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Anomalous non-linear pressure dependence of T_C for MgB$_2$

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Abstract

We report the pressure dependence of the superconducting transition temperature of MgB$_2$ from 0 to 4.5 GPa, investigated using a diamond-anvil cell. We found that under non-hydrostatic pressure, T_C at first decreases linearly at a rate $\mathrm{d}T_C/\mathrm{d}P = -1.1$ K GPa^{-1} from 0 to 0.5 GPa. But as pressure is increased from 0.5 to 1.3 GPa, $\mathrm{d}T_C/\mathrm{d}P$ decreases drastically, passing through a small minimum and then starting to increase at about 1.3 GPa. At this pressure, T_C increases slightly. From about 1.4 to 4.5 GPa, T_C, decreases with a subsequent *quadratic*, as opposed to the expected linear, dependence on P. Our results are discussed and compared with others for this superconducting material. The results are analysed in terms of theoretical predictions.

The new binary superconducting compound MgB$_2$ discovered by the Akimitsu group [1] has generated a great deal of experimental and theoretical interest in the past few months. According to the experimental evidence, this compound seems to behave like a conventional BCS electron–phonon-mediated superconductor. However, some discrepancies still exist and more information related to the dominant mechanism in this MgB$_2$ superconductor is thus required. For instance, new measurements of the isotope effect have shown that this coefficient is quite reduced from the BCS prediction of 1/2; values of about 0.32 have been reported by Hinks *et al* [2]. These results might indicate that the system is limited to high values of λ and μ^* (the McMillan electron–phonon interaction parameters), and/or other features such as anharmonic effects, as was pointed out by the Hinks group in the same paper. In addition, data determining the order of magnitude of the phonon energies involved in the pairing mechanism are needed. Hence, further evidence will be necessary to define more precisely the underlying physics of this compound. Among the techniques which could provide the required information are tunnelling and point-contact spectroscopies; from these we can extract values of the phonon energies involved in the pairing mechanism, to be studied using Eliashberg theory. On the other hand, the pressure dependence of T_C may provide important information required to prove different theoretical predictions. Hirsch [3,4] for instance, with a hole superconducting model, makes relevant predictions, particularly for experiments where T_C is a function of the uniaxial pressure. BCS theory [5,6] and Hirsch's models predict different pressure coefficients for T_C. The former predicts that when the pressure is increased, T_C must drop linearly with P, whereas Hirsch proposes that T_C will increase or decrease depending on the number of carriers or the doping level.

Earlier high-pressure experiments on MgB_2 [7–11] revealed negative values for the dT_C/dP coefficient, although all of them found different magnitudes for the dependence. These differences have been attributed to variations in sample quality, stoichiometry [11], and defects [10]. Moreover, as far as we know there is no clear recent report on pressure dependence of T_C in the regime from 0 to 5 GPa, under non-hydrostatic conditions. In this work we concentrate on experiments performed under non-hydrostatic pressure using a diamond-anvil cell. We have performed measurements on MgB_2, and our data show an anomalous behaviour of $T_C(P)$ consisting of a *shoulder shape*. We analyse that dependence and compare it with recent results obtained by other workers. We believe that this might give additional information on the underlying nature of the pairing mechanism in this compound.

A high-frequency induction furnace (Stanelco type STA 30 KW) with a 120 mm work coil as the heat source was used to produce polycrystalline samples of MgB_2. The initial materials were small magnesium chunks with 99.9% purity and boron powders with 99.8% purity. These materials were mixed in 1 g stoichiometric ratios with 5 and 10% excess of Mg to compensate for evaporation losses. The materials were put into tantalum tubes, and placed on the cold copper crucible area of the radio-frequency oven. The reaction was maintained in a nonreactive atmosphere of clean argon of high purity. At first, the tantalum tubes with the initial reactants were heated at approximately 700, 800, and 950 °C for a total time of 6–10 h. After this period, the sample was cooled to room temperature. Figure 1 shows the x-ray diffraction data for the sample after different periods of heating. At 950 °C the pattern shows the phase of the MgB_2 compound with a small content of MgO impurities ($<5\%$). The pressure measurements were performed over the temperature range from 2 to 300 K using non-hydrostatic pressure generated with a diamond-anvil cell (a piston-cylinder-type Be–Cu cell) [12]. This consists of two diamonds, each of 0.7 mm culet size. The MgB_2 sample had dimensions of approximately $100 \times 100 \times 40\ \mu m^3$ and was placed in the steel gasket hole of 200 μm diameter and filled with pyrophyllite powder as the pressure-transmitting medium. The metallic gasket was electrically insulated, by pressing over it Al_2O_3 powder of 1 μm grain size. Four gold wires of 10 μm diameter were used as the electrical leads. A thin foil of aluminium was placed under the gold wires to prevent ruptures during the experiment. A thin Mylar film was used in order to prevent motion of the sample and of the electrical leads when pressure was applied. To measure the pressure at low temperature we used a Pb manometer [13]. The Pb wire with dimensions of about $50 \times 100 \times 50\ \mu m^3$ was placed along with the MgB_2 sample in the same pressure chamber. Four gold wires were used as electrical leads. The superconducting transition temperature of the Pb manometer was measured for each pressure application.

A selection of $R(T)$ versus P curves covering the entire pressure range from 0 to 4.5 GPa are presented in figure 2. All of them were measured in an Optistat Bath Oxford cryostat. The R versus T curve at ambient pressure is linear from 300 to about 175 K. The residual resistance ratio (RRR) for the sample was 4.2 and the transition temperature $T_C(R = 0)$ was found to be 39.8 K (see the inset of figure 2). The transition width of the MgB_2 between the 10 and 90% values of the resistance drop was 0.8 K, which increases slightly with pressure. The behaviour of $T_C(P)$ for the sample can be appreciated in figure 2.

The pressure dependence of T_C up to 4.5 GPa for the sample is shown in figure 3. Initially, T_C (black triangles) decreases linearly with pressure from 0 to 0.5 GPa at a rate $dT_C/dP = -1.1$ K GPa^{-1}. This result is quantitatively in agreement with previous

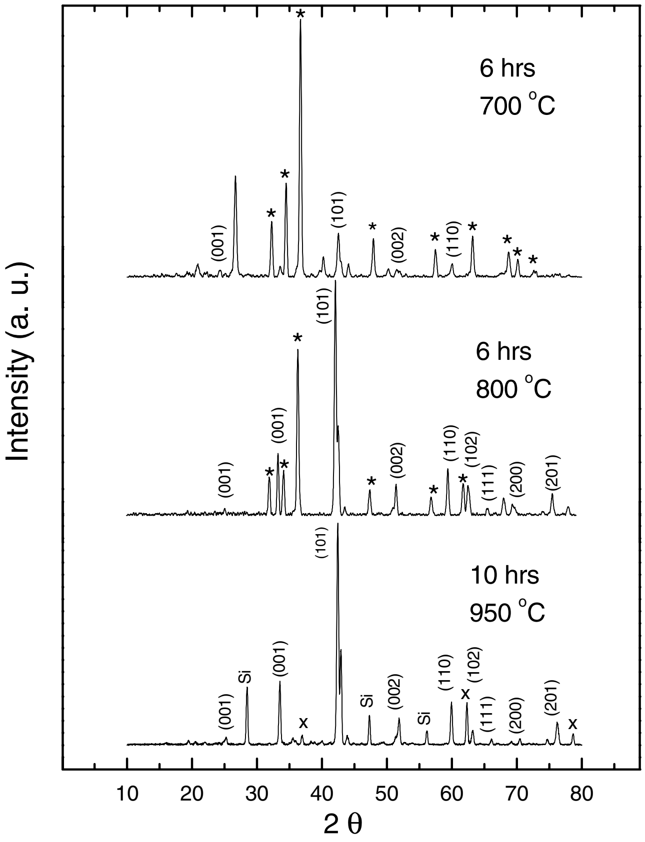

Figure 1. Diffraction patterns for three MgB$_2$ samples with different purities. They show Mg ($*$) and MgB$_2$ as the main phases at 700 and 800 °C with different annealing times. The diffraction pattern for the other sample (bottom graph) shows mainly MgB$_2$ phase after an annealing for 10 h at 950 °C. Si was used as a standard in order to find the lattice parameters; we found $a = 3.0864 \pm 0.0006$ Å and $c = 3.5205 \pm 0.00145$ Å.

measurements carried out under hydrostatic conditions in the same pressure range by various authors [9, 11]. With additional increase in pressure, $\mathrm{d}T_C/\mathrm{d}P$ decreases, passing through a minimum at about 0.5 GPa and then showing a small increase for T_C up to about 1.3 GPa. As the pressure increased beyond 1.3 GPa, T_C started decreasing again but with a quadratic as opposed to the expected linear dependence. After the sample had been exposed to pressure, it was taken out and then pressed again in order to verify the presence of the minimum at about 0.5 GPa. The second $T_C(P)$ curve is shown in the same figure 3 (open squares). We observed that it follows the same behaviour as in the first measurement, showing a rate $\mathrm{d}T_C/\mathrm{d}P = -1.1$ K GPa^{-1} from ambient pressure to about 0.5 GPa, and again showing a minimum of $T_C(P)$ at about 0.5 GPa. Monteverde *et al* [10] have studied several MgB$_2$ samples with different qualities up to 25 GPa using non-hydrostatic conditions. They found different pressure behaviour of T_C depending on the sample, which they have attributed to the particular nature of the band structure of MgB$_2$. In particular, a quadratic behaviour was reported by them. However, the part of the $T_C(P)$ curve corresponding to such behaviour, from atmospheric pressure to about 2 GPa, is not accurately determined. Our results indicate that under non-hydrostatic conditions, there is indeed a dramatic decrease of $\mathrm{d}T_C/\mathrm{d}P$ at about 0.5 GPa, preceded by

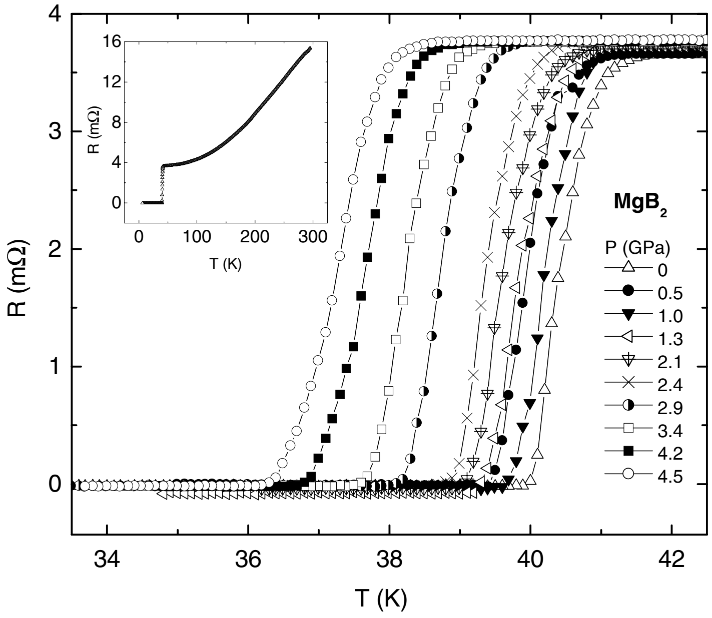

Figure 2. Electrical resistance for a sample as a function of temperature at different quasi-hydrostatic pressures from 0 to 4.5 GPa, shown in the interval close to the transition temperature. The inset shows the electrical resistance versus temperature measured under normal atmospheric pressure.

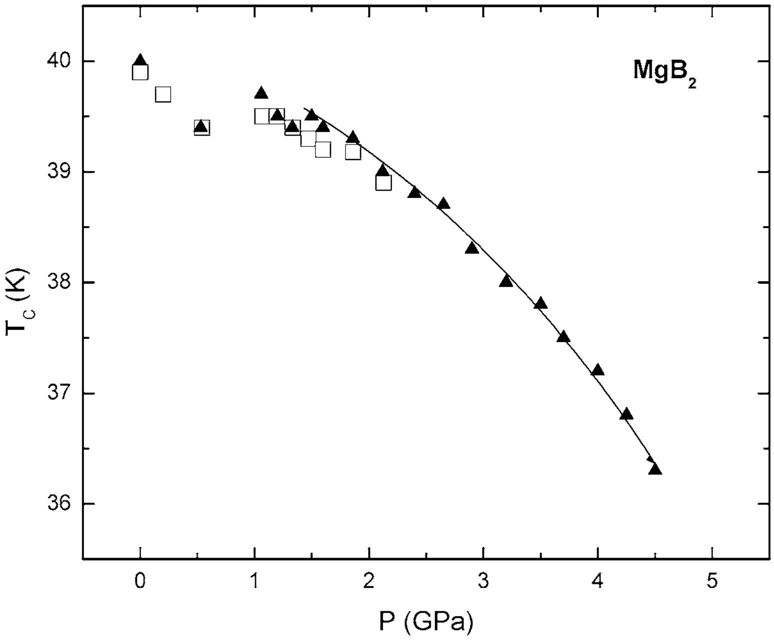

Figure 3. Curves of $T_C(P)$ for MgB$_2$. Note the change of the pressure coefficient, dT_C/dP, at about 0.5 GPa. At high pressure (from 1.3 to 4.5 GPa) the curve shows a P^2-dependence (black triangles are for the first pressure cycle and white squares for the second one for the same sample).

a linear behaviour, in the MgB_2 compound. Lorentz *et al* [11] performed quasi-hydrostatic pressure experiments in the low-pressure regime from 0 to 1 GPa using solid He as the pressure-transmitting medium, and they conclude that the $\mathrm{d}T_C/\mathrm{d}P$ coefficient is independent of whether the pressure is hydrostatic or quasi-hydrostatic. However, it is known that He gas, which is the best hydrostatic pressure-transmitting medium, starts to freeze at about 40 K under 0.5 GPa; thus the quasi-hydrostatic environment could not be significant. Recently, another anomaly has been observed, at 9 GPa, in the $T_C(P)$ curve of MgB_2, which has been interpreted as a result of a pressure-induced electronic transition [14]. On the other hand, electronic structure studies [15] of MgB_2 under uniaxial and hydrostatic compression have predicted that the two pressure environments affect the electronic band differently. The band-structure change is small under hydrostatic compression, but it is not so small under uniaxial pressure applied along the c-axis. This latter pressure environment modifies the σ-bands at the Γ–A line which tends to become completely occupied with increasing pressure. This occupation has been explained by an increase in the number of σ-bonding electrons in the B layer, which results in a strengthening of the B–B bond. According to the 'dressed-hole theory', any decrease of the B–B distance caused by pressure must increase T_C.

A possible hypothesis related to the anomalous change observed is that it may be due to decreases in the Mg–B and B–B distances with applied pressure. Such behaviour is predicted by the 'dressed-hole theory' of Hirsch and Marsiglio [4]. The fact that the increase of T_C has not been seen in other recent high-pressure experiments could be due to the degree of hole doping in the material—which in turn is, in a way, controlled by the purity of the compound. It is possible that from 0.5 to about 1.3 GPa the dominant effect of pressure is in reducing the intraplanar distances rather than the interplanar distances. Such an effect would mainly be caused by shear stress or uniaxial stress due to the non-hydrostatic pressure environment.

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