Hydrostatic Pressure Effect on the Superconducting Transition Temperature of MgB$_2$

B. Lorenz, R. L. Meng and C. W. Chu$^1$

Department of Physics and Texas Center for Superconductivity, University of Houston,

Houston, Texas 77204-5932

$^1$also at Lawrence Berkeley National Laboratory, 1 Cyclotron Road,

Berkeley, California 94720

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Abstract

The pressure effect on the superconducting transition temperature of MgB$_2$ has been determined using gas pressure up to 1 GPa. The transition temperature $T_c$ was found to decrease linearly at a constant rate over the whole pressure range. The recently observed dramatic decrease of $|dT_c/dp|$ at the 40 K freezing pressure (0.5 GPa) cannot be confirmed. The pressure coefficient was also found to be independent of the hydrostatic or nonhydrostatic He environment. The differences in recently reported values of $dT_c/dp$ may be attributed to variations in the sample conditions, e.g. stoichiometry.

74.60.-w, 74.62.Fj, 74.25.Ha
The recent discovery of superconductivity in MgB$_2$ at temperatures as high as 40 K has generated great interest. MgB$_2$, which exhibits an AlB$_2$ structure with honeycomb layers of boron atoms, appears to be electrically and mechanically three-dimensional and its grain boundaries have a far less detrimental effect on superconducting current transport. The new compound may provide a way to a higher superconducting transition temperature $T_c$ and an easier avenue for devices. Shortly after the discovery of this exciting compound a still ongoing discussion was initiated whether the superconductivity in MgB$_2$ is better described by a BCS-like theory or by heavily dressed holes in an almost completely filled conduction band. The boron isotope effect on $T_c$ and a BCS-like superconducting gap structure favor the BCS-type pairing mechanism. The pressure effect on $T_c$ is of special interest since the dressed hole theory predicted an increase of $T_c$ with pressure as long as there is no charge transfer between the boron and magnesium planes. First high pressure measurements revealed a negative pressure coefficient of $dT_c/dp \sim -1.6 \, K/GPa$ indirectly supporting the BCS-mechanism. Subsequent band structure calculations are in good agreement with the experimental pressure effect and could explain the decrease of $T_c$ within the BCS model by a pressure induced change of the density of states and the phonon frequency. The negative sign and the order of magnitude of the pressure coefficient were later confirmed but the absolute value $dT_c/dp$ varied from $-1.1 \, K/GPa$ to $-2 \, K/GPa$.

Compressibility measurements performed at room temperature show consistently that the $c/a$ ratio changes very little under pressure (about 1 % at 10 GPa) indicating nearly isotropic compression. The same conclusion was drawn from band structure calculations under pressure. Jorgensen et al. recently found that the compression along the c-axis is 64 % larger than along the a axis. As a result, they proposed that a truly hydrostatic pressure is indispensable to obtain correct results of MgB$_2$. Using a He-gas pressure system to generate the best hydrostatic environment for MgB$_2$, Tomita et al. found that $dT_c/dp = -1.11 \, K/GPa$ up to 0.5 GPa but drops to almost zero above 0.5 GPa. This is in strong contrast to what previously was observed, namely, $T_c$ decreases with pressure linearly up to 1.8 GPa at a greater rate of $-1.6$ to $-2.0K/GPa$. Liquid He is known to freeze at
about 40 K under 0.5 GPa. Consequently, Tomita et al. proposed their smaller $|dT_c/dp|$ below 0.5 GPa should be the ”true hydrostatic value” and the nearly zero $|dT_c/dp|$ above 0.5 GPa should be a result of the non-hydrostaticity associated with the freezing of the liquid He pressure medium. These observations have been cited as supports for the proposed sensitive role of hydrostaticity in the $T_c$-behavior of $MgB_2$ under pressure. Unfortunately, the reduction of $|dT_c/dp|$ above 0.5 GPa due to the proposed shear-stress effects cannot reconcile with the larger $|dT_c/dp|$ previously observed in a less-than-ideal hydrostatic environment. It should be noted that, despite of the greater compression along the c-axis than along the a-axis reported, the overall fractional changes in $c/a$, c and a up to 0.6 GPa are very small, $\sim 10^{-4}$. Given the small compressibility of $MgB_2$, the drastic $|dT_c/dp|$ change upon the freezing of liquid He is rather puzzling in the absence of any phase transition in a quasihydrostatic pressure up to 8 GPa, especially in view of the fact that solid He is the softest material at low temperature. An experimental artifact due to a failure to deliver pressure to the sample chamber after freezing of liquid He is therefore suspected.

We have, therefore, carried out high pressure experiments on $MgB_2$ samples with different $T_c$’s up to 1 GPa using helium as pressure medium. The pressure coefficient of $T_c$ is carefully monitored in the hydrostatic ($p < 0.5$ GPa) region and at higher pressure where the He freezes above $T_c$. $T_c$ was found to decrease linearly with pressure over the whole pressure range at a rate depending on the sample. We conclude that nonhydrostatic pressure environment has no or only minor effect on the superconducting transition temperature of $MgB_2$. We also conclude that the observed differences in the value of $dT_c/dp$ are due to subtle differences in sample purity, porosity, or stoichiometry.

For the high pressure measurement we prepared a high quality polycrystalline $MgB_2$ sample using the standard synthesis. Small Mg chips (99.8 % pure) and B powder (99.7 %) with a ratio of Mg:B = 1.25:2 were sealed inside a Ta tube in an Ar atmosphere. The magnesium was added in excess of the stoichiometric amount in order to compensate for any Mg loss during the synthesis. The sealed Ta ampoule was in turn enclosed in a quartz tube. The ingredients were heated slowly up to 950 °C and kept at this temperature for
2 hours, followed by furnace-cooling to room temperature. The samples so-prepared were dense and x-ray spectra show a very minor amount of MgO phase. The resistivity and thermoelectric power of this sample show very sharp transitions to zero at 39.3 K (midpoint of the superconducting transition) with a width of less than 0.14 K. The ac susceptibility, $\chi_{ac}$, at ambient pressure exhibits an equally sharp diamagnetic drop at 39.2 K (midpoint) as shown in Fig. 1. For comparison, we have also re-measured another $MgB_2$ sample previously studied under the same conditions in the helium environment. This sample exhibits a lower $T_c$ ($< 38$ K) and a broader transition. This sample was previously investigated in the Fluorinert FC 77 pressure medium.

The superconducting transition was detected by ac susceptibility measurements. The sample was placed in a transformer in the He gas pressure cell (UNIPRESS) which was connected to a 1.5 GPa gas compressor (UNIPRESS) by a beryllium copper capillary (0.3 mm ID). The gas pressure cell and part of the capillary was inserted into a Model 8CC Variable Temperature Cryostat (CRYO Industries) for cooling and temperature control. Special care was taken in cooling at high pressure ($p > 0.5 \text{ GPa}$) to avoid freezing of helium in the capillary before it solidifies in the pressure cell. If frozen helium blocks the capillary first and then solidifies inside the pressure cell a large drop of pressure (about 13 % at 0.7 GPa) in the cell is usually observed which may not be recognized if the manometer is located in the room temperature pressure reservoir. This can easily lead to large errors in the pressure measurement. Therefore, the pressure cell was cooled very slowly by controlling the temperature of the cooling He gas to guarantee that the helium freezes from the bottom of the cell towards the upper end connected to the gas supply capillary. The cooling process was monitored by two thermometers mounted to the top and the bottom of the gas pressure cell. Furthermore, a semiconductor pressure gauge was placed inside the pressure cell close to the sample position and the pressure was measured in situ also in the solid state of the pressure medium. The pressure values used in Figs. 1 and 2 are measured right at the superconducting transition temperature.

In the first pressure cycle the cell was loaded to 1 GPa at room temperature. After
cooling and solidification of the helium the pressure decreased to 0.843 GPa at $T_c = 38.29 \, K$. The cell was heated to above 150 $K$ before changing pressure. $\chi_{ac}$ was measured during cooling and warming through the transition. Fig. 1 shows a set of data taken at different pressures. The pressure values indicated in the figure refer to $p(T_c)$. The diamagnetic drop of $\chi_{ac}$ shifts in parallel to lower temperature with increasing pressure. $T_c(p)$ was determined as the midpoint temperature of this drop. As shown in Fig. 2, $T_c$ is a linear function of $p$ over the whole pressure range. The pressure coefficient of $-1.07 \, K/GPa$ is very close to the value of Tomita et al.\textsuperscript{9} in the hydrostatic range ($p < 0.5 \, GPa$), however, the drastic decrease of $|dT_c/dp|$ observed by them at higher pressures is not detected in our experiments.

There remains the question if the larger absolute value of $dT_c/dp$ observed in the piston-cylinder clamp using quasi hydrostatic pressure media may be a consequence of pressure induced shear stress. We repeat the He gas pressure measurement with our $MgB_2$ sample that was shown to yield a pressure coefficient of $-1.6 \, K/GPa$ using the Fluorinert FC77 as pressure medium.\textsuperscript{7} Again, $T_c$ decreases linearly with $p$ over the pressure range to 0.84 GPa and no anomaly is detected in passing the freezing pressure of He. A pressure coefficient $dT_c/dp = -1.45 \, K/GPa$ is obtained and is in agreement with our previous data (within the experimental uncertainty). As mentioned above, this sample shows a lower $T_c$ and a broader transition. We propose that the spread of $dT_c/dp$ reported by different groups\textsuperscript{7,10,9} is rather due to subtle differences in the sample condition, e.g. composition, than to shear stress in quasi hydrostatic pressure environment.

In conclusion we have shown that the pressure effect on the superconducting transition temperature of MgB$_2$ is linearly negative to the highest pressure studied and is insensitive to small deviations from truly hydrostatic pressure conditions. Our results support the view that MgB$_2$, despite its layered structure, is nearly isotropic with respect to compression. The variation in the value of $|dT_c/dp|$ by various groups results from the differences in sample conditions such as composition.
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FIGURES

FIG. 1. $\chi_{ac}$ vs. $T$ at various pressures.
open symbols: data taken in liquid He; closed symbols: data taken in solid He

FIG. 2. $T_c$ as function of pressure.
