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Thermal transport properties of MgB_2

T. Muranaka,[1] J. Akimitsu,[1] and M. Sera[2]
[1]*Department of Physics, Aoyama-Gakuin University, Tokyo, Japan*
[2]*ADMS, Hiroshima University, Higashi-Hiroshima, Japan*
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The thermal transport properties and magnetic susceptibility of MgB_2 have been studied. The thermoelectric power with a positive sign shows a nearly T-linear increase up to ~150 K but shows a saturated behavior at higher temperatures, which suggests the existence of two types of charge carrier. The magnetic susceptibility above T_c shows a Pauli paramagnetism but shows a substantial T dependence which should include the information of the band structure of MgB_2. The thermal conductivity shows a suppression below T_c without a clear anomaly at T_c which is a result of the reduction of the normal charge carrier concentration below T_c.

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Since the recent discovery of the superconductivity of MgB_2 with $T_c = 39\,\mathrm{K}$,[1] extraordinary intensive studies have been performed to clarify the origin of the superconductivity. The crystal structure of MgB_2 is that of the hexagonal AlB_2 type, which consists of the hexagonal Mg layer and graphitelike B layer. The band structure calculation indicates that 30% of the density of states at the Fermi energy originates from the $2p_{x,y}$ bonding orbital of B in the B layer.[2] The covalency between B-B is so strong as to induce a large splitting between the bonding and antibonding bands.[2] In such a case, a strong electron-phonon interaction is expected in the B layer. This leads to the idea of the *s* wave electron-phonon mechanism of the superconductivity in this compound.[2] In fact, the substantial B isotope effect was observed.[3] The results of the tunneling spectroscopy,[4] the spin-lattice relaxation rate,[5] and photoemission[6] suggest the *s* wave superconductivity.

On the other hand, as for the normal state above T_c, there exist less reports. The electrical resistivity shows saturated behavior below ~150 K and an increase with increasing temperature above ~150 K. The spin-lattice relaxation rate exhibits a $T_1T = \mathrm{constant}$ behavior above T_c.[5] The sign of the thermoelectric power[7] and Hall coefficient[8] is positive, which is consistent with the prediction of the hole superconductivity. It is noted that although the Hall coefficient exhibits its anomalous temperature dependence,[8] detailed studies of the physical properties in the normal state have not yet been performed. It is important to perform such studies to clarify whether or not the normal state of MgB_2 is really normal.

In order to clarify if the normal state above T_c is really normal or not, we have investigated the temperature (T) dependence of the thermoelectric power and magnetic susceptibility of MgB_2. In order to clarify the heat conduction mechanism in the superconducting state, we have measured the thermal conductivity of MgB_2 in a temperature range between 4.5 and 250 K.

The sample was prepared as in Ref. 1. The thermoelectric power was measured by the usual dc method and the thermal conductivity by the conventional dc method. The dc magnetization was measured using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. All the measurements were performed on the sintered sample cut from the same ingot.

Figure 1 shows the temperature dependence of the thermoelectric power (S) of a MgB_2 sintered sample. The inset of Fig. 1 shows the temperature dependence of the electrical resistivity of the sintered sample. The residual resistivity ratio is approximately 3.5. The sign of S above T_c is positive as reported by Lorenz *et al.*[7] S shows a discontinuous increase at T_c and shows a nearly T-linear increase up to 150 K. Near 150 K, a bending with a negative curvature is observed and a saturated behavior is observed at higher temperatures. The T dependence up to ~150 K is consistent with that reported by Lorenz *et al.*[7] The high temperature saturated behavior was not observed clearly in the result by Lorenz *et al.*[7] but their data also show a deviation above ~150 K from the low temperature T-linear behavior. The T-linear behavior is usually observed in the conventional metallic compounds and its slope is proportional to $1/\epsilon_F$. In the present case, this slope is three or four times larger than those of the noble metals such as Cu and Au, etc. Assuming that the T-linear behavior is that of a metallic compound, the present result suggests that ϵ_F of MgB_2 is roughly $1 \sim 2\,\mathrm{eV}$. However, it is not consistent with a large carrier concentration of $\sim 10^{23}\,\mathrm{cm}^3$ estimated from the Hall coefficient (R_H) assuming a single carrier.[8] It is reported that the T

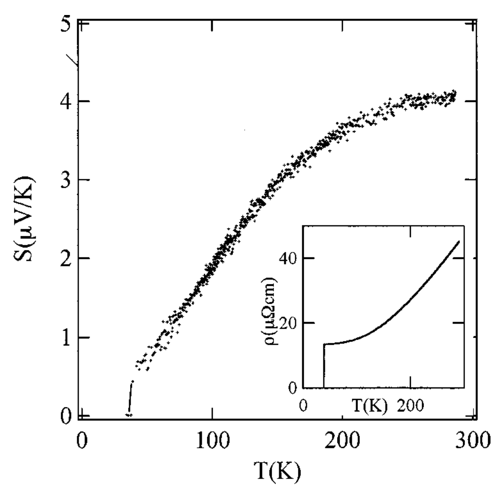

FIG. 1. Temperature dependence of the thermal conductivity of MgB_2. The inset shows the temperature dependence of the electrical resistivity of MgB_2.

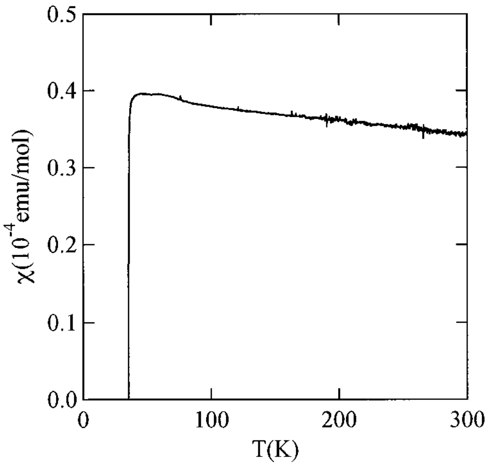

FIG. 2. Temperature dependence of the magnetic susceptibility of MgB_2 measured at $H=1$ T.

dependence of R_H changes below and above ~150 K. R_H shows an increase with decreasing temperature below ~150 K and is nearly constant above ~150 K. The temperature of ~150 K where the anomalies are observed in S and R_H seems to correspond to the temperature where $d\rho/dT$ changes. Thus, the temperature dependences of all the transport properties mentioned above change form at $T\sim150$ K. Here, we note that the carrier concentration of $\sim10^{23}/\mathrm{cm}^3$ estimated from R_H may be too large for the conventional metallic compounds. This strongly suggests that there exist two types of carriers, i.e., hole and electron. Quite recently, the importance of the existence of the different types of charge carriers was discussed by Voelker *et al.*[9] The fact that the T dependences of R_H and S show the anomaly at $T\sim150$ K suggests that the balance of the conduction by hole and electron is changed at ~150 K. When there exist two types of carriers, both R_H and S are dominated by the carrier with a larger electrical conductivity. The results of R_H and S suggest that the hole conduction is dominant below ~150 K but the electron conduction becomes comparable with the hole conduction.

In order to check if the anomaly is observed at $T\sim150$ K in the normal state, we have measured the magnetic susceptibility of a MgB_2 sintered sample at $H=1$ T. The result is shown in Fig. 2. The magnetic susceptibility shows a Pauli paramagnetism, as expected. Although an anomaly is not seen at $T\sim150$ K, it shows a substantial decrease with increasing temperature. Such a temperature dependence may be due to a less than large Fermi energy in this compound. This result includes the information on the band structure. It is interesting to investigate the magnetic susceptibility of $Mg_{1-x}Al_xB_2$ in the normal state where a larger temperature dependence is expected because the electron filling by Al doping is expected to reduce the Fermi energy.[10,11] The detailed study in the normal state is important to understand the origin of the high T_c of MgB_2.

Figure 3 shows the T dependence of the thermal conductivity (κ) of a MgB_2 sintered sample below 60 K. The insets (a) and (b) of Fig. 3 show those up to 10 K and 250 K, respectively. The reduced Lorentz number L/L_0 is 1.45, 1.15, 1.09, 1.20, and 1.36 at $T=40$, 100, 150, 200, and 250 K, respectively. $L_0=24.5\,\mathrm{nW}\,\Omega\,\mathrm{K}^{-2}$. A broad minimum is formed at $T\sim150$ K. This suggests that the phonon contribution to the heat conduction increases with decreasing temperature below 150 K and with increasing temperature above 150 K. The log-log plot of Fig. 4 illustrates this. The inset of Fig. 4 shows κ as a function of T_c/T. κ shows a suppression below T_c. Such a T dependence in different from the enhancement below T_c in the high-T_c cuprate which is observed independent of the sample quality.[12] κ of YNi_2B_2C with $T_c=16$ K shows a suppression below T_c accompanied with a clear kink at T_c.[13] κ of the present sample does not show a clear anomaly but a smooth shoulder at T_c. In the early stage of the high-T_c cupurate, the enhancement of κ below T_c was attributed to the enhancement of the phonon thermal conductivity.[14] Such an enhancement is expected to take place when the phonon is strongly scattered by the

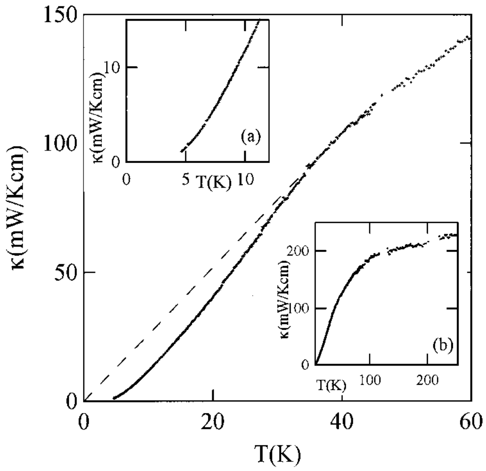

FIG. 3. Temperature dependence of the thermal conductivity of MgB_2 below 60 K. The dashed straight line is a guide to the eye. The insets (a) and (b) show those up to 10 K and 250 K, respectively.

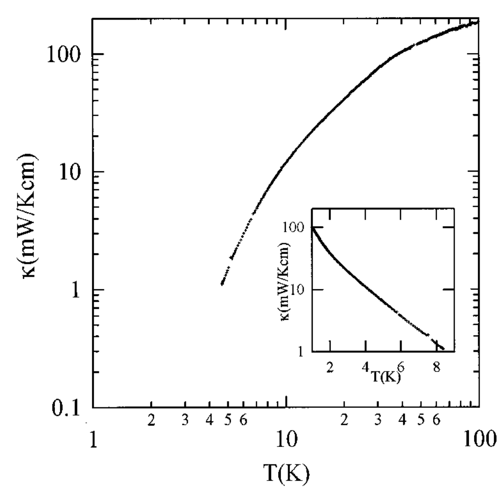

FIG. 4. Temperature dependence of the thermal conductivity of MgB_2 by a log-log plot. The inset shows the thermal conductivity of MgB_2 as a function of T_c/T.

charge carrier above T_c. In such a case, the concentration of charge carrier is decreased by forming a Cooper pair and the mean free path of the phonon becomes longer. However, the above explanation was denied by the discovery of the thermal Hall conductivity where the temperature gradient is induced perpendicular to both thermal current and applied magnetic field.[15] In the case of YNi_2B_2C, the enhancement of κ was observed below $\sim T_c/3$, which was attributed to the enhancement of the phonon thermal conductivity as is discussed above. The magnitude of the enhancement of the phonon thermal conductivity below T_c depends on the purity of the sample. The observation of the enhancement of κ below $\sim T_c/3$ in YNi_2B_2C is due to a rather small residual resistivity of $\sim 2\ \mu\Omega\,\mathrm{cm}$. In the present sample, such an enhancement of κ is not seen below T_c and no anomaly is seen at T_c. This indicates that the impurity or defect scattering of the charge carrier and phonon is large in the present polycrystalline sample. If there exists only the contribution from the charge carrier, κ is expected to decrease below T_c due to a reduction of a normal charge carrier concentration as a result of the Cooper pair formation. Then, κ is expected to exhibit the exponential T dependence with an energy gap in the case of the s wave superconductivity. If we estimate the energy gap (Δ) below $\sim$6 K by fitting the data to a semi-log plot of κ vs. $1/T$, Δ is estimated to be one-third of the BCS value. Such an unreasonable value of Δ indicates that the phonon thermal conductivity is substantial at this temperature range. At present, it is difficult to separate the contribution to the heat conduction from the charge carrier and phonon. The investigation using a higher quality sample is necessary. At high temperature above 100 K, κ shows a small increase with increasing temperature. The overall T dependence of κ is that of a conventional metal and the phonon contribution is expected to be small. A small increase of κ above 100 K is possibly due to the small increase of the phonon contribution which originates from the growing increase of a phonon specific heat irrespective of the decrease of the phonon lifetime with increasing temperature.

In conclusion, we have investigated the thermal transport properties and magnetic susceptibility of MgB_2. The thermoelectric power with a positive sign shows a nearly T-linear increase up to $\sim$150 K, as is expected for the conventional metallic compounds but shows a saturated behavior at higher temperatures. This suggests the existence of two types of charge carrier. The magnetic susceptibility above T_c shows a Pauli paramagnetism but shows a substantial T dependence which may include the information of the band structure of MgB_2. The thermal conductivity shows a suppression below T_c without a clear anomaly at T_c, being different from the enhancement below T_c observed in the high-T_c cuprate. The suppression of κ below T_c is a result of the reduction of the normal charge carrier concentration below T_c.

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