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Structural and electronic properties of superconductor MgB_2 under high pressure

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Abstract

The superconductivity and the lattice properties of a sintered MgB_2 material have been investigated under high pressure up to 10 GPa. The transition temperature was found to decrease linearly with increasing hydrostatic pressure at a rate of 1.03 K GPa^{-1}, which can be explained with the classical Bardeen–Cooper–Shrieffer theory based on an electron–phonon coupling mechanism. The crystal lattice exhibits an anisotropic compressibility characterized by a larger compressibility along the c-direction than the a/b-directions. The anisotropy is attributed to a weaker inter-plane bonding along the c-axis in comparison with a stronger intra-plane bonding perpendicular to the c-axis. The bulk modulus of the measured material was deduced to be 172 GPa.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The simple MgB_2 crystal structure with the space group of $P6/mmm$ ($a = 3.086$ Å and $c = 3.524$ Å) consists of a layered stacking with alternating Mg and B layers on top of each other. The surprising discovery of superconductivity in magnesium diboride (MgB_2) at 39 K by Nagamatsu *et al* [1] has stimulated great interest in this material. This represents the highest T_c discovered so far in a binary system, much higher than that for the Nb_3Ge alloy ($T_c = 23.2$ K). A few theoretical analyses have been presented to consider the fundamental mechanism responsible for the superconductivity in this material. The conduction bands of the sp^2 bonded boron sheets have been suggested to contribute to the superconductivity, which results from the strong electron–phonon interaction and the high phonon frequency associated with the honeycomb-like boron structure [2].

Since the announcement of the discovery, several reports have been released reporting measurements of pressure effects on MgB_2 materials prepared under various conditions. Vogt *et al* [3] characterized the bulk modulus of commercially available high-purity MgB_2 powders using high pressure and obtained $B_0 \approx 151$ GPa based on the first-order Murnaghan equation of state

$$V = V_0\left(1 + P\frac{B_0'}{B_0}\right)^{-1/B_0'},$$

where V is the unit-cell volume at pressure P, $B_0' = 4$, and V_0 and B_0 are the unit-cell volume and the bulk modulus at ambient conditions, respectively. This measurement gives rise to a value for the volume compressibility, defined as

$$\kappa = \frac{1}{B_0} = -\frac{1}{V_0}\frac{\mathrm{d}V}{\mathrm{d}P},$$

of about 6.7×10^{-3} GPa^{-1} to a good approximation. Prassides *et al* [4] measured the compressibility of the material used by Nagamatsu *et al* [1] and obtained a volume compressibility of 8.3×10^{-3} GPa^{-1}. Lorenz *et al* [5] prepared their MgB_2 material by a solid-state reaction method and measured the dependence of T_c on hydrostatic pressure up to 1.84 GPa; they obtained $\mathrm{d}T_c/\mathrm{d}P \approx -1.6$ GPa^{-1}. Loa and Syassen [6] gave a theoretical analysis and suggested a rate of decrease of 1.6 K GPa^{-1} in response to pressure. Jorgensen *et al* [7] reported their results on the crystal lattice properties of MgB_2 versus both temperature (11–297 K) and pressure (0–0.62 GPa) using neutron diffraction.

Here we report the results on an experimental measurement of the electrical resistivity and lattice compressibility, under hydrostatic pressure up to 10 GPa, of the MgB_2 superconducting material.

2. Experimental details

The MgB_2 material employed in the present investigation was prepared using a higher pressure (3.5 GPa compared with the pressure of 0.2 GPa used by Nagamatsu *et al* [1]) and higher sintering temperature (1000 °C). This material shows a substantial improvement in its superconducting performance [8]. Since it was high-pressure sintered, no porosity was observed in the material and it has a mass density of 2.66 g cm^{-3}, close to the theoretical value of MgB_2 of 2.63 g cm^{-3}.

For the *in situ* measurement of the lattice parameters under pressure, a diamond anvil cell was employed and synchrotron x-ray diffractograms were collected on imaging plates, using the Photon Factory beamline 18 C at the High Energy Accelerator Research Organization (KEK), Tsukuba, Japan.

The electrical resistivity of the MgB_2 superconducting material was measured as a function of temperature at each fixed pressure with the DC four-probe method using a cubic anvil apparatus operated up to 10 GPa. The cubic anvil device is compressed evenly from six directions with six anvil tops, thereby producing a hydrostatic pressure on the sample material sealed in a gasket. The gasket material is a mixture of amorphous boron and epoxy resin in 4:1 weight ratio. Fluorinert liquid is used as the pressure medium to maintain the hydrostatic pressure. Low temperature down to 7 K is attained with continuous flows of liquid nitrogen and helium, successively.

3. Results and discussion

We measured the dependence of the superconductivity transition temperature T_c on pressure up to 10 GPa. The sharp drop of resistivity observed under ambient conditions was retained

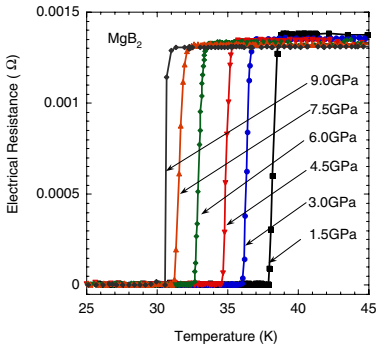

Figure 1. Electrical resistivity versus temperature when the sample is subjected to various pressures up to 10 GPa. The transition from the normal state to the superconducting state takes place within a very narrow temperature range of about 0.2 K.

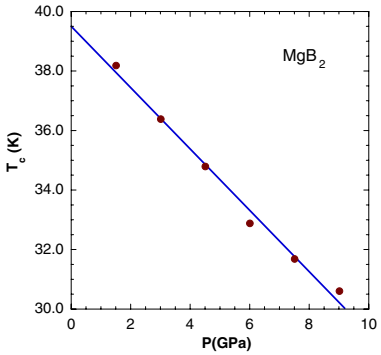

Figure 2. A plot of transition temperature T_c versus pressure P. A linear behaviour is observed and T_c decreases with increasing pressure at a rate of about 1.03 K GPa^{-1}.

even when the pressure was applied, as is shown in figure 1. A remarkable linear behaviour is observed up to the full range of 10 GPa and figure 2 shows the experimental data with a fitted linear curve that is expressed by

$$T_c = 39.6 - 1.03P.$$

The rate of decrease of T_c, 1.03 K GPa^{-1}, is much lower than the value of 1.6 K GPa^{-1} obtained by Lorenz *et al* [5], and the negative slope of the T_c–P relationship goes against the theoretical predictions by Hirsch [9]. On the other hand, the experimental observation suggests that T_c could be increased further if a chemical substitution could increase the lattice parameter while maintaining the same electronic structures, which is equivalent to producing a negative pressure.

The transition range is about 0.5 K between the normal state and the superconducting state, demonstrating the superb quality of the MgB_2 material. Even when the material was subjected to high pressure, the transition width changed only slightly. For example, at the pressure 7.5 GPa, the transition width is measured to be 0.6 K, giving rise to an increase of the transition width at a rate less than 0.1 K GPa^{-1}. In addition, before the transition to superconducting behaviour occurred, the material showed a metallic behaviour upon cooling with a ratio of electrical resistivities

$$\frac{\rho(300\ \mathrm{K})}{\rho(40\ \mathrm{K})} = 1.93$$

at 1.5 GPa pressure and the ratio increased linearly with pressure, reaching 2.05 at 9.0 GPa pressure.

Figure 3 shows the dependence of the lattice parameters a and c and the unit-cell volume V on the applied pressure.

There is a significant difference between the contraction rates of the intra-planar periodicity a and the inter-planar periodicity c; the latter reduces about 1.4 times as fast as the former, signifying that the structure is more compressible in the c-direction than in the a/b-directions. A least-squares linear fitting of the experimental data yielded a value for the bulk modulus $B_0 = 172$ GPa while a value of $B_0 = 174$ GPa was obtained using a least-squares fitting of the Murnaghan equation. The volume compressibility κ was also deduced from the experimental data and showed a weak dependence on the pressure.

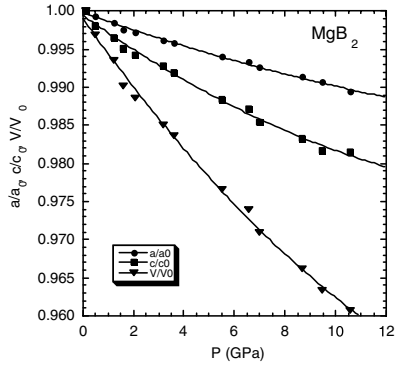

Figure 3. Dependences of the lattice parameters a and c and the unit-cell volume V on the applied pressure up to 10 GPa. The plots are given as ratios to the corresponding values at ambient conditions. The deduced bulk modulus of the MgB_2 material is $B_0 = 172$ GPa.

The boron isotope effect has indicated that the superconductivity in the MgB_2 material is phonon mediated, and can be described within the framework of the BCS theory [10]. Using the McMillan formula [11], we estimated the pressure dependence of the transition temperature T_c within the framework of the BCS theory [12] and obtained

$$\frac{\mathrm{d}T_c}{\mathrm{d}P} \approx -1.22 \text{ K GPa}^{-1},$$

which is very close to our experimental value of -1.03 K GPa^{-1}. However, we should note here that since the McMillan formula was deduced for isotropic structures, given the anisotropic nature of the MgB_2 structure, the above analysis has limits on its validity [13].

4. Conclusions

The MgB_2 material synthesized under high pressure and high temperature shows an improved superconducting performance characterized by (a) a sharp transition from the normal state to the superconducting state, (b) a lower rate of decrease ($\sim$1.03 K GPa^{-1}) of the transition temperature T_c with increasing pressure, and (c) lower ratios of electrical resistivities $\rho(300\text{ K})/\rho(40\text{ K})$. These characteristics of the transport properties are preserved under pressure at least up to 10 GPa and the pressure effect on T_c is well explained within the framework of the classical BCS theory based on the electron–phonon coupling mechanism.

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