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# Thermal conductivity of superconducting MgB<sub>2</sub>

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## Abstract

Thermal conductivity of superconducting MgB<sub>2</sub> was studied in both the superconducting and the normal state region. The latter is almost equally determined by the electronic - and the lattice contribution to the total thermal conductivity. In the superconducting state, however, the lattice contribution is larger. The electronic thermal conductivity below  $T_c$  was derived from the experimental data considering the Bardeen-Rickayzen-Tewordt theory together with the model of Geilikman. The analysis shows that electron scattering on static imperfections dominates.

**key words:** MgB<sub>2</sub> superconductivity, thermal conductivity, specific heat

# 1 Introduction

Superconductivity with a remarkably high transition temperature  $T_c \approx 39$  K was recently discovered in  $\text{MgB}_2$  [1]. The subsequent investigation of the boron isotope effect by Bud'ko *et al.* [2] revealed a partial isotope exponent  $\alpha_B \approx 0.26$  (corresponding to  $\Delta T_c = 1$  K) which appears to be consistent with a phonon-mediated BCS superconducting (SC) mechanism. Also other experimental reports as e.g. on specific heat [3, 4] argued that their data can be accounted for by a conventional, s-wave type BCS-model. Investigations of the SC gap of  $\text{MgB}_2$  by means of Raman scattering are also consistent with an isotropic s-wave gap with a moderate coupling  $2\Delta \approx 4.1k_B T_c$  [5]. However, a theoretical analysis of the temperature dependence of the upper critical field  $H_{c2}(T)$  in terms of Eliashberg type models by Shulga *et al.* [6] demonstrates that the shape and the magnitude of the upper critical field of  $\text{MgB}_2$  can definitely not be accounted for by an isotropic single band model, but may successfully be described within a multi-band Eliashberg model with various options. A careful calorimetric investigation of the SC parameters of  $\text{MgB}_2$  by Wang *et al.* [7] gave even more direct evidence against the arguments for simple isotropic BCS type superconductivity: “The nearly quadratic dependence of  $C(T)$  versus  $T$  at  $T \ll T_c$ , its non-linear field dependence, and the discrepancy between the electron-phonon coupling constant  $\lambda_{ep}$  as determined by the renormalization of the electron density of states ( $\lambda_{ep} \sim 0.6$ ) and by McMillan’s equation for isotropic superconductors ( $\lambda_{ep} \sim 1.1$ ), are inconsistent with a single isotropic gap”. Direct hints for a non BCS temperature dependence of the gap energy  $\Delta(T)$  were also obtained by tunneling experiments on  $\text{MgB}_2/\text{Ag}$  and  $\text{MgB}_2/\text{In}$  junctions [8].

Thermal conductivity  $\lambda$  is one of those transport coefficients which exhibits non-zero values in both the normal and the SC state. The temperature dependence of  $\lambda$  allows to distinguish between the most important interactions present in a superconductor. In particular, the interaction of electrons with phonons are recorded in the magnitude of  $\lambda(T)$ . Moreover, scattering of these particles by static imperfections like impurities, defects or grain boundaries are reflected.

The aim of the present work is to derive the temperature dependent thermal conductivity of  $\text{MgB}_2$  and to analyse the data with respect to the electronic - and the lattice thermal conductivity both in the normal and the SC state. Moreover, we present resistivity and specific heat measurements in order to characterise the investigated sample.

## 2 Experimental

A  $\text{MgB}_2$  sample of about 1.3 g was synthesized by direct reaction of the elements. The starting materials were elemental magnesium (rod 99.9 mass % nominal purity) and boron (99.5 % powder, crystalline, < 60 mesh, 99.5 mass %). The elements in a stoichiometric ratio were enclosed in a cylindrical tantalum crucible sealed by arc welding under argon atmosphere. The tantalum crucible was then sealed in an iron cylinder and heated for one hour at 800° C and two hours at 950° C in a furnace. The sample characterized by x-ray diffraction show pure  $\text{MgB}_2$  phase; only one very weak peak due to extra phase has been found.

The thermal conductivity measurement was performed in a flow cryostat on a cuboid-shaped sample (length: about 1 cm, cross-section: about 2.5 mm<sup>2</sup>), which was kept cold by anchoring one end of the sample onto a thick copper panel mounted on the heat exchanger of the cryostat. The temperature difference along the sample, established by electrical heating, was determined by means of a differential thermocouple (Au + 0.07 % Fe/Chromel). The measurement was performed under high vacuum and 3 shields mounted around the sample reduced the heat losses due to radiation at finite temperatures. The innermost of these shields is kept on the temperature of the sample via an extra heater maintained by a second temperature controller.

Resistivity data were taken from bar shaped samples applying a standard 4-probe d.c. technique at temperatures down to 0.5 K and in magnetic fields up to 12 T.

Specific heat measurements were carried out on a sample of about 1 g in the temperature range 5 K–50 K using a quasi adiabatic step heating technique.

## 3 Results and discussion

In order to give a direct proof of the SC bulk properties of our  $\text{MgB}_2$  sample prior to the transport measurements we checked the specific heat . These specific heat measurements performed in zero-field, 1 and 9 T showed reasonable agreement with results previously reported [3, 4, 7]. The thermodynamic mean transition temperature  $\overline{T}_c$  of our sample is 37.5 K. As already noted in the introduction there have been distinctly different and partly controversial

conclusions suggested in Refs. [3, 4, 7], although their raw data are in fair agreement with each other. Therefore, we show in figure 1 the temperature dependence of the electronic specific heat,  $C_{el}(T)$  versus  $T$ , obtained by subtracting the lattice heat capacity deduced from the 9 T specific heat data. Of course, 9 T are insufficient to obtain a complete suppression of superconductivity in MgB<sub>2</sub>, but there is already a large reduction of the order parameter combined with a dramatic broadening of the transition (see Ref. [7] for comparison of 10, 14 and 15 T data). Thus, we obtained a Sommerfeld coefficient of the normal-state electronic specific heat  $\gamma \simeq 2.4(2)$  mJ/mol K<sup>2</sup> by extrapolating the 9 T data in  $C/T$  versus  $T^2$  from 30 – 100 K<sup>2</sup> to zero temperature which is already close to  $\gamma = 2.7 \pm 0.15$  mJ/mol K<sup>2</sup> obtained from the 14 and 16 T measurement by Wang *et al.* [7]. The important point to emphasize in figure 1 is the non BCS-like temperature dependence of the SC-state electronic specific heat. In fact, we observed a similar deviation from a simple BCS temperature dependence (solid line, figure 1) as previously reported by Wang *et al.* [7] ( $C_{el}^{BCS} = 8.5\gamma T_c \exp[-0.82\Delta(0)/k_B T]$  for  $T < 0.4T_c$ ). This discrepancy is supposed to be indicative for the opening of an additional gap below about 10 K. The solid line in figure 1 indicates that a fraction of electrons corresponding to a normal state  $\gamma \sim 1.4$  mJ/mol K<sup>2</sup> is tentatively accounted for by the BCS-fit with  $\Delta(0)/k_B T_c \simeq 1.9$  while a second fraction corresponding to  $\gamma \sim 1.0$  mJ/mol K<sup>2</sup> contributes to the smaller gap opening at temperatures well below  $T_c/2$ .

To further screen the quality of the sample, temperature and magnetic field dependent resistivity measurements  $\rho(T, H)$  were performed from 0.5 K up to room temperature. Shown in figure 2 (right axis) is  $\rho(T)$  at zero field. The transition into the SC state occurs at  $T_c = 38.9$  K, which is in fine agreement with already published data. The RRR ratio of this polycrystalline sample is about 6. The resistivity behaviour in the normal state region matches a dependence according to  $\rho(T) = \rho_0 + AT^2$  with the residual resistivity  $\rho_0 = 12.5 \mu\Omega\text{cm}$  and the coefficient  $A = 9 \times 10^{-4} \mu\Omega\text{cm}$ . A  $T^2$  behaviour of  $\rho(T)$  was recently reported [9] and it seems to reflect interactions between charge carriers. A study of the Hall coefficient implies that electrical transport is dominated by holes [10]. The value of the coefficient  $A$ , however, is significantly smaller than that known e.g., for highly correlated electron systems, but seems to reflect the modest density of states at Fermi energy [11]. It should be mentioned that other power laws with an exponent close to 3 were reported for sintered material of MgB<sub>2</sub> [12, 13]. Measurements of the resistivity down to 0.5 K and in fields up to 12 T indicate that  $H_{c2}$  is well

above that limit. Interestingly, the transition region becomes much broader with increasing fields, but different values of imprinted currents (from 5 to 40 mA) do not change the width of the transition.

The temperature dependent thermal conductivity  $\lambda$  of MgB<sub>2</sub> is shown in figure 2. The overall behaviour of  $\lambda(T)$  is typical of an intermetallic compound where scattering on static imperfections prohibits a pronounced maximum to occur, as it is the case in pure and simple metals. Moreover, the absolute magnitude appears to be of that order, as usually found for intermetallics. Anomalous behaviour of  $\lambda(T)$  in the proximity of  $T_c$  is not observed and a local maximum or a pronounced shoulder below  $T_c$  do not occur in the investigated sample.

Generally, the total thermal conductivity of metals consists of a sum of an electronic contribution  $\lambda_e$  and a lattice contribution  $\lambda_l$ :

$$\lambda = \lambda_e + \lambda_l. \quad (1)$$

In order to separate both contributions from the total measured effect, the Wiedemann-Franz law is applied, assumed to be valid, at least, in simple metals. This model relates the electrical resistivity  $\rho$  with the electronic contribution to the thermal conductivity  $\lambda_e$  and can be expressed as

$$\lambda_e(T) = \frac{L_0 T}{\rho(T)}, \quad (2)$$

where  $L_0 = 2.45 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$  is the Lorenz number.

Using equation 1 and taking into account the appropriate values of the electrical resistivity in the normal state region of MgB<sub>2</sub> (compare figure 2, right axis) allows to split  $\lambda$  into  $\lambda_e$  (dashed line, figure 2) and  $\lambda_l$  (dashed-dotted line, figure 2). This type of analysis indicates that both contributions are almost equal in the entire temperature range of the normal state region of MgB<sub>2</sub>.

According to Matthiessen's rule both  $\lambda_e$  and  $\lambda_l$  are limited owing to various scattering processes, which can be expressed in terms of a thermal resistivity  $W$ . In the case of non-magnetic materials, the following temperature dependence of the electronic contribution to the total measured quantity is assumed to be valid [14]:

$$1/\lambda_e(T) \equiv W_e(T) = W_{e,0}(T) + W_{e,ph}(T) = \frac{\alpha}{T} + \beta T^2, \quad (3)$$

where the subscripts (e,0) and (e,ph) refer to interactions of the conduction electrons with static imperfections and thermally excited phonons, respectively;  $\alpha$  and  $\beta$  are material constants.

Equation 3 allows to determine  $W_{e,0}$  and  $W_{e,ph}$ . Shown in figure 3 is the electronic thermal resistivity  $W_e$  of MgB<sub>2</sub> displayed in the normal state region up to about 80 K. The solid line is a least squares fit of the data according to equation 3 and the dashed and the dashed-dotted lines represent  $W_{e,0}$  and  $W_{e,ph}$ , respectively. Thus, the deduced parameters are  $\alpha = 0.55 \text{ cmK}^2/\text{mW}$  and  $\beta = 2.8 \times 10^{-7} \text{ cm/mWK}$ . Obviously from figure 3, the scattering of electrons with static imperfections of the crystal becomes dominant as the temperature approaches  $T_c$ .

The relative weight of  $\lambda_e$  and  $\lambda_l$  at the SC transition temperature  $T_c$ , as defined from the Wiedemann Franz law, also serves to determine the temperature dependence of  $\lambda_e$  and subsequently of  $\lambda_l$  below  $T_c$ . Since within the BCS theory, Cooper pairs do not carry heat and entropy, the scattering terms of equation 3 have to be modified in order to account for the decreasing number of unpaired electrons.

In the SC state, the thermal resistivity  $W_e^s$  can be represented as

$$W_e^s \equiv 1/\lambda_e^s = W_{e,0}^s + W_{e,ph}^s = \frac{\alpha}{Tf(t)} + \frac{\beta T^2}{g(t)}, \quad (4)$$

with  $t = T/T_c$  [15]. The functions  $f(t)$  and  $g(t)$  were calculated repeatedly and agree well with experimental findings [16, 17, 15]. In the dirty limit of a superconductor, the first term of equation 4 dominates, i.e.,  $1/\lambda_e^s \equiv W_e^s \approx W_{e,0}^s$ . In terms of the BRT theory [16]  $\lambda_e^s(t)/\lambda_e^n$  is a universal function of  $t$ , dependent on the value - and the temperature dependence of the SC gap  $\Delta$ .

On the contrary, clean limit superconductors are dominated by the second term of equation 4, revealing  $1/\lambda_e^s \equiv W_e^s \approx W_{e,ph}^s$ . Geilikman et al. [15] have calculated and tabulated  $g(t)$  which yields again a universal behaviour on  $t$ . Differently to scattering on imperfections,  $\lambda_e^s(t)/\lambda_e^n(T = T_c)$  in the BCS limit increases initially with decreasing values of  $t$  in spite of a rapid decrease of electronic excitations. A maximum occurs at  $t \approx 0.28$  with  $\lambda_e^s(t = 0.28)/\lambda_e^n(T = T_c) = 2.44$ . Various high temperature superconductors are found to exhibit a maximum in  $\lambda(T)$  below  $T_c$  and thus the origin of that feature is, at least partly, attributed to a significant scattering strength of electrons on thermally excited lattice vibrations (compare e.g. [18]).

Figure 3 evidences that slightly above  $T_c$ ,  $W_{e,0}$  exceeds  $W_{e,ph}$  by more than one order of magnitude and contributes at this temperature about 95 %

to  $W_e$ . This implicitly favours a description of the thermal conductivity of MgB<sub>2</sub> based on scattering of electrons on impurities. Nevertheless, for the present analysis of the data both terms of equation 4 are considered in order to analyse the electronic contribution to the total thermal conductivity. Taking into account the functions  $f(t)$  and  $g(t)$  and the numerical values of  $\alpha$  and  $\beta$  allows us to determine  $\lambda_e^s$  below  $T_c$  (compare figure 4, panel (a)).  $\lambda_e^s$  decreases with decreasing temperature and is primarily determined by the BRT function  $f(t)$ . The difference  $\lambda - \lambda_e^s$  represents the phonon-originated thermal conductivity  $\lambda_l^s$  in the SC state. The latter appears to be larger in the SC state than  $\lambda_e^s$ . For temperatures  $T/T_c < 0.4$ , the lattice thermal conductivity of MgB<sub>2</sub> becomes dominant. This nicely agrees with theoretical considerations [15]. The lattice term  $\lambda_l^s$  is constrained by various scattering processes; among them are interactions of the phonons with electrons, point defects, dislocations or sheetlike faults.

To account for observed deviations of the SC gap of MgB<sub>2</sub> from the BCS theory, the experimental data from tunnel experiments on MgB<sub>2</sub>/Ag and MgB<sub>2</sub>/In [8] have been used to modify the function  $f(t)$  and thus  $W_{e,0}^s$ . The smaller the value of  $\Delta(T)$  with respect to the BCS theory, the less steep is the decrease of  $f(t)$  when the temperature is lowered. Since  $\Delta(0)$  of MgB<sub>2</sub> as obtained from that study ( $2\Delta(0)/k_B T_c \approx 2.4$ , [8]) is well below the BCS value ( $2\Delta(0)/k_B T_c = 3.5$ ),  $\lambda_e^s$  - in this type of analysis - becomes larger in the SC state down to  $T/T_c \approx 0.2$ . Still,  $\lambda_l^s > \lambda_e^s$ . Taking  $\lambda_l^s$  as derived from the BCS-like gap, however, provides a slightly smoother crossover from the SC - to the normal state region of MgB<sub>2</sub>.

Panel (b) of figure 4 shows the temperature dependent thermal resistivity  $W_e^s \equiv 1/\lambda_e^s$ . Here, the data derived from the BCS model are used. Obviously, scattering of electrons by phonons in the SC state contributes just a fraction to the thermal resistivity  $W_e^s$ , and therefore  $W_{e,0}^s$  is the most significant term below  $T_c$ . This, of course, will not change if the actual dependence of  $\Delta(T)$  is considered.

## 4 Summary

The measurement of the thermal conductivity of MgB<sub>2</sub> reveals no pronounced anomaly at  $T = T_c$  and furthermore no local maximum occurs at temperatures well below  $T_c$ . Such a behaviour is most likely caused by the dominance of electron (hole) scattering on static imperfections present in the investi-



gated MgB<sub>2</sub> sample. In fact, an analysis based on the BRT model and the model of Geilikman evidences that  $W_{e,0}^s$  is the predominant term. Beside the classical BCS behaviour of the SC gap, an attempt was made to incorporate the actual gap behaviour reported for MgB<sub>2</sub> [8]. The subsequent analysis shows that the electronic contribution to the thermal conductivity becomes larger than in the former case. The application of the Wiedemann Franz law to the experimental data indicates that both the electronic - and the lattice contribution to the total thermal conductivity are almost of the same size over a large temperature range.

## 5 Acknowledgments

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## 6 Figure captions

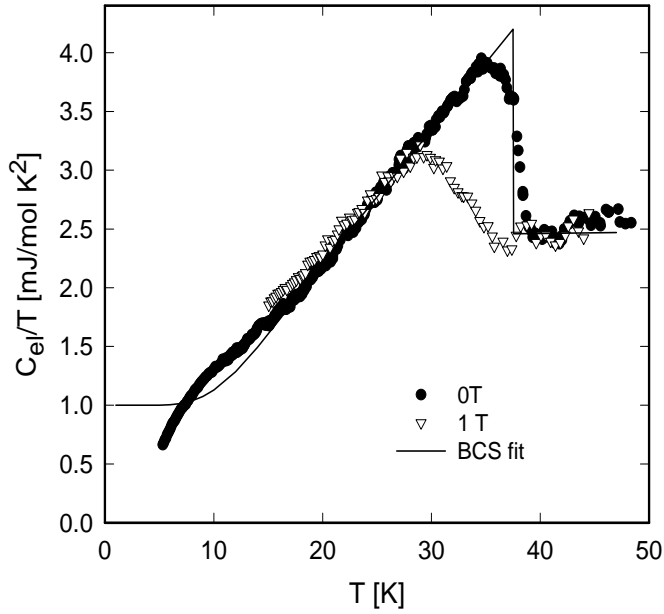


Figure 1: The electronic specific heat of MgB<sub>2</sub>,  $C_{el}(T)/T$  versus  $T$  obtained by subtracting the lattice contribution,  $C_{lat} = C^{9T} - \gamma T$ , where  $\gamma \simeq 2.4$  mJ/mol K<sup>2</sup>. The full line indicates a BCS temperature dependence of  $C_{el}$  as explained in the text.

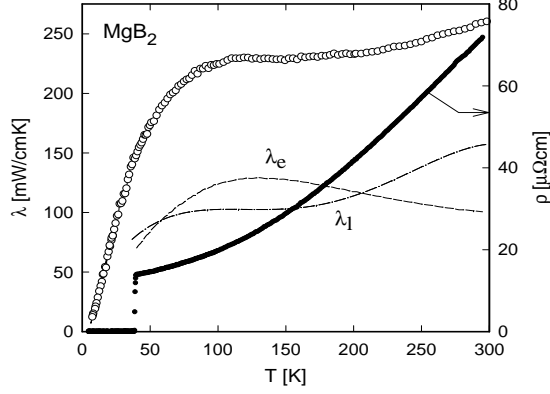


Figure 2: Temperature dependent thermal conductivity  $\lambda(T)$  (left axis), and electrical resistivity  $\rho(T)$  (right axis) of  $\text{MgB}_2$ . The dashed and the dashed-dotted lines are the electronic - and the lattice contributions  $\lambda_e$  and  $\lambda_l$ , respectively.

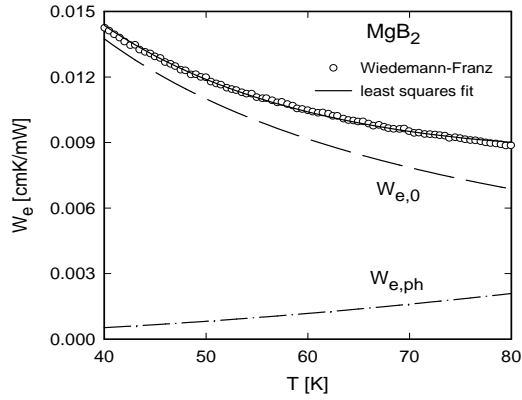


Figure 3: Temperature dependent electronic thermal resistivity  $W_e(T)$  of  $\text{MgB}_2$  in the normal state region. The dashed and the dashed-dotted lines are the contributions due to electron-imperfection - and electron-phonon scattering  $W_{e,0}$  and  $W_{e,ph}$ , respectively.

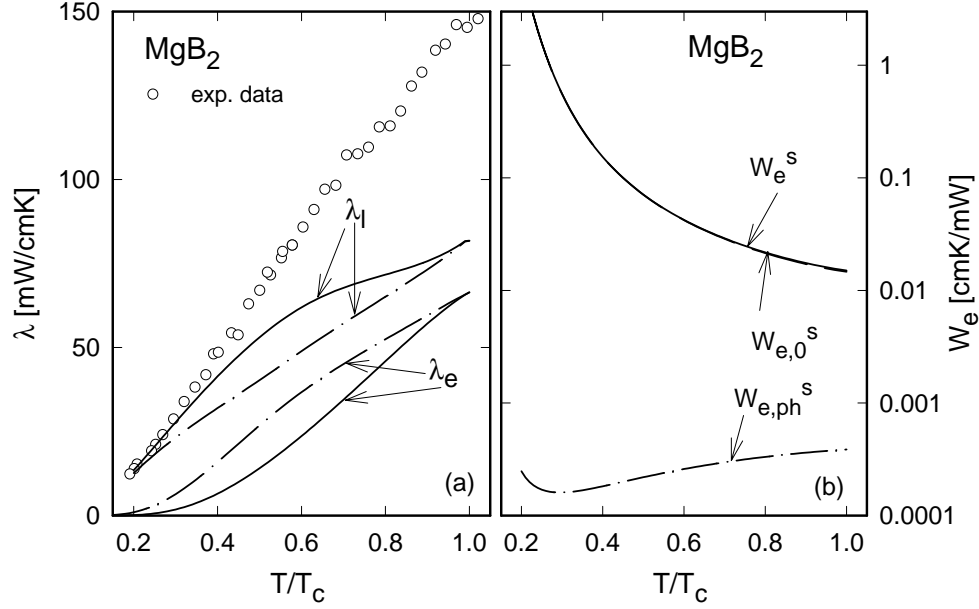


Figure 4: (a): Temperature dependent thermal conductivity  $\lambda(T)$  of MgB<sub>2</sub> in the SC state. The solid and the dashed-dotted lines represent electron- and lattice contributions  $\lambda_e^s$  and  $\lambda_l^s$ , derived from the BCS - and the modified model as discussed in the text, respectively. (b): Temperature dependent electronic thermal resistivity,  $W_e^s(T)$  of MgB<sub>2</sub> in the SC state derived from the BCS model. The dashed and the dashed-dotted lines are the contributions due to electron-imperfection - and electron-phonon scattering  $W_{e,0}^s$  and  $W_{e,ph}^s$ , respectively.