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Magnetotransport study of MgB_2 superconductor

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Abstract

Precise magnetotransport studies of heat and charge carriers in polycrystalline MgB_2 show that magnetic fields up to 8 T remarkably influence electrical resistivity, thermoelectric power and thermal conductivity. The superconducting transition temperature shifts from 39 K to 19 K at 8 T as observed on electric signals. The temperature transition width is weakly broadened. Electron and phonon contributions to the thermal conductivity are separated and discussed. The Debye temperature calculated from a phonon drag thermoelectric power component is inconsistent with values derived through other effects.

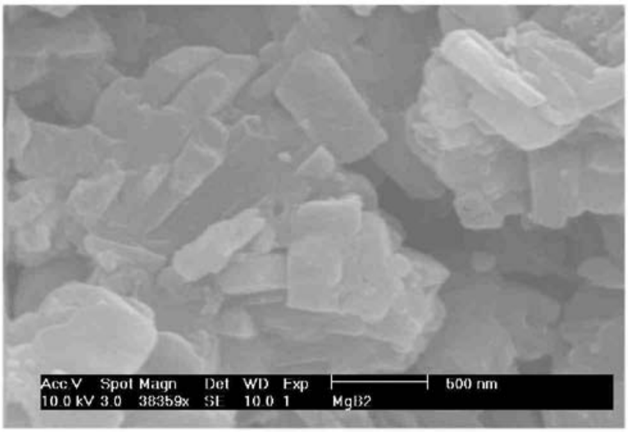
1. **Introduction**

Superconductivity in a simple compound, i.e. MgB_2, has recently been discovered [1] at a remarkably high transition temperature near 40 K. Most experiments on MgB_2, such as the isotope effect [2, 3], the T_C pressure effect [4, 5], tunnelling spectroscopy [6-8], magnetic susceptibility [9] and also thermal conductivity [9-12], indicate that the superconducting properties of MgB_2 can be consistent with a phonon-mediated BCS electron pairing with two gaps in the quasi-particle excitation spectrum. Indeed, there are two distinctive Fermi surfaces: one is a two-dimensional (2D) cylindrical Fermi surface arising from σ-orbitals due to p_x and p_y electrons of B atoms and the other is a three-dimensional (3D) tubular Fermi surface network coming from π-orbitals due to p_z electrons of B atoms. These are weakly hybridized with Mg electron orbitals. The two Fermi surfaces have different superconducting energy gaps: a large bandgap (LBG) Δ_L on the 2D Fermi surface sheets and a small bandgap (SBG) Δ_S on the 3D Fermi surface. From an electron-phonon interaction point of view, note that electrons in the σ-orbital are strongly coupled to the in-plane B-atom vibration with E_{2g} symmetry while those in the π-orbital are weakly coupled with this phonon mode. From a magnetic field point of view, it seems reasonable to expect that a weak field more easily suppresses the energy SBG than the LBG. Therefore superconductivity is maintained at high fields mainly because the LBG survives.

These results indicate that phonons play an important role in the electrical and heat carrier interaction in MgB_2, while electron scattering can be quite particular, the more so in a magnetic field. This can be confirmed through studies of related transport (non-equilibrium) phenomena, such as the thermoelectric power (TEP), or Seebeck coefficient. It has been used previously and widely in order to study superconductors, offering important clues about heat and charge carrier nature and their scattering processes, especially in the superconducting state(s) where traditional electrical probes such as the electrical resistivity (even in presence of an external magnetic field) and Hall effect are weakly operative. Moreover, studies of the TEP (and the related electrothermal conductivity) in applied magnetic fields have been an interesting new probe of the vortex state of high-T_C superconductors [13, 14] (and the order parameter symmetry [15, 16]). The TEP of MgB_2 has received little attention [17-23] but no investigation seems to have been reported in the literature about the influence of magnetic field on the TEP of MgB_2.

In this paper we present a study in particular of the TEP of polycrystalline MgB_2, at low and moderate temperatures both in the absence and in the presence of moderate magnetic fields, together with electrical resistivity and thermal conductivity data under similar conditions in a homemade specially built system. The temperature range extends between 10 and 300 K and the applied magnetic field goes up to 8 T. The transport coefficient behaviours are discussed; the role of the two gaps is sought near the superconducting transition.

Figure 1: *An electron microscope image of a part of the studied MgB_2 sample.*



2. Sample preparation and characterization

MgB_2 samples were synthesized under argon pressure from stoichiometric amounts of high-purity powdered Mg and B, as described elsewhere [1]. X-ray diffraction measurements show that the samples are single-phased. The samples are very dense although polycrystalline as observed by an electron microscope PHILIPS ESEM XL30. The superconducting volume fraction is about 60%, as estimated from susceptibility measurements [1]. A high-resolution electron microscope micrograph is shown in figure 1.

3. Transport measurements

The TEP and thermal conductivity were measured in the specially designed cryostat inserted into the PPMS system. This small cryostat could be operated in two modes: either at the required constant temperature, i.e. that of the sample plate, or at a slow drift of about 0.02 K min^{-1}. In both modes the temperature difference, equal to a few kelvin, between the sample plate and the PPMS connector, was electronically kept constant during the measurements. Such a technique allows us to stabilize the temperature of the sample cold end with an accuracy of ±0.005 K.

The thermal conductivity was measured using the stationary heat flux method in a broad temperature range, i.e. 10-300 K. The sample temperature and temperature gradient were measured by a constantan-chromel thermocouple. The temperature gradient of about 0.1-0.5 K along the sample was created by a small heater dissipating about 5 mW. Particular care was taken to avoid any parasitic heat transfer between the sample and its environment, i.e. the sample was placed inside the cylindrical screen and the temperature of the screen was electronically stabilized at the level of the sample. Moreover, all current and voltage leads were thermally anchored to the screen [24]. The radiation and gas conductivity losses are negligible, being less than 0.1% even at 300 K. The total measurement error of absolute values of thermal conductivity is estimated to be below 2%. The relative surplus error, estimated as maximum deviations of experimental points from an average curve, did not exceed ±0.3%. The temperature gradient measured along the sample was used to calculate both the thermal conductivity and the TEP. The electrical resistivity of each investigated sample was measured by the standard four-probe method within a relative accuracy estimated to be about 3%. The current density was usually about 0.1 A cm^{-2}. The temperature sweep rate was about 0.2 K min^{-1}. The experimental set-up and the measurement procedure have been described in detail in [24-26]. The fitting of experimental data to formulae was based on the least-squares method and the calculated parameters correspond to 95% confidence level.

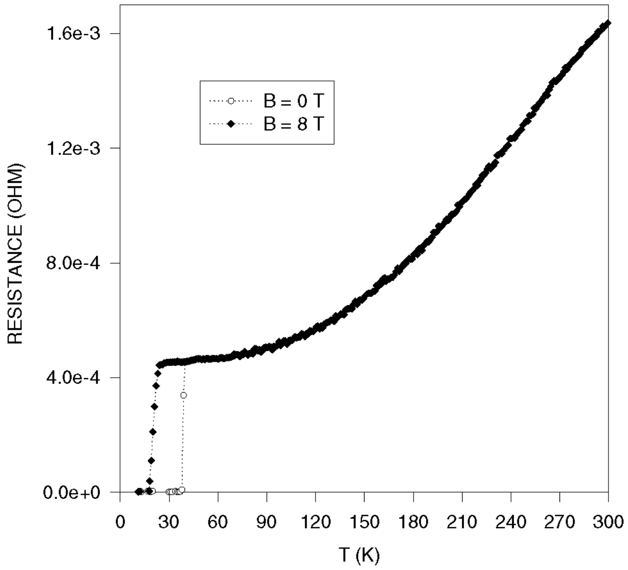
4. Electrical resistivity

The electrical resistivity is relatively low, being about 0.5 x 10^{-6} Ω m at room temperature as derived from the electrical resistance of the samples studied (figure 2(a)). This confirms a high quality of the sample and good intergrain connectivity. The relatively large value of the residual resistance ratio, defined as RRR = $R_{RT}/R(40\text{ K})$ equal to about 3.5, reveals that only a low density of defects and impurities is likely to be present in the sample. The temperature variation of the normal state electrical resistivity in the temperature range of interest is similar to that found in the literature and may be fitted to a function such as

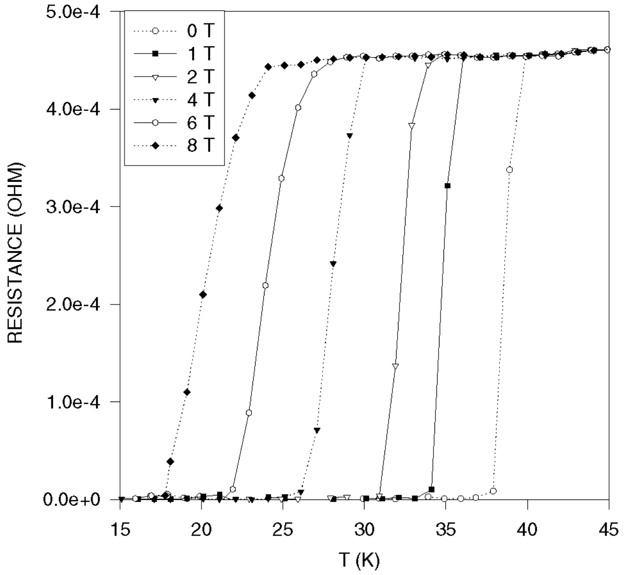
$$\rho(T) = \rho_0 + AT^N \qquad (1)$$

where N equals 2.52 ± 0.1, which falls in the range of typical values spread between 2 and 3 as found in [4], The ρ_0 term covers the temperature-independent electron scattering on impurities and defects. This ρ_0 term is equal to 1.5×10^{-7} Ω m at 40 K, proving again the high quality of the sample. The second temperature-dependent term is usually thought to be due to electron-phonon scattering and the complicated density of states for such excitations. No measurable magnetoresistance was observed in magnetic fields up to 8 T, which allows us to exclude the presence of unreacted magnesium. The middle point of the superconducting transition is at a temperature T_C equal to 38.5 K. This agrees with the Testardi correlation between T_C and RRR as recalled in [27]. The superconducting transition interval DT is narrower than 1.5 K in the absence of an externally imposed magnetic field (figure 2(b)). The magnetic field affects strongly the superconducting transition shifting the transition temperature down to 20.5 K in an 8 T external magnetic field. This shift is accompanied by an increase of the transition width DT, up to about 6 K at 8 T.

Figure 2: *(a) Temperature variation of electrical resistance at various magnetic field strengths, (b) Low-temperature variation of electrical resistance at various magnetic field strengths.*

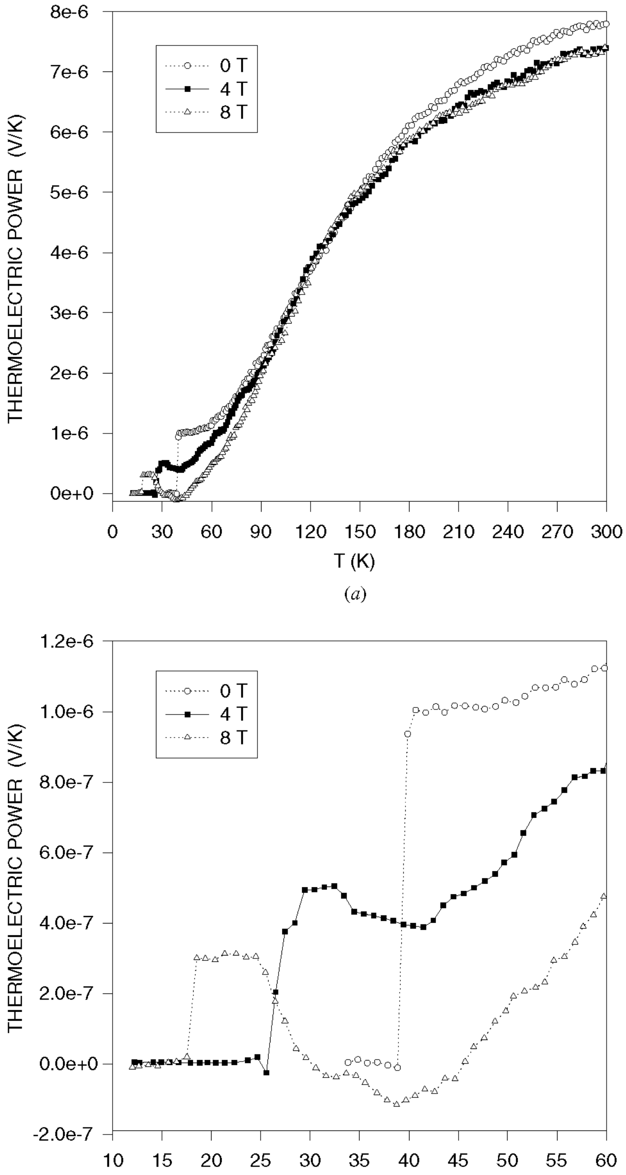


(*a*)



(*b*)

Figure 3: *(a) Temperature variation of thermoelectric power at magnetic fields from 0 to 8 T. (b) Low-temperature variation of thermoelectric power at magnetic fields from 0 to 8 T.*



(*a*)

(*b*)

5. Thermoelectric power

The normal state TEP (figure 3(a)) is a smooth function of temperature almost constantly rising. Between 70 and 130 K the TEP is a linear function of temperature varying with a mean rate equal to 4 x 10^{-8} V K^{-2}. At higher temperatures, TEP slowly approaches a saturation level estimated to be about 8 μV K^{-1}. Such a shape is similar to that reported in [23].

The influence of the magnetic field is remarkably manifested both below 70 K and above 130 K. In both cases TEP values are shifted down. As expected, although not reported in the literature, the transition temperature, i.e. the inflection point at T_C, shifts down to a lower temperature, when the magnetic field increases (figure 3(*b*)). The TEP vanishes at T_0 = 27 and 16 K in external magnetic fields of 4 and 8 T, respectively. The width of the transition interval is about 1.5 K and remains almost unaffected by the magnetic field strength. Such behaviour of TEP is in contrast to that found for the bismuth- and yttrium-based superconductors [30], where the transition width extends up to dozens of degrees in comparable magnetic fields. However, something similar to what is found in d-wave order parameter TEP behaviour seems to take place here as well. The vanishing of the TEP occurs after a change in sign (figure 3(*b*)). This has been discussed as due to either internal temperature gradients and electric forces, perhaps related to the material polycrystallinity, or due to intrinsic effects related to the complicated gap structure of the Fermi surface [31]. An extra feature is the change in behaviour between 0 and 4 T on one hand and between 4 and 8 T on the other hand. It could be argued that this is similar to what happens in the thermal conductivity [33], and results from the disappearance of the SBG near 4 T.

Table 1: *Magnetic field (T),A and C parameters of the TEP fit, Fermi energy E_F and Debye temperature T_D.*

Magnetic field (*T*)	***A* (V K^{-2})**	***C* (V K^{-4})**	**E_F (eV)**	**T_D(K)**
0	1.50	1.2	2.5	1450
4	1.35	1.57	2.7	1330
8	1.3	1.79	2.8	1270

The low-temperature normal state variation of TEP between 40 and 95 K was attempted to be fitted by

$$Q(T) = Q_D(T, H) + Q_G(T, H) = A(H)T + C(H)T^3 \quad (2)$$

where the terms on the right-hand side might correspond to a diffusion and phonon drag contributions, respectively. A cubic temperature term may also occur when the material has a complicated electronic potential and Fermi surface [32], as is the case in MgB_2 indeed. The zero magnetic field *A* and *C* parameters approximately coincide with those reported by Putti *et al* [22] for the temperature range below 100 K. The *A* and *C* parameters evolve with the magnetic field as reported in table 1 for $B = 0$, 4 and 8 T. The data of table 1 show that a strong magnetic field affects the electron-phonon coupling. At high magnetic fields and moderate temperature, the distinctive two superconductivity gaps on the MgB_2 Fermi surface have probably disappeared [33, 34]. Therefore, we could argue that the contributions are mainly controlled by electron-phonon scattering. In a first approximation, assuming isotropic carrier scattering and an energy-independent relaxation time [35, 36], we have

$$Q_D = \pi^2 K_B^2 T / 2\,eE_F \qquad (3)$$

and

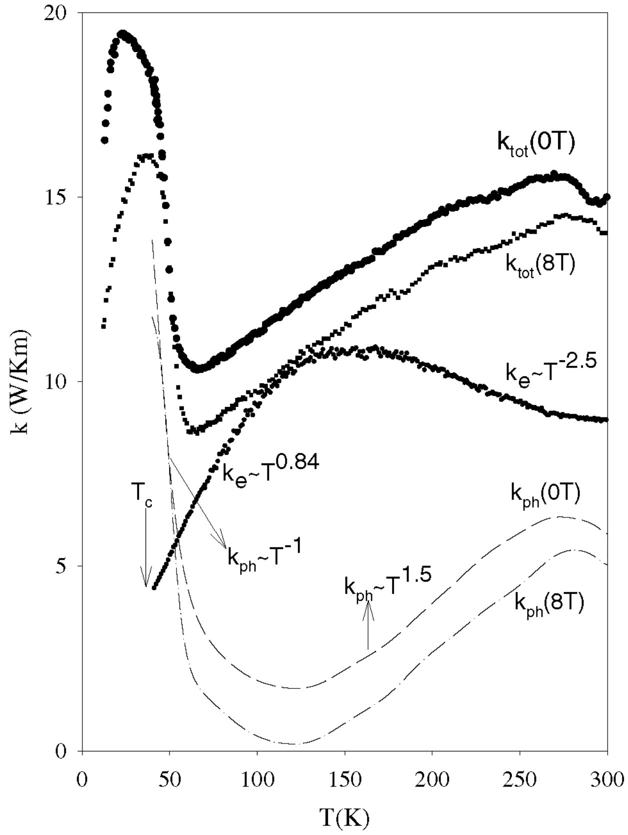
$$Q_G = 4\pi^4 K_B T^3 / 5\,e n_v T_D^3, \qquad (4)$$

where υ_n is the number of conduction electrons in the valence band and T_D is the Debye temperature. This in turn allows us to conclude that the Fermi energy (E_F) increases to 2.8 eV for a magnetic field of 8 T at these moderate temperatures above T_C. This is accompanied by a lowering of the Debye temperature T_D from 1450 to 1270 K (table 1). The values of T_D, as calculated from equation (4), are remarkably higher than those determined from the thermal conductivity of the same sample (see below) and seem to be overestimated as compared to T_D measured through the specific heat. In the latter case, T_D is usually about 800-900 K [37, 38]. This is usual for T_D, which is very much a property-dependent parameter. It is worth to noting that, if a phonon drag effect exists in MgB_2, it seems so weak that it does not generate the specific bump in the TEP like that observed in various metals and compounds. Thus, a more rigorous theoretical approach will be needed to analyse any possible phonon drag effect in MgB_2 and its usefulness to determine the Debye temperature.

6. Thermal conductivity

The temperature variation of the total thermal conductivity k_{tot}(0 T) and k_{tot}(8 T) measured in magnetic fields of 0 and 8 T, respectively is shown in figure 4. Values of k_{tot}(0 T) are 10-15% higher than k_{tot}(8 T), with the exception of the interval ranging from T_C to 50 K, where they are approximately equal. The temperature variations of the thermal conductivity measured in both magnetic fields exhibit a common shape. Both k_{tot}(0 T) and k_{tot}(8 T) decrease when the temperature decreases from 270 to 66 K. A very similar variation was also reported for intermetallic rare-earth compounds [39]. Below 66 K, both k_{tot}(0 T) and k_{tot}(8 T) start to rise quickly and achieve a maximum of 19.5 W K m^{-1} at T_{max} = 24 K and 16 W K m^{-1} at T_{max} = 38 K, respectively. Below T_{max} the thermal conductivity varies as $k_{tot}(0\ T) \sim T^{0.5}$ and $k_{tot}(8\ T) \sim T^{0.35}$. The sharp maximum of the thermal conductivity of MgB_2 at a lower temperature is similar to that reported for borocarbide superconductors [40]. The thermal conductivity anomaly seen above 280 K is also similar to that of $La_{1.98}Y_{0.02}CuO_4$ single crystals [41]. This cannot be explained without additional studies.

Figure 4: *Temperature variation of thermal conductivity at magnetic fields of 0 and 8 T*



Usually the thermal conductivity of metallic alloys may be written as the sum of electron and phonon contributions

$$k_{tot} = k_e + k_{ph}. \qquad (5)$$

In order to separate both components, the Wiedemann-Franz law is used

$$k_e(T) = L_0 T / \rho(T) \qquad (6)$$

where $L_0 = 2.44 \times 10^{-8}$ W Ω K^{-2} is a Lorentz number and $\rho(T)$ is electrical resistivity. The temperature variation of calculated electron thermal conductivity of MgB_2 is plotted in figure 4. We can see that the electron component of thermal conductivity is overwhelming at all temperatures above 50 K. A broad maximum of k_e (T) appears between 125-175 K. The temperature variations below and above this maximum may be approximated by $k_e \sim T^{0.84}$ and $k_e \sim T^{2.5}$, respectively. These exponents calculated for electron thermal conductivity correspond to the best fit only and cannot be directly related to specific models. The exponent equal to 0.84 is close to a value of 1 predicted for elastic electron scattering on impurities in a Wiedeman-Franz approach. The difference may be due to the possible contribution from, for example, inelastic processes. According to the Matthiessen rule, the electron thermal resistivity

$$k_e^{-1} \equiv W_e = W_e^p + W_e^i \qquad (7)$$

where W_e^p and W_e^i correspond to the high-temperature thermal resistivity, due to the electron-phonon scattering, and the low-temperature thermal resistivity, due to electron scattering on chemical impurities and defects, respectively. Following [31] they may be expressed as

$$W_e^i = \rho_0 / L_0 T \qquad (8)$$

and

$$W_e^p \approx (C/L_0 T)(T/T_D)^3. \qquad (9)$$

The separated phonon thermal conductivity is higher at 0 T than at 8 T magnetic fields (figure 4), with an exception for the range from T_C to 50 K, where they are almost equal, likely indicating again that the SBG has disappeared (much) before 8 T. A broad minimum in phonon thermal conductivity is located at 120 K. The temperature dependence of phonon thermal conductivity may be approximated by power laws $\sim T^{1}$ and $\sim T^{1.5}$ below and above the minimum, respectively. Such a temperature variation of the phonon thermal conductivity is characteristic for the phonon-phonon interaction [42].

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 centres for heat carriers. The phonons, which can inelastically interact with the bound quasi-particles in the vortex cores, are only those for which the phonon wavelength λ, is of the size of a vortex core [43]. In MgB_2, due to the two sorts of bandgaps, we may expect two types of vortices; thus the vortex-phonon interaction might be of a double type, when calculating its effect in the sense of the Tewordt-Wolkhausen picture [44, 45]. It seems obvious that due to the overlapping features of the low-energy states, the SBG vortices become less effective scattering centres 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-particles associated with the LBG moderately contribute to the thermal conductivity at high field and low temperature. This picture has provided a qualitative explanation of the field-induced decrease in $k(H)$ observed in the vortex states of conventional superconductors and high T_C cuprates in the investigated regions of the field-temperature diagram [46-48].

Following Wilson [49] the electronic component of the thermal resistivity may be written as

$$k_e^{-1} \equiv W_e = \beta T^{-1} + \alpha T^2 \qquad (10)$$

with $\beta = \rho_0/L_0$ and $\alpha = 4\rho' T_D \frac{3}{\pi^2} n_a^{2/3}$, where ρ_0 is a residual electrical resistivity, ρ' is temperature derivative of electrical resistivity, and n_a is the effective number of conduction electrons approximately equal to 0.15 and 0.36 per atom for the σ and π conduction bands, respectively. The first term describes the thermal resistivity due to electron scattering on impurities and defects. The second term corresponds to the electron-phonon scattering. Fitting the electron thermal conductivity to equation (10) supplies $\alpha = 7.66 \times 10^{-7}$ with 1.5% accuracy. Then we arrive at the Debye temperature $T_D = 844$ K, which agrees with values usually reported for MgB_2 and is remarkably lower than T_D estimated from thermoelectric data.

7. Conclusion

The electrical resistivity and TEP of polycrystalline MgB_2 show that a magnetic field of 8 T shifts the superconducting transition temperature and the mid-point transition temperature down but only weakly affects the width of the transition interval. The influence of the magnetic field on the electron and phonon contributions to the thermal conductivity have been separated and discussed, together with the possible magnetic field effect on the two bandgaps and the resulting consequences for scattering processes. The Debye temperature calculated from the thermal conductivity confirms the usually reported values. To concur that a phonon drag term occurs in the TEP in the absence or presence of a magnetic field is not proven, surely not in view of the deduced value of the Debye temperature through a phonon drag argument. It seems that the order parameter symmetry might influence the behaviour of the TEP tail near the superconducting transition as found in the superconducting cuprates. Due to the two bandgap features, the TEP behaviour near its vanishing point might receive further interest.

Acknowledgments

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