Unconventional phonon-mediated superconductivity in MgB$_2$

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Abstract. We have evaluated the total carrier mass enhancement factor $f_t$ for MgB$_2$ from two independent experiments (specific heat and upper critical field). These experiments consistently show that $f_t = 3.1 \pm 0.1$. The unusually large $f_t$ is incompatible with the measured reduced gap ($2\Delta(0)/k_B T_c = 4.1$) and the total isotope-effect exponent ($\alpha = 0.28 \pm 0.04$) within the conventional phonon-mediated model. We propose an unconventional phonon-mediated mechanism, which is able to quantitatively explain the values of $T_c$, $f_t$, $\alpha$ and the reduced energy gap in a consistent way.

The recent discovery of superconductivity near 40 K in MgB$_2$ [1] has led to a remarkable excitement in the solid-state physics community. Such high-$T_c$ superconductivity in this simple intermetallic compound immediately raises a question of whether mechanisms other than the conventional electron–phonon interaction are responsible for the superconductivity. A significant boron isotope effect ($\alpha_B = -d \ln T_c/d \ln M_B = 0.26 \pm 0.03$, where $M_B$ is the mass of boron) [2, 3]† and nearly zero magnesium isotope effect ($\alpha_{Mg} = 0.02 \pm 0.01$) [3] suggest that electron–phonon coupling plays an important role in the pairing mechanism. To explain the 40 K superconductivity, an electron–phonon coupling constant of about 1 is needed. First-principles calculations give the coupling constant of 0.7–0.9 [4]–[7], which appears to be sufficient to explain the observed high-$T_c$ superconductivity. On the other hand, recent specific heat data [8] indicate a very strong electron–phonon coupling with a total mass enhancement factor of about 3.2. This would suggest a coupling constant of 2.2 and $2\Delta(0)/k_B T_c \approx 5$

† The measured boron isotope shift is $1.0 \pm 0.1$ K [2] and $1.12 \pm 0.05$ K [3]. Using our definition $\alpha_B = -d \ln T_c/d \ln M_B$, we evaluate $\alpha_B = 0.26 \pm 0.03$. With $\alpha_{Mg} = 0.02 \pm 0.01$ [3], we obtain the total isotope exponent $\alpha = 0.28 \pm 0.04$. 

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within the conventional phonon-mediated mechanism \[9\], in contradiction with the bulk-sensitive Raman scattering experiments, which show \(2\Delta(0)/k_B T_c = 4.1\) \[10\]. Moreover, the conventional phonon-mediated mechanism alone cannot quantitatively explain the significantly reduced isotope exponent unless one assumes a very large Coulombic pseudopotential, which is not realistic for the \(sp\)-band electrons. Adding other electronic pairing interactions can in principle explain the reduced isotope exponent, but the mass enhancement factor of about 3 is difficult to be reconciled with.

Here we evaluate the carrier mass enhancement factor \(f_t\) from the measured upper critical field. The deduced \(f_t\) is \(3.1 \pm 0.1\), in remarkably good agreement with that deduced from the independent specific heat data \[8\] as well as from the penetration depth \[11\]. The large mass enhancement factor is not compatible with the measured gap amplitude and the reduced total isotope-effect exponent within the conventional phonon-mediated model. We thus propose an unconventional phonon-mediated mechanism where both the long-range Fröhlich-type and short-range electron–lattice interactions are considered and treated distinctively. Within this scenario, we are able to quantitatively explain the \(T_c\) value, the mass enhancement factor, the total isotope-effect exponent and the reduced energy gap.

For a clean superconductor, as is the case for MgB\(_2\) \[12, 13\], the zero-temperature coherence length \(\xi(0)\) is equal to the BCS coherence length \[9\], that is,

\[
\xi(0) = \frac{\hbar v_F}{\pi \Delta(0)},
\]

(1)

Here \(v_F\) is the renormalized Fermi velocity, i.e. \(v_F = v_F^b / f_t\) (\(v_F^b\) is the bare Fermi velocity). Then

\[
\xi(0) = \frac{\hbar v_F^b}{f_t \pi \Delta(0)}.
\]

(2)

Using \(H_{c2}(0) = \Phi_0 / 2\pi [\xi(0)]^2\) and \(H_{c2}(0) = 15 \pm 2 \text{ T}\) \[13\], we obtain \(\xi(0) = 47 \pm 2 \text{ Å}\). With \(v_F^b = 4.85 \times 10^5 \text{ m s}^{-1}\) \[4\], \(\Delta(0) = 6.9 \text{ meV}\) \[10\] and \(\xi(0) = 47 \pm 2 \text{ Å}\), we find from equation (2) that \(f_t = 3.1 \pm 0.1\). Remarkably, the deduced mass enhancement factor \(f_t\) here is in excellent agreement with that found from specific heat data \[8\]. We should mention that the electronic specific heat deduced in \[8\] is reliable because the deduced minimum superconducting gap (3.9 meV) \[10\] is the same as that deduced from independent scanning tunnelling spectroscopic (STS) experiments \[14\] and because the inferred mass enhancement is the same as that extracted from the critical field as well as from the penetration depth \[11\].

It is clear that the large mass enhancement factor is not compatible with the measured energy gap (\(2\Delta(0)/k_BT_c = 4.1\)) \[10\] and the reduced isotope exponent (\(\alpha = 0.28 \pm 0.04\)) within the conventional phonon-mediated model. In order to resolve the above controversy, one needs to consider a long-range Fröhlich-type electron–phonon interaction that results from the Coulomb interactions of electronic charge carriers with the ions of ionic (polar) materials \[15\]. This interaction is distinct from the short-range electron–lattice interaction (e.g. deformation-potential interaction, Holstein interaction) that has a carrier’s energy only depending on the positions of the atoms with which it overlaps. The Fröhlich electron–phonon interaction is particularly strong in an ionic solid or a perovskite oxide, where its static dielectric constant \(\epsilon_0\) is always much larger than its high-frequency dielectric constant \(\epsilon_\infty\), whereas the short-range electron–lattice interaction (including the electron–lattice interaction with acoustic phonons) is present in all the materials \[15\]. Recent quantum Monte Carlo simulation \[16\] has shown

\[
\text{New Journal of Physics 4 (2002) 3.1–3.7 (http://www.njp.org/)}
\]

† The bare Fermi velocity \(v_F^b\) is a simple geometric average of the calculated Fermi velocities along three axes \[4\].
that the Fröhlich-type electron–phonon interaction is nonretarded and can always lead to a mass enhancement factor of \( f_p = \exp(g^2) \) no matter how weak this interaction is. Here \( g^2 = A/\omega_0 \), \( A \) is a constant and \( \omega_0 \) is the characteristic frequency of optical phonons. It is apparent that this mass enhancement factor \( f_p \) will strongly depend on the isotope mass if \( g^2 \) is substantial, in contrast to the conventional retarded electron–phonon coupling model where the mass enhancement factor is essentially isotope-mass independent [9]. On the other hand, the short-range component of the electron–lattice interaction is retarded and can be modelled within the conventional Eliashberg equations when the coupling constant is less than 1 and the phonon energies are much lower than the bare hopping integral [16, 17]. When only a short-range electron–lattice interaction is present and the coupling constant is far larger than 1, the interaction becomes nonretarded and small polarons can be formed [15, 17]. In this case, the Migdal adiabatic approximation breaks down [17] and one cannot use the Eliashberg equations to describe superconductivity [15, 17].

Since the Fröhlich-type electron–phonon interaction is nonretarded, we can assume that the role of this interaction is simply to enhance the density of states [17] and reduce the direct Coulomb interaction between two carriers [15, 17]. In other words, we can model the retarded short-range electron–phonon coupling within the conventional Eliashberg equations, but the effective density of states and the retarded electron–phonon coupling constant \( \lambda \) are enhanced by a factor of \( f_p \) due to the presence of the nonretarded Fröhlich-type electron–phonon interaction. More explicitly, superconductivity arises from the \( k \)-space pairing of Fröhlich polarons through the retarded short-range electron–phonon coupling. This superconductivity mechanism is in contrast to the bipolaronic superconductivity where the real-space pairing is an essential feature [17]. Similar theoretical approach was applied to explain the superconductivity in doped C_{60} [18] as well as in MgB_{2} [19]. In their model [18, 19], they considered the electron–phonon coupling with high-energy phonons to be nonretarded and the coupling with low-energy phonons to be retarded.

The effective Coulomb pseudopotential \( \mu^* \) in the Eliashberg equations will not reduce significantly when the Fröhlich electron–phonon interaction sets in. This is because this interaction enhances the density of states, which tends to increase \( \mu^* \), while it produces an attractive potential that effectively reduces the direct Coulombic repulsion by a factor of about \( \epsilon_0/\epsilon_{\infty} \) [15] and thus tends to decrease \( \mu^* \) (see equation (4) below). Therefore, the Fröhlich-type electron–phonon interaction can enhance superconductivity mainly through increasing the effective coupling constant for the retarded electron–phonon interaction. For ionic materials (e.g. MgB_{2}), this type of electron–phonon coupling should be rather strong, so that the superconductivity can be enhanced substantially.

Within this simplified approach, the effective coupling constant for the retarded short-range electron–phonon interaction is \( \lambda = \lambda_b f_p \), where \( \lambda_b \) is the bare short-range retarded electron–phonon coupling constant in the absence of the Fröhlich-type electron–phonon interaction. The value of \( \lambda_b \) for MgB_{2} has been calculated to be 0.7–0.9 [4–7]. The total carrier mass enhancement factor is then given by

\[
f_t = f_p (1 + \lambda_b f_p) \tag{3}
\]

The factor \( 1 + \lambda_b f_p \) is the mass enhancement factor due to the retarded electron–phonon interaction with the enhanced coupling constant \( \lambda = \lambda_b f_p \). The Coulomb pseudopotential \( \mu^* \) is

\[
\mu^* = \frac{\mu}{1 + \mu \ln(E_F/h\omega_{\text{ph}})} \tag{4}
\]
Here $\mu \propto U f_p$ ($U$ is the effective Coulomb interaction between two carriers, which is renormalized by the Fröhlich interaction); $E_F^h = E_F^b / f_p$ ($E_F^b$ is the bare Fermi energy); $\omega_{ln}$ is the logarithmically averaged frequency, which is normally lower than the Debye frequency $\omega_D$ by a factor of about 1.2. Indeed, the calculated $\hbar \omega_{ln}$ is 53.8 meV [21], which is a factor of 1.2 smaller than the measured $\hbar \omega_D$ (64.3 meV) [2, 8].

Since $f_p = \exp(g^2)$, which depends on the isotope mass [16], the quantities $\lambda$, $\mu^*$ and $f_l$ are all isotope-mass dependent. One can easily deduce that the total exponents of the isotope effects on $\lambda$ ($\alpha = -\sum d \ln \lambda / d \ln M_j$, where $M_j$ is the mass of the $j$th atom in the unit cell) and on $\mu^*$ ($\alpha_{\mu^*} = -\sum d \ln \mu^* / d \ln M_j$) are given by

$$\alpha = -\frac{1}{2} \ln f_p,$$

and

$$\alpha_{\mu^*} = \alpha [1 - \mu^* \ln(E_F/\hbar \omega_{ln})] + \mu^*(\alpha + \frac{1}{2}).$$

Furthermore, from the McMillian formula [9],

$$k_B T_c = \frac{\hbar \omega_{ln}}{1.2} \exp \left[ \frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right],$$

we can also determine the total exponent of the isotope effect on $T_c$ ($\alpha = -\sum d \ln T_c / d \ln M_j$),

$$\alpha = \frac{1}{2} + \frac{1.04(1 + 0.38\mu^*)\lambda}{[\lambda - \mu^*(1 + 0.62\lambda)^2]^{\alpha_{\mu^*}}} - \frac{1.04(1 + \lambda)(1 + 0.62\lambda)\mu^*}{[\lambda - \mu^*(1 + 0.62\lambda)^2]}.$$

From the above equations, we can calculate $f_p$, $\alpha$, $\hbar \omega_{ln}$, $\alpha_{\mu^*}$ and $\alpha$ as a function of $\lambda_b$ using fixed parameters $f_l = 3.2$, $T_c = 40$ K, $E_F = 0.57$ eV [20] and $\mu^* = 0.1$. In figure 1, we plot the calculated $\alpha$, $\hbar \omega_{ln}$ and $f_p$ as a function of $\lambda_b$. One can see that the calculated $\alpha$ is 0.3 for $\lambda_b = 0.8$, a value lying within the first-principles calculations ($\lambda_b = 0.7–0.9$) [4]–[7]. The calculated isotope exponent $\alpha$ is in quantitative agreement with the measured one (0.28 ± 0.04) (see footnote 1). The reduction in the isotope exponent is due to the fact that the coupling constant $\lambda$ has a negative isotope effect, which partially cancels out the positive isotope effect on the prefactor of equation (7). As $\lambda_b$ increases, $f_p$ must decrease to keep $f_l$ a constant (see equation (3) and figure 1(c)). The decrease of $f_p$ reduces the magnitude of the negative isotope effect on the coupling constant, and thus increases the isotope effect on $T_c$, as seen clearly in figure 1(a).

Meanwhile, the calculated $\hbar \omega_{ln}$ is about 40 meV for $\lambda_b = 0.8$ (see figure 1(b)), which appears to be lower than the one (53.8 meV) determined from the phonon density of states [21]. However, inelastic neutron scattering experiments [22] show that a low-energy phonon mode at about 17 meV is strongly coupled to conduction electrons; the intensity of the 17 meV peak increases with decreasing temperature for $T > T_c$, whereas below $T_c$ it starts decreasing. The strong coupling to the low-energy mode is not expected from the theoretical calculations [4]–[7], and may be related to some kind of structural instability [22]. If this is true, the calculated $\hbar \omega_{ln}$ in [21] should be substantially overestimated. Indeed, from the measured $2\Delta(0) / k_B T_c = 4.1$, one can determine the magnitude of $\hbar \omega_{ln}$ using a formula [9]

$$\frac{2\Delta(0)}{k_B T_c} = 3.53 \left[ 1 + 12.5 \left( \frac{k_B T_c}{\hbar \omega_{ln}} \right)^2 \ln \left( \frac{\hbar \omega_{ln}}{2k_B T_c} \right) \right].$$

Substituting $T_c = 40$ K and $2\Delta(0) / k_B T_c = 4.1$ into equation (9), we obtain $\hbar \omega_{ln} = 40$ meV, in quantitative agreement with the above independent calculation.
Figure 1. The total isotope exponent $\alpha$, the logarithmically averaged frequency $\hbar\omega_{ln}$ and the mass enhancement factor $f_p$ (due to the Fröhlich electron–phonon interaction) as a function of the bare retarded electron–phonon coupling constant $\lambda_b$. 

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From figure 1(c), one can see that $f_p$ is about 1.5 at $\lambda_b = 0.8$, that is, the Fröhlich-type electron–phonon interaction enhances the density of states and the coupling constant by a factor of 1.5. Without this interaction, the material would have a transition temperature of about 22 K. Therefore, the Fröhlich-type electron–phonon interaction in ionic materials can indeed enhance superconductivity.

Although the conventional phonon-mediated model could account for the observed $T_c$ value [4]–[7], it cannot consistently explain the $T_c$ value, the reduced isotope exponent ($\approx 0.3$), the large carrier mass enhancement factor (3.1) and the reduced energy gap ($2\Delta(0)/k_B T_c = 4.1$). The accurate determination of $2\Delta(0)/k_B T_c = 4.1$ places a strong constraint on $h\omega_{ln}$, that is, $h\omega_{ln} \approx 40$ meV. With $h\omega_{ln} = 40$ meV, and $T_c = 40$ K, one cannot find any parameters that could lead to $\alpha \approx 0.28$. Further, one would never get a carrier mass enhancement factor larger than 3 within the conventional model. On the other hand, the proposed unconventional phonon-mediated mechanism naturally resolves these controversies and is able to quantitatively explain these experiments in a consistent way. In addition, an important prediction of this scenario is that both electronic specific heat $\gamma$ and London penetration depth $\lambda_L(0)$ depend on the isotope mass of boron. One can easily show that, upon replacing $^{10}$B with $^{11}$B, both $\gamma$ and $\lambda_L^2(0)$ will decrease by about 4%. The isotope effect on $\gamma$ should be observable if one could measure the specific heat of the isotope samples down to a low temperature ($<1$ K) under a magnetic field higher than $H_{c2}(0)$. On the other hand, a special cation must be taken in determination of the intrinsic London penetration depth since the extrinsic contribution to the measured penetration depth due to defects [23] would mimic the isotope effect if two isotope samples had different densities of defects.

In summary, the carrier mass enhancement factor $f_t$ has been determined for MgB$_2$ from the measured upper critical field and the calculated bare Fermi velocity. It is found that $f_t$ is $3.1 \pm 0.1$, in remarkably good agreement with that deduced from the independent specific heat data. The unusually large $f_t$ is inconsistent with the measured reduced gap and the total isotope-effect exponent ($\alpha \approx 0.3$) within the conventional phonon-mediated model. We thus propose an unconventional phonon-mediated mechanism where long-range Fröhlich electron–phonon interaction and short-range retarded electron–phonon interaction are modelled separately. Within this scenario, we are able to quantitatively explain the values of $T_c$, $f_t$, $\alpha$ and $2\Delta(0)/k_B T_c$.

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**References**

[1] Nagamatsu J, Nakagawa N, Muranaka T, Zenitani Y and Akimitsu J 2001 Nature 410 63
[2] Bud’ko S L, Lapertot G, Petrovic C, Cunningham C E, Anderson N and Canfield P C 2001 Phys. Rev. Lett. 86 1877
[3] Hinks D G, Claus H and Jorgensen J D 2001 Nature 411 457
[4] Kortus J, Mazin I I, Belashchenko K D, Antropov V P and Boyer L L 2001 Phys. Rev. Lett. 86 4656
[5] Kong Y, Dolgov O V, Jepsen O and Andersen O K 2001 Preprint cond-mat/0102499
[6] Bohnen K-P, Heid R and Renker B 2001 Phys. Rev. Lett. 86 5771
[7] Liu A Y, Mazin I I and Kortus J 2001 Phys. Rev. Lett. 87 087005
[8] Wälti Ch, Felder E, Degen C, Wigger G, Monnier R, Delley B and Ott H R 2001 Phys. Rev. B 65 172515
[9] Carbotte J P 1990 Rev. Mod. Phys. 62 1027
[10] Chen X K, Konstantinovic M J, Irwin J C, Lawrie D D and Franck J P 2001 Preprint cond-mat/0104005
[11] Zhao G M 2001 unpublished
[12] Canfield P C, Finnemore D K, Bud’ko S L, Ostenson J E, Lapertot G, Cunningham C E and Petrovic C 2001 Phys. Rev. Lett. 86 2423
[13] Bud’ko S L, Petrovic C, Lapertot G, Cunningham C E, Canfield P C, Jung M-H and Lacerda A H 2001 Phys. Rev. B 63 220503R
[14] Gibubileo F, Roditchev D, Sacks W, Lamy R and Klein J 2001 Phys. Rev. Lett. 87 177008
[15] Emin D 1989 Phys. Rev. Lett. 62 1544
   Emin D and Hillery M S 1989 Phys. Rev. B 39 6575
   Emin D 1995 Phys. Rev. B 52 13 874
[16] Alexandrov A S and Kornilovitch P E 1999 Phys. Rev. Lett. 82 807
[17] Alexandrov A S and Mott N F 1995 Polaron and Bipolarons (Singapore: World Scientific)
[18] Alexandrov A S and Kabanov V V 1996 Phys. Rev. B 54 3655
[19] Alexandrov A S 2001 Physica C 363 231
[20] Lorenz B, Meng R L, Xue Y Y and Chu C W 2001 Preprint cond-mat/0104041
[21] Osborn R, Goremychkin E A, Kolesnikov A I and Hinks D G 2001 Phys. Rev. Lett. 86 017005
[22] Sato T J, Shibatal K and Takano Y 2001 Preprint cond-mat/0102468
[23] Zhao G M 2001 Phys. Rev. B 63 024503