Temperature- and magnetic-field-dependent resistivity of MgB₂ sintered at high temperature and high pressure condition

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We report the temperature- and magnetic-field-dependent resistivity of MgB₂ sintered at high temperature and high pressure condition. The superconducting transition width for the resistivity measurement was about 0.4 K, and the low-field magnetization showed a sharp superconducting transition with a transition width of about 1 K. The resistivity in the normal state roughly followed \( T^2 \) behavior with smaller residual resistivity ratio (RRR) of 3 over broad temperature region above 100 K rather than reported \( T^3 \) behavior with larger RRR value of \( \sim 20 \) in the samples made at lower pressures. Also, the resistivity did not change appreciably with the applied magnetic field, which was different from previous report. These differences were discussed with the microscopic and structural change due to the high-pressure sintering.

I. INTRODUCTION

Very recently, superconductivity at about 40 K was discovered in MgB₂. Conventional BCS superconductivity has been proposed for this compound and due to the enhanced phonon frequency from the light ionic masses, the superconducting transition temperature is expected to be high. A shift in the \( T_c \) due to the boron isotope has been reported with an isotope critical exponent of \( \alpha_B \sim 0.26 \). In addition, several thermodynamic parameters have been measured in this compound and these include a upper critical field \( H_{c2} \), its slope \( dH_{c2}/dT \), the Ginzburg-Landau parameter \( \kappa \), zero-temperature coherence length \( \xi(0) \), and penetration depth \( \lambda(0) \) etc. It is quite interesting that these thermodynamic parameters are nearly the same as those for Sr\(_{0.5}\)La\(_{0.1}\)CuO\(_2\) whose infinite-layer structure consisting of a conducting plane (CuO\(_2\)) and a metallic spacer layer (Sr,La) is quite similar to the structure of MgB\(_2\).

The type of carrier was predicted to be positive with boron planes acting like the CuO\(_2\) planes in cuprate high-temperature superconductors, which was confirmed by a Hall measurement by us. With high carrier density (>10\(^{23}\) cm\(^{-3}\)) revealed by the Hall measurement, the material have been reported to have characteristic metallic transport behaviors. The transport measurements for MgB\(_2\) synthesized at lower pressures (\( p \ll 1 \) GPa) showed that a residual resistivity ratio between 300 K and 40 K was more than 20 with room temperature resistivity of about 10 \( \mu \Omega \text{cm} \), and the resistivity value increases by several tens of percent in the magnetic field of 5 Tesla, and overall temperature dependence followed a \( T^3 \) behavior in the normal state. However, samples made at lower pressures were reported to be rather porous and mechanically weak, thus the concrete transport properties should be established for compact form of samples especially for device application.

In this paper we report the physical properties of hard and dense MgB\(_2\) sintered at high temperature and high pressure (\( p \sim 3 \) GPa). This sample was strong enough to prepare an optically clean surface for the reflectivity measurement by polishing. We found that \( T_c \) onset decreased by about 0.5 K due to the high-pressure sintering and overall temperature dependence of the resistivity in the normal state followed a \( T^2 \) and residual resistivity ratio was less than 3. And near \( T_c \), the change of resistivity with magnetic field up to 5 Tesla All these resistivity behaviors were different from those for samples made at lower pressures. These differences were discussed with the microscopic and structural changes caused by the high-pressure sintering.

II. EXPERIMENTAL

High-pressure sintering was performed with a 12-mm cubic multi-anvil-type press. Commercially available powder of MgB\(_2\) (Alfa Aesar) was used to make pellets. The pellets were put into a Au capsule in a high-pressure cell. One group of pellets were pressurized up to 3 GPa without subsequent heat treatment, to make a ‘cold-pressed’ sample(CP-sample) for resistivity measurement and the other group of the pellets was heated after pressurization to make a ‘hot-pressed’ sample(HP-sample). A D-type thermocouple was inserted near the Au capsule to monitor the temperature. It took about 2 hours to pressurize the cell to 3 GPa. After the pressurization, the heating power was increased linearly and then maintained constant for 2 hours. The sample was sintered at a
temperature of 850 ~ 950°C and then quenched to room temperature. The weight of the sample obtained in one batch was about 130 mg, and the size was about 4.5 mm in diameter and 3.3 mm in height.

A SQUID magnetometer (Quantum Design, MPMSXL) was used to measure the low-field magnetization of the samples. A scanning electron microscope (SEM) was used to investigate the surface morphology. For the resistance measurement, we cut the HP-sample by using a diamond saw with a coolant and then polished it into a rectangular solid shape with dimension. The resistance curve, $\rho(T)$, was measured using the standard 4-probe technique.

III. DATA AND DISCUSSION

Figure 2 shows the normalized magnetic susceptibility, $\frac{4\pi}{M}\chi(T)$, from the measured low-field magnetization data for two kinds of MgB$_2$. The curve with the broad transition is for as-purchased powder, the other curve with the symbols is for the HP-sample. The magnetic susceptibility data show that the superconducting transition width decreased from about 10 K to 1 K after the high-pressure sintering. The decreased field-cooling signal in the magnetic susceptibility for HP-sample indicates that the flux pinning is greatly enhanced and suggests a higher possibility of high current superconducting applications in the compact bulk form, which was also verified from the bulk pinning behaviors in the magnetic hysteresis $M(H)$.

The transition temperature of the HP-sample was about 37.5 K, was slightly lower than 38 K for the as-purchased powder.

Figure 3 shows SEM pictures for both samples. In Fig. 3 (a), the grain size of the CP-sample is much less than 1 µm. The grains in the HP-sample are well connected as shown in Fig. 3 (b). We cannot even distinguish the grain boundaries over wide regions. The microscopic connections between the grains may be the reasons for HP-sample being strong and dense macroscopically.

The above resistivity behaviors for HP-samples sintered at high pressure $p \gg 1$ GPa were also observed by other group thus might be somehow intrinsic. Most probable extrinsic origin for the different resistivity behaviors for samples made at higher pressures would be the appearance of inter-grain impurities during the high-pressure sintering which wrap around the grains and block the inter-grain current transport. However our recent study using a high-resolution transmission electron microscope showed that the impurities were well isolated from the major MgB$_2$ phases, not forming the inter-grain layer. Then the strong connectivity of the grains without inter-grain impurity might suggest that intra-grain contribution itself dominates the different resistivity behavior. The lattice parameters directly obtained from high resolution TEM images were the nearly same as those of lower-pressure samples measured near 3 GPa. Thus the strain caused by the high-pressure seems to remain appreciably after the sintering, which could answer partly the different transport behavior in some way. The observed difference of $T_c$ between HP-sample and CP-sample was less than about 0.5 K, which is much smaller than reported $T_c$ reduction by 4.8 K upon applying 3 GPa. The different $T_c$ reduction may be partly due to different pressurization method and/or the different distribution of strains inside the sample. The identification of the exact origin for the different resistivity behaviors still needs more studies.

IV. SUMMARY

In summary, we report the temperature- and magnetic-field-dependant resistivity of a hard and dense MgB$_2$ sintered at high temperature and high pressure ($p \sim 3$ GPa). The superconducting transition width for the resistivity measurement was about 0.4 K, and the resistivity in the normal state followed a behavior of $T^2$. The absolute values of the resistivity at room temperature and at just above $T_c$ were 50 to 21 µΩcm respectively. Also, the resistivity in the normal state did not change appreciably with the applied magnetic field up to 5 T. These behaviors are different from those for samples made at lower pressures and maybe are partly due to the presence of strain caused by high-pressure condition for the sintering.
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FIG. 1. Normalized magnetic susceptibility from the low-field magnetization $M(T)$ of MgB$_2$. $M(T)$ for zero-field-cooling and field-cooling states were measured at 20 Oe. The curve with a broad transition was for the as-purchased powder (50 mg), and the curve with a sharp transition was for the HP-sample (120 mg).

FIG. 2. Resistivity of HP-sample of MgB$_2$. Symbols denote the data and the two lines are fitting lines. Above $T_c$, the temperature dependence of the resistance followed a $T^2$ behavior (solid line through the data) rather than reported previously $T^3$ behavior (dashed line). The transition width was about 0.4 K. The inset shows the dependence of the resistivity on the external magnetic field. This dependence is different from the previous results that the resistivity near $T_c$ doubles upon applying high magnetic field $H = 9$ T.
FIG. 3. SEM pictures of (a) the CP-sample and (b) the HP-sample which is a heat treated MgB$_2$ at 3 GPa. The scale bars indicate 1 µm for both pictures.