonons on vortices, and negligible electronic contribution, both of these features being at odds with the expectations for unconventional superconductors.[35] Formation of the normal cores close to H_{c2} and on field inclination restores both finite phonon scattering and electronic conductivity.

II. EXPERIMENT

Single crystals of β-(BDA-TTP)$_2$SbF$_6$ were grown by a standard electrochemical technique, as described in detail elsewhere.[28] Contrary to the crystals of most of organic compounds, the samples of β-(BDA-TTP)$_2$SbF$_6$ are very bulky in nature, with typical dimensions in the range $2\times2\times0.5$ mm^3. For thermal conductivity measurements we selected crystals with a shape close to an elongated bar. A notable problem in measurements on β-(BDA-TTP)$_2$SbF$_6$ comes from the difficulty of making low resistance contacts. Even the best contacts, made by gold evaporation on the side surface of fresh samples, gave resistance values at low temperatures of about 100 Ω, which is two to three orders of magnitude higher than usual contact resistance obtained with this technique on another conducting organic charge-transfer salts.[18,36] Therefore the thermal resistance of the contacts was determined mainly by phonon conductivity. This limited the temperature range of thermal conductivity measurements down to 0.3 K and *in situ* resistance measurements in the vacuum cell down to approximately 1.5 K, which was caused by strong sample overheating with measuring current.

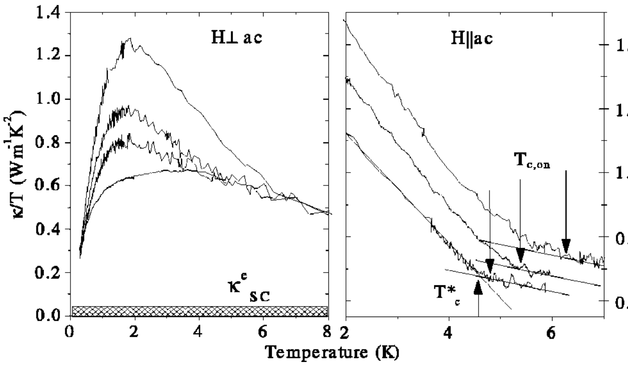

FIG. 3. (a) Temperature dependence of thermal conductivity in magnetic fields of 0, 0.5, 1, and 4.3 T (top to bottom) applied perpendicular to the conducting plane. The shaded area at the bottom shows estimated electronic contribution in the normal state. (b) Temperature dependence of thermal conductivity in parallel fields of 0, 4, and 6 T (top to bottom); the curves are offset to avoid overlapping. The determination of T_c was made in two ways: as a crossing point of linear extrapolations of the $\kappa(T)$ above and below T_c, we designate this as T_c^*, and from the point where $\kappa(T)$ starts to deviate from the curve in the normal state, $T_{c,on}$.

Measurements of thermal conductivity were made by a standard steady-state one-heater-two-thermometers technique, using RuO_2 thick-film resistors[37] as both heater and thermometers. The vacuum ambient was created by a miniature rotatable cell.[38] The cell was set into a goniometer allowing double-axis rotation in magnetic fields up to 17 T in a top-loading ^{3}He refrigerator. The alignment of the field direction parallel to the conducting plane was done by measuring an angular dependence of sample resistance in a magnetic field slightly below $H_{c2\parallel}$. The resistive minimum was sharp enough to ensure alignment with the accuracy of about 0.1°. More precise alignment was done by measuring $\kappa(H)$ at 0.3 K with an angular step of 0.02°, corresponding to the angular resolution of our system. Measurements were performed for three field orientations within the plane with respect to the heat current Q (with H and Q perpendicular, parallel and at 45° to each other, respectively) with the results being the same within experimental scatter. In the following we discuss all the data for $H \parallel Q$ configuration.

III. RESULTS

A. Temperature dependence

In Fig. 3(a) we show the temperature dependence of thermal conductivity, taken in zero field and under magnetic fields of 0.5, 1, and 4.3 T (normal state) applied perpendicular to the conducting plane. Above ~6.5 K all curves overlap within experimental scatter. On cooling, the curves at 0, 0.5, and 1 T deviate upward from the normal-state curve, with the point of deviation corresponding to the superconducting T_c. In zero field, κ/T increases more than two times on cooling from T_c (~6.5 K) down to 2 K, below which κ/T decreases steeply. This behavior is very similar to that observed in another organic superconductors, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$,[19,39] κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br,[40] λ-(BETS)$_2$GaCl$_4$,[18] in perpendicular fields.

In the simplest form, the thermal conductivity can be represented as $\kappa^i = \frac{1}{3} v^i C_V^i l^i$, where superscript i represents different groups of heat carriers (electrons and phonons), v is Fermi velocity for electrons and sound velocity for phonons, l is mean free pass and C_V contribution to the specific heat of respective carriers. The superconducting condensation reduces the density of electronic carriers participating in the heat transport (the condensate itself does not carry heat), while the density of phonons remains unaltered. Therefore the increase of κ/T below T_c originates only from an increase of the mean free path of heat carriers (both electrons and phonons), which is caused by the suppression of scattering on conduction electrons. The density of phonons changes smoothly through T_c and since the phonon-electron scattering is the dominant scattering mechanism in the normal state for most of the metals, κ^g usually increases below T_c.[1] The increase of κ^e in the superconducting state is more subtle. The condensation decreases the density of electronic heat carriers, so the increase of κ^e/T below T_c can be observed only if the rate of the mean free path increase is higher than the rate of the density of quasiparticles (QP) decrease. This requires domination of an inelastic scattering at T_c, a condition met only in very clean samples with relatively high T_c. The electronic maximum was first well-documented in the cuprates,[8] and was recently observed in some other clean superconductors with strong inelastic scattering: $CeCoIn_5$,[41] and the purest samples of Sr_2RuO_4.[42]

Since in organic superconductors the inelastic electron-electron scattering is usually strong, as seen in the T^2 temperature dependence of the resistivity immediately above T_c,[34,43] we cannot disregard *a priori* the possibility of the electronic nature of the κ/T increase below T_c. However, in β-(BDA-TTP)$_2$SbF$_6$, similar to other organic compounds, the phonon contribution κ^g is strongly dominating measured κ at all temperatures covered by our experiment. This is caused by a combination of low κ^e, due to low density of electronic carriers in these materials, and a rich phonon spectrum leaving the density of phonons high even at low temperatures, ensuring high κ^g. In β-(BDA-TTP)$_2$SbF$_6$ this general trend can be verified from both the estimation of κ^e through the resistivity value in the normal state via the Wiedemann-Franz law[44] and from the magnitude of $\kappa(H)$ increase near H_{c2} at low temperatures [shown with dashed area in Fig. 3(a)], giving κ^e of about 16% of the normal state κ at 0.3 K. In the superconducting state κ^g is always higher than in the normal state, so we can conclude that here the thermal conductivity is arising mostly from phonons.

On application of magnetic field perpendicular to the plane, the rate of initial κ/T increase below T_c is rapidly diminished, resulting in suppression of the maximum. This clearly shows that perpendicular field rapidly restores phonon scattering on conduction electrons. On the contrary, the rate of κ/T increase is influenced only slightly by parallel magnetic field, Fig. 3(b).

As it can be seen from Fig. 3, the change of κ at T_c is not sharp, resulting in a notable difficulty in determination of a position of the normal-superconductor boundary from temperature dependence of thermal conductivity at high temperatures. The most justified approach, using the onset point

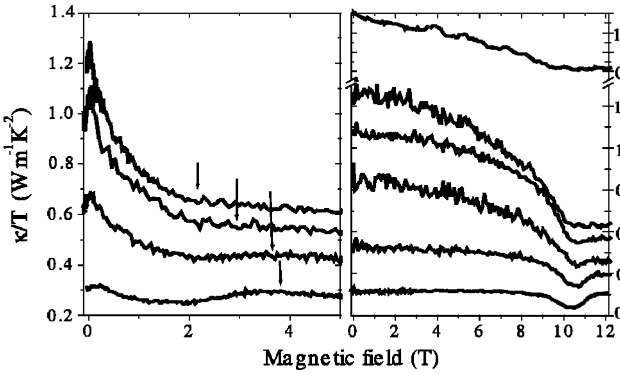

FIG. 4. Field dependence of thermal conductivity. (a) Field applied perpendicular to the conducting *ac* plane, from top to bottom, T=1.55, 1.05, 0.64, and 0.29 K. Arrows show $H_{c2\kappa}$ for these curves. (b) Field parallel to the conducting plane, from top to bottom, T=3.11, 1.55, 1.06, 0.83, 0.45, and 0.29 K. The curve for 3.11 K is shown out of scale to avoid overlapping.

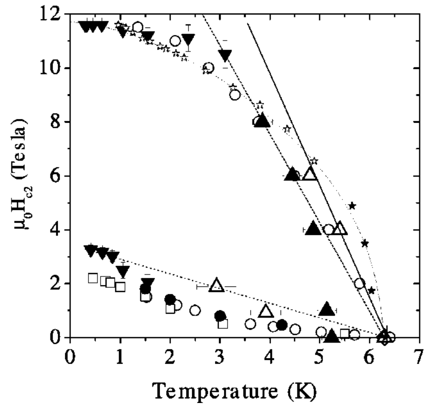

FIG. 5. H-T phase diagram of the superconducting state of β-(BDA-TTP)$_2$SbF$_6$. Filled down-triangles are from $\kappa(H)$ sweeps; filled and open up-triangles are T_c^* and $T_{c,on}$ from $\kappa(T)$ sweeps. The data from resistivity (open circles) and specific-heat (open diamonds) measurements by Shimojo *et al.*, Ref. 33, and from resistivity (filled circles) and tunnel diode oscillator (open squares) measurements by Choi *et al.*, Ref. 30, are shown for comparison. The dotted line is a guide for eyes for thermal conductivity data in the perpendicular field. The solid line shows evaluation of the slope of $H_{c2\parallel}(T)$ from $T_{c,on}$ in thermal conductivity measurements, the dashes show a conservative estimation of $H_{c2}(T)$ slope from specific heat (zero field) and thermal conductivity T_c^* data. For comparison we show with the dash-dotted line the theoretical curve for pure Pauli limiting without orbital contribution (Ref. 47) and experimental data for κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br from specific-heat measurements (filled stars) by Kovalev *et al.* (Ref. 48) and pulsed field resistivity measurements (open stars) by Shimojo *et al.* (Ref. 49). Both the theoretical curve and the data were normalized to match T_c and $H_{c2}(0)$ of β-(BDA-TTP)$_2$SbF$_6$.

of deviation from the normal-state curve, is not very precise due to slow takeoff and suffers badly from the experimental noise. Another way to follow variation of T_c is to use a crossing point of lines representing linear approximations of $\kappa(T)$ in the normal state and in the region of increase below T_c, as shown in Fig. 3(b) for H parallel to the conducting plane. We designate this value as T_c^* in the following. It is clear that this procedure gives a value systematically below real bulk T_c, determined from both the onset of deviation in κ/T and specific-heat measurements[33] (see Fig. 5 below).

B. Field dependence

In Fig. 4 we show evolution with temperature of the field dependence of thermal conductivity in perpendicular (a) and parallel (b) field orientations. In a perpendicular field, κ decreases rapidly above H_{c1}.[45] The derivative of the $\kappa(H_\perp)$ has maximum magnitude at H_{c1} and decreases monotonically towards H_{c2} [arrows in Fig. 4(a)] until reaching almost constant value in the normal state. Small variation of the background in the normal state is caused by the electronic contribution, namely by its change due to orbital magnetoresistance.[34,46] On cooling, total variation of κ/T in the range from H_{c1} to H_{min} rapidly decreases, reflecting the decrease of the phonon conductivity. On the contrary, variation of the electronic contribution with field becomes larger on cooling, so at 0.29 K the curve shows a clear double maximum structure, with maxima at H=0 and H_{c2}, in line with schematic presentation of Fig. 1.

In the parallel field, the shape of the field dependence changes notably on cooling. At temperatures close to T_c, $\kappa(H_\parallel)$ has a usual shape with the slope of the curve gradually decreasing towards $H_{c2\parallel}$, though at 3.11 K, $\sim 0.5T_c$, this dependence already does not resemble sharp "$1/H$ behavior" observed in the perpendicular field. On cooling, the slope of the curve at H=0 gradually diminishes. At high temperatures, the slope increases with field until the very vicinity of $H_{c2\parallel}$, where it starts to decrease rapidly towards zero value in the normal state. At low temperatures the $\kappa(H_\parallel)$ shows a broad plateau up to about 8 T, then shows a rapid decrease until H_{min}, followed by a rapid increase near $H_{c2\parallel}$. As can be seen from Fig. 4(b), the width of the plateau rapidly decreases on warming, until it transforms into a region of slow κ decrease above 1.5 K.

IV. DISCUSSION

A. *H*-*T* phase diagram

In Fig. 5, we plot the H-T phase diagram, determined from temperature and field dependence of κ/T in parallel and perpendicular fields. For comparison we show the values of H_{c2} determined from resistivity measurements by Shimojo *et al.*[33] and by Choi *et al.*[30] We note that the H_{c2} determined from $\kappa(T)$ and $\kappa(H)$ are in good correspondence. In the following we designate H_{c2} determined from thermal conductivity measurements as $H_{c2\kappa}$, to distinguish it with the resistively determined $H_{c2\rho}$. Comparison of these two determinations shows a systematic trend in the behavior. In the perpendicular field, $H_{c2\kappa}$ is always higher than $H_{c2\rho}$, which is determined by vortex lattice melting.[31] The $H_{c2\kappa}(T)$ is almost linear, while $H_{c2\rho}$ shows a clear upturn on going to T=0, with a tendency of both curves to a close $H_{c2}(0)$ in the T=0 limit. A similar trend was shown previously in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$.[50]

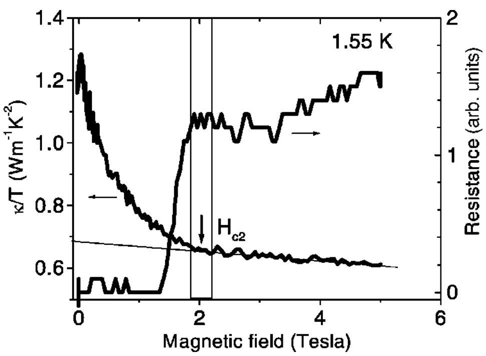

FIG. 6. Field dependence of resistance and of thermal conductivity in a magnetic field perpendicular to the conducting plane, measured *in situ* at 1.55 K.

To verify that the difference between $H_{c2\kappa}$ and $H_{c2\rho}$ is not caused by the difference in the experimental conditions between two sets of measurements, in Fig. 6 we show the two curves measured *in situ* at 1.5 K. Despite notable problems in the quality of the resistive data, it is clear that $\kappa(H)$ curve flattens above the midpoint of resistive transition, most closely to the complete restoration of the normal-state resistivity. This comparison clearly shows that thermal conductivity is not sensitive to the flux flow in the mixed state, which actually determines the position of the $H_{c2\rho}(T)$ line.

In parallel field, the values of $H_{c2\rho}$ and $H_{c2\kappa}$ are in much closer correspondence, except for the very vicinity of T_c. This clear difference with the perpendicular field orientation is presumably caused by a much stronger intrinsic pinning of vortices in parallel configuration by the lattice.[48,51] $H_{c2\parallel\rho}(T)$ and $H_{c2\parallel\kappa}(T)$ show a clear saturation at low temperatures, implying Pauli paramagnetic limiting, as was pointed out by Shimojo *et al.*[33] Indeed the value of $H_{c2\parallel}(0)=11.6$ T coincides within experimental accuracy with the value expected for Pauli limiting in the case of a weak-coupling BCS superconducting gap, $H_{c2}(0)=1.84T_c$ (for $T_c=6.3$ K from specific-heat measurements this gives 11.6 T).

For our analysis of the field dependence of thermal conductivity, it is essential to know the degree of order parameter suppression inside the vortices in parallel field. This strongly depends on the ratio between the interplane distance d and the coherence length $\xi_\perp$.[25] Shimojo *et al.*[33] determined coherence lengths from the extrapolated values of $H_{c2\rho}(T)$ curves to $T=0$, getting the $\xi_\perp=2.6$ nm, which is about 50% larger than the interplane distance $d=1.76$ nm. Since the value in parallel field determined in this way can be affected by the paramagnetic Pauli limiting, $\xi_\perp$ may be overestimated.

There were several proposals to go around Pauli limiting problem in determining ξ for parallel field (see Ref. 31 for a review). The most standard approach is to determine $\xi_\perp$ from the slope of H_{c2} line in a temperature range close to T_c. This approach is valid, if orbital effect is determining the upper critical field, as is always the case for perpendicular orientation. Here we find in our experiment the slope of bulk $H_{c2\kappa}(T)$ of 0.55 T/K. For parallel orientation this is true only if the anisotropy of a superconductor is finite, otherwise $H_{c2\parallel}$ may be determined by paramagnetic Pauli limiting all the way up to T_c in zero field. To verify if the upper critical field close to T_c is Pauli limited, we compare in Fig. 5 the shape of the experimentally determined $H_{c2\parallel}(T)$ line in β-(BDA-TTP)$_2$SbF$_6$ with that expected for pure paramagnetic limiting[47,52] and with the one observed experimentally in highly anisotropic κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br.[48,49] In the case of Pauli limiting, the upper critical field is determined from the solution of the equation $\ln[T_c(H)/T_c(H=0)]=\mathrm{Re}\left[\psi\left(\frac{1}{2}+ig\mu_B H/4\pi k_B T_c(H)\right)-\psi\left(\frac{1}{2}\right)\right]$; here μ_B is the Bohr magneton, g is the electron g factor, and ψ is the digamma function. Close to T_c in zero field this dependence can be well approximated by $\Delta T_c \sim H^2$, which is indeed very close to observation in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br,[48] but is strongly overshooting the H_{c2} data in β-(BDA-TTP)$_2$SbF$_6$ from onset of thermal conductivity rise. Combining the data from thermal conductivity onset and specific-heat measurements, we can conclude that the slope of bulk $H_{c2\parallel}(T)$ stays in the range between 3.3 and 4.2 T/K (dashed and solid lines in Fig. 5, respectively). For comparison, the same slope in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, which is Pauli limited close to T_c, was estimated as ~ 14 T/K.[48] Thus, close to T_c, the $H_{c2\parallel}$ in β-(BDA-TTP)$_2$SbF$_6$ is determined by the orbital effect and we can use Ginzburg-Landau (GL) theory approach for the determination of the coherence length.

For anisotropic superconductors the values of the coherence length can be found from the formulas

$$\mu_0 T_c \frac{dH_{c2\parallel}(T)}{dT} = \frac{\Phi_0}{2\pi\xi_\perp(0)\xi_\parallel(0)},$$

$$\mu_0 T_c \frac{dH_{c2\perp}(T)}{dT} = \frac{\Phi_0}{2\pi\xi_\parallel(0)^2},$$

where $\xi_\parallel(0)$ and $\xi_\perp(0)$ are the coherence lengths parallel and perpendicular to the layer, respectively, μ_0 is the magnetic constant, and $\Phi_0=2.07\times10^{-15}$ Wb is the magnetic-flux quantum. It is assumed that ξ is isotropic in the plane, which is in line with our observation of negligible variation of $\kappa(H)$ curves for different field directions within conducting plane. Substituting the values of the slopes we get $\xi_\parallel=9.7$ nm and $\xi_\perp=1.28$–1.63 nm. The value of $\xi_\perp$ is slightly lower than the interplane distance of 1.76 nm. These values of the coherence length show that the superconducting state of β-(BDA-TTP)$_2$SbF$_6$ is actually on the border between truly two-dimensional and anisotropic three-dimensional.

B. Field dependence of thermal conductivity

The dependence of thermal conductivity on a magnetic field parallel to the plane is the central result of this work. As we already noted, the field dependence of κ in perpendicular field is quite usual. In line with the scheme of Fig. 1, κ decreases rapidly with field due to suppression of κ^g, while at $H=H_{min}$ the decrease of κ^g becomes slower than the increase of κ^e, so that the measured κ increases towards $H_{c2\perp}$. As it is clear from the comparison of the curves for parallel and perpendicular fields, the difference is caused mainly by complete disappearance of initial decrease of κ in parallel field, so that even at relatively high temperatures of about 1.5 K the initial slope of the $\kappa(H)$ curves is close to zero.

Here we should notice that the lack of variation of thermal conductivity with field can in no way be explained by a compensation of a decrease of κ^g by an increase of κ^e, so that $\kappa=\kappa^e+\kappa^g$ remains constant. It is clear from Fig. 4 that the first signs of flattening are seen at temperatures as high as 3 K, the slope of κ/T vs H curves at $H=0$ becomes negligibly small at 1.5 K and the curves at 0.45 and 0.29 K become completely flat, despite very large variation of κ/T at $H=0$. Since $\kappa^e\sim T$ and $\kappa^g\sim T^{\alpha}$ (α=2–3 depending on dominant scattering mechanism, Ref. 3) vary in a very different way with T, their exact compensation can never be achieved for almost fivefold variation of T. Therefore, to have constant κ, neither κ^e nor κ^g should vary with field at low T and H.

A negligible variation in κ^g is natural to relate with the lack of scattering on vortices. Indeed, linear increase of the phonon thermal resistivity $W^g=1/\kappa^g$ in conventional superconductors[4] is frequently viewed as gradual restoration of phonon-electron scattering to a value typical of the normal state. In this case the phonon thermal resistivity W should be proportional to the density of scatterers, i.e., to the electronic specific heat $C^e\sim H$. Recent studies of the phonon thermal resistivity of Sr_2RuO_4 as a function of direction of the field with respect to the conducting plane have shown that the cross section of phonon scattering on vortices is proportional to the coherence length.[17] Simultaneously it was shown for this unconventional superconductor that the relation between phonon resistivity and the electronic specific heat still holds due to phonon scattering on quasiparticles in the bulk. Following this line, zero phonon scattering in our experiment can be considered as a limiting case of this behavior, typical for superconductors without nodal quasiparticles. It indicates that the electronic quasiparticles inside the core are the actual cause of phonon scattering in the superconducting state, completely similar to the conduction electrons in the normal state.

An understanding of the lack of increase of κ^e is not as straightforward. At low temperatures, the field dependence of κ^e is determined by several factors. First of all, it is different in clean ($\xi\ll l$) and dirty ($\xi\gg l$) limits. For β-(BDA-TTP)$_2$SbF$_6$, $\xi_\perp$ is of the order of lattice constant, while observation of quantum oscillations in the material[30] implies a rather long mean free path. Therefore it is natural to use the clean limit. This approach finds justification in our experiment as well: in the dirty limit κ^e is expected to increase linearly with field,[3] which is not seen in our experiment even in the perpendicular orientation. In the clean limit, we have to make a clear distinction between conventional *s*-wave superconductors with isotropic gap and unconventional superconductors with line nodes. In conventional superconductors activated behavior of thermal conductivity is natural in low fields, since quasiparticles are localized in the cores. In unconventional superconductors, application of magnetic field should increase the density of the delocalized quasiparticles. In perpendicular field this is caused by the Volovik effect, while in parallel field by Zeeman splitting of the Fermi surface.[35] As a result we should expect an increase of the electronic thermal conductivity and of the phonon scattering, both of which are opposite to our observation.

According to a Ginzburg-Landau theory of second-order phase transitions, the coherence length should become infinite at the normal-superconductor boundary (at T_c and at H_{c2}). This situation corresponds to a rapid suppression of the order parameter inside the vortex on approaching H_{c2} at low temperatures and restoration of the normal core. A consequence of this crossover can be easily recognized in our experiment as a restoration of phonon scattering at high $T\sim T_c$ and $H\sim H_{c2}$. Indeed, this crossover reveals itself at low T as a downturn of $\kappa(H)$ from the plateau value (Fig. 4), followed by a rapid takeover of the total variation of κ by the increase of κ^e close to $H_{c2\parallel}$. Of note that the values of κ at H_{min} are close for parallel and perpendicular orientations of the field, which indicates complete restoration of phonon scattering, while a small difference can be assigned to variation of κ^e at different values of H/H_{c2}. At 0.3 K, the restoration of phonon scattering happens only at the high fields, so that all variation of $\kappa(H)$ occurs in a field range spanning over just 30% of the $H_{c2\parallel}$.

Both the lack of phonon scattering at low fields and its restoration above the crossover show directly that coreless vortices do not affect phonon conductivity. This is in striking contrast to the electronic conductivity, in which case Josephson vortices introduce scattering of quasiparticles due to Andreev reflection on the supercurrent circulating around the vortex.[53]

Another way to track the effect of the restoration of the normal cores is to incline the magnetic field from parallel orientation. For small angles, when a perpendicular component of the field is less than $H_{c1\perp}$, magnetic flux penetrates into the sample only along the planes and does not follow the direction of the external field. With further increase of the inclination, the perpendicular component of the field exceeds $H_{c1\perp}$. However, though the flux enters the sample, it is still energetically unfavorable to create inclined vortex lines,[26] so two mutually perpendicular systems of vortices are formed, one with the normal core (for H component perpendicular to the plane) and one without normal core (parallel to the plane). Formation of normal cores rapidly restores phonon scattering, Fig. 7(a), giving exceptionally sharp angular dependence, Fig. 7(b).

From the comparison of $\kappa(H)$ curves for small inclination angles (Fig. 7) it is clear that the increase of κ^e above H_{min} on approaching H_{c2} remains practically unchanged, which is in striking contrast to strongly angle-dependent decrease below H_{min}. This clearly shows that the rapid increase close to H_{c2} is not related to formation of crossed vortex lattices. Since the increase of κ with field near H_{c2} is purely electronic in origin, it is instructive to search for a mechanism giving a sharp rise of κ^e near H_{c2} in the parallel field, as opposed to a gradual change in the perpendicular field.

The most obvious possibility is involvement of Pauli limiting. The rapid increase of thermal conductivity close to H_{c2} is associated with filling of the whole volume of the superconductor with vortex cores. In the case of orbital limiting this happens gradually, while in the case of Pauli limiting vortex cores occupy only a fraction of the sample volume, and the transition between superconducting and normal states becomes discontinuous (first order). Rapid increase of thermal conductivity at H_{c2} below $\sim$1 K is indeed observed

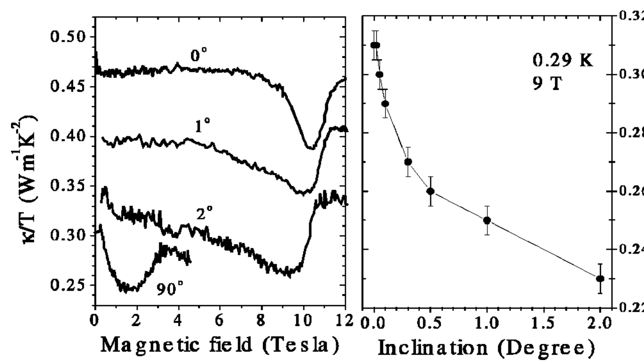

FIG. 7. (a) Field dependence of thermal conductivity at 0.29 K for various inclination angles to the conducting plane. From top to bottom: parallel field, 0°, 1°, 2°, and perpendicular field, 90°. The curves are offset up from the 90° curve to avoid overlapping. (b) Thermal conductivity at 0.29 K and 9 T as a function of the inclination angle from parallel to the plane orientation.

in a range where the $H_{c2\parallel}(T)$ line flattens (see Fig. 5), as naturally expected for Pauli limiting. The sharp increase of the electronic thermal conductivity at H_{c2} in the Pauli limiting range was observed in a number of materials.[11,18,54] It finds a natural explanation in an almost first-order character of transition at H_{c2}. The variation of Pauli limiting field on magnetic-field direction is determined by anisotropy of the g factor [usually in organic superconductors this is of the order of 1% (Ref. 29)], which is presumably the reason for the lack of measurable anisotropy of H_{c2} within the conducting plane. Inclination of field direction from the conducting plane should gradually switch the mechanism from Pauli to orbital, as discussed for a number of layered materials.[31,55] As can be seen in Fig. 7, for inclination angles up to 2° variation of H_{c2} is quite small, which implies that the orbital effect is still weak. Therefore the increase of κ close to H_{c2} remains sharp, and is unaltered by the transformation of the vortex lattice. This observation is in line with previous observations for several systems where the thermal conductivity and the magnetization were studied in the same conditions, including three-dimensional UPt_3,[11,56] and two-dimensional Sr_2RuO_4.[12,57,58]

V. CONCLUSION

Thermal conductivity of anisotropic superconductors reveals a notable difference in response to magnetic field, depending on field direction with respect to the superconducting plane. In the organic superconductor β-$(BDA\text{-}TTP)_2SbF_6$, we found anomalous independence of thermal conductivity on magnetic-field strength in parallel field, in stark contrast to the usual dependence, observed in identical conditions when the field was perpendicular to the plane. The anomalous behavior in the parallel field is observed over a notable portion of the superconducting domain, to the fields as high as $0.6H_{c2}$. Evolution of the plateau regime, with temperature and inclination of the field direction from the parallel to the plane, points to its close relation to the special coreless structure of vortices in the parallel field. The lack of quasiparticle states in the cores diminishes phonon scattering in the mixed state, and simultaneously suppresses electronic tunneling between cores. Formation of the normal cores close to H_{c2} and on field inclination restores the usual behavior with finite phonon scattering and electronic conductivity. The field dependence of thermal conductivity of this type can be expected neither for phonon, nor for electron contributions, if quasiparticles are spread in the bulk of a superconductor, as expected for unconventional superconductors.[35]

ACKNOWLEDGMENTS

The work has been done under support by the CREST program from Japan Science and Technology Corporation. The authors acknowledge valuable discussions with H. Shimahara, K. Machida, A. Vishwanath, A. E. Koshelev, and L. Taillefer.

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