The increase in T_c for MgB_2 superconductor under high pressure

2002 J. Phys.: Condens. Matter 14 11301

(http://iopscience.iop.org/0953-8984/14/44/471)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **14** (2002) 11301–11304 PII: S0953-8984(02)39619-X

The increase in T_c for MgB_2 superconductor under high pressure

Z-X Liu, C-Q Jin, J-Y You, S-C Li, J-L Zhu, R-C Yu, F-Y Li and S-K Su

Laboratory for Extreme Condition Physics, Institute of Physics, and Beijing High Pressure Research Centre, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

Received 1 June 2002
Published 25 Octobr 2002
Online at stacks.iop.org/JPhysCM/14/11301

Abstract

We report *in situ* high-pressure studies up to 1.0 GPa on MgB_2 superconductor which had been synthesized at high pressure. The as-prepared sample is of high quality as regards having a sharp superconducting transition (T_c) at 39 K. The *in situ* high-pressure measurements were carried out using a Be–Cu piston–cylinder-type instrument with a mixed oil as the pressure-transmitting medium, which provides a quasi-hydrostatic pressure environment at low temperature. The superconducting transitions were measured using the electrical conductance method. It is found that T_c increases with pressure in the initial pressure range, leading to a parabolic-like T_c–P evolution.

1. Introduction

The discovery of 39 K superconductivity in MgB_2 [1] has raised interest in the intermetallic compounds considerably. MgB_2 has the highest T_c ever reached in an intermetallic compound. MgB_2 crystallizes into the so-called AlB_2 structure which forms honeycomb-like B layers alternating with Mg layers as shown in figure 1. Generally, it is believed that MgB_2 is a BCS-like conventional superconductor but with unusual phonon–electron structures, which consequently may give rise to the high T_c of binary boride. Theoretical works indicate that the electronic structure at the Fermi level is primarily of boron character, e.g. B $\sigma(\mathrm{sp}_x\mathrm{p}_y)$ is believed to offer hole-like conducting carriers while Mg^{2+} lowers the nonbonding B $\pi(\mathrm{p}_z)$ causing $\sigma \rightarrow \pi$ charge transfer [2]. MgB_2 can be made at ambient conditions, but high-pressure synthesis has proven to be much effective as regards getting high-quality bulk samples (sharp transition, improved intergrain links and higher critical current density) [3, 4]. As an effective way to investigate modulation of the transition temperature and to test the theoretical prediction, *in situ* high-pressure experiments were performed by several groups [5]. They all found that the transition temperature of MgB_2 decreases with increasing pressure, which implies a crucial role for the phonon-mediated pairing mechanism in the MgB_2 superconductor. $\mathrm{d}T_c/\mathrm{d}P$ at lower pressure (∼2.0 GPa) was found to be -1.0 to -2.0 K GPa^{-1}. This is in agreement with the expectations within the traditional framework of BCS theory, where the

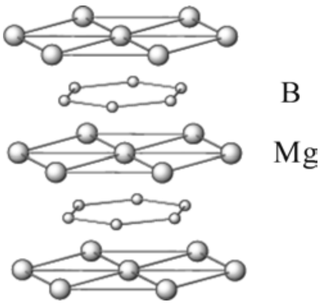

Figure 1. The crystal structure of MgB_2.

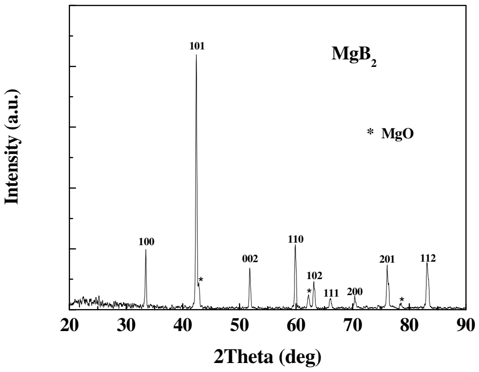

Figure 2. The powder x-ray diffraction pattern of the MgB_2 sample synthesized under high pressure.

stiffening phonon frequency will decrease T_c. The theoretical calculation indicates that it is a combination of increasing phonon frequency and decreasing electronic density of states at the Fermi level which leads to the observed decrease of the critical temperature under pressure within the context of BCS theory [6, 7]. However, Hirsch *et al* [8] proposed a novel hole superconducting mechanism which predicts the increase of T_c with pressure in MgB_2. Here we report experimental results of *in situ* high-pressure studies of the change in T_c for MgB_2 superconductor which had been synthesized at high pressure directly from the elements.

2. Experimental details

The MgB_2 sample was directly synthesized from the elements Mg and B under high pressure. Mg (99% purity) and B (5 N purity, 300 mesh) powders were used as the starting materials. The carefully mixed powder, in the stoichiometric molar ratio Mg:B = 1:2, was subjected to high pressure synthesis. The product was released from the high pressure and then quenched from high temperature. The high-pressure synthesis was carried out using a cubic-anvil-type high-pressure apparatus. Pyrophyllite was used as the pressure-transmitting medium and graphite tubes as the electric heater. Details of the sample preparation are described in [4]. The sample obtained was checked using powder x-ray diffraction to confirm the phase purity. The superconductivity was checked by magnetic susceptibility measurements using a SQUID magnetometer and by electrical measurements using the four-probe method. The *in situ* high-pressure measurements were performed using a piston–cylinder-type apparatus. The apparatus is made of Be–Cu with a kind of mixed silicon oil and kerosene as the pressure-transmitting media, which provide a good quasi-hydrostatic environment at low temperature. The pressure was first loaded at room temperature and then screw-locked. The pressure and temperature were calibrated using the well known superconducting transition point of Pb metal. The low-temperature experiments were carried out in a Dewar. The temperature was read from a RhFe thermometer. In view of the high electrical conductivity of MgB_2, a high resolution linear research type digital ac bridge (accuracy better than 0.1 mΩ) was used to carry out the four-probe measurements with a 1 mA measuring current. The sample was measured in both cooling and heating processes at a very slow rate, e.g. 2–3 h between the transition processes (10 K below and above T_c), to ensure a better thermal equilibrium state at each temperature.

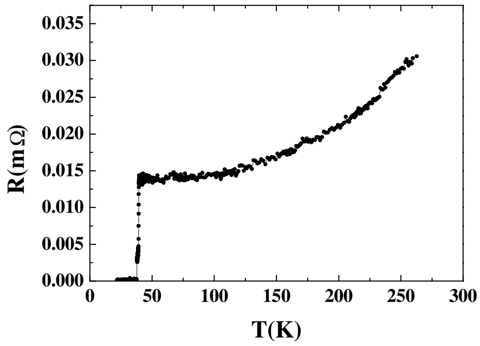

Figure 3. The R–T relation of the MgB_2 sample.

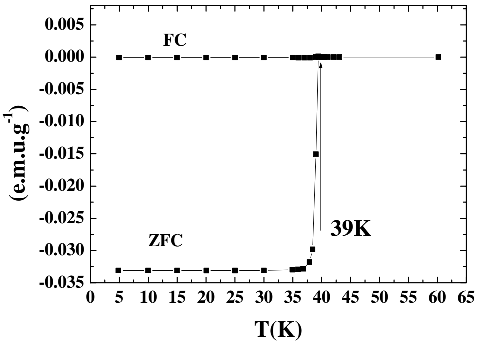

Figure 4. The dc magnetic susceptibility of MgB_2 superconductor as a function of temperature in both ZFC and FC modes.

3. Experimental results and discussion

Figure 2 shows the powder x-ray diffraction pattern of the high-pressure-synthesized MgB_2 sample. The major phase of the sample can be indexed to MgB_2 with the AlB_2 structure, but a small amount of MgO impurity was also present. Figure 3 shows the resistivity versus temperature for the sample, showing the superconducting transition around T_c (39 K) with a transition width less than 1 K.

Figure 4 shows the dc magnetic susceptibility versus temperature for both zero-field-cooling (ZFC) and field-cooling (FC) modes in a 20 Oe applied field obtained using a Quantum Design SQUID magnetometer. It indicates that a sharp superconducting transition occurred at T_c equal to 39 K. The nearly flat ZFC curve up to 35 K and the large disparity between the ZFC and FC signals imply bulk superconductivity and improved intergrain links in the high-pressure-synthesized MgB_2 sample. We adopted the sample to make further *in situ* measurements of the resistivity versus temperature at various pressures up to $\sim$1.0 GPa. Figure 5 shows the R–T curves around T_c for the sample at various pressures up to $\sim$1.0 GPa. The shifts of the superconducting transition with pressure are clearly visible. It is noted that the transition shifts monotonically to high temperature as pressure increases from ambient to $\sim$0.5 GPa before it shifts back with further increasing pressure. Figure 6 plots the change of T_c (the temperature at which the conductivity shows a sudden jump during the slow heating process) with pressure. The tendency shown in figure 6 is different from those of the experimental results in [5]. We repeated the measurements using a second high-pressure sample and got similar results. We will try to provide a possible explanation for this phenomenon. The relatively strong Mg–B bonding and the change in B–B bonding may responsible for the increase of T_c in some pressure region. The high-pressure strain resulting from the high-pressure synthesis may lead to complex behaviour in *in situ* pressure measurements. The intercalation of some organic molecules or clusters from the pressure-transmitting oil may possibly increase the a-axis dimension and consequently give rise to a higher T_c according to the calculation of [6]. The uniaxial pressing of the sample may result in the c-axis being under compression while the a-axis is being stretched and consequently raise T_c [6]. Moreover, the superconducting hole mechanism proposed by Hirsch *et al* supports the current experimental results. However, in order to make clear the physical origin, further experiments such as ones using different pressure-transmitting media are needed.

In summary, we observed T_c increasing with applied pressure in a MgB_2 sample which had been synthesized at high pressure directly from the elements. A parabolic-like T_c–P relation

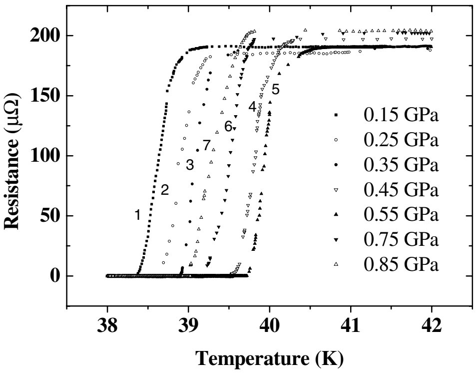

Figure 5. The superconducting transitions for various applied pressures.

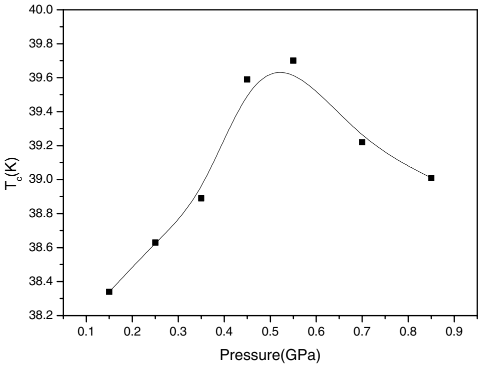

Figure 6. The dependence of T_c for MgB_2 superconductor on the applied pressure.

was found. The enhancement of T_c by more than 1 K in the experiments means that it becomes the highest superconducting transition temperature recorded so far for MgB_2 superconductor.

Acknowledgment

This work was supported by the Natural Science Foundation of China, and the 'Hundreds of Talents Programme' of the Chinese Academy of Sciences.

References

[1] Nagamatsu J, Nakagawa N, Muranaka T, Zenitani Y and Akimitsu J 2001 *Nature* **410** 63–4
[2] An J M and Pickett W E 2001 *Phys. Rev. Lett.* **86** 4366
[3] Takano Y, Takeya H, Fujii H, Kumakura H, Hatano T and Togano K 2001 *Appl. Phys. Lett.* **78** 29 134–6
[4] Li S-C, Zhu J-L, Yu R-C, Li F-Y, Liu Z-X and Jin C-Q 2001 *Chin. Phys.* **10** 338–9 (in English)
[5] Lorenz B, Meng R L and Chu C W 2001 *Phys. Rev.* B **64** 12 507
Monteverde M, Nunez-Regueiro M, Rogado N, Regan K A, Hayward M A, He T, Loureiro S M and Cava R J 2001 *Science* **292** 75
Tang Jie, Qin Lu-Chang, Matsushita A, Takano Y, Togano K, Kito H and Ihara H 2001 *Phys. Rev.* B **64** 132509
[6] Loa I and Syassen K 2001 *Solid State Commun.* **118** 279
[7] Medvedeva N I, Ivanovskii A L, Medvedeva J E and Freeman A J 2001 *Phys. Rev.* B **64** 20 502
[8] Hirsch J E 2001 *Phys. Lett.* A **282** 392
Hirsch J E and Marsiglio F 2001 *Phys. Rev.* B **64** 144523