Hole carrier in MgB$_2$ characterized by Hall Measurements

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The longitudinal resistivity $\rho_{xx}$ and Hall coefficient $R_H$ were measured for MgB$_2$ sintered under high pressure. We found that $R_H$ is positive like cuprate high-$T_c$ superconductors, and decreases as temperature increases for 40 K $< T < 300$ K. The cotangent of Hall angle was found to follow $a + bT^2$ behavior from $T_c$ to 300 K. At $T = 100$ K, $R_H = 4.1 \times 10^{-11}$ m$^3$/C from which hole carrier density was determined to be $1.5 \times 10^{21}$/cm$^3$. This carrier density is 2 - 3 orders of magnitude larger than those of Nb$_3$Sn and optimally doped YBa$_2$Cu$_3$O$_y$ superconductors.

Recently, MgB$_2$ was found to be metallic superconductor with transition temperature ($T_c$) of about 40 K, and has provided great scientific interest. Several thermodynamic parameters have been estimated, such as a upper critical field $H_{c2} = 13 - 18$ T, a Ginzburg-Landau parameter $\kappa \sim 26$, and the critical supercurrent density $J_c(0) \sim 10^5$ A/cm$^2$. In order to probe the nature of gap, tunneling spectroscopy measurements have been reported, and they observed superconducting energy gap ($\Delta$) of 5 - 7 meV in the framework of the BCS model. The conventional BCS electron-phonon interaction was proposed as the origin of the superconductivity based on a band calculation. The possible origin of the enhanced $T_c$ is suggested to originate from a strong electron-phonon interaction and a enhanced phonon frequency due to the light boron mass in MgB$_2$. Most of the charge carrier density at the Fermi level comes from the boron band. Indeed, the boron isotope effect has been reported with an exponent of $\alpha_B \sim 0.26$. No experimental study on the electronic structure has been reported yet.

Another interesting feature concerning the normal-state Hall effect in high-$T_c$ cuprates is the universal temperature dependence of the cotangent of the Hall angle ($\cot \theta_H$). Anderson and Chien et al. have proposed that the charge transport is governed by two different scattering times with different temperature dependences. In this model, the $\cot \theta_H$ should be proportional to $T^2$ since the Hall angle is proportional to the inverse of the Hall scattering time $\tau_H \propto T^{-2}$, and has been observed for most high-$T_c$ superconductors. In the mixed-state, the flux-flow Hall effect is also quite interesting. A puzzling sign anomaly has been observed in some conventional superconductors as well as in most of high-$T_c$ superconductors. Even double or triple sign changes have been observed in some high-$T_c$ superconductors. Furthermore, a universal scaling behavior between the Hall resistivity and the longitudinal resistivity has attracted much experimental and theoretical interest. However, these Hall effects in the mixed state are not well understood.

To understand the superconductivity in MgB$_2$, it is essential to know the type of charge carrier and its density, but these have not been reported yet. Theoretically, Hirsch proposed that the 40 K superconductivity of MgB$_2$ originates mainly from the hole carriers with boron planes acting like the CuO$_2$ planes in cuprate high-temperature superconductors. He proposed that pairing of hole carriers leads to hole undressing, which is driven by Coulomb interactions. To the best of our knowledge, the Hall coefficient ($R_H$) for MgB$_2$ has not been reported.

To obtain reliable results from transport measurements by using polycrystalline samples, one must make the sample strong and dense. In this case, samples sintered under high pressure suitable. In our previous report, we showed that the mechanical properties, as well as the superconducting properties, were vastly enhanced for samples sintered at 950 °C under high pressure (3 GPa range).

In this paper, we report the first measurement of the $R_H$ of MgB$_2$, which was carried out using carefully prepared sample with a thin bar shape. We found that the sign of $R_H$ is positive like those of cuprate high-$T_c$ superconductors. This is contrary to most other metal diborides with the same structure as MgB$_2$. Also the $R_H$ decreased as the temperature increased, and the cotangent of the Hall angle follows $a + bT^{1.8}$ for most of the measured temperature region from 40 to 300 K. At $T = 100$ K, $R_H = 4.1 \times 10^{-11}$ m$^3$/C, and the calculated hole carrier density is $1.5 \times 10^{21}$/cm$^3$.

The polycrystalline samples (4.5 mm in diameter and 3.3 mm in height) used in this study were sintered at 950 °C under 3 GPa. The fabrication method was reported in detail by Jung et al. The sample purity was more than 99% as determined by X-ray diffraction analysis. No grain boundaries were observed using the scanning electron microscopy. In order to obtain a higher Hall voltage signal, we cut the sample into a bar shape with a length.
of 4 mm and a width of 2.4 mm, and then mechanically polished it until it was very thin (50 - 100 µm). The standard photolithography technique was adopted to align the electrical pads shown in the upper inset of Fig. 1. To obtain good ohmic contacts (<1 Ω), we coated Au film on contact pads after cleaning the sample surface with an Ar ion beam. This process was done in situ in a high vacuum chamber. The voltage noise, which is detrimental to precise measurements, was successfully reduced to a lower level by preparing a very thin and optically clean specimen polished from a strong, dense samples. After installing a low noise preamplifier (N11, EM Electronics) prior to the nanovoltmeter (HP 34420A), we achieved a voltage resolution of below 1 nV under a bias current of 50 -100 mA. Fine temperature control was crucial since the Hall signal was very small. The longitudinal and Hall voltages were measured simultaneously by using the standard dc 6-probe method. The magnetic field was applied perpendicular to the sample surface by using a superconducting magnet system (Maglab2000 Oxford Ltd.) and the applied current density was ~ 42 A/cm². The Hall voltage was extracted from the antisymmetric parts of the transverse voltages measured under opposite directions to remove the longitudinal component due to the misalignment of the Hall voltage pads. The Hall voltage was found to be linear in both the current and the magnetic field.

Fig. 1 shows the temperature dependence of the longitudinal resistivity, \( \rho_{xx} \). The low-field magnetization in the zero-field-cooling state for the original bulk sample is shown in the lower inset of Fig. 1. The diamagnetism is 100% to almost \( T_c \); thus we normalized to the value at lower temperature. The superconducting transition temperature is 38.4 K with a narrow transition width of \( \sim 0.6 \) K, as judged from the 10 to 90% superconducting transition. The resistivity value of \( \rho \sim 70 \) µΩcm at 300 K is comparable to that of single crystalline intermetallic superconductors. As reported in our previous work, the normal-state \( \rho_{xx} \) follows a \( T^2 \) behavior rather than a \( T^3 \) behavior, for the entire temperature region below room temperature. No magnetoresistance was observed from \( T_c \) to 300 K, which is consistent with the previous results by Jung et al. and Takano et al., but different from the data by Finnemore et al.

The temperature dependence of the Hall coefficient is shown in Fig. 2. The two curves in the inset represent the Hall voltage measured at 100 K for opposite magnetic fields up to 5 T. The clearly symmetric and linear shape demonstrates that the signal to noise ratio for our measurement is high. The Hall coefficient was positive for all temperatures above \( T_c \). At 100 K, \( R_H = 4.1 \times 10^{-11} \) m²/C, and the hole carrier density was calculated to be \( 1.5 \times 10^{23} \) /cm³. The absolute value of the hole carrier density is two orders of magnitude larger than that of NbSn superconductors and nearly three orders of magnitude larger than that of optimally doped YBa₂Cu₃O₆. [1]

Hirsch offers an explanation based on a universal mechanism by assuming that superconductivity in MgB₂ is similar to that in cuprate superconductors and is driven by pairing of heavily dressed hole carriers in a band that is almost full, whereby they gain enough kinetic energy to overcome the Coulomb energy. [2] Based on this assumption, he claimed that the type of the charge carrier is positive.

In Fig. 3, we show the temperature dependence of \( \cot \theta_H \) at 5 T. A good linear fit to \( a + b T^{1.8} \), rather than \( a + b T^2 \), is observed for the temperature range from \( T_c \) to 300 K. In high-\( T_c \) cuprates, the charge transport is governed by two different scattering times with different temperature dependences. According to this two-scattering-time model, the longitudinal conductivity (\( \sigma_{xx} \)) is governed by the transport scattering time \( \tau_r \), which is proportional to \( 1/T \), whereas the Hall conductivity (\( \sigma_{xy} \sim \tau_H \tau_r \)) follows \( 1/T^3 \) since the Hall relaxation rate is proportional to \( 1/T^2 \). As a result, the \( \cot \theta_H = \sigma_{xy}/\sigma_{xx} \) should follow an \( \sim T^2 \) law. Our data is in fair agreement with a \( \cot \theta_H \sim T^2 \) law, which is consistent with the observations in most high-\( T_c \) superconductors. However, this observation cannot be explained by the above model because the data show \( \rho_{xx} \sim T^2 \), as shown in Fig. 1.

In summary, we report the temperature dependence of \( R_H \) for the recently discovered binary superconductor MgB₂ which has a remarkably high transition temperature. We find that \( R_H \) is positive like those for cuprate high-\( T_c \) superconductors and that the \( \cot \theta_H \) follows \( a + b T^2 \) from \( T_c \) to 300 K. At \( T = 100 \) K, \( R_H = 4.1 \times 10^{-11} \) m²/C, and the hole carrier density was \( 1.5 \times 10^{23} \) /cm³. We discussed the implication of the hole superconductivity based on a recent model.

**ACKNOWLEDGMENTS**

We appreciate valuable discussion with J. L. Tallon. This work is supported by the Ministry of Science and Technology of Korea through the Creative Research Initiative Program.

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FIG. 1. $\rho_{xx}$ - $T$ shows overall $T^2$ behavior and a sharp transition near $T_c$ for MgB$_2$ sample. The lower inset shows the low-field magnetization curve measured in the zero-field-cooling state and the upper inset shows the configuration of the measurement, namely six-terminal method. The two electrical pads at both sides are for current path and the other four leads for longitudinal and transverse voltage measurement.

FIG. 2. Hall coefficient measured at 5 T. The two lines in the inset represent the Hall voltage measured at 100 K for opposite two directions of the applied field up to 5 T.
FIG. 3. Cotangent of Hall angle measured at 5 T. The curves show nearly $T^{1.8}$ behavior over the entire temperature region measured.