Two-band Eliashberg equations and the experimental Tc of the diboride Mg1-xAlxB2

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G.A. Ummarino, R.S. Gonnelli
INFM−Dipartimento di Fisica, Politecnico di Torino,
Corso Duca degli Abruzzi 24, 10129 Torino, Italy
E-mail address: giovanni.ummarino@infm.polito.it

S. Massidda
INFM−Dipartimento di Fisica, Università di Cagliari,
Cittadella Universitaria SP Monserrato-Sestu, km 0,700 I-09042 Monserrato, Italy

A. Bianconi
INFM - Dipartimento di Fisica,
Università di Roma "La Sapienza"
Piazzale Aldo Moro 2, 00185 Roma, Italy

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 σ and π-band gap has been found and is in agreement with experiments.

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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 π bonding and antibonding bands, and two nearly cylindrical sheets from the two-dimensional σ 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 $Mg_{1-x}Al_xB_2$ alloys show [4, 5, 6] a continuous evolution through a complicated mixed phase from MgB_2 (x=0) to the final member $AlMgB_4$ (x=0.5) where an ordered superlattice 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, 5, 6] 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 $x \cong 0.33$, of the topology of the Fermi surface [5, 6]. Theoretically, the transition temperature T_c in Al-substituted MgB_2 has been studied [7] within the two-band BCS formalism, as a function of the aluminum content; the variation of the effective interband 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.*[8] and by de la Peña *et al.*[9], 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.

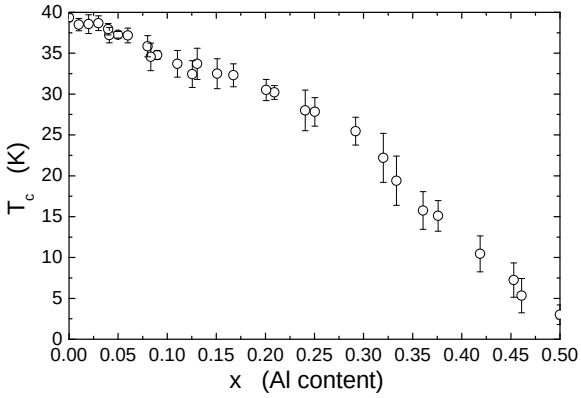

FIG. 1: The value of the experimental T_c (open circle) as a function of x, taken from ref. 4-6.

In this work we further investigate $Mg_{1-x}Al_xB_2$ (for $0 \leq x \leq 0.5$) by computing the variation of the critical temperature as a function of x by means of the Migdal-Eliashberg two bands theory. For the peculiar characteristics of our system we must use a generaliza-

tion of the Eliashberg theory [10, 11, 12] for systems with two bands [13, 14]. The two bands Eliashberg equations have already been used for studying the MgB_2 system [16, 17, 18, 19]. Here we have eight input parameters: four (but only three independent [14]) electron phonon spectral functions $\alpha^2_{ij}(\omega)F_{ij}(\omega)$ 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 $\Delta_i(i\omega_n)$ and the renormalization functions $Z_i(i\omega_n)$. In the approximation of a flat normal densities of states and infinite bands, the s-wave two bands Eliashberg equations are:

$$\omega_n Z_i(i\omega_n) = \omega_n + \pi T \sum_{m,j} \Lambda_{ij}(i\omega_n - i\omega_m) N^j_Z(i\omega_m) + + \sum_j \Gamma^{ij} N^j_Z(i\omega_n) \quad (1)$$

$$Z_i(i\omega_n)\Delta_i(i\omega_n) = \pi T \sum_{m,j} [\Lambda_{ij}(i\omega_n - i\omega_m) - \mu^*_{ij}(\omega_c)] \cdot \cdot\theta(|\omega_c| - \omega_m) N^j_\Delta(i\omega_m) + \sum_j \Gamma^{ij} N^j_\Delta(i\omega_n) \quad (2)$$

where θ is the Heaviside function, ω_c is a cut-off energy, Γ^{ij} is the non magnetic impurity scattering rate in the Born approximation and

$$\Lambda_{ij}(i\omega_n - i\omega_m) = \int_0^{+\infty} \frac{d\omega \alpha^2_{ij} F(\omega)}{(\omega_n - \omega_m)^2 + \omega^2} \quad (3)$$

$$N^j_\Delta(i\omega_m) = \frac{\Delta_j(i\omega_m) Z_j(i\omega_m)}{\sqrt{\omega^2_m Z^2_j(i\omega_m) + \Delta^2_j(i\omega_m) Z^2_j(i\omega_m)}} \quad (4)$$

$$N^j_Z(i\omega_m) = \frac{\omega_m Z_j(i\omega_m)}{\sqrt{\omega^2_m Z^2_j(i\omega_m) + \Delta^2_j(i\omega_m) Z^2_j(i\omega_m)}} \quad (5)$$

where $\omega_n = \pi T(2n-1)$ and $n, m = 0, \pm 1, \pm 2....$ In writing the Eliashberg equations we have neglected the asymmetric part of the self-energy $\chi_i(i\omega_n)$ (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 σ and π 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 π-band (half-bandwidth $W_\pi(x=0) \simeq 3$ eV) while we can have some doubt for the σ-band ($W_\sigma(x=0) \simeq 0.5$ eV)[23]. If we solve the Eliashberg equations with these last finite values of the bands

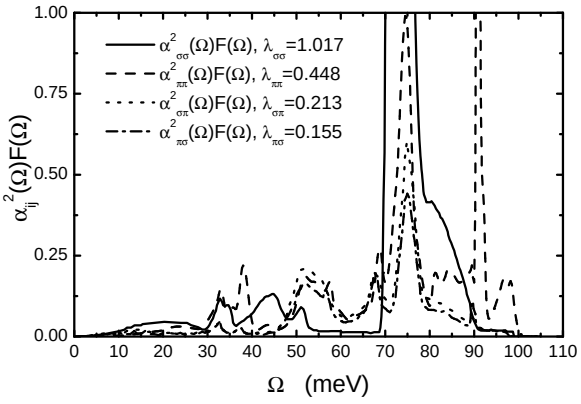

FIG. 2: The spectral functions of the two-band model for the MgB_2: $\sigma\sigma$ (solid line), $\pi\pi$ (dashed line), $\sigma\pi$ (dotted line) and $\pi\sigma$ (dashed dotted line), taken from ref. 15.

[12, 24] for the MgB_2 ($x = 0$ case), by using the same parameters of the infinite bands approximation, we find T_c =40 K. For having T_c =39.4 K it must be $\mu(\omega_c)$=0.0333 (see eq. 10) instead of 0.0326. Since in the first case T_c is only slightly lower than the experimental one and $\mu(\omega_c)$, 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 [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, $\lambda_{ij}(x)/\lambda_{ji}(x) = \mu^*_{ij}(x)/\mu^*_{ji}(x) = N^j_N(\omega = E_F, x)/N^i_N(\omega = E_F, x)$ where $N^j_N(\omega = E_F, x)$ 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 $\alpha^2_{ij}F(\omega, x)$ does not change with x but only the λ_{ij} value, that is:

$$\alpha^2_{ij}F(\omega, x) = \frac{\lambda_{ij}(x)}{\lambda_{ij}(x=0)} \alpha^2_{ij}F(\omega, x=0) \quad (6)$$

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

From the definition of electron-phonon coupling con-

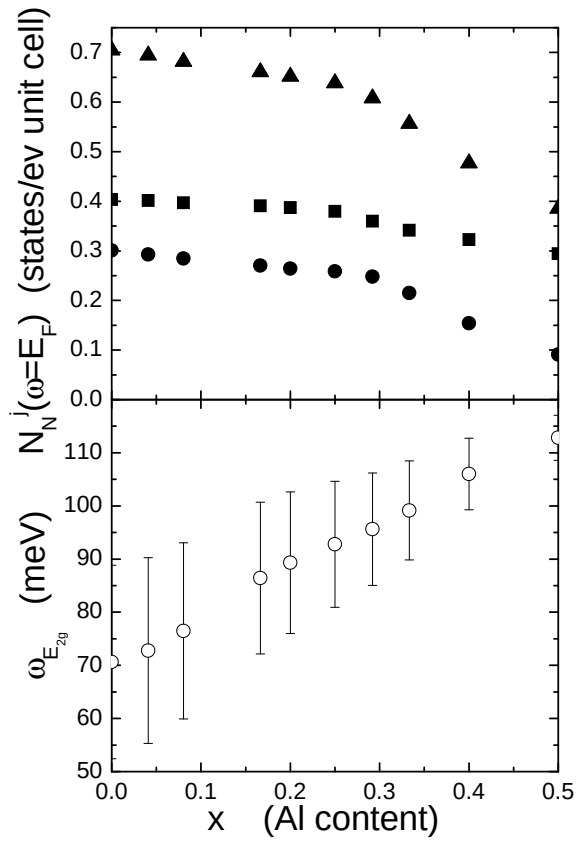

FIG. 3: Upper panel: calculated density of states at the Fermi energy $N_N(\omega = E_F, x)$ in the σ-band (filled circles), in the π-band (filled squares) and total (filled up triangles) as a functions of x. Lower panel: the phonon frequency $\omega_{E_{2g}}$ obtained from experimental data as a function of x, see ref. 6.

stant 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 Ω_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 [8] 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 Ω_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

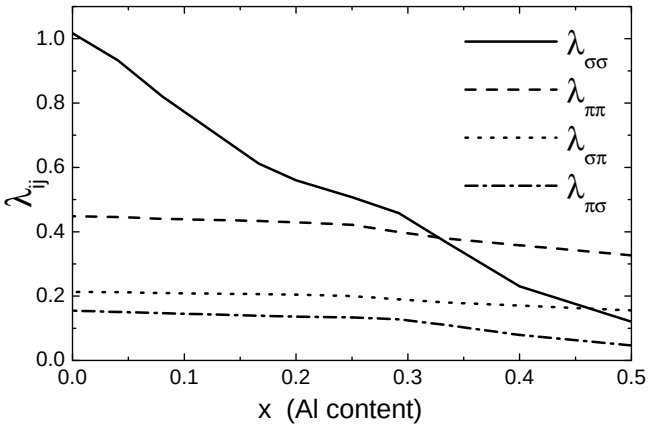

FIG. 4: Calculated electron phonon coupling constants λ_{ij} as a function of x in the four cases: $\sigma\sigma$ (solid line), $\pi\pi$ (dashed line), $\sigma\pi$ (dotted line) and $\pi\sigma$ (dashed dotted line).

system, the electron-phonon coupling constant $\lambda_{\sigma\sigma}$ is:

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

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 [26], we neglect its effects on the shape of the spectral function. For checking this hypothesis we have also used $\alpha^2_{\sigma\sigma}F_{\sigma\sigma}(\omega, x) = L(\omega, \Omega_0, \Upsilon) - L(\omega, -\Omega_0, \Upsilon)$ 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] $\lambda_{\sigma\sigma}(x = 0) = 1.017$, $\lambda_{\pi\pi}(x = 0) = 0.448$, $\lambda_{\sigma\pi}(x = 0) = 0.213$ and $\lambda_{\pi\sigma}(x = 0) = 0.155$.

In Fig. 4 we can see the calculated electron phonon coupling constants λ_{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]

$$\mu^*(p) = \begin{vmatrix} \mu^*_{\sigma\sigma} & \mu^*_{\sigma\pi} \\ \mu^*_{\pi\sigma} & \mu^*_{\pi\pi} \end{vmatrix} = \\ = \mu(\omega_c)N_{\rm N}^{tot}(E_{\rm F}, p)\begin{vmatrix} \frac{2.23}{N_{\rm N}^\sigma(E_{\rm F},p)} & \frac{1}{N_{\rm N}^\sigma(E_{\rm F},p)} \\ \frac{1}{N_{\rm N}^\pi(E_{\rm F},p)} & \frac{2.48}{N_{\rm N}^\pi(E_{\rm F},p)} \end{vmatrix} \tag{10}$$

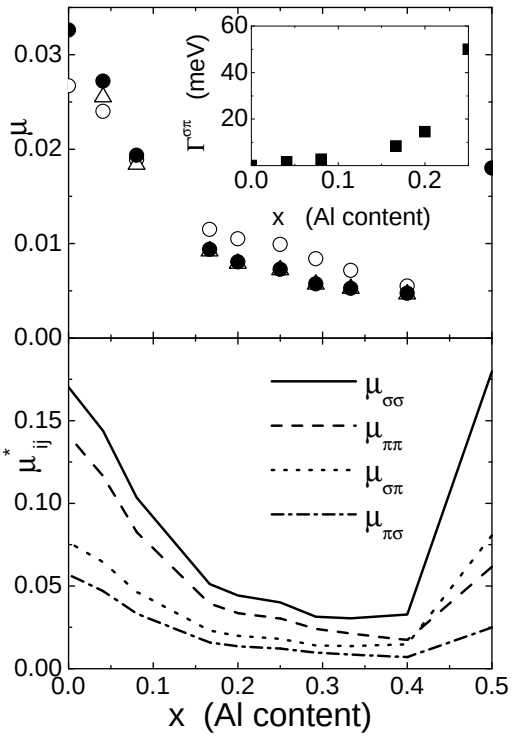

FIG. 5: Upper panel: the prefactor of the Coulomb pseudopotential μ as a function of x when the $\alpha^2_{\sigma\sigma}F(\omega,x)$ is a lorentzian (open circles), when is MgB_2-like (filled circles) and when is MgB_2-like and in presence of interband impurities, $\Gamma^{\sigma\pi}=1$ meV, (open up 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 μ^*_{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 $\alpha^2_{\sigma\sigma}F(\omega,x)$ spectral function case.

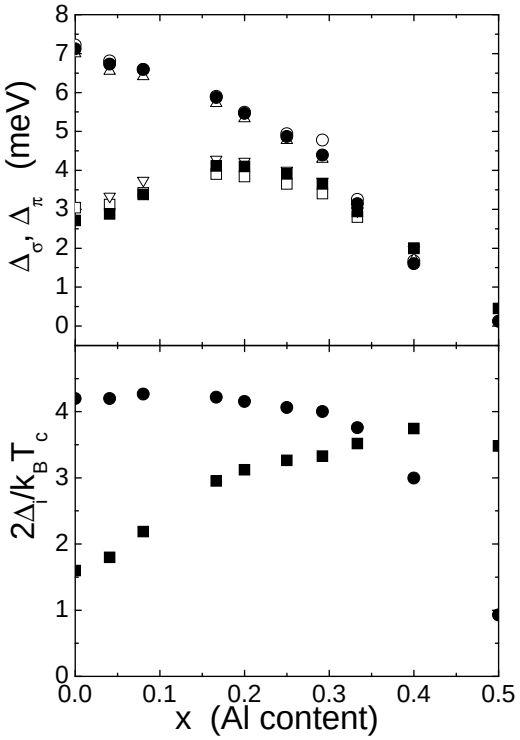

FIG. 6: Upper panel: the value of $\Delta_i(i\omega_{n=0})$ of the σ-band (open circles) and of the π-band (open squares) calculated on imaginary axis, at $T=T_c/4$, versus x when the $\alpha^2_{\sigma\sigma}F(\omega,x)$ is a lorentzian, when is MgB_2-like (filled circles and squares) and when is MgB_2-like and in presence of interband impurities, $\Gamma^{\sigma\pi}=1$ meV (open up and down triangles). Lower panel: calculated rate $2\Delta_i/k_BT_c$ for the σ-band (filled circles) and of the π-band (filled squares) in the MgB_2-like $\alpha^2_{\sigma\sigma}F(\omega,x)$ spectral function case.

where $\mu(\omega_c,x)$ is a free parameter and and $N_N^{tot}(\omega=E_F,x)$ 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 Γ^{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, $\Gamma^{\sigma\pi}$, because $\Gamma^{\sigma\sigma}$ and $\Gamma^{\pi\pi}$ don't affect [27] 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 $\Gamma^{\sigma\pi}$ growths 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 $\Gamma^{ii}\gg\Gamma^{ij}\approx 1$ meV i.e. Γ^{ij} values found are completely to outside of the range expected. As second possibility we examine the case [27] $\Gamma^{\sigma\pi}=1$ 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 $\mu(\omega_c,x)$

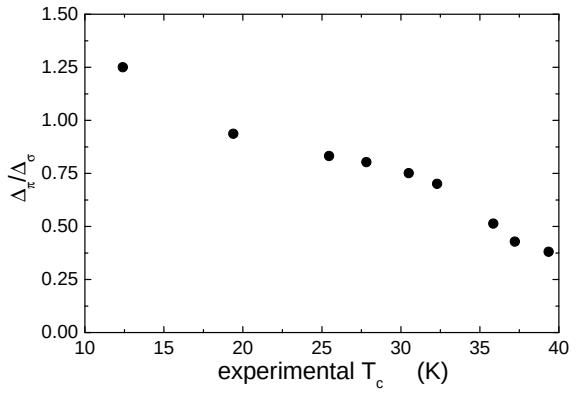

FIG. 7: The calculated rate of Δ_π/Δ_σ as a function of experimental T_c.

(upper panel) while in the lower panel we can see the values of $\mu^*_{ij}(x)$ 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 $\Delta(i\omega_{n=0})$, at $T = T_c/4$, of the σ-band and the π-band (solid circles and solid squares respectively) versus x. The values calculated are almost the same of those found by solution of the equation $\Delta^0_i = \Delta_i(\omega = \Delta^0_i)$ 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 π-gap, as a function of x has a maximum for $0.164 < x < 0.198$ while the σ-gap decreases and, for $x \geq 0.4$, 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 $x \simeq 0.4$. In the lower panel we show the calculated ratio $2\Delta_i/k_BT_c$. Finally in Fig. 7 we can see the calculated rate of Δ_π/Δ_σ as a function of experimental T_c (which is preferable due to the loch 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 σ-gap is almost unchanged, while the one of the π-gap changes, and decreases monotonically. We have no arguments to justify the omission of the dependence of $\lambda_{\sigma\sigma}$ on $\omega_{E_{2g}}$.

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 π-band).

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