Two band gap field-dependent thermal conductivity of $MgB_2$

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The thermal conductivity $\kappa(H,T)$ of the new superconductor $MgB_2$ was studied as a function of the temperature and a magnetic field. No anomaly in the thermal conductivity $\kappa(H,T)$ is observed around the superconducting transition in absence or presence of magnetic fields up to 14 Tesla; upon that field the superconductivity of $MgB_2$ persisted. The thermal conductivity in zero-field shows a $T$-linear increase up to 50K. The thermal conductivity is found to increase with increasing field at high fields. We interpret the findings as if there are two subsystems of quasiparticles with different field-dependent characters in a two ($L$ and $S$)-band superconductor reacting differently with the vortex structure. The unusual enhancement of $\kappa(H,T)$ at low temperature but higher than a ($H_{c2S} \simeq 3T$) critical field is interpreted as a result of the overlap of the low energy states outside the vortex cores in the $S$-band.

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I. INTRODUCTION

Superconductivity at a remarkably high transition temperature closed to 40K was recently discovered in $MgB_2$ [1,2]. $MgB_2$ has a Debye temperature about 800K, several times larger than that of $Nb_3Sn$, as seen from specific heat measurements [3-7]. The phonon density of states of $MgB_2$ has been obtained by inelastic neutron scattering [8-11]. These results indicate that phonons are playing an important role in the interaction of the new superconductor $MgB_2$. Most experiments in $MgB_2$, such as the isotope effect [12,13], $T_c$ pressure dependence [14,15] and tunneling spectroscopy [16,17] indicate that the superconductivity of $MgB_2$ can be consistent with a phonon-mediated BCS electron pairing. Beside [5-7,17] reports have also shown some evidence for two gaps in the quasiparticle excitation spectrum of $MgB_2$ [18-20].

The thermal conductivity has been used widely to study superconductors [21], offering important clues about the nature of heat carriers and scattering processes between them, especially in $H=0$, and in unconventional superconductors [22-26], the more so in the superconducting state for which traditional electrical probes such as the electrical resistivity, Hall effect, and thermopower are inoperative. The study of the thermal conductivity in applied magnetic fields has also been an interesting new probe of the vortex state of high-$T_c$ superconductors [27-37] and in $MgB_2$, in absence [38-40] or presence [41-42] of a magnetic field.

In this paper we present a new study of the thermal conductivity $\kappa(H,T)$ of polycrystalline $MgB_2$, as a function of temperature in both absence and presence of magnetic fields in order to probe the anomalous dependences [40,41] in light of new vortex structure considerations [7]. The temperature range goes between 4-50K and the applied magnetic field goes up to 13.9 Tesla. The superconductivity of $MgB_2$ persisted over the investigated condition though no sharp anomaly in the thermal conductivity $\kappa(H,T)$ was observed around the superconducting transition. The thermal conductivity almost monotonously increases with increasing temperature in zero and low magnetic fields whereas a significant enhancement of the values of the thermal conductivity is found for high magnetic fields. A model based on two different gaps and two subsystems of quasiparticles with different field-dependent characters in superconducting $MgB_2$ is argued to be consistent with these results.

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II. SAMPLE PREPARATION AND MEASUREMENTS

Polycrystalline \( MgB_2 \) samples were prepared by the conventional solid state reaction method. A mixed powder of high purity \( Mg \) and \( B \) with an appropriate ratio was ground and pressed into pellets. The pellets were wrapped in a tantalum foil and sealed in a stainless steel reactor, then heated at 950\(^\circ\)C for 4 hours under an argon gas flow. Repeating the process we got a high-density bulk sample. The phase purity was confirmed by X-ray diffraction analysis. No impurity phase was observed within the usual X-ray resolution. For the electrical and thermal conductivity measurements one 8 x 1.5 x 0.8 mm\(^3\) bar was cut from a bulk sample pellet.

In order to check the sample superconductivity we have measured the electrical resistivity by the standard four-probe d.c. technique in the 4 to 50 K temperature range and for magnetic fields up to 14 T. Fig. 1 shows the temperature dependence of the electrical resistivity, \( \rho(H, T) \), of polycrystalline \( MgB_2 \) for several magnetic fields under heating condition. The applied field was 0, 1, 4, 7, 10 and 14T in turn applied at low temperature after zero field cooling. A narrow superconductivity transition occurs at \( T_c = 38.1 \)K in zero-field. With increasing fields \( T_c \) shifts to lower temperature and the transition width increases as expected. It should be noted that the superconductivity of \( MgB_2 \) persisted over the investigated fields.

The thermal conductivity of the sample was measured by the longitudinal steady-state thermal flow method in the temperature range 4-50 K and for several magnetic field up to 13.9 T. The thermal flow and the magnetic field were parallel to the long axis of the sample. A thin-film chip resistor was used as an end heater, and thin-wire differential thermocouples for monitoring the thermal gradients on the sample. The measurements were performed under high vacuum with two additional shields mounted around the sample in order to reduce the heat losses due to radiation at finite temperature [43]. A carbon-doped glass resistance thermometer was used to measure the temperature of sample in a magnetic field. The sensitivity of the thermocouple as well as the resistance of the thin-film resistor were previously calibrated for magnetic field conditions. The whole experiment was computer controlled [44].

Fig. 2 shows the temperature dependence of the thermal conductivity of polycrystalline \( MgB_2 \), in applied 0.0, 0.02, 0.5, 1.0, 5.0, 10.0 and 13.9 Tesla field after zero-field cooling. Data were recorded while warming the sample up at a fixed magnetic field. The thermal conductivity is seen to increase monotonically with increasing temperature in the investigated ranges. The temperature dependence of \( \kappa(0, T) \) is linear (correlation coefficient \( R = 0.9995 \)). With increasing fields, \( \kappa(H, T) \) has an almost linear temperature dependence at low fields. A significant enhancement in \( \kappa(H, T) \) is obvious in the high-field region at low temperature, as in [41,42]. All \( \kappa(H, T) \) over \( T \) curve intersect each other at 19K above which temperature the curves of \( \kappa(H, T) \) for different fields merge on each other, except that for 13.9T.

III. DISCUSSION OF MEASUREMENTS

As shown in Fig.2, the absence of anomaly in the thermal conductivity in zero-field near the superconducting transition \( T_c \), is first in reasonable agreement with other results reported for polycrystalline [37-39] and single crystalline \( MgB_2 \) [41,42]. Such a smoothness is often attributed to charge carrier and phonon scattering by a distribution of localized impurities or extended defects. However, the \( MgB_2 \) thermal conductivity markedly differs in behavior at \( T_c \) from that of conventional superconductors and high temperature superconductors. The thermal conductivity of such superconducting materials generally shows an anomalous behavior at the superconducting transition temperature, either a decrease or an increase below \( T_c \) depending on the nature of the heat carrier and their interaction. In conventional superconductors \( \kappa(T) \) decreases below \( T_c \) due to the condensation of electrons. In the case of the high temperature superconductors \( \kappa(T) \) shows a peak below \( T_c \) that is explained by considering either an increase in the phonon mean free path due to carrier condensation or an increase in the quasi-particles relaxation time in the superconducting state or to both. Our results show no such a response in the thermal conductivity for \( MgB_2 \) around \( T_c \) in both absence (and presence of magnetic fields) within our experimental resolution. It means that \( MgB_2 \) thermal conduction is not much sensitive to any Cooper pairing below \( T_c \) nor to the breaking effect introduced by a field. However, the lattice contribution is substantial in both the superconducting and the normal state region. Indeed, in zero field, the thermal conductivity \( \kappa \) in a metal is generally considered to be the sum of electrons \( \kappa_e \) and phonon contributions \( \kappa_{ph} \), i.e. \( \kappa(T) = \kappa_e(T) + \kappa_{ph}(T) \).

The electronic contribution to \( \kappa(T) \) in the superconducting state has been calculated by Bardeen, Rickayzen, and Tewordt [21]. The phonon thermal conductivity is usually represented within the Debye-type relaxation rate approximation [45]

\[
\kappa_{ph} = \frac{k_B}{2\pi^2 v} \left( \frac{k_B}{\hbar} \right)^3 T^3 \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} \tau(\omega, T) \, dx,
\]
where $\omega$ is the frequency of a phonon, $\tau(\omega, T)$ is the corresponding relaxation time, $v = \Theta_D (k_B/\hbar) (6\pi^2 n)^{-1/3}$ is the average sound velocity, $n$ is the number density of atoms, and $x = \hbar \omega / k_BT$. For our analysis, we use $\Theta_D = 750$ K, as deduced from specific heat measurements [4,9]. The total phonon relaxation rate

$$\tau^{-1} = L/v + B\omega^4 + CT\omega^2 \exp(-\Theta_D/bT) + D\omega + E\omega g(x, y)$$  \hspace{1cm} (2)$$
can be represented as a sum of terms corresponding to independent scattering mechanisms. The individual terms, dominating in different temperature intervals, introduce phonon scattering by sample boundaries, point defects, phonons, dislocations, and electrons, respectively. The constants $L, B, C, b, D,$ and $E$ are a measure for the intensity of corresponding phonon relaxation processes. The function $g(x, y)$ is given in Ref.[21].

The $\kappa_c(T)$ magnitude can be estimated from the electrical resistivity and the Wiedemann-Franz (W-F) rule $\kappa_c\rho = LT$ where $L = L_0$, the Lorentz constant, if the scattering is elastic; $L < L_0$ if it is inelastic. Since the value of $\kappa_c$ is an upper limit when obtained by using the W-F rule and $L_0$, we estimate that the value of $\kappa_c$ is about 20% of the total value of the thermal conductivity in the normal state, $\kappa_n$ e.g. at $T=50K$ for our sample. The best fit value for $L$ being significantly smaller than the expected Lorentz constant $L_0$ calls attention to the influence of inelastic scattering, and to a large phonon (80%) contribution.

Next, consider the field dependence of the thermal conductivity $\kappa(H)/\kappa(H = 0)$ at several temperatures (Fig.3). In low fields, the thermal conductivity seems insensitive to the magnitude of the applied magnetic fields whereas it becomes variably enhanced at higher fields. Notably, the value $\kappa(H)$ is higher than that of $\kappa(H = 0)$ in the field region larger than 0.5 T at low temperature (below 19K). The non-linear field dependence and the field-dependence enhancement in $\kappa(H)$ imply that the effect of vortex structure, induced by the field, over heat carrier in the mixed state of $MgB_2$ is no longer a beaten track. According to standard thinking for conventional superconductors the introduction of a magnetic field would lead to a decrease in heat conductivity because vortices constitute new scattering centers for heat carriers. This picture provided a qualitative explanation of the field-induced decrease in $\kappa(H)$ observed in the vortex states of conventional superconductors and high $T_c$ cuprates in the investigated regions of the field-temperature plane [27-37].

To interpret quantitatively the field dependence of the thermal conductivity in superconductors one again assumes that phonons and electrons contribute independently as thermal carriers, such that

$$\kappa(B, T) = \kappa_e(B, T) + \kappa_{ph}(B, T),$$  \hspace{1cm} (3)$$
where $\kappa_e(B, T)$ and $\kappa_{ph}(B, T)$ are the electronic and phononic contribution to $\kappa(B, T)$, respectively.

Again the mean free path of both, electrons and phonons, can change below $T_c$, because the scattering of phonons on electrons is reduced when electrons condense into the superconducting state. Moreover the electron-electron interaction can change when the superconducting gap opens below $T_c$ and modify the mobility because of the condensate presence. Therefore, it is not immediately clear a priori which type of thermal carriers and which interactions are responsible for the behavior of the thermal conductivity in various temperature and field ranges.

More recently experiments have shown unequivocally that the two gaps structure is an intrinsic Fermi property of $MgB_2$. According to band structure calculations and tunneling experiments, there are two distinctive Fermi surfaces: one is a two dimensional cylindrical Fermi surface arising from $\sigma$-orbitals due to $p_z$ and $p_x$ electrons of $B$ atoms and the other is a three dimensional tubular Fermi surface network coming from $\pi$-orbitals due to $p_z$ electrons of $B$ atoms. They are weakly hybridized with $Mg$ electron orbitals. These two Fermi surfaces have different superconducting energy gaps: A large band gap (LBG) $\Delta_L$ on the 2D Fermi surface sheets and a small band gap (SBG) $\Delta_S$ on the 3D Fermi surface surface. The ratio $\Delta_S / \Delta_L$ is estimated to be around 0.3–0.4 [7,17,41]. Electrons on these two Fermi surfaces own different characters since the $\sigma$-orbital is strongly coupled to the in-plane $B$-atom vibration with $E_{2g}$ symmetry but the $\pi$-orbital is weakly coupled with this phonon mode.

Consider first the possible electronic contribution and let us analyze the field dependence of the thermal conductivity at low temperature (19K in our study) within a vortex lattice structure picture for a two band gap superconductor. The single vortex state has been studied by Nakai et al. [7]. In each unit cell and within a two-band superconducting model [7], the vortex core radius is found to be narrow for the LBG and large for the SBG electrons. When increasing $H$ the vortex core radius widens (and the order parameter is suppressed). This widening $H$ effect is stronger for the SBG than for the LBG electrons. Therefore the LBG vortex bound states are highly confined, due the narrow core radius, while the S-band vortex core states are more loosely bound. In the vortex core of the L-band, the local density of states (LDOS) at site $r$, $N_L(r, E \sim O)$, is thus highly concentrated, while the $N_S(r, E \sim O)$ LDOS is rather spread out in the S-band case. In fact, the low energy states extending from the S-band vortex cores are even expected to overlap with neighboring ones. For increasing $H$ the overlap is expected to become more pronounced and the LDOS to reduce to a flat profile, i.e. $N_S(r, E \sim O)/N_S(E_F) \simeq 0.5$ for the S-band, - where $N_S(E_F)$ is the total DOS in
the normal state at the Fermi level. This picture of the vortex structure in $MgB_2$ is in fine agreement with the field dependence of the electronic specific heat [5-7].

From a scattering point of view, we have also to consider two subsystems of quasiparticles characterized by $\Delta_L$ and $\Delta_S$ band gaps. Let $\kappa_e(B,T) = \kappa_{e,L}(B,T) + \kappa_{e,S}(B,T)$, thereby taking into account two sets of electrons with different characters.

According to the analysis on single crystalline $MgB_2$ [41] the electrons experiencing the large gap provide approximately 2/3 of the total electronic heat conduction, and the dominant part of the interaction between quasiparticles and low-frequency phonons is provided by that part of the electronic excitation spectrum experiencing the small gap [25,41].

The function $g(x,y)$ in Eq.(2) is then given by the sum

$$g(x,y) = g(x,\Delta_L(T)/k_BT) + \alpha g(x,\Delta_S(T)/k_BT).$$

The parameter $\alpha$ characterizes the relative weight of phonon scattering by quasiparticles in the two $S$ and $L$ subsystems.

It seems reasonable to expect that a weak field more easily suppresses the energy SBG than the LBG. Therefore superconductivity is maintained in a field mainly because the LBG survives, up to $H_{c2}$, as seen in Fig.1, i.e $\alpha = 0$. The $N_s(r,E \sim O)$ domains with low-energy excitations survive, but for increasing fields the SBG LDOS reaches a flat profile $N_s(r,E \sim O)/N_s(E_F) \simeq 0.5$, thereby not contributing to heat conduction. This $H_{c2S}$ seems to be ca. 3.0 T. These SBG electrons become heat carrier prone, inducing above $H_{c2S}$ an increase in the electronic thermal conductivity. Nakai et al. [7] claim that $H_{c2S}$ and $H_{c2L}$ are the same because they do not observe a hump in the specific heat at some intermediary field, i.e. the lower one. This maybe due to the fact that the specific heat is on one hand a bulk quantity in contrast to the thermal conductivity rather probing a percolation path. Moreover, we have shown elsewhere that it is sometimes very hard to obtain accurately kinks and phase transition lines form specific heat data [46], even with high precision measurements [47], in contrast to magnetotransport data [48].

Consider the phonon contribution next. Phonons are accepted to be the dominant heat carriers in $MgB_2$, especially down to $T/T_c \simeq 0.4$ [37-39]. The study of the phonon spectrum in $MgB_2$ [8-11] has clearly shown an anomalous phonon behavior at the low energy transfer of -24 meV on cooling through $T_c$ reflecting a strong relation between these phonon states and superconductivity origin in $MgB_2$.

From the thermal conductivity in a magnetic field behavior point of view, the phonon-(double type) vortex structure interaction should be expected to depend also on the nature of the phonon spectrum, in the sense of Tewordt-Wolkhausen picture [22]. The phonons which can inelastically interact with the bound quasiparticles in the vortex cores are only those for which the phonon wavelength $\lambda < \xi$, is of the size of a vortex core [27]. Thus it seems obvious that due to the overlapping features of the low energy states, the SBG vortices become less effective scattering centers for phonons as well. Therefore the mean free path of low frequency phonons may also increase, resulting in an increase of the thermal conduction process at high field, in agreement with the above data. We conjecture that the quasi particle associated with the LBG contribute moderately to the thermal conductivity at high field and low temperature. In fact interactions involving the quasiparticles associated with the LBG and thermal phonons have not been well described. Nevertheless they could contain an elastic component of the scattering process. Such a picture seems a fine way of interpreting the decrease of the thermal conductivity at high field and high temperature (Fig.2).

Finally, an alternative view considers that electrons self-organize into collective textures rather than point-like entities [49]. Such an image of electronic collective textures in an applied magnetic field is complex and not immediately ready for interpreting the (electron or phonon in fact) scattering process within the framework of a double vortex structure, as discussed above. We cannot further debar the possibility that a field-induced shift of the phonon spectrum itself could produce effects similar to temperature. Some softening of the phonon spectrum indeed exists, as seen in the thermal expansion coefficient [50,51]. This equivalency, to be fair, may be also at the origin of the enhanced behavior of the field-temperature dependent thermal conductivity in superconducting $MgB_2$.

**IV. CONCLUSION**

In conclusion, we have investigated the heat transport in $MgB_2$ in both absence and presence of magnetic fields (up to 14T) in the temperature range between 4-50K. In these regimes, the superconductivity of $MgB_2$ persisted as seen from electrical resistivity measurements; no clear anomaly appeared in the thermal conductivity at $T_c$. The thermal conductivity in absence of field shows a $T$-linear increase up to 50 K. In weak magnetic fields the thermal conductivity is insensitive to the magnitude of the applied field; an enhanced thermal conductivity appears at higher fields and low temperature, but not at high temperature. We argue that the existence of two quasiparticle subsystems with different field-dependent characters explain the results. The field dependence of the thermal conductivity for such a two-band
superconductor indicates that the enhancement of $\kappa(H, T)$ at low temperature in high fields is a result of the overlap of the low energy states outside the small band gap vortex cores, above a critical $H_{c2S}$ field.

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Figure 1:
Electrical resistivity of polycrystalline MgB$_2$ versus temperature in 0, 1, 4, 7, 10 and 14 Tesla applied magnetic fields

Figure 2:
Thermal conductivity, $\kappa(H, T)$, of polycrystalline MgB$_2$ vs. temperature $T$ in zero $H$-field and in 0.02, 0.5, 1.0, 5.0, 10.0 and 13.9 Tesla applied magnetic fields

Figure 3:
Field dependence of MgB$_2$ reduced thermal conductivity $\kappa(H)/\kappa(H = 0)$) at several temperatures
Fig. 2
Fig. 3

$k(H)/k(0)$ vs. $\mu_0H [T]$ for different temperatures: $T = 8$ K, $T = 11$ K, $T = 15$ K, $T = 20$ K, $T = 30$ K.