Two-band Eliashberg equations and the experimental $T_c$ of the diboride $Mg_{1-x}Al_xB_2$

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The recent discovery of superconductivity at $T=40$ K in magnesium diboride [1] has stimulated intense investigation, both from the theoretical and the experimental point of view. Now the electronic structure of $MgB_2$ is well understood and the Fermi surface consists of two three-dimensional sheets, from the $\pi$ bonding and anti-bonding bands, and two nearly cylindrical sheets from the two-dimensional $\sigma$ bands [2]. There is a large difference in the electron-phonon coupling on different Fermi surface sheets and this fact leads to a multiband description of superconductivity. The superconductivity in $MgB_2$ has been deeply studied in the past two years. More recently the substitutions of $Mg$ with aluminum [3] have been investigated to understand the evolution of the pairing process by tuning the Fermi level toward the top of the sigma band edge. The electronic structure of boron layers intercalated by alternating layers of Al and Mg is formed. Even though the alloys with intermediate $x$ are rather disordered, their $T_c$ is well defined and drops with decreasing $x$, becoming zero [4] for $x > 0.5$. We can see in Fig. 1 the experimental data of critical temperature taken from ref. 4-6. Two ranges of variation are present: a "high-$T_c$" range for $0 < x < 0.3$ that shows a slow variation with $x$, while a rapid variation occurs in the "low-$T_c$" range $0.3 < x < 0.5$. This variation has been interpreted as to a 2D-3D cross-over, at $x \approx 0.33$, of the topology of the Fermi surface [5]. Theoretically, the transition temperature $T_c$ in Al-substituted $MgB_2$ has been studied [6] within the two-band BCS formalism, as a function of the aluminum content; the variation of the effective inter-band coupling as a function of $x$ has been obtained from the experimental values of $T_c(x)$. Ab-initio calculations have been performed on Al-substituted $MgB_2$ by Profeta et al. [7] and by de la Peña et al. [8], within the virtual crystal approximation. These works correctly describe the experimental trends; in particular [8], the behavior of the $E_{2g}$ phonon frequency and of the critical temperature agree reasonably well with experiment.

The variation of the superconducting critical temperature $T_c$ as a function of $x$ in the diboride $Mg_{1-x}Al_xB_2$ has been studied in the framework of the two-bands Eliashberg theory and traditional phonon coupling mechanism. We have solved the two-bands Eliashberg equations using first-principle calculations or simple assumptions for the variation of the relevant physical quantities. We have found that the experimental $T_c$ curve can be explained only if the Coulomb pseudopotential changes with $x$ by tuning the Fermi level toward the sigma band edge. In polycrystal samples the $x$ dependence of the $\sigma$ and $\pi$-band gap has been found and is in agreement with experiments.

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FIG. 1: The value of the experimental $T_c$ (open circle) as a function of $x$, taken from ref. 4-6.
tion of the Eliashberg theory \cite{10,11,12} for systems with two bands \cite{13,14}. The two bands Eliashberg equations have already been used for studying the MgB$_2$ system \cite{16,17,18,19}. Here we have eight input parameters: four (but only three independent \cite{14}) electron-phonon spectral functions $\alpha_{ij}^2(\omega)F_{ij}(\omega)$ and four (but only three independent) Coulomb pseudopotential $\mu_{ij}(\omega_c)$ to reproduce the experimental values of $T_c$ versus $x$ by solving the Eliashberg equations. In this case there are four equations to be solved for the calculation of the gaps $\Delta_i(\omega_n)$ and the renormalization functions $Z_i(\omega_n)$. In the approximation of a flat normal densities of states and infinite bands, the s-wave two bands Eliashberg equations are:

\begin{equation}
\omega_n Z_i(\omega_n) = \omega_n + \pi T \sum_{m,j} \Lambda_{ij}(\omega_n - \omega_m) N^j_Z(\omega_m) + \\
\sum_j \Gamma^{ij} N^i_Z(\omega_n)
\end{equation}

\begin{equation}
Z_i(\omega_n) \Delta_i(\omega_n) = \pi T \sum_{m,j} [\Lambda_{ij}(\omega_n - \omega_m) - \mu_{ij}(\omega_c)] \cdot \\
-\theta(|\omega_c| - \omega_m) N^j_N(\omega_m) + \sum_j \Gamma^{ij} N^i_N(\omega_n)
\end{equation}

where $\theta$ is the Heaviside function, $\omega_c$ is a cut-off energy, $\Gamma^{ij}$ is the non magnetic impurity scattering rate in the Born approximation and

\begin{equation}
\Lambda_{ij}(\omega_n - \omega_m) = \int_0^{+\infty} \frac{d\omega \alpha_{ij}^2 F(\omega)}{(\omega_n - \omega_m)^2 + \omega^2}
\end{equation}

\begin{equation}
N^j_N(\omega_m) = \frac{\Delta_j(\omega_m) Z_j(\omega_m)}{\sqrt{\omega_n^2 Z^2_j(\omega_m) + \Delta^2_j(\omega_m) Z^2_j(\omega_m)}}
\end{equation}

\begin{equation}
N^j_N(\omega_m) = \frac{\omega_m Z_j(\omega_m)}{\sqrt{\omega_n^2 Z^2_j(\omega_m) + \Delta^2_j(\omega_m) Z^2_j(\omega_m)}}
\end{equation}

where $\omega_n = \pi T(2n - 1)$ and $n, m = 0, \pm 1, \pm 2, \ldots$. In writing the Eliashberg equations we have neglected the asymmetric part of the self-energy $\chi_i(\omega_n)$ (which is always equal to zero in the half-filling case) and the equation that represents the conservation of the particles necessary for calculating the shift of the chemical potential. These assumptions are correct if the normal density of states is symmetric or constant as function of energy and the bands are infinite. We have calculated the normal densities of state of the $\sigma$ and $\pi$ band and, with good approximation, they are constant around the Fermi energy for all $x$-values. Finally to use the infinite band approximation is correct for the $\pi$-band (half-bandwidth $W_{\pi}(x = 0) \approx 3$ eV) while we can have some doubt for the $\sigma$-band ($W_{\sigma}(x = 0) \approx 0.5$ eV) \cite{20}. If we solve the Eliashberg equations with these last finite values of the bands

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{The spectral functions of the two-band model for the MgB$_2$: $\sigma$ (solid line), $\pi\pi$ (dashed line), $\sigma\pi$ (dotted line) and $\pi\pi$ (dashed dotted line), taken from ref. 15.}
\end{figure}

\[ \text{for the MgB}_2 \ (x = 0 \text{ case}), \text{by using the same parameters of the infinite bands approximation, we find } T_c = 40 \text{ K. For having } T_c = 39.4 \text{ K it must be } \mu(\omega_c) = 0.0333 \text{ (see eq. 10) instead of 0.0326. Since in the first case } T_c \text{ is only slightly lower than the experimental one and } \mu(\omega_c), \text{ in the second case, is almost the same as in the infinite band case so we can conclude that to assume the bands as infinite is a plausible approximation. The situation can be different, of course, for large values of } x. \]

Finally, one can think that it would be simpler to use the Suhl formula \cite{25} for explaining the $T_c$ versus $x$ curve without solving the complete two-band Eliashberg equations. The problem of the Suhl formula is the prefactor before the exponential: it is a phononic energy but in a two-band theory it is difficult to specify it exactly and so it is considered as another free parameter.

For solving the two-band Eliashberg equations we must know how the six input quantities vary with $x$ because, as shown in ref. 14, $\lambda_{ij}(x)/\lambda_{ji}(x) = \mu_{ij}(\omega_c)/\mu_{ji}(\omega_c) = N_{N}(\omega = E_F, x)/N_{N}(\omega = E_F, x)$ where $N_{N}(\omega = E_F, x)$ is the density of states at the Fermi level in the $j$-band and $\lambda_{ij}(x)$ are the electron-phonon coupling constants. For carrying out this task we must adopt some drastic approximations because almost all input quantities are, at the moment, difficult to determine.

We begin with the four spectral functions of the $MgB_2$ that we can see in Fig. 2 as calculated in ref. 15. We assume that the general shape of $\alpha_{ij}^2 F(\omega, x)$ does not change with $x$ but only the $\lambda_{ij}$ value, that is:

\begin{equation}
\alpha_{ij}^2 F(\omega, x) = \frac{\lambda_{ij}(x)}{\lambda_{ij}(x = 0)} \alpha_{ij}^2 F(\omega, x = 0)
\end{equation}

Since it is known that the details of $\alpha^2 F(\omega)$ do not affect the resulting $T_c$ significantly \cite{26}, this approximation is expected to be reasonable.

From the definition of electron-phonon coupling con-
constant we have \[20\]:

\[
\lambda = \frac{N_N(\omega = E_F) < I^2 >}{M \Omega_0^2} \tag{7}
\]

\(M\) is the ion mass, the frequency \(\Omega_0\) is the frequency representative of phonon spectrum, \(N_N\) is the density of states at the Fermi level and \(< I^2 >\) is the average matrix element of the electron-ion interaction \[20\]. The mass \(M\) is the boron mass \[21\] and does not depend on \(x\). We assume that the average matrix element of the electron-ion interaction \(< I^2 >\) is, in the first approximation, constant because it is basically determined by the deformation potential which is almost \[3\] independent of \(x\). We have calculated \[8\], within the virtual crystal approximation, the dependence on \(x\) of \(N_N^J\) (see Fig. 3, upper panel) and we have identified the representative phonon frequency \(\Omega_0\) with the \(E_{2g}\) phonon mode obtained by experimental data \[6\] (see Fig. 3 lower panel). So we have that, the more important contribution to superconductivity in our

\[
\text{system, the electron-phonon coupling constant } \lambda_{\sigma\sigma} \text{ is:}
\]

\[
\lambda_{\sigma\sigma}(x) = \frac{N_N^J(\omega = E_F, x)\omega_{E_{2g}}^2(x = 0)}{N_N^J(\omega = E_F, x = 0)\omega_{E_{2g}}^2(x)} \lambda_{\sigma\sigma}(\omega, x = 0) \tag{8}
\]

In this way we assume that the change of \(E_{2g}\) influence only the value of the electron-phonon coupling constant while, in the first approximation \[20\], we neglect its effects on the shape of the spectral function. For checking this hypothesis we have also used \(\alpha_{ij}^2 F_{ij}(\omega, x) = L(\omega, \Omega_0, \Upsilon) - L(\omega, -\Omega_0, \Upsilon)\) where \(L(\omega, \Omega_0, \Upsilon)\) is a lorentzian curve with the peak in \(\Omega_0(x) = \omega_{E_{2g}}(x)\) and half-width \(\Upsilon(x)\) equal to the error bar of Fig. 2 (lower panel). The result is almost the same as we can see in Figs. 5 (upper panel) and 6 (upper panel).

For the other coupling constants, only for simplicity, we assume:

\[
\lambda_{ij}(x) = \frac{N_N^J(\omega = E_F, x)}{N_N^J(\omega = E_F, x = 0)} \lambda_{ij}(x = 0) \tag{9}
\]

with \[15, 16\] \(\lambda_{\sigma\sigma}(x = 0) = 1.017\), \(\lambda_{\sigma\pi}(x = 0) = 0.448\), \(\lambda_{\sigma\pi}(x = 0) = 0.213\) and \(\lambda_{\pi\pi}(x = 0) = 0.155\).

In Fig. 4 we can see the calculated electron phonon coupling constants \(\lambda_{ij}\) as a function of \(x\). We can see that, with our approximations, only \(\lambda_{\sigma\sigma}\) depends strongly on \(x\) and, for \(x > 0.33\), \(\lambda_{\sigma\sigma} < \lambda_{\pi\pi}\). The Coulomb pseudopotential, calculated, for the first time, in ref. 15 and 16 is \[22\]

\[
\mu^*(p) = \begin{vmatrix}
\mu_{\sigma\sigma}^* & \mu_{\sigma\pi}^* & \mu_{\pi\sigma}^* & \mu_{\pi\pi}^*
\end{vmatrix}
\]

\[
= \mu(\omega_c) N_N^{\text{tot}}(E_F, p) \begin{vmatrix}
0.23 & 1 & 1 & 2.48
\end{vmatrix}
\]

\[
N_N(E_F, p) N_N(\omega = E_F, p) \]

\[
\begin{vmatrix}
0.23 & 1 & 1 & 2.48
\end{vmatrix}
\]

\[
\begin{vmatrix}
\mu_{\sigma\sigma}^* & \mu_{\sigma\pi}^* & \mu_{\pi\sigma}^* & \mu_{\pi\pi}^*
\end{vmatrix}
\]
as a function of $x$ when the $\alpha_{\sigma\sigma}^2 F(\omega, x)$ is a lorentzian, when is $MgB_2$-like (filled circles) and when is $MgB_2$-like and in presence of interband impurities, $\Gamma^{\sigma\pi} = 1$ meV, (open up triangles); in the insert the impurity scattering rate $\Gamma^{\sigma\pi}$, necessary to reproduce the experimental $T_c$, without change the $MgB_2$ physical parameters, versus $x$. Lower panel: the values of Coulomb pseudopotential $\mu_{ij}$ as a function of $x$ in the four cases: $\sigma\sigma$ (solid line), $\pi\pi$ (dashed line), $\sigma\pi$ (dotted line) and $\pi\sigma$ (dashed dotted line) in the $MgB_2$-like $\alpha_{\sigma\sigma}^2 F(\omega, x)$ spectral function case.

where $\mu(\omega_c, x)$ is a free parameter and and $N_N^{\text{tot}}(\omega = E_F, x)$ is the total normal density of states at the Fermi level. The numbers 2.23 and 2.48 in the Coulomb matrix have been calculated in the $MgB_2$ case and we suppose that, as first approximation, they do not depend on $x$ i.e. the Coulomb pseudopotential depend by $x$ only via the density of states at the Fermi level while an outer possible dependence by $x$, included the effect of the disorder, is hidden in $\mu(\omega_c, x)$ and is the same for the four values of $\mu_{ij}^*(x)$. In conclusion we put the cut-off energy $\omega_c = 700$ meV. So we have now only one free parameter, $\mu(\omega_c, x)$ that must be determined by reproducing exactly the experimental critical temperatures.

In our discussion we have neglected the effects of the disorder caused by Al doping because we don’t know the dependence of $\Gamma^{ij}$ by $x$ and this fact lead to have a new free parameter in the problem. We seek of justifying this approximation and we examine two possibilities. One extreme case is one of thinking the $Mg_{1-x}Al_xB_2$ as a contamination with Al impurities of the $MgB_2$. In this case also we have only a free parameter for reproduce the experimental critical temperatures, $\Gamma^{\sigma\sigma}$, because $\Gamma^{\sigma\pi}$ and $\Gamma^{\pi\pi}$ don’t affect $T_c$ and $\Gamma^{\pi\sigma} = N_N^\sigma(\omega = E_F)/N_N^\pi(\omega = E_F)\Gamma^{\sigma\pi}$. In this way we find that $\Gamma^{\sigma\pi}$ grows fast in function of $x$ as we can see in the insert of the upper panel of Fig. 5. The minimum critical temperature that we can obtain in this way is $T_c \approx 25.8$ K and an isotropic gap $\Delta_\sigma = \Delta_\pi \approx 4.1$ meV but we cannot explain the lower critical temperatures. Moreover the authors of ref. 27 predict the $\Gamma^{\sigma\sigma} \gg \Gamma^{ij} \approx 1$ meV i.e. $\Gamma^{ij}$ values found are completely to outside of the range expected. As second possibility we examine the case $\Gamma^{\sigma\pi} = 1$ meV and we find the dependence of $\mu(\omega_c)$ by $x$. As we can see in Figs. 5 and 6 (upper panel) the differences are almost imperceptible. For all these reasons we neglect the effects of impurities in the Eliashberg equations.

In the case of $MgB_2$ ($x = 0$) we find $\mu(\omega_c, x = 0) = 0.0326$ ($\omega_c = 700$ meV and maximum of energy equal to 1 eV). In Fig. 5 we show the dependence on $x$ of $\mu(\omega_c, x)$.
The calculated rate of $\Delta_\pi/\Delta_\sigma$ as a function of experimental $T_c$. (upper panel) while in the lower panel we can see the values of $\mu_\omega^c(x)$ calculated following eq. 10. We note a monotonic parabolic decrease until $x = 0.4$ while after it is present a rapid increase. This result shows that the coulomb pseudo-potential obtained in this way decreases, approaching the “shape resonance” determined by the superlattice of boron monolayers as predicted in several papers [28]. The reduction of the effective Coulomb repulsion in the pairing seems to be a driving term for rising the critical temperature. The final sharp increase can be due to an inadequateness of the model: probably, for $x \approx 0.5$, the Migdal’s theorem fails [29] because there is a growth of the representative phonon frequencies and a strong reduction of the Fermi energy.

After determining the dependence of the free parameter $\mu(\omega_c)$ on $x$, we can calculate all the physical quantities by solving the Eliashberg equations. In Fig. 6 we show the value of $\Delta(\omega_n=0)$, at $T = T_c/4$, of the $\sigma$-band and the $\pi$-band (solid circles and solid squares respectively) versus $x$. The values calculated are almost the same of those found by solution of the equation $\Delta_\sigma^0 = \Delta_\pi(\omega = \Delta_\pi^0)$ on the real axis with Padé approximants because we are in a weak coupling regime. In strong coupling regime instead the differences can be remarkable [30]. We can see that the behaviour of the $\pi$-gap, as a function of $x$ has a maximum for 0.164 < $x$ < 0.198 while the $\sigma$-gap decreases and, for $x > 0.4$, we find that $\Delta_\pi > \Delta_\sigma$. The experimental data [31] exist only for $T_c \geq 20$ K ($x \leq 0.33$) and are in good agreement with our theoretical predictions. For higher aluminum content, at present there are no measurements that confirm or deny the fact that the gaps merge for $x \approx 0.4$. In the lower panel we show the calculated ratio $2\Delta_\pi/k_B T_c$. Finally in Fig. 7 we can see the calculated rate of $\Delta_\pi/\Delta_\sigma$ as a function of experimental $T_c$ (which is preferable due to the lack of agreement between the different experiments on the true aluminum content of the sample).

It is worth to note that if, for the all four coupling constants (including the $\sigma\sigma$ component), we use eq. 9, while the parameter $\mu(\omega_c)$ is fixed to the value $\mu(\omega_c, x = 0) = 0.0326$ used in the $MgB_2$ case, i.e. all input quantities of the Eliashberg equations depend by $x$ only via the densities of states at the Fermi level, we have a model without free parameters. This model produces a very good fit of the experimental critical temperatures for $x < 0.33$, but for larger $x$ the predicted $T_c$ is much lower than the experimental values. The value $x = 0.33$ corresponds a the transition from a pure two-dimensional Fermi surface to an overall three dimensional dispersion regime [8]. The behavior of the $\sigma$-gap is almost unchanged, while the one of the $\pi$-gap changes, and decreases monotonically. We have no arguments to justify the omission of the dependence of $\lambda_{\sigma\sigma}$ on $\omega_{E_{\pi\pi}}$.

Further experimental work is necessary to determine the variation of the gaps with $x$ in order to understand the correct theoretical approximation and to understand the variation of the pairing process tuning the Fermi level through the “shape resonance” condition.

In conclusion we can affirm that not only the $MgB_2$ but also the same material doped with aluminum are weak coupling two band phononic systems well described by two-bands Eliashberg theory where the Coulomb pseudopotential and the interchannel pairing mechanism are key terms to interpret the superconducting phase (it is the interband pairing that yields the superconductivity in the $\pi$-band).

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