Dependence of the Superconducting Transition Temperature of Single- and Polycrystalline MgB$_2$ Hydrostatic Pressure

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Abstract. The dependence of $T_c$ for MgB$_2$ on purely hydrostatic or nearly hydrostatic pressure has been determined to 23 GPa for single-crystalline and to 32 GPa for polycrystalline samples, and found to be in good agreement. $T_c$ decreases from 39 K at ambient pressure to 15 K at 32 GPa with an initial slope $dT_c/dP \simeq -1.11(2)$ K/GPa. Evidence is presented that the differing values of $dT_c/dP$ reported in the literature result primarily from shear-stress effects in nonhydrostatic pressure media and not differences in the samples. Although comparison of these results with theory supports phonon-mediated superconductivity, a critical test of theory must await volume-dependent calculations based on the solution of the anisotropic Eliashberg equations.

1 Introduction

In spite of considerable efforts during the 20 months since the discovery of superconductivity in MgB$_2$ at 39 K [1], the search for a further binary compound with a higher value of $T_c$ has yet to bear fruit [3]. In contrast, the high-$T_c$ oxide Y-Ba-Cu-O ($T_c \approx 92$ K) was synthesized [3] only a few weeks after the landmark discovery of superconductivity near 35 K in La-Ba-Cu-O [4]. Progress with the binary compounds may have to wait until we first reach a clear understanding of MgB$_2$’s extraordinary superconducting and normal-state properties, most of which are highly anisotropic. The compressibility, for example, is 64% larger along the $c$ axis than along the $a$ direction [5]. Under $c$-axis compression the electronic density of states $N(E_f)$ is predicted to decrease much more rapidly ($\sim 0.6\%$/GPa) than under hydrostatic compression ($\sim 0.1\%$/GPa) [6], the $\sigma$ band shifting downward and the hole number in the $\sigma$ band decreasing [7]. The anisotropy in the upper critical field $H_{c2}$ increases strongly with decreasing temperature [8] [9], approaching a value near 7 at 0 K [8]. A large number
of experiments point to the existence of at least two superconducting energy gaps \[10\] which are predicted \[11, 12, 13\] to open up on different parts of the anisotropic Fermi surface. Gonnelli \textit{et al.}\[14\] have very recently provided direct evidence for two-band superconductivity in MgB\(_2\) where the temperature-dependence of the distinct gaps originating from the \(\sigma\) and \(\pi\) bands were measured using point-contact spectroscopy on single crystals.

The existence of multiple gaps and the highly anisotropic electronic and lattice-vibration properties of MgB\(_2\) have recently prompted Choi \textit{et al.}\[13\] to attempt the solution of the fully anisotropic Eliashberg equations. A comparison of their calculation with experiment has yielded promising results \[12, 13\]. A critical test of their approach would be provided by extending their calculation to reduced lattice parameters, allowing a direct comparison with the results of high pressure experiments.

Following the discovery of a new superconductor, high pressure studies are often among the first to be carried out. The reason for this is that the magnitude and sign of \(dT_c/dP\) help guide the materials scientist how to best modify the superconductor to raise \(T_c\) at ambient pressure, a case in point being the discovery of Y-Ba-Cu-O \[3\]. In addition, the high-pressure technique, sometimes in conjunction with high temperatures, is also invaluable to: (1) create new superconductors (e.g. sulfur \[15\] or oxygen \[16\] which metallize at Mbar pressures); (2) vary the properties of known superconductors (e.g. polycrystalline MgB\(_2\) \[17\] or the synthesis of MgB\(_2\) single crystals \[18, 19, 20\]); (3) induce structural phase transitions (e.g. in Ba, As, Bi, Sn, Ga and Tl \[21\]); and, finally, (4) vary the lattice parameters to help identify the pairing mechanism and critically test theoretical models. Unfortunately, this final application has been traditionally underutilized by theorists.

Hydrostatic or uniaxial pressure experiments on single crystals determine the dependence of a given property solely on the lattice parameters. Most high-pressure experiments, however, are carried out under nonhydrostatic conditions which subject the sample to a distribution of unspecified shear stresses. These shear stresses may be large enough to plastically deform the sample, resulting in lasting distortions and lattice defects. The pressure dependence of \(T_c\) may thus depend on whether the pressure medium is hydrostatic or not, particularly in elastically anisotropic materials like quasi 1D and 2D organic superconductors \[22\] or high-\(T_c\) oxides \[23\]. Studies on single crystals are of particular value since strain effects from grain boundaries in polycrystalline materials are avoided.

Several studies of the dependence of \(T_c\) on pressure for polycrystalline MgB\(_2\) were carried out shortly after the discovery of its superconductivity \[24, 25, 26, 27\]. Using solid steatite pressure medium to 20 GPa, Monteverde \textit{et al.}\[24\] reported that \(T_c\) decreased under pressure at different initial rates (-0.35 to -0.8 K/GPa) for each of the four samples studied. On the other hand, in an experiment in fluid Fluorinert to \(\sim 1.7\) GPa, Lorenz \textit{et al.}\[25\] and Saito \textit{et al.}\[26\] found \(dT_c/dP \approx -1.6\) K/GPa and -1.9 K/GPa, respectively. The first truly hydrostatic measurement of \(T_c(P)\) was carried out by our group to 0.7 GPa using He gas on an isotopically pure \(^{11}\)B)
sample, revealing that $T_c$ decreases reversibly under hydrostatic pressure at the rate $dT_c/dP \simeq -1.11(2)$ K/GPa [27]; later work on isotopically pure $^{10}$B and $^{11}$B samples yielded dependences between -1.09(4) K/GPa and -1.12(3) K/GPa [28]. In further He-gas studies, Lorenz et al. [29] and Schlachter et al. [30] obtained -1.07 K/GPa and -1.13 K/GPa, respectively. Experiments utilizing fluid pressure media were carried out by Razavi et al. (-1.18(6) K/GPa) [31], Choi et al. (-1.36 K/GPa) [32], and Kazakov et al. (-1.5 K/GPa) [33]. The first high-pressure measurements on a single crystal were carried out by Masui et al. [34] using Fluorinert, yielding $dT_c/dP \simeq -2.0$ K/GPa. In the above experiments the values of $dT_c/dP$ reported are seen to differ by more than a factor of two. Experiments to much higher pressures have been carried out by several groups and will be discussed below. The results of all known $dT_c/dP$ measurements on MgB$_2$ are summarized in the Table.

In this paper we provide evidence that the variation in the reported $T_c(P)$ dependences for MgB$_2$ is primarily a result of shear stresses exerted by the solidified pressure media on the sample. From recent measurements on high quality single-crystalline and polycrystalline samples we conclude that the initial dependence of the transition temperature on purely hydrostatic pressure is given by $dT_c/dP \simeq -1.11(2)$ K/GPa. Under nearly hydrostatic (dense He) pressures to 32 GPa ($V/V_0 \simeq 0.855$), we find $T_c$ to decrease monotonically from $\sim 39$ K to 15 K. Although these results appear consistent with phonon-mediated superconductivity in MgB$_2$, a quantitative check must await comparison with calculations based on the fully anisotropic Eliashberg equations.

2 Experimental methods

A wide variety of high-pressure techniques with many different pressure media have been used to study the dependence of $T_c$ on pressure [35]. All techniques discussed below have been used at some time by our group.

Perhaps the most widely used technique in the pressure range 1-2 GPa is a piston-cylinder cell in which two pistons compress a Teflon bucket containing the sample immersed in a fluid pressure medium such as Fluorinert, n-pentane/isopentane, or silicon oil. For studies to higher pressure a diamond-anvil-cell (DAC) may be used with a 4:1 methanol:ethanol mixture as pressure medium since this mixture remains fluid to approximately 10 GPa at ambient temperature. All these fluid pressure media freeze upon cooling and subject the immersed sample to shear stresses of varying strength which depend on the pressure medium used, the details of the pressure technique, and the rate at which the pressure cell is cooled, among other factors.

The only pressure medium that remains fluid under high pressure ($P \leq 0.4$ GPa) near 40 K, where MgB$_2$ superconducts, is helium and thus only high-pressure experiments on MgB$_2$ in liquid helium are able to determine the dependence of $T_c$ on purely hydrostatic pressure. At pressures above 0.5 GPa, helium is frozen below 39 K, but the pressure is still very nearly hydrostatic since solid helium is the
softest solid known; in addition, if the proper cooling procedure is followed, a single crystal of helium can be grown around the sample, resulting in negligible shear stresses which permit even de Haas van Alphen measurements on single crystals at very low temperatures [36]. High-pressure studies in liquid helium are thus the measurements-of-choice for exacting quantitative studies of the properties of solids under high-pressure conditions.

At the other end of the spectrum, in some high pressure techniques solid pressure media (e.g., steatite, NaCl or NaF) are used [35]. With a solid pressure medium, the application of pressure may subject the sample to relatively large shear stresses, possibly strong enough to plastically deform the sample or even crush it, thus introducing a large number of lattice defects. The magnitude and direction of the shear stresses depend, among other things, on the pressure medium used, the temperature at which the pressure is changed, the pressure range, and whether the ring containing the solid pressure medium is supported by a “belt” or not. With this so-called “quasihydrostatic” technique $T_c$ may not be a reversible function of pressure. For these reasons high-pressure techniques using solid pressure media should be avoided in quantitative studies, particularly when studying elastically anisotropic materials such as the high-$T_c$ oxides [23], organic superconductors [22], and MgB$_2$.

In the present experiments, dense helium is used as pressure medium both in a He-gas pressure cell to 1 GPa and in a DAC to over 30 GPa. The pressure in the cell is measured at temperatures within a few degrees of the transition temperature $T_c$; the superconducting transition is detected in a sensitive ac susceptibility measurement. Details of these pressure techniques have been published elsewhere [37]. Isotopically pure polycrystalline MgB$_2$ samples were synthesized at the Argonne National Labs [5, 38]. Single crystals were grown at the Superconductivity Research Laboratory (ISTEC) in Tokyo, Japan; following the crystal growth at elevated temperatures, “type A” (“type B’) crystals were quenched (slow cooled) [18]. The dimensions of the crystals studied in the He-gas cell were approximately $0.15 \times 0.4 \times 0.03$ mm$^3$ and in the DAC $\sim 0.09 \times 0.09 \times 0.03$ mm$^3$.

### 3 Results of experiment

In Figs. 1-3 we show the results of our measurements of the pressure dependence of $T_c$ on MgB$_2$ single crystals. The midpoint of the superconducting transition in the real part of the ac susceptibility $\chi'$ is used to define $T_c$ [39], as seen in Fig. 1. The two “type B” crystals have higher values of $T_c$ ($\sim 38.2$ K) than the “type A” crystal ($\sim 37.2$ K). For both “type A” and “type B” crystals $T_c$ is seen in Fig. 2 to decrease reversibly and linearly with hydrostatic (He-gas) pressure. As with the polycrystalline samples, the same $T_c(P)$ dependence is obtained whether the pressure is changed at low temperatures or at room temperature. As seen in the Table, the pressure dependence of $T_c$ in the He-gas experiments is very nearly the same whether MgB$_2$ is in single- or polycrystalline form.
In Fig. 3 we extend the He-gas results on the same “type B” single crystal \[40\] to much higher pressures using a He-loaded DAC. The initial \(T_c(P)\) dependence agrees well with the data in Fig. 2, showing a positive curvature at higher pressures. As seen in Fig. 3, this curvature is removed if \(T_c\) is plotted versus the relative unit-cell volume \(V/V_o\) \[41\]. The linearity of the \(T_c\) versus \(V/V_o\) data over such a wide range of pressure is remarkable.

In Fig. 4 are shown our \(T_c(P)\) data on an isotopically pure \(^{11}\)B polycrystalline sample to 32 GPa from three separate experiments in a He-loaded DAC \[42\]; the sample used is from the same synthesis batch as in our earlier He-gas studies to 0.7 GPa \[27\]. \(T_c\) decreases monotonically and reversibly with pressure from \(\sim 39\) K at ambient pressure to \(\sim 15\) K at 32 GPa. The initial slope, \(dT_c/dP \approx -1.1\) K/GPa, is the same as in the He-gas data. As will be discussed below (see Fig. 7), the single-crystal and polycrystalline data are in good agreement.

The only other \(T_c(P)\) measurements on polycrystalline MgB\(_2\) to very high pressures using dense He as pressure medium were carried out by Struzhkin et al. \[43\] and are included in Fig. 4 for the \(^{11}\)B isotope; these authors use a double-modulation ac susceptibility technique which determines the superconducting onset rather than the superconducting midpoint. As seen in Fig. 4, the agreement with our data is remarkably good to 20 GPa, but begins to deviate at higher pressures. Parallel studies by the same authors \[43\] to 44 GPa in dense He on a \(^{10}\)B isotopic sample yield a similar \(T_c(P)\) dependence which lies \(\sim 1\) K above our \(^{11}\)B data below 20 GPa, but gradually merges at higher pressures. Unlike our data in Figs. 3 and 4, where \(T_c\) is seen to be a linear function of \(V/V_o\), the data of Struzhkin et al. \[43\] show a break in slope \(dT_c/dV\) near 15-20 GPa. For a full discussion of the latter data see the paper by Goncharov and Struzhkin in this special edition.

Non-He-gas studies to very high pressures include those of Tang et al. \[44\] to 9 GPa using a cubic-anvil cell with Fluorinert pressure medium; the \(T_c(P)\) dependence is in good agreement with that in Fig. 4. However, in DAC studies using methanol-ethanol, Tissen et al. \[45\] found a much larger initial slope \((dT_c/dP \approx -2\) K/GPa\) accompanied by a relatively large drop in \(T_c\) to approximately 6 K at 28 GPa; these authors interpret a break in slope \(dT_c/dP\) near 10 GPa as evidence for a topological transition. We note that the present \(T_c(P)\) data lie 1-2 K below those of Razavi et al. \[31\] to 11 GPa obtained using steatite pressure medium. In further DAC measurements using steatite Bordet et al. \[10\] and Monteverde et al. \[24\] report \(T_c(P)\) dependences for four different samples which generally lie well above those in the present measurement.
4 Discussion

4.1 Intrinsic Dependence of $T_c$ on Pressure

Before attempting a quantitative analysis of the present data, we would like to first discuss possible origins for the differing values of $dT_c/dP$ for MgB$_2$ reported in the literature (see the Table). Tissen et al. [45] and, somewhat later, Lorenz et al. [47] have presented data indicating a strong inverse correlation between the magnitude of the initial slope $dT_c/dP$ and the value of $T_c$ at ambient pressure, i.e. $|dT_c/dP|_0$ is larger if $T_c(0)$ is smaller. To reexamine this possible correlation, we have plotted in Fig. 5 the initial slope $dT_c/dP$ using the high-pressure data in the Table. Taken as a whole, the data in Fig. 5 would appear to give some support to the proposed strong inverse correlation. Such a correlation is, however, not supported by the hydrostatic He-gas data. With the exception of the single data point of Lorenz et al. [29], all known $(dT_c/dP)_0$ values for single- or polycrystalline samples obtained using He pressure medium lie between -1.07 and -1.2 K/GPa. Although the He-gas data do not support a strong correlation between $|dT_c/dP|_0$ and $T_c(0)$, samples with lower values of $T_c(0)$ (37.16 K versus 39.1 K) do appear to exhibit slightly (~ 5%) larger initial slopes $dT_c/dP$ (-1.17 K/GPa versus -1.11 K/GPa). In contrast, the $(dT_c/dP)_0$ values obtained using other less hydrostatic pressure media are often larger (up to 50-80%) in magnitude. This indicates that differences in the values of $(dT_c/dP)_0$ in the literature may depend more on the pressure medium used than on differences between samples, at least as reflected in their $T_c(0)$ values.

One way to help resolve this issue is to carry out parallel $T_c(P)$ measurements on the same sample using different pressure media. We carried out two such studies. In the first, we placed a sample provided by V. Tissen in our He-gas system, obtaining under purely hydrostatic pressure conditions $dT_c/dP \simeq -1.2$ K/GPa [48]. This value is 40% less than that (-2 K/GPa) from Tissen et al.’s DAC study [45] on the same sample with methanol/ethanol pressure medium (see the appropriate vertical arrow in Fig. 5). In the second experiment we took the same isotopically pure $^{11}$B sample studied previously in both our He-gas and He-loaded DAC (where $dT_c/dP \simeq -1.1$ K/GPa) and replaced the He pressure medium with Fluorinert, obtaining the DAC data shown in Fig. 4 with initial slope $dT_c/dP \simeq -1.6$ K/GPa, a nearly 50% increase in slope (see the appropriate vertical arrow in Fig. 5). Similar effects are observed with single crystals; the slope -2.0 K/GPa observed in Fluorinert [31] is much larger than that (-1.1 K/GPa) found in the present He-gas study on crystals from the same source. The large values of $|dT_c/dP|_0$ reported in the literature thus appear to arise from the use of fluid pressure media, such as Fluorinert or methanol/ethanol, which freeze solid at temperatures well above $T_c \approx 40$ K. The present results thus do not support the existence of the originally proposed [45, 47] strong inverse correlation between $|dT_c/dP|_0$ and $T_c(0)$.

It is interesting to note that the DAC measurements of Schlachter et al. [30]
with solid NaF pressure medium yield a large negative slope with increasing pressure (-1.6 K/GPa) which reduces to -1.13 K/GPa as the pressure is reduced to ambient, leaving an ambient pressure value $T_c(0)$ permanently suppressed by $\sim 12\%$ from the initial value. The authors infer that, because of shear stress effects from the solid pressure medium, the sample is degraded when pressure is applied, with no further degradation upon pressure release. It is also noteworthy that the value $dT_c/dP \simeq -1.13$ K/GPa is the same to 0.4 GPa as that obtained on the same sample in a He-gas experiment by the same group (see Table).

With these results in mind, it is difficult to understand the relatively small values of $|dT_c/dP|_0$ reported in the resistivity studies by Monteverde et al. [24] and Bordet et al. [46] using solid steatite pressure medium. Since their temperature-dependent resistivity data at different pressures was not published, it is difficult to speculate on possible origins for this difference. The use by these authors of the resistivity onset to define $T_c$ could lead to substantial errors in estimating $(dT_c/dP)_0$, particularly if significant broadening occurs in the transition under pressure. We also note that in this type of pressure cell it is particularly difficult to obtain reliable resistivity data and pressure values in the lower 1-2 GPa pressure range. In these papers [24, 46] no information was given whether $T_c(P)$ was reproducible for identical samples, reversible in pressure, or whether the transition broadened significantly under pressure. We note that Struzhkin et al. [43] remeasured $T_c(P)$ in their DAC with no pressure medium whatsoever, pressing the diamond anvils directly onto the sample and gasket, and reported results resembling the two lower curves of Monteverde et al. [24]. It is also noteworthy that Razavi et al. [31] also used steatite pressure medium, and obtained the initial slope -1.03 K/GPa; in their published resistivity data there is little change in the transition width with pressure so that the estimate of the shift in $T_c$ is relatively straightforward.

In a further experiment, we left out the pressure medium entirely and pressed two WC Bridgman anvils with 6 mm dia. flats for 5 minutes directly onto $\sim 5$ mg MgB$_2$ powder with 10 tons force; this typically results in a pressure distribution across the center of the disc resembling a bell-shaped curve with $P \approx 8$ GPa at the center and $P = 0$ at the edge. The sample was thoroughly compacted by this procedure, resulting in a disc-shaped sample approximately 70 $\mu$m thick. After removing the compacted disc from between the anvils, the disc was separated into three separate regions (center, middle, and outside), as seen in Fig. 6; the sample from each region was then gently broken up and placed into our ac susceptibility coil system. As seen in Fig. 6, the sharp superconducting transition of the virgin powder sample is broadened by a significant amount ($\sim 10$ K) by the compaction procedure, the broadening being somewhat less from the center to the middle to the outside. This broadening is presumably the result of strong plastic deformation and the resulting internal strains in the material. If strong internal strains can lead to broadening as large as 10 K at ambient pressure, it is not unreasonable to assume that such strains are capable of causing enhanced downward shifts in $T_c$ under nonhydrostatic pressure...
conditions.

The important role that defects and strains play in MgB$_2$’s superconducting state is emphasized by the fact that the value of $T_c$ in a large number of thin-film and bulk MgB$_2$ samples appears to follow a Testardi correlation [4, 17]: $T_c$ is lower for samples in which the conduction electrons are strongly scattered. Lorenz et al. [17] also report that the value of $T_c$ is degraded with increasing lattice strain, a result confirmed by Serquis et al. [19]. $T_c$ in MgB$_2$ has also been found to be lowered following mechanical milling [50] and after irradiation by fast neutrons [51]. In contrast to the work of Lorenz et al. [17] and Serquis et al. [19], Hinks et al. [38] have recently reported that accidental impurity doping can have an effect on $T_c$ much larger than that of lattice strain. Additionally, they show that grain-interaction stresses can significantly alter the lattice parameters of MgB$_2$ depending on the impurity phases present with the MgB$_2$. For example, the largest strains are seen for samples where MgB$_4$ is present as an impurity phase, imparting to the bulk sample the properties of an MgB$_2$/MgB$_4$ composite. One might hypothesize that grain-interaction stresses could significantly alter the response of MgB$_2$ grains to pressure in such samples, and, in this way, modify the observed response of $T_c$ to pressure.

In contrast to the poor agreement between the large body of nonhydrostatic $T_c(P)$ data on MgB$_2$, the agreement between the purely hydrostatic He-gas data to 0.7 GPa and the nearly hydrostatic dense He data to 20 GPa (see Fig. 4) is remarkably good for both single-crystalline and polycrystalline samples. It is thus reasonable to assert that the intrinsic initial pressure dependence for MgB$_2$ samples with the highest values of $T_c$ (39.1 K for $^{11}$B) is given by $dT_c/dP \simeq -1.11(2)$ K/GPa, the $T_c(P)$ dependence to 32 GPa being given by the present data shown in Figs. 3 and 4. We would now like to compare these experimental results with theoretical models.

### 4.2 Comparison with Theory

For most known superconductors, including MgB$_2$, $T_c$ is found to decrease with pressure. In fact, $dT_c/dP$ is negative for all simple-metal superconductors (e.g. Pb, Al, Sn, and In [52]) due to pressure-induced lattice stiffening (higher phonon frequencies) which weakens the electron-phonon coupling [23]. Rb$_3$C$_{60}$ and other alkali-doped fullerenes are exceptions to this “rule”: here the rapid decrease in $T_c$ with pressure originates not from lattice stiffening but rather from a sharp decrease in the electronic density of states $N(E_f)$ [53]. What is the origin of the negative pressure dependence of $T_c$ for MgB$_2$ - is it lattice stiffening, a decrease in the electronic density of states, or something else?

Theoretical models are calculated in terms of the dependence of the relevant properties on the lattice parameters or unit cell volume $V$. From the intrinsic initial slope $dT_c/dP \simeq -1.11$ K/GPa for MgB$_2$ we can calculate the logarithmic volume derivative

$$\frac{d\ln T_c}{d\ln V} = -\frac{B}{T_c(0)} \left( \frac{dT_c}{dP} \right) = +4.18,$$

(1)
where we use $T_c(0) = 39.1$ K from above and the bulk modulus $B = 147.2$ GPa from He-gas neutron diffraction studies on the same sample [5].

We now convert the $T_c(P)$ data for polycrystalline MgB$_2$ in Fig. 4 to $T_c(V)$ data using the Murnaghan equation of state [11]. In Fig. 7 we compare the resulting $T_c(V)$ dependences to that for the type “B” single crystal from Fig. 3; the agreement is excellent considering that $T_c(0)$ for the single crystal is nearly 1 K lower than for the polycrystalline sample.

We now attempt a fit to the data in Fig. 7 using the well known McMillan formula [54]

$$T_c \approx \langle \omega \rangle \exp \left\{ \frac{-1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right\}, \tag{2}$$

which is valid for strong coupling ($\lambda \lesssim 1.5$) and connects the value of $T_c$ with the electron-phonon coupling parameter $\lambda$, an average phonon frequency $\langle \omega \rangle$, and the Coulomb repulsion $\mu^*$. We should not expect too much from this fit since the McMillan formula is a solution of the isotropic Eliashberg equations and thus ignores the strong anisotropies in the vibrational, electronic, and superconducting properties of MgB$_2$. In the following we draw heavily on the detailed analysis of Chen et al. [55] who consider the effect of pressure on the three relevant parameters in Eq. (2) $\langle \omega \rangle$, $\lambda$, and $\mu^*$.

Taking the logarithmic volume derivative of both sides of Eq. (2), we obtain the simple relation

$$\frac{d \ln T_c}{d \ln V} = -\gamma - \Delta_1 \left\{ \frac{\partial \ln \mu^*}{\partial \ln V} \right\} + \Delta_2 \left\{ \frac{\partial \ln \eta}{\partial \ln V} + 2\gamma \right\}, \tag{3}$$

where $\gamma \equiv -\partial \ln \langle \omega \rangle / \partial \ln V$ is the Grüneisen parameter, $\eta \equiv N(E_f) \langle I^2 \rangle$ is the McMillan-Hopfield parameter given by the product of the electronic density of states and the average squared electronic matrix element, and the dimensionless prefactors are given by $\Delta_1 = 1.04\mu^*(1 + \lambda) \left[ 1 + 0.62\lambda \right] / \left[ \lambda - \mu^*(1 + 0.62\lambda) \right]^2$ and $\Delta_2 = 1.04\lambda \left[ 1 + 0.38\mu^* \right] / \left[ \lambda - \mu^*(1 + 0.62\lambda) \right]^2$. Both $\Delta_1$ and $\Delta_2$ are calculated using the values of $\lambda$ and $\mu^*$ at ambient pressure.

Eq. (3) looks rather formidable, but has a very simple interpretation. Fortunately, the first two terms on the right are usually small relative to the third term, as we will see below, so that $d \ln T_c/d \ln V \approx \Delta_2 \left\{ \partial \ln \eta / \partial \ln V + 2\gamma \right\}$. Since $\Delta_2$ is always positive, the sign of the logarithmic derivative $d \ln T_c/d \ln V$ is determined by the relative magnitude of the two terms $\partial \ln \eta / \partial \ln V$ and $2\gamma$. The first “electronic” term is negative ($\partial \ln \eta / \partial \ln V \approx -1$ for simple metals (s,p electrons) [23], but may equal -3 to -4 for transition metals (d electrons) [56]), 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, the sign of $d \ln T_c/d \ln V$ is the same as that of $\{ \partial \ln \eta / \partial \ln V + 2\gamma \}$, 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\[56].

We now apply the McMillan equation to the above results on MgB$_2$ using at
ambient pressure the logarithmically averaged phonon energy from inelastic neutron
studies\[57] $\langle \omega \rangle = 670$ K, $T_c(0) \simeq 39.1$ K, and $\mu^* = 0.1$ \[58], yielding $\lambda \simeq 0.898$, $\Delta_1 = 0.558$ and $\Delta_2 = 1.76$. From the expression derived by Chen et al.\[55] for s,p metals $\partial \ln \eta / \partial \ln V = -[\partial \ln N(E_f)/\partial \ln V] - 2/3$ and the value $\partial \ln N(E_f)/\partial \ln V = +0.46$ from Loa and Syassen\[59], the dependence of the Hopfield parameter is estimated to be $\partial \ln \eta / \partial \ln V = -1.13$, a value reasonably close to the generic value (-1) used above in the analysis for the simple-metal superconductors and to the value (-0.81) obtained from first-principles electronic structure calculations on MgB$_2$ by Medvedera et al.\[60]. Chen et al.\[55] find that $\mu^*$ increases only weakly with pressure at a rate $\phi = \partial \ln \mu^* / \partial \ln V = -0.1 \gamma - 0.035$; note that this derivative is very small ($\sim -0.3$) so that the second term on the right side of Eq. (3) is relatively unimportant. We now have estimates of all quantities on the right side of Eq. (3) except $\gamma$ which we use as a fit parameter. Setting the left side of Eq. (3) equal to the experimental value $+4.18$ from Eq. (1), we find $\gamma = 2.39$, in reasonable agreement with the value $\gamma \approx 2.9$ from Raman spectroscopy studies\[61] or $\gamma \approx 2.3$ from ab initio electronic structure calculations on MgB$_2$\[62]. Note that for the present “type B” crystal $T_c(0) \simeq 38.24$ K and $dT_c/dP \simeq -1.10$ K/GPa which gives $d \ln T_c / d \ln V = +4.23$, $\lambda \simeq 0.887$, and $\gamma = 2.36$.

Eq. (3) is only valid for small changes in the parameters, i.e. for experiments to
a few GPa pressure where the change in unit cell volume is only a few percent. To
attempt to fit the very high pressure $T_c(V)$ data in Figs. 3 and 7, we need to use the full McMillan equation and insert explicitly the change in the parameters with
relative volume. As suggested by Chen et al.\[55], we set

$$\langle \omega \rangle = \langle \omega \rangle_0 (V/V_0)^{-\gamma}, \quad \lambda = \lambda_0 (V/V_0)^{\phi}, \quad \text{and} \quad \mu^* = \mu^*_0 (V/V_0)^{\phi},$$

where $\phi \equiv \{\partial \ln \eta / \partial \ln V + 2\gamma\}$. Using the values of the parameters for the sample
with $T_c(0) = 39.1$ K used above to fit the initial pressure dependence ($\gamma \simeq 2.4$, $\phi = -1.13 + 2 \times 2.4 = 3.67$, and $\phi = -0.1 \times 2.4 - 0.035 = -0.275$ and inserting the volume dependences from Eq. (4) into the McMillan equation, the lower solid
fit curve in Fig. 7 is obtained. Note that this curve clearly lies below the data at
higher pressures. A reasonably good fit to the data over the pressure range to 32
GPa ($V/V_0 = 0.855$) is found for $\gamma = 2.2$ (see Fig. 7). As with the simple s,p
metal superconductors, $T_c$ in MgB$_2$ appears to decrease under pressure due to lattice
stiffening. Note that within this approximation $T_c$ approaches 0 K asymptotically
at very high pressures. The fit curve for $\gamma = 2.2$ is predicted to fall below 1 K for
$V/V_0 = 0.73$ which corresponds to an applied pressure of 93 GPa. The fact that the
experimental data can be well fit by the McMillan formula with reasonable values of
the parameters lends support to the view that MgB$_2$ is a BCS superconductor with
moderately strong electron-phonon coupling. However, the relatively small value of
the fit parameter $\gamma = 2.2$ compared to experiment is cause for concern.

As discussed in the Introduction, the binary compound MgB$_2$ is a quasi 2D system
with highly anisotropic electronic and lattice properties, including multiple supercon-ducting gaps. The above analysis of experimental data using the McMillan formula,
which represents a solution to the isotropic Eliashberg equations, is a good first step
but does not permit unequivocal conclusions. What is needed is an extension of
the solution of the fully anisotropic Eliashberg equations to reduced lattice pa-
rameters. The initial dependences $dT_C/dP \simeq -1.11(2)$ K/GPa and $d\ln T_C/d\ln V \simeq
+4.18$, and the $T_C(P)$ and $T_C(V)$ data to 32 GPa in Figs. 3, 4, and 7 stand ready to
provide a stringent test of such a calculation.

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We use the midpoint of the superconducting transition in $\chi'$ to define $T_c$ rather than the onset. Since the transition broadens somewhat at higher pressures due to pressure inhomogeneity and/or strain effects from the solid He pressure medium, the midpoint would be expected to give a temperature closer to the average in the sample than the onset.

The small “type B” crystal (approximately $0.09 \times 0.09 \times 0.03$ mm$^3$) used in the DAC experiment was actually cut off from the larger crystal used to obtain the He-gas data (●) in Fig. 2.

We use the Murnaghan equation-of-state $V/V_o = [1 + B'P/B]^{-1/B'}$ with the value $B = 147.2$ GPa from Ref. [3] and the canonical value $B' \equiv dB/dP = 4$ supported by the $V(P)$ data to 40 GPa [16] and a recent calculation [59].

The $T_c(P)$ data to 24 GPa are taken from Ref. [28] after correcting the pressure values for a spectrometer miscalibration.

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5 Figure Captions

Fig. 1. Real part of the ac susceptibility $\chi'$ (1023 Hz, 1 Oe (rms)) versus temperature for a “type B” MgB$_2$ single crystal at three different pressures. The position of the superconducting midpoint is indicated for ambient pressure data.

Fig. 2. Dependence of the superconducting transition temperature of one “type A” ($\bullet$) and two “type B” ($\bullet$, $\times$) MgB$_2$ single crystals on hydrostatic He-gas pressure. Numbers give order of measurement. Pressure was normally changed at room temperature; for data with primed numbers the pressure was changed at low temperatures ($\sim$ 50 K). Solid and dashed lines are guides to the eye.

Fig. 3. Dependence of the superconducting transition temperature of “type B” MgB$_2$ single crystal on nearly hydrostatic pressure in a He-loaded DAC. All data taken in order of increasing pressure, where pressure was only changed at ambient
temperature. \( T_c \) is determined from the midpoint of the superconducting transition in the temperature-dependent ac susceptibility \( \chi' \) (1003 Hz, 3 Oe (rms)). Dashed line gives slope \( dT_c/dP \approx -1.10 \text{ K/GPa} \) of He-gas measurement on same crystal (Fig. 2). The “error bars” give the temperatures of the onset and end of the superconducting transition; note that the transition broadens for \( P \geq 14 \text{ GPa} \). \( T_c \) versus relative volume \( V/V_0 \) is also shown; straight solid line is guide to the eye.

**Fig. 4.** Dependence of the superconducting transition temperature of isotopically pure \( ^{11}\text{B} \) polycrystalline MgB\(_2\) on nearly hydrostatic pressure in He-loaded DAC measurements: three different experiments (\( \bullet, □, ▲ \)), closed symbols (increasing pressure), open symbols (decreasing pressure); measurements from Struzhkin et al. \[43\] (\( × \)). Measurements in our DAC on same sample with Fluorinert pressure medium (\( ♦ \)) are also shown; solid and dashed lines gives slopes (-1.11 K/GPa) and (-1.6 K/GPa) from He-gas \[27\] and Fluorinert data, respectively, on same sample.

**Fig. 5.** Initial pressure derivative \( (dT_c/dP)_0 \) versus value of \( T_c \) at ambient pressure; data are taken directly from the Table. Measurements with He (\( \bullet, ◦ \)) and non-He (\( +, × \)) pressure media; both polycrystalline (\( \bullet, + \)) and single-crystalline (\( ◦, × \)) samples are represented. Vertical arrows show change in measured value of \( (dT_c/dP)_0 \) for a given sample upon changing to He pressure medium (see text).

**Fig. 6.** Relative change in the real part of the ac susceptibility \( \chi' \) of MgB\(_2\) versus temperature for both loose powder and flat, compacted samples. For the compacted disc, samples were taken from the center, middle, and outside regions.

**Fig. 7.** \( T_c \) values from present single-crystalline (open stars) and polycrystalline (\( \bullet, □, ▲ \)) data from Figs. 3 and 4 plotted versus relative volume. Solid lines are calculated curves using McMillan’s equation (Eq. 2) for three different values of the Grüneisen parameter \( \gamma \) (see text).
Table. Summary of available high-pressure $T_c(P)$ data on MgB$_2$ single-crystals (first 4 rows) and polycrystals (remaining rows). $T_c$ values are at ambient pressure from the superconducting midpoint in the ac susceptibility $\chi_{ac}$ and electrical resistivity $\rho$ measurements; Struzhkin et al. \cite{43} use a double-modulation technique $\chi_{ac}^{mod}$ which is believed to give the superconducting onset. $(dT_c/dP)_0$ is the initial pressure derivative. $P_{max}^{\text{max}}$(GPa) is the maximum pressure reached in the experiment. Unless otherwise specified, samples with the natural boron isotopic abundance $^{10.81}$B are studied. Arrows indicate increasing (↑) or decreasing (↓) pressure.

| $T_c(0)$ (K) | $(dT_c/dP)_0$ (K/GPa) | $P_{max}^{\text{max}}$(GPa) | measurement | pressure medium | reference |
|---------------|------------------------|-----------------------------|-------------|-----------------|-----------|
| 38.24         | -1.10(3)               | 0.63, 23                    | $\chi_{ac}$, “B” crystal | helium        | this paper |
| 38.27         | -1.14(3)               | 0.61                        | $\chi_{ac}$, “B” crystal | helium        | this paper |
| 37.16         | -1.17(4)               | 0.4                         | $\chi_{ac}$, “A” crystal | helium        | this paper |
| 38.0          | -2.0                   | 1.4                         | $\rho$, crystal          | Fluorinert    | 34        |
| 39.1          | -1.11(2)               | 0.66                        | $\chi_{ac}$, $^{11}$B    | helium        | 22        |
| 39.1          | -1.09(4)               | 0.63                        | $\chi_{ac}$, $^{11}$B    | helium        | 22        |
| 39.2          | -1.11(3)               | 0.61                        | $\chi_{ac}$, $^{10}$B    | helium        | 22        |
| 40.5          | -1.12(3)               | 0.64                        | $\chi_{ac}$, $^{10}$B    | helium        | 22        |
| 37.5          | -1.13                  | 0.4                         | $\chi_{ac}$              | helium        | 30        |
| 39.2          | -1.07                  | 0.84                        | $\chi_{ac}$              | helium        | 29        |
| 37.4          | -1.45                  | 0.84                        | $\chi_{ac}$              | helium        | 29        |
| 37.3          | -1.2                   | 0.6                         | $\chi_{ac}$              | helium        | 30        |
| 39.1          | -1.1                   | 32.3                        | $\chi_{ac}$, $^{11}$B    | helium        | this paper |
| 40.2          | -1.1                   | 33                          | $\chi_{ac}^{mod}$, $^{11}$B | helium        | 43        |
| 39.2          | -1.1                   | 44                          | $\chi_{ac}^{mod}$, $^{10}$B | helium        | 43        |
| 39.1          | -1.6                   | 15                          | $\chi_{ac}^{mod}$, $^{11}$B | Fluorinert    | this paper |
| 37.4          | -1.6                   | 1.84                        | $\chi_{ac}$              | Fluorinert    | 25        |
| 37.3          | -2                     | 28                          | $\chi_{ac}$              | 4:1 methanol/ethanol | 42 |
| 38.2          | -1.36                  | 1.46                        | $\rho$                   | 1:1 daphne/kerosene | 32 |
| 37.5          | -1.9                   | 1.35                        | $\rho$                   | Fluorinert    | 29        |
| 38.3          | -1.5(1)                | 1.1                         | $\chi_{dc}$              | kerosene/mineral oil | 33 |
| 39.6          | -1.03                  | 9                           | $\rho$                   | Fluorinert    | 44        |
| 38            | -1.18(6)               | 0.8                         | $\chi_{dc}$              | silicon oil   | 31        |
| 37.5          | -1.6 (P ↑)             | 7.6                         | $\chi_{ac}$              | NaF           | 30        |
| 37.5          | -1.13 (P ↓)            | 7.6                         | $\chi_{ac}$              | NaF           | 30        |
| 39            | -1.20(9)               | 11                          | $\rho$                   | steatite      | 31        |
| ~ 35          | -0.35 to -0.8           | 33                          | $\rho$                   | steatite      | 24, 42    |

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\( \chi' \) (arb. units)

\( P(\text{GPa}) \)

\( \text{MgB}_2 \) (single crystal)

midpoint

Figure 1
Figure 2

MgB$_2$ (single crystal)

$T_c$ (K)

Pressure (GPa)

"A" crystal

-1.17(4) K/GPa

"B" crystal

-1.14(3) K/GPa

-1.10(3) K/GPa

A crystal

B crystal

1 2 3 4 5 6 7 8

1' 2' 3' 4' 5' 6' 7' 8'

-1 10(3) K/GPa

-1 14(3) K/GPa

-1 17(4) K/GPa
Figure 3
Figure 4
Figure 5
Figure 6
Figure 7