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Theoretical Study of Screening Dependence of Aluminium Doped MgB_2

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The screening dependence of superconducting state parameters (λ, μ^*, T_c, α and N_0V) of six alloys of aluminium doped MgB_2 systems are studied in the BCS–Eliashberg–McMillan framework by employing five forms of dielectric screening function, viz. random phase approximation (RPA), Harrison, Geldart and Vosko (GV), Hubbard and Overhauser in conjunction with Ashcroft's potential. It is observed that electron-phonon coupling strength λ and Coulomb pseudopotential μ^* are quite sensitive to the form of dielectric screening, whereas transition temperature T_c, isotope effect exponent α and effective interaction strength N_0V show weak dependence on the form of dielectric screening function. It is found that the RPA form of dielectric screening function yields the best results for transition temperature T_c for all alloys of the Mg-Al-B system. The results obtained using GV screening are much higher than the experimental results. This shows that all the four dielectric screenings used here almost describe superconductivity in all the alloys of the Mg-Al-B system, but the GV screening is not suitable for such an alloy system.

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MgB_2 initiated an interest of activity aimed at understanding the origin of the large superconducting transition temperature T_c since the discovery of its superconductivity in 2001.[1] Extensive research into substitution chemistry has shown that only a few elements can substitute onto the Mg site[2] (Al,[3–6] Li,[7] and Cu[8]). However, so far there have been no reports on an improvement of superconducting properties with doping on the Mg site.[9,10] It is possible that the doping levels studied have been too high or that poor micro structures have obscured any improved properties. Although the structure of MgB_2 is rather simple, the Al substitution chemistry is quite complicated. Structural investigations of Al substituted MgB_2 polycrystalline samples show the existence of a miscibility gap.[11] A number of groups undertook synthesis and characterization of $(Mg_{1-x}W_x)B_2$ (W = transition metal, Li, Be, Al) materials. Slusky *et al.*[5] have shown that Al can aggressively react with MgB_2 and the substitution into the Mg position leads to a loss of superconductivity.

In this Letter, we report on the effect of various doping levels of Al on the superconducting properties of MgB_2. The empty core model pseudopotential theory due to Ashcroft[12] is a simple one parameter potential, which has been found to work well for metallic systems and binary alloys.[13,14] This potential when used with a suitable form of dielectric screening has also been found to yield good results in predicting superconducting state parameters of metallic systems.[15,16] So we decide to employ this potential in the present work. To determine the best form of dielectric screening used with Ashcroft's potential for predicting the superconducting state parameters for Al doped MgB_2 systems, we use five different forms of dielectric screening in conjunction with Ashcroft's potential, viz. random phase approximation (RPA) screening due to Gellamann and Brueckner[17] Harrison (Ha),[18] Geldart and Vosko (GV),[19] Hubbard (HB)[20] and Overhauser (OH).[21]

We follow McMillan[22] in defining electron-phonon coupling strength by

$$\lambda = 2\int_0^\infty [\alpha^2(\omega)F(\omega)/\omega], \tag{1}$$

where $\alpha^2(\omega)F(\omega)$ is the spectral function. When appropriately evaluated in the plane-wave approximation for scattering on the Fermi surface, it yields[23]

$$\lambda = \frac{m^*}{4\pi^2 k_F MN\langle\omega^2\rangle}\int_0^{2k_F} dqq^3|V_s(q)|^2, \tag{2}$$

where q is the change in the electron wave vector k in scattering on the Fermi surface, m^* is the effective mass of electron, M is the ionic mass, N is the ion number density, k_F is the Fermi radius, $\langle\omega^2\rangle$ is the average square phonon frequency, and $V_s(x)$ is the screened potential. Using $x = q/2k_F$ and $N^{-1} = \Omega_0 = 3\pi^2 z^*/k_F^3$, we obtain

$$\lambda = \frac{12m^*z^*}{M\langle\omega^2\rangle}\int_0^1 dxx^3|V_s(x)|^2. \tag{3}$$

The Coulomb pseudopotential (μ^*)[23] for the alloy of the $Mg_{1-x}Al_xB_2$ system may be written by extending the relevant formula as

$$\mu^* = \frac{\frac{m_b}{\pi k_F}\int_0^1 \frac{dx}{x\epsilon(x)}}{[1+\frac{m_b}{\pi k_F}\ln(\frac{k_F{}^2}{20\theta_D})\int_0^1 \frac{dx}{x\epsilon(x)}]}, \tag{4}$$

where $\varepsilon_H(x)$ is the Hartree function. The relevant expression of Ashcroft's potential[23] and $\varepsilon_H(x)$ are given (in a.u.) by[24]

$$V_s(x) = \frac{-\pi z^*\cos(2k_F r_C x)}{\Omega_0 k_F^2 x^2 \mathcal{E}(x)}, \tag{5}$$

where

$$\varepsilon_H(x) = 1 + \frac{m_b}{\pi k_F x^2}\left\{0.5 + \frac{(1-x^2)}{4x}\ln\left|\frac{(1+x)}{(1-x)}\right|\right\} + 1. \tag{6}$$

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The symbols m^*, z^*, Ω_0, $k_{\rm F}$, M and $\theta_{\rm D}$ denote the effective mass, effective valence, Fermi wave vector, ionic mass and Debye temperature for the alloys, respectively. In the present work we use $m_{\rm b} = m^*$ for simplicity. To obtain the dielectric dependence of superconducting state parameters, we consider the dielectric screening function due to RPA screening, GB,[17] Ha,[18] GV,[19] HB[20] and OH.[21]

In the present work we have chosen the RPA screening function, which is a modified form of the Hartree screening function where the band mass ($m_{\rm b}$) is replaced by the effective mass (m^*),

$$\varepsilon_{\rm RPA}(x) = 1 + \frac{m^*}{\pi k_{\rm F} x^2}\left\{0.5 + \frac{(1-x^2)}{4x}\ln\left|\frac{(1+x)}{(1-x)}\right|\right\} + 1. \tag{7}$$

The relevant form of the modified dielectric screening function is given by[16]

$$\varepsilon(x) = 1 + \{(1 - f(x))(\varepsilon_{\rm H}(x) - 1)\}, \tag{8}$$

where $\varepsilon(x)$ is the modified screening function, and $\varepsilon_{\rm H}(x)$ is the Hartree function.[16] Functions $f(x)$ due to Ha and GV are given as follows:[18,19]

$$f_{\rm Ha}(x) = \frac{1}{2}\left[\frac{x^2}{x^2 + \frac{1}{3}}\right], \tag{9}$$

$$f_{\rm GV}(x) = \frac{2x^2}{4x^2 + v}, \tag{10}$$

with $v = \frac{2}{[1+0.153(\frac{m^*}{\pi k_{\rm F}})]}$.

The relevant forms of dielectric screening function due to HB and OH[20,21] read

$$\varepsilon(x) = 1 + \frac{Q(x)}{1 - f(x)Q(x)}, \tag{11}$$

where

$$Q(x) = \epsilon_{\rm HAR}(x) - 1, \tag{12}$$

$$f_{\rm HB}(x) = \frac{2x^2}{1 + 4x^2 + \frac{4m^*}{\pi k_{\rm F}}}, \tag{13}$$

$$f_{\rm OH}(x) = \frac{1.1x^2}{(1 + 10x^2 + 1.50x^4)^{\frac{1}{2}}}. \tag{14}$$

The relevant expressions of the transition temperature ($T_{\rm c}$), isotope effect exponent (α), and effective interaction strength (N_0V)[24] for the $Mg_{1-x}Al_xB_2$ system are taken as

$$T_{\rm c} = \frac{\theta_{\rm D}}{1.45}\left\{\exp\left(\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right)\right\}, \tag{15}$$

$$\alpha = \frac{1}{2}\left[1 - \left(\mu^* \ln\frac{\theta_{\rm D}}{1.45T_{\rm C}}\right)^2 \frac{1+0.62\lambda}{1.04(1+\lambda)}\right], \tag{16}$$

$$N_0V = \frac{\lambda - \mu^*}{1 + (\frac{10}{11})\lambda}. \tag{17}$$

Thus for different concentrations of x in the $Mg_{1-x}Al_xB_2$ system, we can calculate the superconducting state parameters, viz. λ, μ^*, $T_{\rm c}$, α and N_0V using the formulation mentioned above.

The values of input parameters and constant relevant to the components of the $Mg_{1-x}Al_xB_2$ system have been taken from Sharma *et al.*[25] In the present work we use $m_{\rm b} = m^*$ for the sake of uniformity. The values of m^*, $\langle\omega^2\rangle$, z^* and M for the $Mg_{1-x}Al_xB_2$ system under investigation are obtained from the relevant values for the component using Vegard's rule[26]

$$V_{\rm s} = \frac{1}{3}[(1-x)V_{\rm s}({\rm Mg}) + xV_{\rm s}({\rm Al}) + 2V_{\rm s}({\rm B})]. \tag{18}$$

The values of the Debye temperature ($\theta_{\rm D}$) and atomic volume (Ω_0) for the $Mg_{1-x}Al_xB_2$ system are computed from the relevant values using Grimvall's formula[27]

$$\frac{1}{\theta_{\rm D}^2} = \frac{1}{3}\left[\frac{1-x}{\theta_{\rm DMg}^2} + \frac{x}{\theta_{\rm DAl}^2} + \frac{2}{\theta_{\rm DB}^2}\right]. \tag{19}$$

Table 1 lists the present calculated values of the SSPs, viz. electron-phonon coupling strength λ, the Coulomb pseudopotential μ^*, transition temperature $T_{\rm c}$, isotope effect exponent α and effective interaction strength N_0V at various concentrations $x = 0.00$, 0.03, 0.08, 0.16, 0.20 and 0.28 for the $Mg_{1-x}Al_xB_2$ system with available experimental findings.[11]

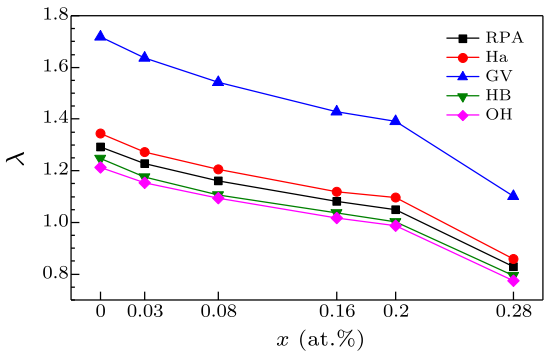

Fig. 1. Variation of electron-phonon coupling strength (λ) with Al-concentration x (at.%) for five dielectric screenings.

The calculated values of the electron-phonon coupling strength (λ) for MgB_2, $Mg_{0.97}Al_{0.03}B_2$, $Mg_{0.92}Al_{0.08}B_2$, $Mg_{0.84}Al_{0.16}B_2$, $Mg_{0.80}Al_{0.20}B_2$ and $Mg_{0.72}Al_{0.28}B_2$ alloys using five different screening functions with Ashcroft's potential are listed in Table 1. It provides the computed values of electron-phonon coupling strength λ for the $Mg_{1-x}Al_xB_2$ system with Ashcroft's potential using five different forms of dielectric screening. Variation of electron-phonon coupling strength λ with different concentrations of aluminium has been shown for five different forms of dielectric screening in Fig. 1. We can find from Fig. 1 that λ is quite sensitive to the form of dielectric screenings. The GV screening yields the highest values of electron-phonon coupling strength λ, whereas the values obtained from the other screenings are lower. The GV screening is suitable for metals and elements whereas it is not suitable for binary glasses and alloys.[23] The magnitude of λ lies between 1.7194 and 0.7735 as the concentration of Al increases, which indicates that the $Mg_{1-x}Al_xB_2$ system has strong coupling behavior to weaken the coupling behavior of

electrons and phonons. MgB_2 superconductors have four bands at the Fermi energy E_F. Al occupies Mg sites[28–30] and dopes in MgB_2 with electrons to form the $Mg_{1-x}Al_xB_2$ system. This doping decreases both the π-band and the σ-band energy gaps. The superconductivity in MgB_2 is mainly due to the σ bands.[31] MgB_2 samples can be understood mainly in terms of a band filling effect due to the electron doping by Al. Mora *et al.*[32] found that the Al extra electron starts to fill up the σ bands, and at 56% of Al replacement these bands become saturated and the system is no longer superconducting,[31] thus SSP λ and T_c decrease with the rising doping level of Al in the $Mg_{1-x}Al_xB_2$ system.[33,34]

The values of Coulomb pseudopotential μ^*, which accounts for the Coulomb interaction between the conduction electrons, obtained from five different forms of dielectric screening are tabulated in Table 1. It is observed from Table 1 that μ^* is quite sensitive to dependence on dielectric screening. The computed results of μ^* for the $Mg_{1-x}Al_xB_2$ system under consideration lie between 0.15 and 0.26, which are in accordance with McMillan who suggested $\mu^* = 0.13$ for transition metals.[22] The graph of μ^* versus concentration x (at.%) for different dielectric screenings is plotted in Fig. 2. It is also observed that the Coulomb pseudopotential μ^* remains almost constant as the concentration of Al increases in the $Mg_{1-x}Al_xB_2$ system.

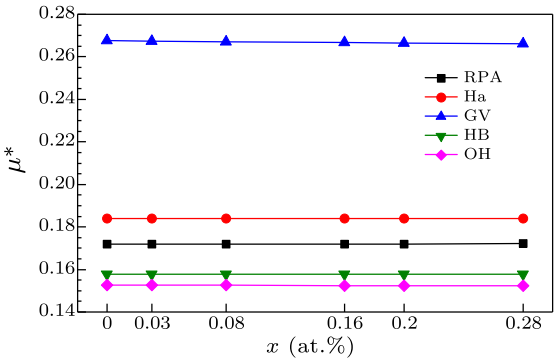

Fig. 2. Variation of Coulomb pseudopotential (μ^*) with Al-concentration x (at.%) for five dielectric screenings.

Table 1. Computed values of superconducting parameters of the $Mg_{1-x}Al_xB_2$ system for five different dielectric screenings.

Alloys	SSP's	Calculated results					Exp.[11]	Others
		RPA	Ha	GV	HB	OH		
	λ	1.2915	1.3447	1.7194	1.2466	1.2134		1.7, 1.4, 1.08, 0.90, 0.87, 0.842[35]
	μ^*	0.1720	0.1840	0.2677	0.1579	0.1526		0.1, 0.15, 0.14, 0.130.12[35]
MgB_2	T_c [K]	38.41	38.6	38.51	38.82	38.19	38.4	39, 39.25, 40, 38, 36.27[35]
	α	0.4341	0.4254	0.3460	0.4447	0.4475		0.462, 0.32, 0.355, 0.30, 0.468[35]
	N_0V	0.5149	0.5222	0.5664	0.5103	0.5044		0.407, 0.477[35]
	λ	1.2278	1.2720	1.6374	1.1748	1.1542		
	μ^*	0.1720	0.1840	0.2675	0.1579	0.1526		
$Mg_{0.97}Al_{0.03}B_2$	T_c [K]	35.58	35.55	35.86	35.49	35.40	35.5	38,[36–38] 35,[29,32]
	α	0.4294	0.4195	0.3359	0.4400	0.4438		
	N_0V	0.4989	0.5045	0.5504	0.4917	0.4888		
	λ	1.1616	1.2050	1.5429	1.1069	1.0949		
	μ^*	0.1720	0.1840	0.2672	0.1579	0.1525		
$Mg_{0.92}Al_{0.08}B_2$	T_c [K]	32.52	32.57	32.66	32.21	32.52	32.5	37,[36] 35,[37] 35.5,[38] 32.5[29,32]
	α	0.4236	0.4131	0.3224	0.4348	0.4395		
	N_0V	0.4813	0.4872	0.5309	0.4730	0.4723		
	λ	1.0815	1.1178	1.4291	1.0359	1.0169		
	μ^*	0.1721	0.1840	0.2667	0.1579	0.1524		
$Mg_{0.84}Al_{0.16}B_2$	T_c [K]	28.59	28.51	28.61	28.66	28.56	28.6	32.5,[36,38,39] 31,[37] 29[29,32]
	α	0.4144	0.4028	0.3022	0.4281	0.4327		
	N_0V	0.4583	0.4631	0.5055	0.4521	0.4492		
	λ	1.0496	1.0968	1.3910	1.0016	0.9878		
	μ^*	0.1721	0.1841	0.2665	0.1579	0.1524		
$Mg_{0.80}Al_{0.20}B_2$	T_c [K]	27.09	27.52	27.2	26.89	27.05	27	31,[36,38,39] 29,[29,32] 27.5[37]
	α	0.4111	0.3999	0.2944	0.4243	0.4297		
	N_0V	0.4490	04570	0.4965	0.4415	0.4401		
	λ	0.8299	0.8593	1.1005	0.7931	0.7735		
	μ^*	0.1721	0.1841	0.2661	0.1579	0.1523		
$Mg_{0.72}Al_{0.28}B_2$	T_c [K]	15.54	15.43	15.5	15.52	15.20	15.5	27,[36,38] 26,[39] 21,[37] 9[32]
	α	0.3683	0.3493	0.1947	0.3884	0.3946		
	N_0V	0.3749	0.3791	0.4171	0.3691	0.3647		

Table 1 contains the computed values of T_c for six alloys obtained from five different forms of dielectric screening along with the experimental data.[11] The values of T_c with varying concentrations of Al in the $Mg_{1-x}Al_xB_2$ system have been plotted for dielectric screening in Fig. 3 with the experimental data. The values of T_c remain approximately the same for different dielectric screenings. It is observed that T_c shows weak dependence on dielectric screening. However, the results of T_c using RPA dielectric screening are in the best agreement with the experimental data of the $Mg_{1-x}Al_xB_2$ system under investigation, as the curve for RPA screening almost overlaps the experimental curve. The values of T_c decrease with the increasing Al concentration in the $Mg_{1-x}Al_xB_2$ system as shown in Table 1 and Fig. 3.[5,29,38,40–42] It is observed from Fig. 3 that T_c for the Al doping decreases linearly from 38.7 K to 28.6 K down to 27 K or 15.5 K as the

concentration x changes from 0.0 to 0.16, whereas a sudden change is observed at concentration $x = 0.20$. Figure 3 shows that the value of T_c remains high at $x = 0.20$ as suggested by Li *et al.*, which shows a kink at $x = 0.20$.[42] Experiments also show an abrupt topological change observed at $x = 0.33$ due to the Fermi surface (FS) topology.

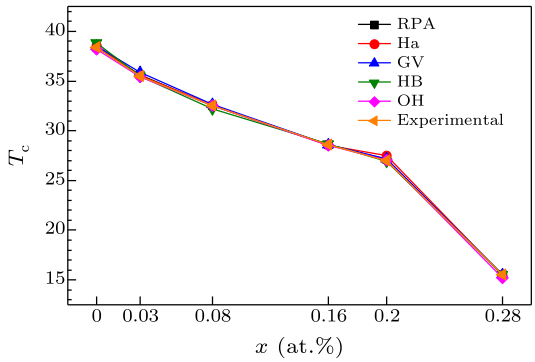

Fig. 3. Variation of transition temperature T_c with Al-concentration x (at.%) for five dielectric screenings.

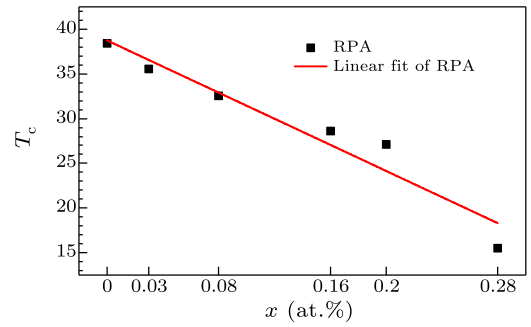

Fig. 4. Fitted T_c equation showing variation of T_c with Al-concentration x (at.%).

The linear regression T_c proposed in the present work also closely resembles the linear regression T_c as suggested by Klie *et al.*[43] The composition dependence can be described by linear regression of the data obtained for RPA screening for different values of x, which yields

$$T_c = -73.04x + 38.75. \tag{20}$$

The plot of fitted T_c equation is represented in Fig. 4, which indicates that T_c drops almost linearly with increasing Al content with a slope $dT_c/dx = -73.04/38.75$. Wide extrapolation predicts a $T_c = 38.01$ K for the hypothetical case of 'amorphous pure AlB_2'. Wide extrapolation also predicts $T_c = 2.23$ K in the hypothetical case for the $Mg_{1-x}Al_xB_2$ system at $x = 0.5$, i.e., $MgAlB_4$ in the end member of this system. This predicts that T_c in the present work is very close to $T_c = 3$ K according to many experimental and theoretical results.[33,44] As a result, the inter-band scattering increases and additionally, DOS in the hole σ band decreases. Transmission electron microscopy (TEM)[11] also shows that the aluminium substitution leads to the formation of superstructure $MgAlB_4$ which inter grows with MgB_2 that leads to the suppression of superconductivity. SSP's of MgB_2, carbon doped MgB_2 and many binary metallic glasses have been calculated using these equations theoretically as well as experimentally.[16,26,35]

The values of isotope effect exponent α for the six alloys of the $Mg_{1-x}Al_xB_2$ system for the five different forms of dielectric screening have been tabulated in Table 1. Figure 5 shows the variation of isotope effect exponent α with aluminium concentration x for different screenings. The computed values of isotope effect α show feeble dependence on the dielectric screening functions. The values obtained using GV screening yield the lowest values than the other screenings. It is observed that the value of isotope effect exponent α decreases continuously by increasing the Al concentration of the $Mg_{1-x}Al_xB_2$ system in all the five forms of dielectric screening, which suggests that the superconductivity is suppressed as the relative concentration of Al increases. The magnitude of α for the $Mg_{1-x}Al_xB_2$ system lies between 0.4447 and 0.1947, which show weak coupling behavior. Decrease in α with increase in x indicates less of a role of ionic vibrations in the superconductivity of these alloys. The experimental value of α has not been reported in the literature so far, the present data of α may be used for studying ionic vibrations in the superconductivity. Since RPA screening yields good results for T_c and λ, it may be observed that results of α due to RPA screening provide the best account of the role of ionic vibrations in the superconducting behavior of this system. Other forms overestimate α.

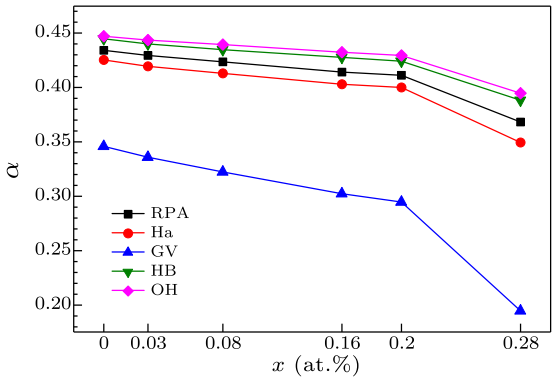

Fig. 5. Variation of isotope effect exponent (α) versus concentration x (at.%) for five dielectric screenings.

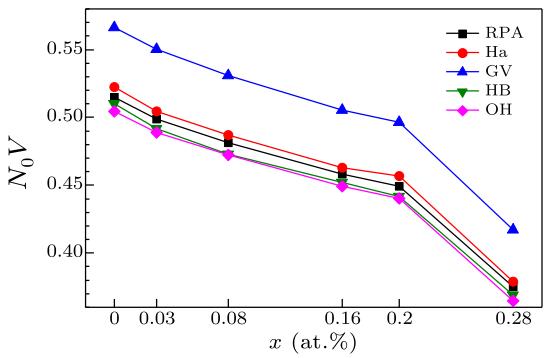

Fig. 6. Variation of interaction strength (N_0V) with Al-concentration x (at.%) for five dielectric screenings.

The values of interaction strength N_0V for the

$Mg_{1-x}Al_xB_2$ system for the five different forms of dielectric screening are tabulated in Table 1. Figure 6 shows the variation of interaction strength N_0V with Al concentration x for different screenings. It is observed from Table 1 as well as Fig. 6 that the values of interaction strength N_0V for the $Mg_{1-x}Al_xB_2$ system decrease continuously by increasing the aluminium concentration for all the five forms of dielectric screening. It is also observed that the magnitudes of interaction strength (N_0V) for the $Mg_{1-x}Al_xB_2$ system under investigation lie between 0.5664 and 0.3647, which are in the range of strong-to-weak interaction superconductors. The result obtained using GV screening yields slightly higher than the other screening, whereas the results obtained using other screenings remain almost the same. The values of interaction strength N_0V show weak dependence to the form dielectric screening. The N_0V values of Al doped MgB_2 are not available in theoretical or experimental data for further comparisons.

In summary, we have shown that electron-phonon coupling strength λ and Coulomb pseudopotential μ^* are quite sensitive to the form of dielectric screening, whereas transition temperature T_c, isotope effect exponent α and effective interaction strength N_0V show weak dependence on the form of dielectric screening function. It is observed that the superconducting parameters of the aluminium doped MgB_2 system is composition dependent, i.e., they vary with the change in the concentration of component metals. The present study also proves that the RPA form of dielectric screening when used with Ashcroft's potential provides the best explanation for the superconductivity in the aluminium doped MgB_2 system. A linear T_c equation is proposed by fitting the present results for RPA screening, which is in conformity with other results for the experimental data.

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