The Dependence of $T_c$ on Hydrostatic Pressure in Superconducting MgB$_2$

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Abstract

The dependence of $T_c$ on hydrostatic (He-gas) pressure for superconducting MgB$_2$ has been determined to 0.7 GPa. We find that $T_c$ decreases linearly and reversibly under pressure at the rate $dT_c/dP \simeq -1.11 \pm 0.02$ K/GPa. These studies were carried out on the same sample used in earlier structural studies under He-gas pressure which yielded the bulk modulus $B = 147.2 \pm 0.7$ GPa. The value of the logarithmic volume derivative of $T_c$ is thus accurately determined, $d\ln T_c/d\ln V = +4.16 \pm 0.08$, allowing quantitative comparison with theory. The present results support the emerging picture that MgB$_2$ is a BCS superconductor with electron-phonon pairing interaction.
The recent discovery\[1\] of superconductivity in MgB\(_2\) at \(T_c \simeq 39\) K has sparked worldwide a torrent of experimental and theoretical activity, reminiscent of the frenzy following the observation \[2\] of superconductivity in La-Ba-Cu-O at a comparable temperature almost 15 years ago. Replacing \(^{10}\)B with \(^{11}\)B results in a sizeable isotope shift\[3\] to lower temperatures which points to BCS superconductivity. Other experiments, such as heat capacity\[4, 5\], photoemission spectroscopy\[6\], and inelastic neutron scattering\[7, 8\] also support the picture that MgB\(_2\) is a phonon-mediated superconductor in the weak-to-moderate coupling regime.

High pressure studies traditionally play an important role in superconductivity. A large magnitude of the pressure derivative \(dT_c/dP\) is a good indication that higher values of \(T_c\) may be obtained through chemical means. It is not widely appreciated, however, that the pressure dependence \(T_c(P)\), like the isotope effect, contains valuable information on the superconducting mechanism itself. For example, in simple-metal BCS superconductors, like Al, In, Sn, and Pb, \(T_c\) invariably decreases under pressure due to the reduced electron-phonon coupling from lattice stiffening \[9\]. More generally, an accurate determination of the dependence of both \(T_c\) and the lattice parameters on pressure yields the functional dependence \(T_c = T_c[a(P), b(P), c(P)]\) which provides a critical test of theoretical models. Hirsch\[10\] and Hirsch and Marsiglio\[11\] have applied a theory of hole superconductivity to MgB\(_2\) and predicted that for an optimally doped sample \(T_c\) should increase with pressure, in contrast to the expected decrease in \(T_c\) from lattice stiffening.

Precise structural data on MgB\(_2\) at room temperature (RT) have recently been obtained by Jorgensen et al.\[12\] for hydrostatic pressures to 0.6 GPa in a He-gas neutron diffraction facility which yield the anisotropic compressibilities \(d\ln a/dP = -1.87 \times 10^{-3}\) GPa\(^{-1}\), \(d\ln b/dP = -3.07 \times 10^{-3}\) GPa\(^{-1}\), and the bulk modulus \(B = 147.2 \pm 0.7\) GPa; the compressibility along the \(c\) axis is thus significantly (64%) larger than that along the \(a\) axis. The binding within the boron layers is evidently much stronger than between the layers. These results are in reasonable agreement with electronic structure calculations by Loa and Syassen\[13\].

Recent synchrotron x-ray diffraction studies at RT in a diamond-anvil-cell (DAC) to much higher pressures (8 - 12 GPa) using dense He\[14\] or methanol-ethanol\[15\] as hydrostatic pressure media yield the bulk moduli \(B = 155 \pm 10\) GPa and \(151 \pm 5\) GPa, respectively, in agreement, within experimental error, with the He-gas study\[12\]. A further DAC study\[16\] to 7 GPa with silicon oil as pressure medium gives the significantly smaller value \(B = 120 \pm 5\) GPa, even though the reported 2.9% decrease in unit cell volume upon applying 6.5 GPa pressure is less than the 4% decrease found in the other two DAC studies\[14, 15\]. The strong deviation of the “silicon oil” data\[16\] may arise from difficulties in extrapolating the data to zero pressure to obtain the bulk modulus and/or from shear stresses arising from the solidification of the silicon oil under pressure. The relatively large compressibility anisotropy in MgB\(_2\) mandates the use of hydrostatic pressure in quantitative studies, since shear stresses applied by non-hydrostatic pressure media to an elastically anisotropic sample can
lead to erroneous results.

Several studies of the dependence of $T_c$ on pressure have appeared for MgB$_2$. In the first experiment utilizing a fluid pressure medium (Fluorinert), Lorenz et al.\cite{17} report that $T_c$ decreases linearly with pressure to 1.8 GPa at the rate $dT_c/dP \simeq -1.6$ K/GPa. On the other hand, using the same pressure medium, Saito et al.\cite{18} find the more rapid decrease $dT_c/dP \simeq -2.0$ K/GPa. Both groups cite their results to argue that MgB$_2$ is a BCS phonon-mediated superconductor\cite{17, 18}, as is also argued by Loa and Syassen\cite{13}. In further experiments to 25 GPa utilizing the solid pressure medium steatite, Monteverde et al.\cite{19} find that $T_c$ decreases under pressure at differing initial rates (-0.35 K/GPa to -0.8 K/GPa), $T_c(P)$ showing a quadratic behavior for two of the four samples studied; each of the four samples was prepared using a different synthesis procedure.

In the present experiment, $T_c(P)$ is determined to 0.7 GPa in an ac susceptibility measurement using a He-gas pressure system; the helium pressure medium remains fluid at $T_c \simeq 39$ K up to 0.5 GPa and thus applies true hydrostatic pressure (no shear stresses) to the sample. We find that $T_c$ decreases linearly and reversibly with pressure at the rate $dT_c/dP \simeq -1.11 \pm 0.02$ K/GPa. Implications for the nature of the superconducting state are discussed.

The powder sample for this study was taken from the same mother sample used in parallel neutron diffraction studies\cite{12}. It is made using isotopically-enriched $^{11}$B (Eagle Picher, 98.46 atomic % enrichment). A mixture of $^{11}$B powder (less than 200 mesh particle size) and chunks of Mg metal were reacted for 1.5 hours in a capped BN crucible at 800°C under an argon atmosphere of 50 bar. As seen in Fig. 1, the resulting sample displays sharp superconducting transitions in the ac susceptibility with full shielding and an onset temperature at ambient pressure $T_c(0) \simeq 39.25$ K. Both x-ray and neutron diffraction data show the sample to be single phase with the AlB$_2$-type structure.

The present high pressure studies were carried out using a He-gas high-pressure system (Harwood) to 1.4 GPa; the pressure is determined by a calibrated manganin gauge in the compressor system at ambient temperature. The superconducting transition of the 8.12 mg MgB$_2$ powder sample is measured by the ac susceptibility technique using a miniature primary/secondary coil system located inside the 7 mm I.D. bore of the pressure cell. A small Pb sphere with 1.76 mm dia. (38.58 mg) is also inserted in the coil system for susceptibility calibration purposes; for selected data the superconducting transition temperature of this Pb sphere serves as an internal manometer\cite{20} to check the pressure indicated by the manganin gauge. The CuBe pressure cell (Unipress), which is connected to the compressor system by a 3 mm O.D. \times 0.3 mm I.D. CuBe capillary tube, is inserted into a two-stage closed-cycle refrigerator (Leybold) operating in the temperature range 2 - 320 K. The pressure can be changed at any temperature above the melting curve $T_m$ of the helium pressure medium (for example, $T_m \simeq 13.6$ K at 0.1 GPa and $T_m \simeq 38.6$ K at 0.50 GPa\cite{21}). For pressures above 0.5 GPa, $T_m$ lies above $T_c$; the slight pressure drop (few 0.01 GPa’s)
on cooling from $T_m$ to $T_c$ is estimated using the isochores of He$^{[21]}$. All pressures are determined at the temperature $T_c \simeq 39$ K. Further details of the experimental techniques are given elsewhere$^{[22]}$.

In Fig. 1 are shown representative examples of the superconducting transition in the ac susceptibility at both ambient and high pressure. With increasing pressure the narrow transition is seen to shift bodily to lower temperatures, allowing a determination of the pressure-induced shift in $T_c$ to within ± 10 mK. Remarkably, close inspection of the data for 0.50 GPa reveals a slight shift in the transition curve near its midpoint, accurately marking the position of the melting curve of helium ($T_m \simeq 38.6$ K) at this pressure.

In Fig. 2, $T_c$ is plotted versus applied pressure to 0.7 GPa and is seen to follow a highly linear dependence $dT_c/dP \simeq -1.11 \pm 0.02$ K/GPa. The first data point 1′ was obtained after first applying a pressure of $\sim 0.7$ GPa at RT before cooling down to low temperatures ($\sim 60$ K) and reducing the pressure, yielding $T_c \simeq 38.88$ K at 0.341 GPa. Point 2′ was measured after releasing the pressure at low temperature, giving $T_c \simeq 39.25$ K at ambient pressure (0 GPa); no change in $T_c$ occurred after intermittently warming the sample to RT (point 3). Further data were obtained following pressure changes at both RT (unprimed data) and low temperature (primed data). As is observed for the vast majority of superconducting materials without phase change, the dependence of $T_c$ on pressure for MgB$_2$ is single-valued and does not depend on the pressure/temperature history of the sample; such history effects do occur in certain high-$T_c$ oxides containing mobile species at RT$^{[23]}$. We thus find that for He-gas pressure changes at both ambient and low temperature, $T_c(P)$ for MgB$_2$ is a linear, reversible function of pressure to 0.7 GPa$^{[24]}$.

In the present experiments the sample is surrounded by fluid helium near $T_c \simeq 39$ K for all data taken at pressures $P \leq 0.50$ GPa so that the slope $dT_c/dP \simeq -1.11$ K/GPa gives the true hydrostatic pressure dependence of $T_c$. The fact that the sample is in solid helium for $P > 0.5$ GPa is seen in Fig. 2 to have no effect on the pressure dependence $T_c(P)$; indeed, solid helium is the softest solid known. Our value of $dT_c/dP$ differs significantly from those of other groups$^{[17, 18, 19]}$ (see discussion above) using pressure media which are either solid at RT or freeze upon cooling down at temperatures well above $T_c$.

In view of the strong compressibility anisotropy$^{[12]}$ and the sizeable anharmonicity and non-linear electron-phonon coupling$^{[8]}$ anticipated for MgB$_2$, it is likely that shear stresses of sufficient magnitude will cause appreciable changes in the pressure dependence of $T_c$, as observed for other anisotropic substances such as the superconducting oxides$^{[25]}$ and organic superconductors$^{[26]}$. The largest shear stresses are generated by changing the pressure on a solid pressure medium, such as steatite, or using no pressure medium at all. The shear stresses generated in cooling Fluorinert or other comparable liquids through the melting curve are admittedly much smaller and depend on details of the individual experimental procedures used, such as the cooling rate, change in applied force upon cooling, etc. Only experiment
can determine whether or not the $T_c(P)$ dependences measured in the Fluorinert experiments\cite{17,18} are influenced by shear stresses. To exclude the possibility of sample-dependent effects, such experiments should be carried out on a single sample. Lorenz \textit{et al.}\cite{27} have very recently carried out He-gas high-pressure studies on the same sample studied by them previously in Fluorinert\cite{17} and find a value of $dT_c/dP$ equal to their previous value (-1.6 K/GPa), within experimental error; further measurements in the same He-gas system on a second, high-quality sample yielded the dependence $dT_c/dP \simeq -1.07$ K/GPa, a value very close to our present result. This finding lends support to the observation by Monteverde \textit{et al.}\cite{19} that $dT_c/dP$ in MgB$_2$ may be sample dependent.

The present $T_c(P)$ studies and parallel high-pressure structural studies by Jorgensen \textit{et al.}\cite{12} were both carried out on the same MgB$_2$ sample over the same He-gas pressure range, thus allowing an accurate determination of the change in $T_c$ with lattice parameter. The change in $T_c$ with unit cell volume, for example, is given by

$$d\ln T_c/d\ln V = B T_c \left( dT_c/dP \right) = +4.16 \pm 0.08,$$

(1)

using the above values $dT_c/dP \simeq -1.11 \pm 0.02$ K/GPa, $B = 147.2 \pm 0.7$ GPa, and $T_c = 39.25$ K.

We will now discuss the implications of this result for the nature of the superconducting state in MgB$_2$. First consider the McMillan equation\cite{28}

$$T_c \simeq \left( \langle \omega \rangle / 1.20 \right) \exp \left\{ \left[ -1.04(1 + \lambda) \right] / \left[ \lambda - \mu^*(1 + 0.62\lambda) \right] \right\},$$

valid for strong coupling ($\lambda \lesssim 1.5$), which connects the value of $T_c$ with the electron-phonon coupling parameter $\lambda$, an average phonon frequency $\langle \omega \rangle$, and the Coulomb repulsion $\mu^*$, which we assume to be pressure independent. Taking the logarithmic volume derivative of $T_c$, we obtain the simple relation

$$d\ln T_c/d\ln V = -\gamma + \Delta \left\{ d\ln \eta/d\ln V + 2\gamma \right\},$$

(2)

where $\gamma \equiv -d\ln \langle \omega \rangle /d\ln V$ is the Grüneisen parameter, $\eta \equiv N(E_f) \langle I^2 \rangle$ is the Hopfield parameter\cite{23} given by the product of the electronic density of states and the average squared electronic matrix element, and $\Delta = 1.04 \lambda \left[ 1 + 0.38 \mu^* \right] / \left[ \lambda - \mu^*(1 + 0.62\lambda) \right]^2$.

Eq. (2) has a simple interpretation. The first term on the right, which comes from the prefactor to the exponent in the McMillan expression for $T_c$, is usually small relative to the second term, as will be demonstrated below. The sign of the logarithmic derivative $d\ln T_c/d\ln V$, therefore, is determined by the relative magnitude of the two terms in the curly brackets. The first “electronic” derivative is negative ($d\ln \eta/d\ln V \approx -1$ for simple metals (s,p electrons)\cite{31}, but equals -3 to -4 for transition metals (d electrons)\cite{29}), whereas the second “lattice” term is positive (typically $2\gamma \approx 3 - 5$). Since in simple-metal superconductors, like Al, In, Sn, and Pb, the lattice term dominates over the electronic term, and $\Delta$ is always positive,
the sign of $d\ln T_c/d\ln V$ is the same as that in the curly brackets, namely positive; this accounts for the universal decrease of $T_c$ with pressure due to lattice stiffening in simple metals. In selected transition metals the electronic term may become larger than the lattice term, in which case $T_c$ would be expected to increase with pressure, as observed in experiment\cite{29}.

Let us now apply Eq. (2) in more detail to a canonical BCS simple-metal superconductor. In Sn, for example, $T_c$ decreases under pressure at the rate $dT_c/dP \simeq -0.482$ K/GPa which leads to $d\ln T_c/d\ln V \simeq +7.2$ \cite{20}. Inserting $T_c(0) \simeq 3.73$ K, $\langle \omega \rangle \simeq 110$ K \cite{31}, and $\mu^* = 0.1$ into the above McMillan equation, we obtain $\lambda \simeq 0.69$ from which follows that $\Delta \simeq 2.47$. Inserting these values into Eq. (2) and setting $d\ln \eta/d\ln V \approx -1$ from above for simple metals, we can solve Eq. (2) for the Grüneisen parameter to obtain $\gamma \simeq +2.46$, in reasonable agreement with experiment for Sn ($\gamma \approx +2.1$)\cite{20}. Similar results are obtained for other simple-metal superconductors.

We now repeat the same calculation with the McMillan equation for MgB$_2$ using the logarithmically averaged phonon energy from inelastic neutron studies\cite{7} $\langle \omega \rangle = 670$ K, $T_c(0) \simeq 39.25$ K, and $\mu^* = 0.1$, yielding $\lambda \simeq 0.90$ and $\Delta \simeq 1.75$. From Eq. (1) we have $d\ln T_c/d\ln V = +4.16$. Since the pairing electrons in MgB$_2$ are believed to be s,p in character\cite{32}, we set $d\ln \eta/d\ln V \approx -1$ \cite{33}. Inserting these values into Eq. (2), we find $\gamma \simeq 2.36$, in reasonable agreement with the value $\gamma \approx 2.9$ from Raman spectroscopy studies\cite{14} or $\gamma \approx 2.3$ from \textit{ab initio} electronic structure calculations on MgB$_2$\cite{34}. If, on the other hand, one were to use the same bulk modulus but the pressure derivative $dT_c/dP \simeq -2.0$ K/GPa from Saito et al.\cite{18}, one obtains from Eq. (2) the unusually high value $\gamma \approx 3.7$.

In extensive specific heat\cite{5} and high-resolution photoemission studies\cite{35} on MgB$_2$, evidence is found for a multicomponent superconducting gap; the latter study also reports an inconsistency in the values of the electron-phonon coupling constant from McMillan’s equation and the renormalization of the electronic density-of-states. These results call into question the suitability of the isotropic McMillan equation for describing this system.

Under the above assumptions, we thus conclude that the rate of decrease of $T_c$ with pressure found in the present experiments on MgB$_2$ is consistent with BCS phonon-mediated superconductivity. The authors hope that the accurate determination of the volume dependence of $T_c$ in this work, $d\ln T_c/d\ln V = +4.16 \pm 0.08$, will stimulate \textit{ab initio} theoretical calculations. At first glance the present results appear to be inconsistent with the model of Hirsch and Marsiglio\cite{10,11} which predicts that $T_c$ should increase with pressure for optimally doped samples. However, within their model, $T_c$ for a non optimally doped sample may decrease if sufficient change in the carrier concentration occurs when pressure is applied. Further studies, such as high-pressure Hall effect measurements, are necessary to clarify this possibility.

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The volume dependence of the Hopfield parameter \( \frac{d \ln \eta}{d \ln V} \approx -1 \) used here is in reasonable agreement with that \( \frac{d \ln \eta}{d \ln V} = B \frac{d \ln \eta}{d P} \approx -0.81 \) obtained from first-principles electronic structure calculations on MgB\(_2\) by N. I. Medvedeva, A. L. Ivanovskii, J. E. Medvedeva, A. J. Freeman, and D. L. Novokov (preprint cond-mat/0104346) which obtain \( \frac{d \ln \eta}{d P} \approx +0.55 \%/\text{GPa} \) and setting \( B = 147.2 \text{ GPa} \) from Ref. 12. The resulting change in the estimate \( \gamma \approx 2.23 \) is small.

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1 Figure Captions

**Fig. 1.** Real part of the ac susceptibility of MgB$_2$ versus temperature at ambient and high pressures. The applied magnetic field is 0.113 Oe (rms) at 1,023 Hz. Intercept of straight tangent lines defines superconducting onset at ambient pressure $T_c(0) \simeq 39.25$ K. No correction is made for demagnetization effects.

**Fig. 2.** Superconducting transition temperature versus applied pressure. Numbers give order of measurement. Data for pts. 2', 6, 8', and 11 are shown in Fig. 1. A typical error bar for $T_c$ ($\pm 0.01$ K) is given in lower left corner; the error in pressure is less than the symbol size. Pressure was either changed at RT (unprimed numbers) or at low temperatures $\sim 60$ K (primed numbers).
Figure 1
Figure 2