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Heat and charge transport properties of MgB_2

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Abstract

A polycrystalline sample of the MgB_2 superconductor was investigated by measurements of the electrical resistivity, the thermopower and the thermal conductivity in the temperature range between 1.8 and 300 K in zero magnetic field. The electrical resistivity shows a superconducting transition at $T_c = 38.7$ K and, similarly to borocarbides, a $T^{2.4}$ behaviour up to 200 K. The electron diffusion thermopower and its band-structure-derived value indicate the dominant hole character of the charge carriers. The total thermopower can be explained by the diffusion term renormalized by a significant electron–phonon interaction and a phonon drag term. In the thermal conductivity, for decreasing temperature, a significant decrease below T_c is observed resulting in a T^3 behaviour below 7 K. The reduced Lorenz number exhibits values smaller than 1 and a characteristic minimum which resembles the behaviour of non-magnetic borocarbides.

Keywords: Superconductivity; Thermopower; Thermal conductivity; MgB_2

1. Introduction

After the surprising discovery of superconductivity up to about 40 K in MgB_2 [1] extensive investigations of its physical properties have been performed. Special interest is focused on the electronic structure and in particular, on the type of the charge carriers, and their relationship to the superconducting pairing mechanism. Numerous studies are devoted to thermodynamic properties such as the specific heat [2–5] and the upper critical field [6–10].

However, there are less reports published on the transport properties of MgB_2 and only few on the heat transport. Results of previous investigations of the electrical resistivity ρ differ not only in the residual resistivity but also in the temperature dependence [6,11–13]. First measurements of the thermopower S [14–16] and the thermal conductivity κ [17] show a significant non-linearity in $S(T)$ in the temperature range close to room temperature and rather high values for the Lorenz number

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derived from the reported data of $\kappa(T)$ and $\rho(T)$. Such measurements are of general interest since they provide additional insight into the electronic structure and the electron–phonon (el–ph) interaction. In the present paper, zero magnetic field measurements of thermal and charge transport properties of MgB_2 are reported. Since at present the pairing mechanism has not been settled yet the comparison with related superconductors might be helpful to elucidate further details of the superconductivity in MgB_2. In this context also similarities and differences with the behaviour of well studied non-magnetic borocarbides will be discussed.

2. Experimental details

The experiments were performed on a polycrystalline sample of MgB_2 of about $5 \times 1.2 \times 1.2$ mm^3. It was cut from a pellet which was prepared by a conventional solid state reaction as described elsewhere [7]. The X-ray diffraction pattern of powder ground from this sample batch have shown that the material is single phased.

To investigate the thermopower S, two copper wires were fixed to the sample by an electrically conducting epoxy resin. The temperature gradient along the sample of about 1% of the temperature was generated by a small strain gauge heater. The temperature differences between the copper wires and between sample and cold copper plate were measured by two AuFe–Chromel thermocouples, the absolute temperature was detected by a germanium and at higher values by a platinum thermometer.

The thermal conductivity κ was measured by the standard steady-state method. For a first measurement up to 100 K the thermocouples were fixed directly to the sample by a low-temperature varnish. The same varnish was used to connect the sample with the cold copper plate. A second measurement, with a better contact by electrically conducting epoxy resin Eccobond 56C, was performed together with the investigation of S. No significant deviations between both results were found.

The electrical resistance was measured by the usual four probe method. Unfortunately, the attempt to fix additional current contacts to the sample prepared for the thermal conductivity measurement failed. Therefore, the electrical resistivity ρ was determined in a separate run, resulting in higher errors for the reduced Lorenz number because of the higher uncertainty in the distance between the voltage leads.

3. Results and discussion

As shown in Fig. 1, the resistivity of the investigated sample decreases from room temperature down to 40 K from a value of 38.2 to 7.1 μΩcm, i.e. the resistivity ratio RRR amounts to 5.4. According to the uncertainties of the cross-section of the sample and the distance between the voltage contacts, the error of the ρ values is about 20%; the uncertainty of RRR is much smaller. Müller et al. reported RRR = 4.5 for another sample cut from the same pellet [7]. A sharp superconducting transition is found at 38.7 K (midpoint value of the normal-state resistivity).

To analyse the temperature dependence of $\rho(T)$, the normal-state data below 200 K can be fitted to the expression

$$\rho(T) = \rho_0 + aT^b, \tag{1}$$

rather than to a Bloch–Grüneisen formula as proposed by Gasparov et al. [13]. Fig. 2 shows the

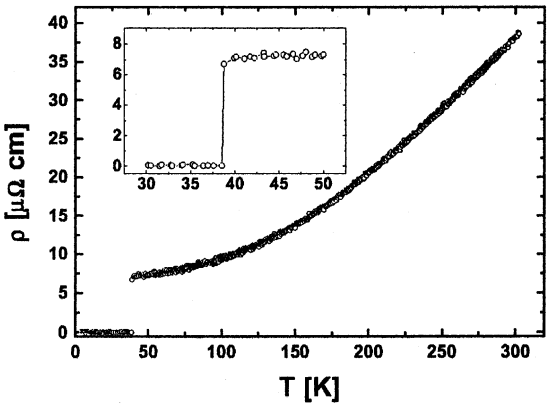

Fig. 1. Temperature dependence of the electrical resistivity ρ of MgB_2. The inset shows the range near T_c.

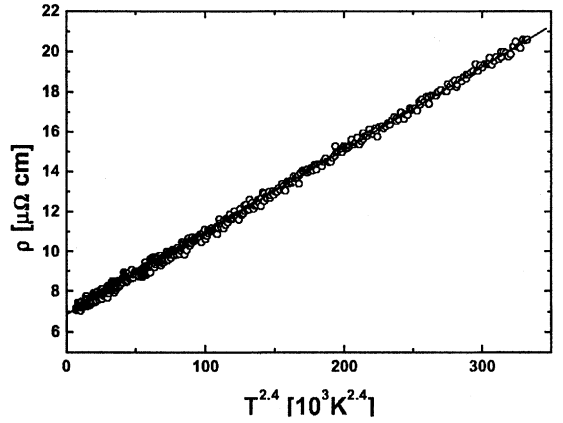

Fig. 2. Resistivity ρ of MgB_2 plotted as a function of $T^{2.4}$. The plotted range corresponds to $T_c < T < 200$ K.

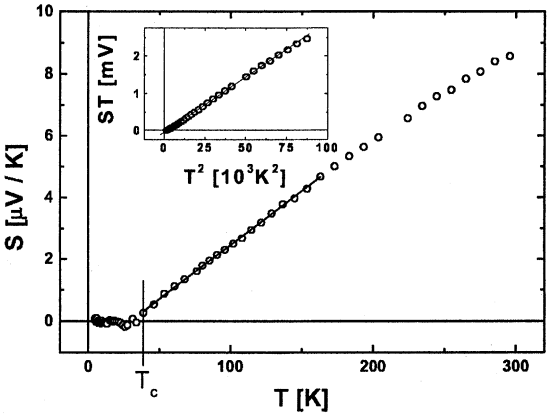

Fig. 3. Thermopower S of MgB_2 in the temperature range between 4 and 300 K. The straight line represents a linear fit to the data in the range 40 K $< T <$ 160 K. The inset shows the product of thermopower and temperature as a function of T^2 in the whole measured range. The straight line in the inset represents a fit according to Eq. (2) to the data in the range 70 K $< T <$ 270 K.

results in the temperature range between T_c and 200 K. The parameter values obtained from this fit are $\rho_0 = 6.8$ μΩ cm, $a = 3.3 \times 10^{-5}$ μΩ cm/K^b and $b = 2.4$. The obtained value of the exponent is in between the reported results $b = 2$ [12] and $b = 3$ [6] and in good agreement with $b = 2.6$ for a dense MgB_2 wire [11]. It is noteworthy that a similar behaviour was also found for other superconducting compounds. Rathnayaka et al. [18] reported exponents b of 2.2 and 2.0 for YNi_2B_2C and $LuNi_2B_2C$ single crystals, respectively. In the temperature range 200 K $< T <$ 300 K the curvature of $\rho(T)$ of MgB_2 decreases with increasing temperature and seems to follow the Bloch–Grüneisen formula.

The thermopower of MgB_2 is shown in Fig. 3. The room temperature value of 8.7 μV/K is in good agreement with the data of Lorenz et al. [14] who reported a value of about 8.3 μV/K for a MgB_2 sample with a RRR of about 3. Furthermore, the value for the slope of 0.036 μV/K^2, fitted to the measured data in the range 40 K $< T <$ 160 K, is close to the reported $dS/dT =$ 0.042 μV/K^2 below 160 K [14].

A quite smooth behaviour of the thermopower with a small jump of about 0.3 μV/K was found at the cross-over from the superconducting to the normal state. Lorenz et al. reported a higher jump of about 0.7 μV/K [14]. Further investigations of high quality very pure samples are required to clarify this issue.

The data can be described by the expression

$$S(T) = \frac{A}{T} + BT, \tag{2}$$

where A/T is the phonon drag term and BT the electron diffusion term. In the range 70 K $<$ $T <$ 270 K, for B a value of 0.031 μV/K^2 is observed. The positive sign of B is an indication of the hole character of the charge carriers [19–21]. For A, the fit yields a value of about -60 μV. For temperatures below 70 K a systematic deviation from the behaviour according to Eq. (2) is found since the expression A/T is valid only at the high-temperature side of the phonon drag peak. Above 270 K, a sublinear behaviour of ST vs. T^2 is observed as reported by Lorenz et al. [14]. The different magnitude of this deviation might be attributed to differences in the samples.

To get some microscopic insight into the magnitude of the second term of Eq. (2), we have also performed a theoretical calculation of the electronic part using local density approximation (LDA) band structure calculations (FPLO code [23]) and employing the well-known Mott formula, renormalized by the el–ph interaction coupling constant λ_{el-ph} [24]:

$$S_{\mathrm{el}} = \frac{\pi^2 k_{\mathrm{B}}^2 T}{3e} \left[\frac{\partial \ln \sigma(\varepsilon)}{\partial \varepsilon} \right]_{\varepsilon = E_{\mathrm{F}}} (1 + \lambda_{\mathrm{el-ph}}(T)). \qquad (3)$$

Here for the sake of simplicity only the energy dependence of the conductivity $\sigma(\varepsilon)$ in the relaxation time approximation has been taken into account ignoring a possible energy dependence of the scattering rates. Thus we obtain a value of about 2.8 μV/K $\times (1 + \lambda_{\mathrm{el-ph}})$ at room temperature. First of all the correct sign (which corresponds to a dominant hole contribution) should be noted. Then, adopting a strong el–ph interaction $\lambda_{\mathrm{el-ph}} \sim 2$ we would arrive approximately at the experimental results $\sim$8 μV/K. This is in qualitative accord with the intermediate to strong coupling scenario proposed in Ref. [10]. However, a more detailed investigation of each Fermi surface sheet and of the coupling to various phonon (boson) modes are required to extract quantitatively the strength of the el–ph interaction in a more reliable manner. In this context a possible relation of the (decreasing) temperature dependence of $\lambda_{\mathrm{el-ph}}(T)$ at high temperatures to the observed deviation from the behaviour according to Eq. (2) above 270 K is worth to be studied in more detail.

For YNi_2B_2C and $LuNi_2B_2C$ the electron diffusion term is smaller and negative: For YNi_2B_2C, a value of about -0.007 μV/K^2 [22] is reported. Furthermore, much higher negative phonon drag contributions in borocarbides have been found resulting in values for A between -450 and -550 μV [22]. Thus, the phonon drag contribution is less pronounced in MgB_2 than in YNi_2B_2C.

The results of the thermal conductivity measurements are presented in Fig. 4. The data taken in two separate runs as described above lie on top up of each other. Nevertheless, the error of the absolute value is about 20%, mainly caused by the uncertainties of cross-section and distance between thermometers.

The measured values of κ at 300 K are about 20% smaller than those reported by Bauer et al. [17] resulting from uncertainties in the measurements and possible differences in the samples. The positive slope of $\kappa(T)$ in the whole investigated temperature range indicates the limitation of the heat conductivity by crystal defects as in pure

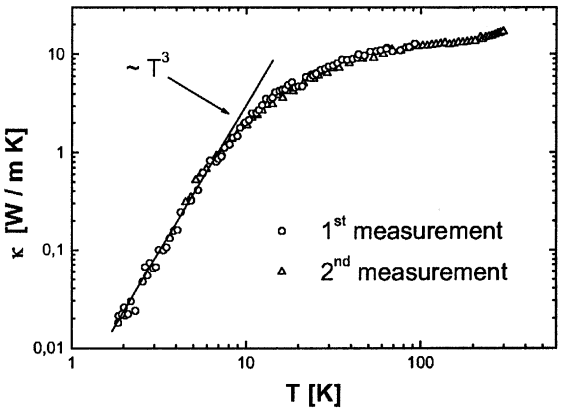

Fig. 4. Thermal conductivity κ of MgB_2 in the temperature range between 1.8 and 300 K in a double logarithmic plot. The straight line represents a T^3 fit to the data at low temperatures.

normal metals κ exhibits a maximum at lower temperatures and then decreases to a constant value with rising temperature.

No kink anomaly of κ could be detected at the superconducting phase transition as reported for the non-magnetic borocarbides [25,26]. The additionally observed peak below T_{c} in $\kappa(T)$ of these compounds should not be regarded as a generic intrinsic feature of clean superconductors since investigations of niobium samples exhibit such a peak only for medium clean samples (RRR $\approx$ 3000) while it disappears, if the sample quality is further increased to RRR $\approx$ 33000 [27]. Furthermore, the reduction of a possible peak is in agreement with the high phonon velocities in MgB_2 [26,28].

According to the decrease of the electronic thermal conductivity below T_{c} a significant change in the slope of $\kappa(T)$ is found. Below 7 K, where the influence of the electrons is negligible, the expected T^3 law for $\kappa(T)$ dominated by phonons at low temperatures is observed. With the measured value of $\kappa/T^3 = 2.9 \times 10^{-3}$ W/K^{4}m a mean free path l of the phonons at low temperatures can be calculated:

$$l = 3 \frac{\kappa(T)}{c(T)} \frac{1}{v} V_{\mathrm{M}}, \qquad (4)$$

where c is the specific heat, v the acoustic sound velocity, and V_{M} the molar volume. For the lattice contribution of the specific heat $c(T)$ a coefficient

$\beta = c/T^3 = 1.04 \times 10^{-5}$ J/K^{4}mol was reported [3]. Using the calculated values for the sound velocity of $v = 10\,600$ m/s for a longitudinal wave [28] and a molar volume of $V_M = 17.5$ cm^3/mol as derived from the unit cell volume of $V = 29.02$ Å^3 [29], Eq. (4) yields $l = 1.4$ μm. This length can be interpreted as an averaged grain size and is in agreement with the results of optical investigations of the pellets.

The reduced Lorenz number

$$\frac{L(T)}{L_0} = \frac{\kappa(T)\rho(T)}{L_0 T}, \tag{5}$$

where $L_0 = 2.44 \times 10^{-8}$ WΩ/K^2 is the Sommerfeld value, was derived from the measured values of $\kappa(T)$ and $\rho(T)$. The results are shown in Fig. 5. The error is about 30%.

At least for temperatures below 250 K the reduced Lorenz number is significantly smaller than the expected value of 1. Similar results shown in the inset of Fig. 5 were found for typical nonmagnetic borocarbides [25,26] and interpreted as the influence of inelastic scattering on the electronic thermal conductivity. The very small values of the Lorenz number confirm the dominating role of the electronic contribution to the heat transport.

Noteworthy, the shape of the Lorenz plots for MgB_2 and YNi_2B_2C/$LuNi_2B_2C$ is very similar.

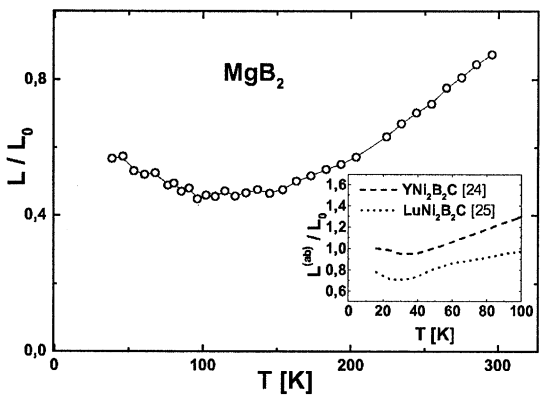

Fig. 5. Reduced Lorenz number L/L_0 of MgB_2 in the temperature range between 40 and 300 K. The inset shows the reduced Lorenz number of YNi_2B_2C and $LuNi_2B_2C$ single crystals of the in-plane heat and charge transport properties taken from Refs. [25,26].

Furthermore, the minima in the reduced Lorenz number occur at temperatures of about 120 and 35 K for MgB_2 and YNi_2B_2C/$LuNi_2B_2C$, respectively. These values correspond to 2–3T_c for that compounds.

Different from these results, the published data of electrical resistivity and thermal conductivity of Bauer et al. [17] yield reduced Lorenz numbers of up to about 2.5 at room temperature mainly caused by the high resistivity values. Their reported $\rho(300\text{ K}) \approx 70$ μΩcm is about twice as high as the value found here. Further investigations are required to clarify these differences in L. However, their reduced Lorenz number shows a similar behaviour with a minimum at about 120 K.

To summarize, in addition to a number of well-known similarities in the superconducting properties of MgB_2 and YNi_2B_2C, in the present work also similarities related to the normal-state transport properties have been found in the temperature dependences of the resistivity and of the reduced Lorenz number. However, the positive sign of the thermopower as observed in the measurement and derived from band structure calculations indicates a dominant hole contribution in MgB_2. The magnitude of the thermopower might indicate an intermediate to strong el–ph coupling scenario. The thermal conductivity strongly deviates from the behaviour expected for clean samples. The averaged grain size of the sample inferred from this measurement is about 1.4 μm.

After completion [30] of the present work, we have learnt about a preprint by Putti et al. [31]. Their resistivity data differ from ours by a significantly higher residual resistivity and a smaller RRR value. The data have been described by a generalized Bloch–Grüneisen formula. The raw thermopower data $S(T)$ looks very similar to ours. However, the interpretation is different. In the narrow interval 45 K $< T <$ 90 K $S(T)$ was fitted to a linear term S_{el} and a *positive* cubic one (i.e. the low-temperature approximation for the phonon drag contribution) which would point to predominant N (normal) scattering processes at low temperature. The high-temperature region was not quantitatively analysed. Our data can be analysed by those terms only in the range 55 K $< T <$ 90 K with clear deviations above 90 K and also *below* 55 K.

Anyhow, this should be compared with our fit (Eq. (2)) in a broader interval 70 K $< T <$ 270 K. It contains a negative high-temperature approximation for the phonon drag contribution which can be interpreted in terms of U (Umklapp) processes [20] and the relevance of soft modes clearly below the Debye energy. In this context the observation of low energy peaks at about 16 and 24 meV in recent neutron scattering [32] and 17 meV in Raman measurements [33] is of interest. Naturally, due to these different adopted approximations for the phonon drag terms with opposite signs, different dressed linear electronic diffusion terms $S_{el}/T = B = 0.0176$ μV/K^2 and $B = 0.031$ μV/K^2 have been derived. Since the band structure result for the bare S_{el}-term, calculated in the same approximation for the scattering rate as we did, seems to coincide with our result, different renormalizations due to many-body effects would be expected.

Furthermore, Muranaka et al. [34] reported a saturation of the thermopower near room temperature at a relatively low level of only 4 μV/K (compared with about 8 μV/K in our work or in Ref. [31]) and a reduced Lorenz number with a similar shape and a magnitude in between that of Bauer et al. [17] and that of the present work. All these different features mentioned above require further investigations especially with respect to the sample quality.

Acknowledgements

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