Effects of Disorder in $\text{Mg}_{1-x} \text{Ta}_x \text{B}_2$ Alloys using Coherent-Potential Approximation

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Abstract

Using Korringa-Kohn-Rostoker coherent-potential approximation in the atomic-sphere approximation (KKR-ASA CPA) method for taking into account the effects of disorder, Gaspari-Gyorffy formalism for calculating the electron-phonon coupling constant $\lambda$, and Allen-Dynes equation for calculating $T_c$, we have studied the variation of $T_c$ in $\text{Mg}_{1-x} \text{Ta}_x \text{B}_2$ alloys as a function of $\text{Ta}$ concentration. Our results show that the $T_c$ decreases with the addition of $\text{Ta}$ for up to 40 at% and remains essentially zero from 60 at% to 80 at% of $\text{Ta}$. We also find $\text{TaB}_2$ to be superconducting, albeit at a lower temperature. Our analysis shows that the variation in $T_c$ in $\text{Mg}_{1-x} \text{Ta}_x \text{B}_2$ is mostly dictated by the changes in the $B\text{p}$ density of states with the addition of $\text{Ta}$.

The experimental [1–4] and theoretical [5–12] efforts aimed at understanding the nature of superconductivity in $\text{MgB}_2$ have made substantial progress since the discovery of superconductivity in it [1]. With an enhanced understanding of superconductivity in $\text{MgB}_2$, attempts are now being made to understand the changes in the electronic structure and the superconducting properties of $\text{MgB}_2$ upon alloying with various elements. Such efforts provide an opportunity to explore the possibility of obtaining $\text{MgB}_2$ alloys with improved superconducting properties.

The changes in the superconducting properties of $\text{MgB}_2$ due to substitution of various elements such as $\text{Be}$, $\text{Li}$, $\text{C}$, $\text{Al}$, $\text{Na}$, $\text{Zn}$, $\text{Zr}$ and others have been studied experimentally [4,13–18]. Some theoretical work has also been reported within the rigid-band model. The main effects of alloying are seen to be (i) a decrease in transition temperature, $T_c$, with increasing concentration of the alloying elements except for $\text{Zn}$ (and possibly $\text{Li}$) where $T_c$ is seen to increase somewhat, (ii) changes in lattice parameters $a$ and $c$, and (iii) a change in crystal structure.
With a view to understand the changes in the electronic structure and the superconducting properties as well as to clarify the reported superconductivity in TaB2 [19,20], we have carried out an ab initio study of Mg$_{1-x}$Ta$_x$B$_2$ alloys. We have used Korringa-Kohn-Rostoker coherent-potential approximation [21] within the atomic-sphere approximation (KKR-ASA CPA) method for taking into account the effects of disorder, Gaspari-Gyorffy formalism for calculating the electron-phonon coupling constant $\lambda$, and Allen-Dynes equation for calculating $T_c$ in Mg$_{1-x}$Ta$_x$B$_2$ alloys as a function of Ta concentration. Such an attempt allows us to examine the possibility of superconductivity in TaB$_2$, given the superconductivity in MgB$_2$. We have analyzed our results in terms of the changes in the total density of states (DOS), in particular the changes in the B $p$ contribution to the total DOS, as a function of Ta concentration.

Based on our calculations, described below, we find that in Mg$_{1-x}$Ta$_x$B$_2$ alloys (i) the $T_c$ decreases with the addition of Ta for upto 40 at%, remains essentially zero from 60 at% to 80 at%, and then rises to $\sim$ 1.8 K for TaB$_2$, (ii) the $T_c$ for TaB$_2$ is much lower than reported earlier [19], and (iii) the variation in $T_c$ is mostly dictated by the changes in the B $p$ densities of states as more and more Ta are added. Before we describe our results, we outline some of the computational details.

The charge self-consistent electronic structure of Mg$_{1-x}$Ta$_x$B$_2$ alloys as a function of $x$ has been calculated using the KKR-ASA CPA method [22]. We have used the CPA rather than a rigid-band model because CPA has been found to reliably describe the effects of disorder in metallic alloys. We parametrized the exchange-correlation potential as suggested by Perdew-Wang [23,24] within the generalized gradient approximation. The Brillouin zone (BZ) integration was carried out using 1215 $k$-points in the irreducible part of the BZ. For DOS calculations, we added a small imaginary component of 2 $m$Ry to the energy and used 3887 $k$-points in the irreducible part of the BZ. The lattice constants for MgB$_2$ and TaB$_2$ were taken from experiments while for other compositions we used the Vegard’s law. The Wigner–Seitz radii for Mg and Ta were slightly larger than that of B. The sphere overlap, which is crucial in ASA, was less than 10% and the maximum $l$ used was $l_{max} = 3$.

The electron-phonon coupling constant $\lambda$ was calculated using Gaspari-Gyorffy [25] formalism with the charge self-consistent potentials of Mg$_{1-x}$Ta$_x$B$_2$ obtained with the KKR-ASA CPA method. Subsequently, the variation of $T_c$ as a function of Ta concentration was calculated using Allen-Dynes equation [26]. The average values of phonon frequencies $\omega_{ln}$ for TaB$_2$ and MgB$_2$ were taken from Refs. [27] and [28] respectively. For intermediate concentrations, we took $\omega_{ln}$ to be the concentration-weighted average of MgB$_2$ and TaB$_2$. 

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The main results of our calculations are shown in Fig. 1, where we have plotted the variation in $T_c$ of $Mg_{1-x}Ta_xB_2$ alloys as a function of $Ta$ concentration. The calculations were carried out as described earlier with the same value of $\mu^* = 0.09$ for all the concentrations. The $T_c$ for $MgB_2$ is equal to $\sim 30\,K$, which is consistent with the results of other works [28] with similar approximations. The corresponding $\lambda$ is equal to 0.73. As a function of $Ta$ concentration we find that the $T_c$ decreases rapidly and goes to zero at around 40 at\% of $Ta$ as shown in Fig. 1. The $T_c$ remains essentially zero until around 75 – 80 at\% of $Ta$, thereafter rising to 1.8 $K$ for $TaB_2$ with $x = 1$. The corresponding $\lambda$ for $TaB_2$ is equal to 0.38..
Fig. 2. The total density of states for $Mg_{1-x}Ta_xB_2$ as a function of Ta concentration $x$, calculated with the charge self-consistent KKR-ASA CPA method, as described in the text. Note that in (f), the vertical scale is different from the rest of the figures. The vertical line denotes the Fermi energy.

In order to get further insight into the variation of $T_c$ as a function of Ta concentration in $Mg_{1-x}Ta_xB_2$ alloys, we have analyzed the total DOS as well as the contribution of $B$ $p$-electrons to the total DOS. In Figs. 2(a) – (f), we show the total DOS of $Mg_{1-x}Ta_xB_2$ alloys for Ta concentration ranging from $x = 0$ to $x = 1$, calculated using the KKR-ASA CPA method as described earlier. The DOS for $MgB_2$ and $TaB_2$, as shown in Figs. 2(a) and 2(f) are similar to previous calculations [28,29]. With small addition of Ta the Fermi energy, $E_F$, moves outward to accommodate the additional electrons resulting in a decrease in the total DOS at $E_F$ for $Mg_{1-x}Ta_xB_2$ alloys, leading to a decrease in $T_c$. Further addition of Ta allows $E_F$ to be pinned in a valley around 50 – 60 at% of Ta, resulting in a decreased donor ability of the metallic
plane \((Mg - Ta)\) with the increase in the number of \(d\) electrons. Thus the overall contribution of \(B\) electronic states decreases as the concentration of \(Ta\) is increased up to around 80 at\% of \(Ta\). Starting with around 80 at\% of \(Ta\) the \(B\) contribution rises and so does the \(T_c\) as shown in Fig. 1. To substantiate these qualitative observations we have listed in Table I the total DOS and the \(B_p\) DOS at \(E_F\). In addition, in Fig. 3, we show the \(B_{px}\) and \(p_z\) partial DOS as a function of concentration. It is clear from Table I and Fig. 3 that the \(B_p\) DOS is responsible for the variation in \(T_c\) of \(Mg_{1-x}Ta_xB_2\) alloys, and the possible loss of superconductivity in these alloys at around 60 at\% of \(Ta\) can be attributed to a very small \(B_{px,y}\) DOS at \(E_F\). In Table I, we have also listed the lattice constants used in our calculations.

In conclusion, we have studied the variation of \(T_c\) in \(Mg_{1-x}Ta_xB_2\) alloys as a function of \(Ta\) concentration. We have used the KKR-ASA CPA method for taking into account the effects of disorder and Gaspari-Georffy formalism for calculating the electron-phonon coupling constant \(\lambda\). The \(T_c\) is then calculated using the Allen-Dynes equation. Our results show that the \(T_c\) decreases with the addition of \(Ta\) for up to 40 at\%, thereafter remains essentially zero up to 80 at\% of \(Ta\). We find \(TaB_2\) to be superconducting, albeit at a low temperature. We have also shown that the variation in \(T_c\) is mostly dictated by the changes in the \(B_p\) density of states.

We would like to thank Dr. I. A. Abrikosov for providing a copy of his locally self-consistent Green’s function code.
Fig. 3. $B\, p$ partial DOS showing $p_x$ (full line) and $p_z$ (dashed line) for $Mg_{1-x}Ta_xB_2$. The vertical line denotes the Fermi energy. Note that for $x = 1$ the vertical scale is different from the rest of the plots.

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