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Difference between Al and C doping in anisotropic upper critical field development in MgB_2

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The temperature dependence of the upper critical field H_{c2} for both field directions in partially substituted polycrystalline MgB_2 was determined. Whereas the suppression of T_c is similar for aluminum and carbon substituted samples, H_{c2} is affected by the substitution in profoundly different ways. In the case of Al substitution changes can tentatively be described by intrinsic effects (shift of the Fermi level). In the C substituted samples, H_{c2} is increased drastically, and extrinsic effects (disorder) have to play a major role. The strong contrast between the two substitutions is discussed, taking into account three relevant scattering rates.

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An unusual temperature dependence of the anisotropic upper critical field H_{c2} is one of the major consequences of two-band superconductivity as realized in magnesium diboride MgB_2.[1] To further explore properties of a given compound it is often helpful to consider the effects of partial chemical substitutions. In the case of MgB_2, partial substitutions with many elements have been attempted, but only two elements are widely recognized to enter the structure: aluminum replacing magnesium[2] and carbon replacing boron.[3]

Both substitutions dope the material with additional electrons, which should similarly affect the superconducting properties, at least to the extent that a rigid band approximation works. According to the detailed band structure calculations[4–6] electron doping most drastically affects the σ bands, which are nearly filled. Furthermore, any partial substitution by small amounts of an additional element increases chemical disorder, leading to increased scattering. In the two-band superconductor MgB_2, at least three different scattering rates have to be distinguished,[7] and the different substitution sites Mg (by Al) and B (by C) are likely to influence these scattering rates in drastically different ways. In general, the upper critical field H_{c2} is influenced by electron-phonon coupling (EPC), Fermi velocities, and by the mean free path ℓ. EPC and Fermi velocities are intrinsic properties altered by electron doping, while ℓ is a function of scattering. It will be interesting to compare the doping and temperature dependence of H_{c2} with substitutions on either the Mg or the B site. This may help in separating electron doping and scattering effects of partial substitutions.

In the case of the B site substitution by carbon a number of studies have presented measurements of H_{c2} on polycrystalline materials[3,8,9] and in a limited range on single crystals.[10–15] All studies agree in significant enhancements of H_{c2}, and the studies on single crystals also indicate a decrease of the H_{c2} anisotropy $\gamma_H \equiv H_{c2}^{\parallel ab}/H_{c2}^{\parallel c}$. Fewer H_{c2} studies exist for Mg site substitution by aluminum, and there is a considerable spread of given values between them.[16–18]

Here, we present a comparison of $H_{c2}^{\parallel c}$ and $H_{c2}^{\parallel ab}$ measured with the same technique on aluminum and carbon substituted polycrystalline MgB_2 with various substitution levels, prepared from the same Mg and (partly) B starting materials with similar procedures. As a function of electron doping, a similar decrease of both the transition temperature T_c and the upper critical field anisotropy γ_H contrasts with the dramatically different development of the magnitude of H_{c2}: Whereas the behavior of the upper critical field in the case of Al substitution can be understood as resulting from a shift of the Fermi level, an increase in scattering has to be taken into account to explain the large increase of H_{c2} upon C substitution. We briefly discuss this different effect on scattering by C and Al substitution.

We investigated carbon substituted polycrystalline MgB_2 samples prepared in two different ways: We synthesized $Mg(B_{0.9}C_{0.1})_2$ at 1200 °C using Mg and B_4C as starting materials, as described in Ref. 3. For low C substitution levels, filaments already studied in Ref. 9 were ground to powder. In the case of aluminum substitution, chemical inhomogeneities are difficult to avoid. Inhomogeneities lead to transition broadening detrimental particularly to the determination of $H_{c2}^{\parallel c}$. In order to maximize sample homogeneity, we tried several techniques, including prealloying Mg and Al, and using AlB_2 and AlB_{12} as Al source. However, the best results were obtained with a two-step synthesis at high temperatures. First, synthesis at constant temperatures from 1000 to 1200 °C from the elements in stoichiometric quantities for up to 10 days, followed by cooling to room temperature in streaming water produced material with large inhomogeneities as visible in x-ray diffraction pattern and particularly in the superconducting transition. In order to improve homogeneity, we finely ground and thoroughly mixed the products of the above synthesis, pressed them into pellets, and then heated them for a second time to 1200 °C for 10 days.

Powder x-ray measurements on MgB_2 samples substituted with up to 20% Al indicate no phase separation, shifts in the lattice parameters close to literature values,[19] and a moderate peak broadening suggesting small variations in the Al content throughout the samples. The broadening becomes significant for $Mg_{0.7}Al_{0.3}B_2$. For this composition, additional small peaks suggest the presence of MgB_2 and $MgAlB_4$ minority phases. The superconducting transition in zero field was measured resistively (on the 60 to 70 % dense pellets) and by magnetization measurements in 20 Oe (after powdering the samples and mixing with epoxy). The magnetization measurements on isolated powder particles makes any T_c variations within the sample well visible as an onset broadening [Fig. 1(a)]. The effective T_c was defined as the crossing point of the steepest slope of the field cooled $M(T)$ with

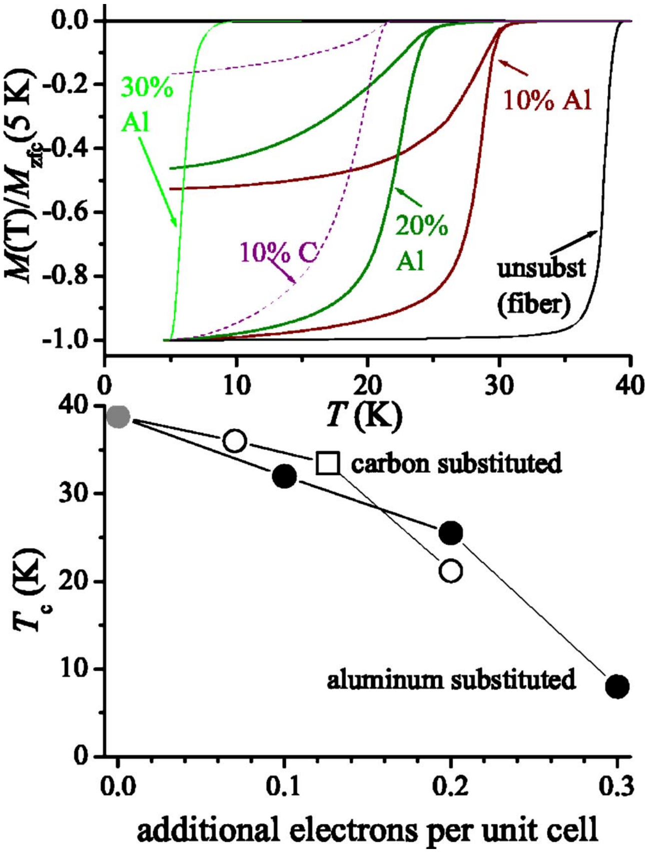

FIG. 1. (Color online.) (a) Magnetization M vs temperature T in 20 Oe (both zero field cooled and field cooled) of Al or C substituted polycrystalline MgB_2 samples. (b) Transition temperature T_c vs number of additional electrons per unit cell due to Al (full symbols) or C (open symbols) substitution. The open square is from Ref. 14.

the $M=0$ axis [Fig. 1(b)]. The height of the "sliver" in the onset above this temperature is a measure for the amount of material with higher T_c due to locally less Al substitution. Such a variation is present in Al substituted samples, but rather small.

The polycrystalline upper critical field, which corresponds to $H_{c2}^{\|ab}$, was determined from resistivity and magnetization in applied fields up to 140 and 70 kOe, respectively. In the overlapping field region, the results agree within error bars. The results on the C substituted samples are also in agreement with the results of Refs. 8 and 9. The "minimum upper critical fields" ($H_{c2}^{\|c}$) were established with a method developed by Bud'ko and co-workers.[20,21] On unsubstituted polycrystalline MgB_2, this method yielded similar results[21] on the temperature-dependent anisotropy as measurements[22] performed on single crystals. Materials were ground to a fine powder and mixed with epoxy. The minimum upper critical field is then visible as pronounced features in the derivatives of the magnetization as a function of temperature or field (see inset of Fig. 2). As an example, the resulting upper critical field of $Mg_{0.9}Al_{0.1}B_2$ is shown in Fig. 2. For this sample, $H_{c2}^{\|c}(0) \simeq 29$ kOe is slightly higher than $H_{c2}^{\|c}(0) \simeq 25$ kOe measured with the same method on an unsubstituted sample.[21] The significance of the increase is questionable, taking into account that even on unsubstituted single crystals from the same source $H_{c2}^{\|c}(0)$ with values of 28 (Ref. 23) up to 31 kOe (Ref. 22) were observed. We note that our result is significantly lower than the value of 52 kOe reported for a $Mg_{0.88}Al_{0.12}B_2$ single crystal.[18] We note that very recent measurements on a $Mg_{0.908}Al_{0.092}B_2$ single crystal from another group gave a value much closer to ours.[24] In contrast to the little change of $H_{c2}^{\|c}(0)$, the upper critical field parallel to the layers, $H_{c2}^{\|ab}(0) \simeq 127$ kOe, is significantly lower than corresponding measurements on unsubstituted MgB_2. Our result is moderately higher than the results reported in Refs. 17 and 18, significantly higher than those reported in Ref. 16. Parts of the discrepancies might be related to different amounts of impurity scattering (see discussion below), whereas other parts may originate from different inhomogeneities in the Al distribution.

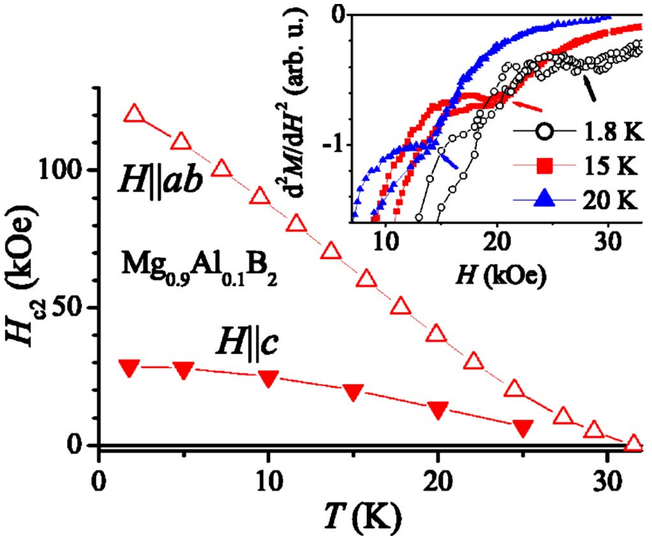

FIG. 2. (Color online.) Upper critical field H_{c2} of $Mg_{0.9}Al_{0.1}B_2$. Inset: Second derivative d^2M/dH^2 of the magnetization as a function of field H at different temperatures on $Mg_{0.9}Al_{0.1}B_2$. The location of the minimum upper critical field (see Ref. 21), i.e., $H_{c2}^{\|c}$ is indicated by arrows.

The analysis on the aluminum doped samples with up to 20% Al substitution[25] yields the following picture: Al substitution first slightly increases, then slightly decreases $H_{c2}^{\|c}$ (essentially constant), whereas H_{c2} parallel to the layers monotonically decreases (Fig. 3, closed symbols). The decrease of $H_{c2}^{\|ab}$ is roughly linearly, extrapolating to 0 for ~30% Al substitution.

The decrease of $H_{c2}^{\|ab}$ and the almost constant behavior of $H_{c2}^{\|c}$ suggests that disorder may not be important in determining the H_{c2} development with Al substitution. In unsubstituted MgB_2, clean limit (two-band) theoretical calculations[26,27] compare rather favorably with experimental data.[27,28] These calculations as well as phenomenological considerations[1] indicate that in the low-temperature limit the π bands are not important for determining H_{c2}: $H_{c2}(0)$ is mostly determined by the σ bands, as first suggested by Bud'ko and co-workers.[20] The upper critical field is related to the coherence length ξ through $H_{c2} \propto \xi^{-2}$. In the clean limit at zero temperature, ignoring the difference between GL and

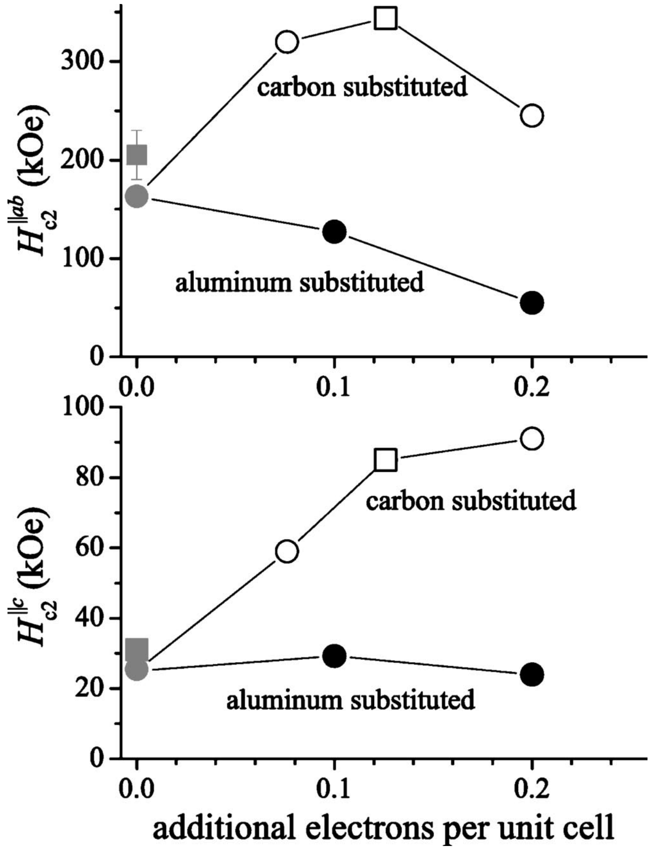

FIG. 3. (Color online.) Zero temperature upper critical field vs number of additional electrons per unit cell due to Al (full symbols) or C (open symbols) substitution. Grey symbols are unsubstituted MgB_2. (a) $H_{c2}^{\|c}(0)$. (b) $H_{c2}^{\|ab}(0)$. Squares in both panels are single crystal results from Refs. 14 and 22, open circles in panel (a) are from Refs. 8 and 9.

BCS coherence length, ξ is related to the superconducting gap and Fermi velocities by $\xi \propto v_F/\Delta$, and the Fermi velocity anisotropy determines the anisotropy of ξ. Here, $v_F(\Delta)$ is defined as the root-mean-squared wave vector dependent Fermi velocity (superconducting gap), averaged over the Fermi surface (in the case here over the σ sheets of the Fermi surface). We may then approximate

$$H_{c2}^{\|c}(0) \propto [\Delta_\sigma(0)/v_{F,\sigma}^{\|ab}]^2, \quad \gamma_H(0) = v_{F,\sigma}^{\|ab}/v_{F,\sigma}^{\|c}. \qquad (1)$$

Apart from disorder effects due to the partial substitution, Al doping modifies charge distribution and decreases the lattice constants (c in particular).[29] The *main* effect, however, is to dope the system with additional electrons, resulting in a shift of the Fermi level E_F to higher energies. For substitution levels well below 30%, where E_F reaches the σ band edge at the Γ point,[29] the changes in the electronic structure are well approximated within a rigid band model. The increase of E_F decreases the density of states (DOS) at E_F and modifies the band averaged Fermi velocities, primarily in the σ bands.[6] For moderate substitution levels, the out-of-plane σ Fermi velocity $v_{F,\sigma}^{\|c}$ remains approximately constant, whereas the in-plane σ Fermi velocity $v_{F,\sigma}^{\|ab}$ substantially decreases. According to Eq. (1) this lowers the H_{c2} anisotropy and increases $H_{c2}^{\|c}$. However, the decreased DOS at E_F weakens the electron-phonon coupling, resulting in decreased superconducting gaps and T_c (see Fig. 1). The additional effects of the substitution on the phonons[30] complicate the theoretical analysis of the development of Δ_σ and Δ_π with substitution level, and experimental reports on the gap development are sparse as of yet.[17,19,34] As an approximation, we can use the experimental values of the transition temperature shown in Fig. 1, and assume $\Delta_\sigma \propto T_c$.

The combination of the decreased Δ_σ and also decreased $v_{F,\sigma}^{\|ab}$ results in little change of $H_{c2}^{\|c}$ as estimated by Eq. (1), in accordance with the experiment. The calculation also yields the substantial decrease of $H_{c2}^{\|ab}$ and of the anisotropy γ_H observed experimentally. For the out-of-plane upper critical fields of $Mg_{1-x}Al_xB_2$, a similar, but slightly more detailed analysis was recently presented by Putti *et al.*[17] The fact that the experimental development of the upper critical field can be accounted for by the clean limit formula (1) clearly suggests that effects of increased scattering are not relevant in our samples of $Mg_{1-x}Al_xB_2$ at low substitution levels. This is different from recently presented results on some single crystal samples,[18] where scattering in the π bands seems to be larger, but we again note that subsequent measurements on crystals from another source[24] are more agreeable with our experimental results and consequently with the conclusion of small effects of disorder by Al substitution.

The development of H_{c2} with partial carbon substitution is also shown in Fig. 3 (open symbols). Our results, which agree qualitatively with the limited results on single crystals available[10,11,13–15] show a drastic increase of H_{c2} both parallel and perpendicular to the layers, in contrast to the Al substitution case. The increase of $H_{c2}^{\|c}$ is monotonous in the substitution range measured, reaching the high value of 91 kOe, whereas $H_{c2}^{\|ab}$ peaks somewhat below 400 kOe for C substitution levels around 5%. Figure 4 displays the temperature-dependent H_{c2} anisotropy for C substituted, Al substituted, and unsubstituted MgB_2. Where $H_{c2}^{\|ab} > 140$ kOe, $H_{c2}^{\|ab}$ results from Refs. 8 and 9 have been utilized to calculate $\gamma_H(T)$. With the exception of $Mg(B_{0.9}C_{0.1})_2$ and $Mg_{0.8}Al_{0.2}B_2$, where the T dependence of γ_H is not obvious, a substantial decrease of γ_H with increasing T is seen for all samples. In accordance with the explanation of the $\gamma_H(T)$ dependence in unsubstituted MgB_2 this indicates that two band effects are still relevant for both Al and C substituted MgB_2 for moderate substitution levels. The preservation of two distinct superconducting gaps has indeed been observed directly on both Al and C substituted MgB_2.[3,17,19,31–34] It is expected for moderate substitution levels from band structure calculations, but also implies that interband scattering cannot be substantially increased by partial substitutions of either of these elements. The anisotropy monotonically decreases with increasing substitution level, down to about 2 for the samples with the highest levels of substitution studied. This decrease is rather similar for substitutions by Al and C. This indicates that as far as the H_{c2} *anisotropy* is concerned, the *main effect* of carbon substitution is (as in the case of Al substitution) a decreased anisotropy of $v_{F,\sigma}$ originating from the shifted Fermi level. The T_c depression (Fig. 1) is also similar for Al and C substitution, suggesting that this too may originate

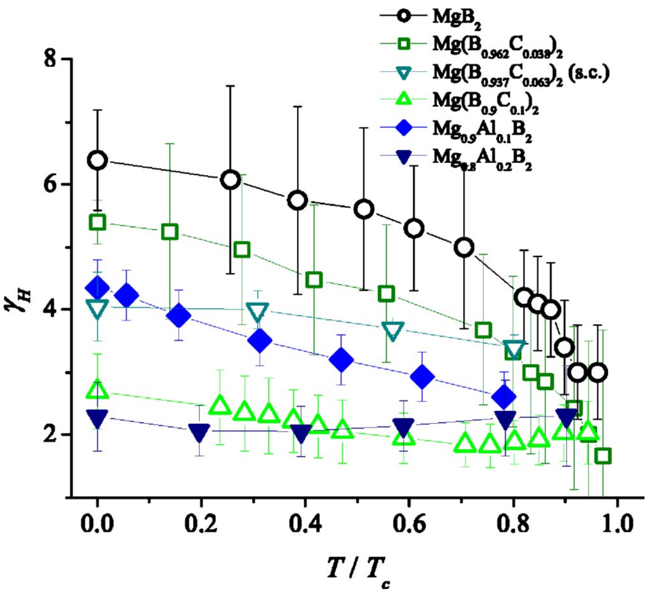

FIG. 4. (Color online.) Upper critical field anisotropy γ_H vs temperature T, for unsubstituted, aluminum substituted, and carbon substituted MgB_2. The results from 6.3% C substituted (single crystalline) MgB_2 are from Ref. 14.

mainly from the shift of E_F and similar changes of phonon modes.

However, the drastically different H_{c2} *magnitude* in C substituted samples cannot be explained within this picture. The difference to the Al substitution case is far too large to be accounted for by different behavior of the lattice constants or phonon modes, particularly given the above similarities in T_c and γ_H vs electron doping level. Rather, the very strong increase of H_{c2} points to the relevance of scattering in the case of carbon substitution. Due to the two band nature of superconductivity, three different scattering rates have to be taken into account: interband scattering and intraband scattering in the σ and π bands.[7] We note that the observation of constant Δ_σ/Δ_π ratios by spectroscopic means[33] indicate that the interband scattering rate is hardly affected by moderate levels of C substitution and may be neglected as in unsubstituted MgB_2. However, a recent point contact study on MgB_2 crystals containing high C substitution levels suggest that the interband scattering rate may be important, particularly for high substitution levels.[35] Calculations within "intraband dirty limit"[36,37] can indeed explain very drastic increases of H_{c2}, much larger than in single band superconductors.

These dirty limit calculations also yield a temperature dependent H_{c2} anisotropy, as the clean limit calculations do. However, here the T dependence of γ_H also depends on the ratio of the scattering in the anisotropic σ and in the nearly isotropic π bands. If the intraband scattering is much larger in the σ bands than in the π bands, a decreasing $\gamma_H(T)$ dependence is expected, whereas the opposite case results in an increasing $\gamma_H(T)$ dependence. Starting from the unsubstituted MgB_2 with a decreasing $\gamma_H(T)$ dependence, a low level partial substitution mainly increasing the $\sigma(\pi)$ bands scattering, should lead to a more (less) pronounced $\gamma_H(T)$ dependence. As we can see from Fig. 4, the $\gamma_H(T)$ dependence becomes less pronounced upon increasing the substitution level. Comparing C and Al substitution, the decrease of the T dependence seems *rather similar*, indicating that it is mostly due to the intrinsic changes discussed above, rather than disorder. For the same electron doping levels, the $\gamma_H(T)$ variation is somewhat less strong for the carbon substitution case. This indicates that upon C doping the scattering is increased more in the π bands than in the σ bands. A similar conclusion was reached for a 6.3% C substituted single crystal[14] and for thin films containing carbon.[38,39] For a more quantitative analysis, a theory treating clean-limit (electron doping) and dirty-limit (scattering) effects *on an equal footing* would be highly desirable.

The effect of higher π band scattering also manifests itself in the form of the H_{c2} curves, most visibly for $H \| c$. In extreme cases this leads to a positive curvature of $H_{c2}^{\|c}(T)$ at low T.[36,38] In contrast to "dirty films" results[38,39] we did not observe such a positive curvature, but compared to unsubstituted MgB_2, the negative curvature of $H_{c2}^{\|c}(T)$ was significantly decreased for the C substituted samples. For 10% C substituted $MgB_2, H_{c2}^{\|c}(T)$ was found to be almost linear at low temperatures. The tendency of decreased negative curvature of $H_{c2}^{\|c}(T)$ with increasing C substitution is also seen in single crystal measurements[11,14,15] and supports the conclusion of mainly additional scattering in the π bands causing the H_{c2} enhancement. In contrast, in the Al substituted samples, the $H_{c2}(T)$ curvature is not significantly affected (see Fig. 2), again indicating less π band scattering.

To account for an upper critical field that is much larger in C substituted MgB_2, C substitution has to increase scattering in the π bands more relative to the σ bands, and much more than Al substitution does. That a substitution within the boron layers would increase scattering more than a Mg site substitution is hardly a surprise. It is, however, surprising that the increase in the scattering is predominantly in the intraband scattering in the isotropic π bands. In partly C substituted thin films, the effects of increased π band scattering are much larger [as visible both in the $H_{c2}(T)$ curve forms and in the T dependence of the anisotropy]. This scattering has been attributed to a buckling of the *ab* planes, tentatively due to nanophase precipitates.[38,39] However, such precipitates are unlikely to be present in our polycrystalline samples or in single crystals, and we therefore conclude that *an increase of the π bands scattering is an intrinsic property of C substitution.* This is in contrast to the aluminum substitution case, where considerable variations of the significance of π bands scattering exist for different samples.[40] A recent first principles electronic structure study[41] on C substituted MgB_2, taking into account disorder effects, found a larger reduction of the mean free path in the σ bands, which is in contrast to our analysis. However, there are a variety of effects that are more involved to include in a calculation, e.g., carbon induced local distortions in the structure, as suggested by a single crystal x-ray diffraction study.[15] Since the large increase of H_{c2} in C substituted MgB_2 and related scattering rates are important for potential applications, additional theoretical studies are clearly desirable, as would be a clear experimental demonstration of a procedure boosting scattering mainly in the σ bands.

In conclusion, whereas the development of H_{c2} with partial aluminum substitution can be understood within a simple rigid band picture, disorder effects are responsible for the large enhancement of the H_{c2} magnitude of carbon substituted MgB_2. In contrast and importantly, the development of the H_{c2} anisotropy and T_c are remarkably similar for the two substitutions. Furthermore, scattering within the π bands is increased much more by carbon than by aluminum substitution, and more than scattering in the σ bands. The origin of this is yet to be resolved, calling for further theoretical work.

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