Comparison of temperature and angular dependence of the upper critical field in $Mg_{1-x}Al_xB_2$ single crystals in dirty-limit two-gap theory

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We studied the temperature and the angular dependences of the upper critical field $[H_{c2}(T,\theta)]$ of $Mg_{1-x}Al_xB_2$ single crystals ($x=0.12$ and 0.21) and compared with the dirty-limit two-gap theory. We found that $H_{c2}(T,\theta)$'s were well described in a unified way by this theory. The values of the parameters obtained by fitting experimental data to the theory indicated that as the Al concentration was increased, anisotropic impurity scattering increased, making the σ bands less anisotropic. Accordingly, the temperature dependence of the anisotropy ratio of H_{c2} (γ_H) systematically decreased, and for $x=0.21$, γ_H was nearly independent of temperature. Our results imply that $Mg_{1-x}Al_xB_2$ single crystals are in the dirty limit and that the two-gap nature survives until $x=0.21$.

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It is now well established that MgB_2 is a two-gap superconductor with two distinct energy gaps: a large gap originating from two-dimensional σ bands and a small gap originating from three-dimensional π bands.[1–3] One of the main consequences of the two-gap nature is the strong temperature dependence of the $H_{c2}(T)$ anisotropy $\gamma_H \equiv H_{c2}^{ab}/H_{c2}^{c}$,[4] which is in contrast to the single-gap Ginzburg-Landau theory. Theoretical calculations show that the strong temperature dependence of γ_H arises from the fact that the anisotropic σ bands dominate at low temperatures while the π bands gradually become important at temperatures near T_c.[5–7] The above anomalous behavior of $\gamma_H(T)$ for MgB_2 single crystals was confirmed by magnetization measurements.[8,9]

When impurity scattering increases, the abovementioned behavior of H_{c2} is modified. Gurevich,[6] and Golubov and Koshelev[7] formulated the dirty-limit two-gap theory for H_{c2} by using the quasiclassical Usadel equations. According to this theory, the shape of the $H_{c2}(T)$ curve essentially depends on the diffusivities of the σ and the π bands. For $T \approx T_c$, $H_{c2}(T)$ is determined by a maximum diffusivity (cleaner bands) between D_σ and D_π while $H_{c2}(0)$ is controlled by a minimum diffusivity (dirtier bands). When the σ bands are dirtier, an upward curvature should appear near T_c, and γ_H should decrease with temperature. In contrast, when the π bands are dirtier, a huge increase in $H_{c2}(T)$ should appear at low temperatures without an upward curvature near T_c, and γ_H should increase with temperature.

Impurity scattering also changes angular dependence of H_{c2} $[H_{c2}(\theta)]$ that was predicted to deviate from the angular dependence of the anisotropic one-gap Ginzburg-Landau (GL) theory, especially near the middle-angle region. This deviation should be most pronounced at $T/T_c \approx 0.95$ when the parameters supplied by band-structure calculations are used.[7,10] Even though these predictions were quantitatively compared with $H_{c2}(\theta)$ for MgB_2 single crystals and reasonable consistency was observed,[10] the problem of whether the dirty-limit theory could be applied to clean MgB_2 single crystals still remained. In this sense, the dirty-limit theory has not yet been verified unambiguously for single crystals in the dirty limit, especially for the orientational dependence of H_{c2}.

In this paper, we report on the effect of Al doping, as deduced from resistance measurements at various angles θ between H and the c axis, on $H_{c2}(T,\theta)$ of Al-doped MgB_2 single crystals. This directional study of the resistance was possible due to success in growing flat and regular-shaped $Mg_{1-x}Al_xB_2$ single crystals with values of x up to 0.21 and with $T_c=25.5$ K. We found that two-gap superconductivity in MgB_2 was drastically affected by the Al doping and that key features predicted by the dirty-limit two-gap theory were observed. Our main observations are the following. (1) As the Al concentration increases, the residual resistivity (ρ_0) greatly increases, implying that Al substitution enhances impurity scattering and that the Al-doped samples are in the dirty region. (2) $H_{c2}(T)$ can be consistently explained within the dirty-limit two-gap theory up to $x=0.21$, even though $H_{c2}(0)$ decreases with Al concentration. (3) The $\gamma_H(T)$ systematically decreases and for $x=0.21$, γ_H is virtually temperature independent. (4) The $H_{c2}(\theta)$ for $x=0.12$ showed a clear deviation from the behavior predicted by the anisotropic GL theory, which is a strong indication of the two-gap nature in MgB_2. However, for $x=0.21$, this deviation became very small. The results suggest that impurity scattering is enhanced in the π bands, especially along the c direction and that the anisotropy of the σ bands is significantly reduced.

$Mg_{1-x}Al_xB_2$ single crystals with $x=0.12$ and 0.21 were grown under high-pressure conditions.[11,12] They were characterized and patterned as reported earlier.[11,12] Two samples with clean, shiny surface were investigated for each Al concentration. For the resistance measurements, well-shaped single crystals with both sides flat were selected from numerous samples. The temperature and the angular dependences of the resistance were measured from 0 to 9 T by using the ac transport option in a PPMS Quantum Design system.

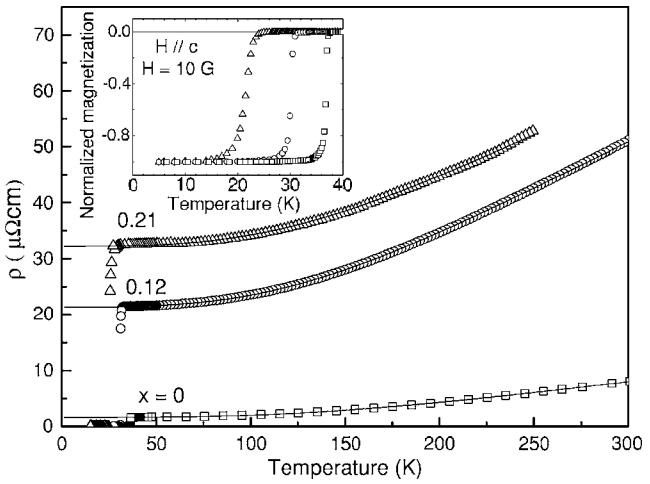

FIG. 1. Temperature dependence of the resistivity for $Mg_{1-x}Al_xB_2$ single crystals (x=0.0, 0.12, and 0.21). The solid lines are theoretical curves of the BG formula. The inset shows the normalized low-field magnetization in the zero-field-cooled state.

Figure 1 shows the resistivity ρ of the $Mg_{1-x}Al_xB_2$ single crystals (x=0, 0.12, and 0.21) as a function of temperature. As the Al concentration increases, T_c decreases. The T_c's are 30.8 and 25.5 K for x=0.12 and x=0.21, respectively. The data for x=0 were taken from Ref. 4 and T_c of this sample was around 37 K. Previously, for MgB_2 single crystals, the resistance was reported to follow the Bloch-Grüneisen (BG) formula with a Debye temperature $\Theta_D \sim 1100$ K.[11] This implied that the normal-state transport properties were well described by electron-phonon interaction and effect of electron-electron interaction could be neglected. To check whether this is the case in Al-doped single crystals, we fitted the $\rho(T)$ data with the BG formula, where fitting parameters are Θ_D and residual resistivity ρ_0. The solid lines in the figure are the BG theoretical curves and are seen to well describe the $\rho(T)$ data. The value of Θ_D in Al-doped single crystals was found to be $\sim$1000 K, which is similar to that of MgB_2 single crystals. ρ_0 increases monotonically with doping, and the fitted values of ρ_0 are 1.63, 21.4, and 32.2 $\mu\Omega$ cm for x=0.0, 0.12, and 0.21, respectively. The inset of Fig. 1 shows the normalized low-field magnetization for the zero-field-cooled state of $Mg_{1-x}Al_xB_2$ single crystals extracted from the same batch of single crystals as were used for the resistivity measurements. The T_c's determined from the resistivity and from the low-field magnetization were virtually the same.

Figures 2(a) and 2(b) show, as an example, the temperature dependences of the resistances of the x=0.12 sample for $H\|c$ and $\|ab$, respectively. As with MgB_2 single crystals, this sample shows surface superconductivity: as the temperature decreases, the resistance first decreases linearly and then suddenly drops to zero. In the region of linear decrease, the resistance depends on the applied current, and a higher current induces a higher resistance. The drop in the resistance indicates the onset of bulk superconductivity. At high currents (I=3 mA) for $H\|c$, a peak, which is absent at low currents (I=1 mA), appears. The current dependence of this peak suggests that it is due to the peak effect, observed in MgB_2 single crystals.[13] The upper critical fields can be determined unambiguously as the points where the resistance drops to zero in the curves for I=1 mA. Those points are indicated by the arrows.

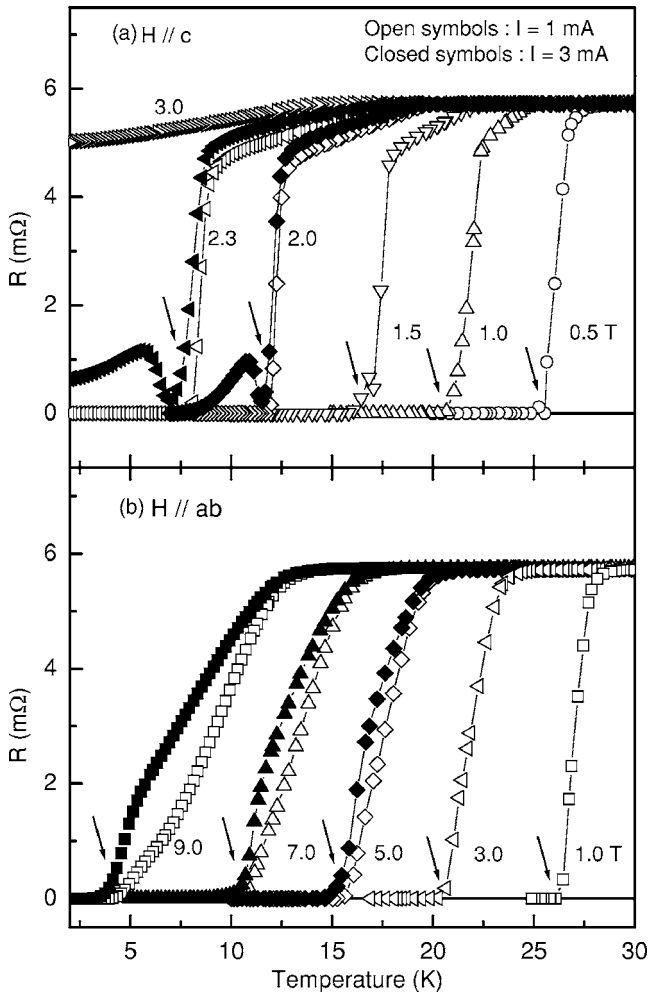

FIG. 2. Temperature dependence of the resistance for (a) $H\|c$ and (b) $H\|ab$.

In Fig. 3(a), $H_{c2}^c(T)$ and $H_{c2}^{ab}(T)$ for x=0.12 and 0.21 are plotted, where $H_{c2}^c(T)$ and $H_{c2}^{ab}(T)$ are $H_{c2}(T)$'s for $H\|c$ and for $H\|ab$, respectively. For comparison, we also insert $H_{c2}(T)$ for x=0.0, which was taken from Ref. 4. Interestingly, both $H_{c2}^c(T)$ and $H_{c2}^{ab}(T)$ decrease with increasing Al doping. As a result, the extrapolated $H_{c2}^c(0)$ and $H_{c2}^{ab}(0)$ are reduced. While the decrease in $H_{c2}^{ab}(0)$ is consistent with the results for polycrystalline samples, the decrease in $H_{c2}^c(0)$ is not. In a study by Angst *et al.*, a small increase in $H_{c2}^c(0)$ was observed at an Al doping of 10%.[14] By comparing $H_{c2}(0)$ in both Al- and C-doped MgB_2, they concluded that in Al-doped samples, the shift in the Fermi level was dominant in determining $H_{c2}(T)$ while in C-doped samples, disorder played a major role. However, in light of the huge increase in ρ_0, the effects of disorder are not negligible and should be taken into account. Another clue to the degree of dirtiness in $Mg_{1-x}Al_xB_2$ single crystals for x=0.12 and x=0.21 can be found in the shape of $H_{c2}^c(T)$ near T_c. While MgB_2 single crystals show a linear decrease in $H_{c2}^c(T)$ near T_c, close inspection reveals that an upward curvature gradually appears

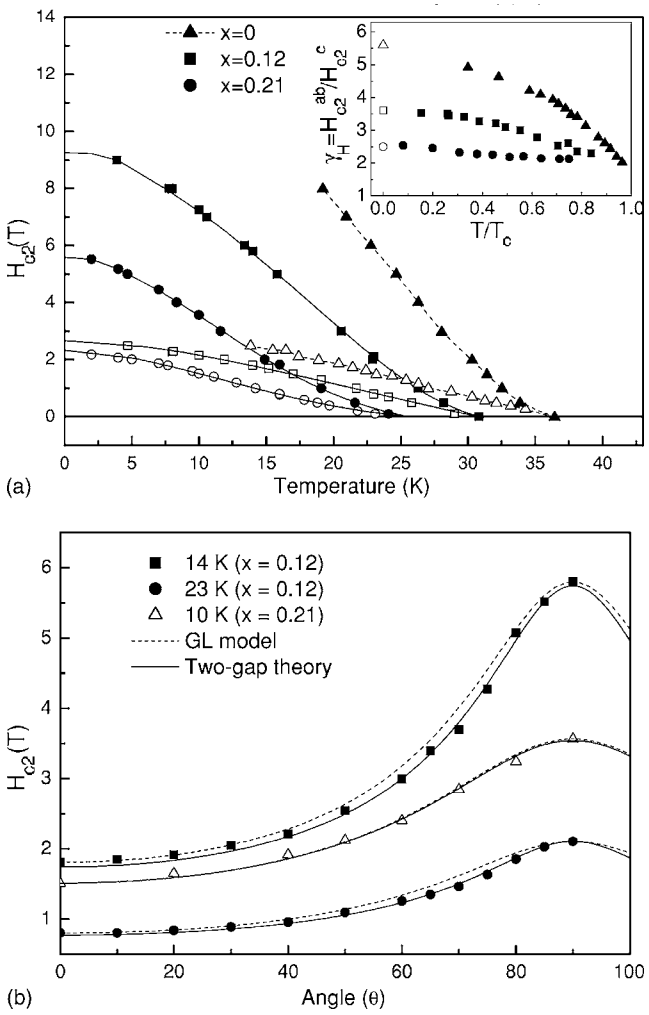

FIG. 3. (a) Temperature dependence of H_{c2} for $Mg_{1-x}Al_xB_2$ single crystals (x=0.0, 0.12, and 0.21). Open symbols represent $H_{c2}(T)$ for $H\|c$ and closed symbols represent $H_{c2}(T)$ for $H\|ab$. The data for x=0.0 were taken from Ref. 4. The inset shows the temperature dependence of γ_H. The open triangle is $\gamma_{v_F}\equiv v_{F,\sigma}^{ab}/v_{F,\sigma}^{c}$, and the open circle and square are $\gamma_\sigma\equiv\sqrt{D_\sigma^{ab}/D_\sigma^{c}}$'s. (b) Angular dependence of H_{c2}. The solid lines are the theoretical curves for the dirty-limit two-gap model, and the dotted lines are those for the Ginzburg-Landau theory.

with Al doping. This becomes even clearer if $H_{c2}^{c}(T)$ for x =0 is compared with that for x=0.21. The upward curvature is consistent with the two-gap dirty-limit theory.

Since the variations in $H_{c2}(T)$ with Al doping appear to agree well with the two-gap theory, we quantitatively analyzed our $H_{c2}(T)$ data by using the dirty-limit theory.[6,7] For x=0, the dirty limit model may be inappropriate because pure MgB_2 crystals are considered to be in the clean limit.[15] If interband impurity scattering is assumed to be zero, $H_{c2}(T)$ for $H\|c$ is given by

$$a_0[\ln t + U(h)][\ln t + U(\eta h)] + a_2[\ln t + U(\eta h)] + a_1[\ln t + U(h)] = 0, \quad (1)$$

where $t=T/T_c$, $U(x)=\Psi(1/2+x)-\Psi(x)$, $\Psi(x)$ is the Euler digamma function, $h=H_{c2}D_\sigma^{ab}/2\phi_0 T$, ϕ_0 is the magnetic flux quantum, $\eta=D_\pi^{ab}/D_\sigma^{ab}$, D_σ^{ab} and D_π^{ab} are the in-plane electron diffusivities of the σ and the π bands, and $a_{0,1,2}$ are constants derived from the electron-phonon coupling constants (λ_{mn}^{ep}) and the Coulomb pseudopotentials (μ_{mn}). The precise definitions of $a_{0,1,2}$ can be found in Ref. 6. For $H\|ab$, the in-plane diffusivities in Eq. (1) can be replaced by $[D_{\sigma,\pi}^{ab}D_{\sigma,\pi}^{c}]^{1/2}$, where $D_{\sigma,\pi}^{c}$ are the out-of-plane electron diffusivities of the σ and the π bands, respectively. Equation (1) can be generalized to the anisotropic case of an inclined field by replacing the diffusivities with the angle-dependent diffusivities $D_\sigma(\theta)$ and $D_\pi(\theta)$ for both bands, where $D_{\sigma,\pi}(\theta)=[(D_{\sigma,\pi}^{ab})^2\cos^2\theta+D_{\sigma,\pi}^{ab}D_{\sigma,\pi}^{c}\sin^2\theta]^{1/2}$. For the four input parameters $\lambda_{mn}=\lambda_{mn}^{ep}-\mu_{mn}$ at each Al doping level, which reflect the change in the electronic structure by electron doping, we used the values determined from first-principle calculations,[16] and we obtained the numerical value of the diffusivity for each band.

In our samples, the interband impurity scattering is believed to be negligible to the first approximation. This is because the interband impurity scattering was predicted to eliminate the distinction of each superconducting gap, and thereby two-gap features.[17] Therefore, the upward curvature, which is a hallmark of the two-gap superconductivity would have not been observed, if interband impurity scattering was significant.

The solid lines in Fig. 3(a) present the theoretical two-gap dirty-limit curves of $H_{c2}(T)$ for x=0.12 and 0.21. The optimized values of $D_\sigma^{ab,c}$, $D_\pi^{ab,c}$, and $H_{c2}^{ab,c}(0)$ from the fits are summarized in Table I. The upward curvature observed near T_c for x=0.12 and 0.21, which is typical when σ bands are dirtier than π bands,[6] may indicate dirtier σ bands. If the π bands are dirtier than the σ bands, the upward curvature near T_c should disappear; instead, a huge increase in $H_{c2}(T)$ should appear at low temperatures. The dashed lines for x =0 are a guide to the eyes.

Quantitatively, the values of $D_\sigma^{ab,c}$ and $D_\pi^{ab,c}$ show that σ bands are dirtier ($D_\sigma^{ab,c}\ll D_\pi^{ab,c}$), which is consistent with the shape of $H_{c2}(T)$. Dirty σ bands were also observed in Al-doped MgB_2 polycrystalline samples.[18] The electron diffusivity along the c direction in the π bands is found to decrease with Al doping while that in the ab plane virtually does not change. This originates from pronounced impurity scattering in the π bands as the Al concentration is increased. The pronounced impurity scattering, however, is not isotropic as is normally assumed. Along the c direction, impurity scattering is more enhanced than in the ab plane. Similarly, Al doping influences impurity scattering in the σ bands. In this case, the electron diffusivity along the c direction increases with Al doping while that in the ab plane is virtually unchanged. Consequently, the σ bands become more isotropic, which is reflected in the ratio $D_\sigma^{ab}/D_\sigma^{c}$ and this value decreases as Al content increases. The isotropization of the σ bands is believed to be due to not only the anisotropic impurity scattering but also the change in the electronic structure that Al doping induces.

The same set of electron diffusivities as in Table I can explain $H_{c2}(\theta)$ for x=0.12 and 0.21, as shown in Fig. 3(b). The solid lines indicate the theoretical curves calculated

TABLE I. Al content x, upper critical fields $H_{c2}^{ab(c)}(0)$, electron diffusivities along the ab plane (the c axis) in the σ and the π bands, and $D_{\sigma}^{ab(c)}$ and $D_{\pi}^{ab(c)}$, obtained by fitting the $H_{c2}(T)$ data to the dirty-limit model.

x	$H_{c2}^{ab}(0)$ (T)	$H_{c2}^{c}(0)$ (T)	D_{σ}^{ab} (m^2s^{-1})	D_{σ}^{c} (m^2s^{-1})	D_{π}^{ab} (m^2s^{-1})	D_{π}^{c} (m^2s^{-1})
0.12	9.3	2.7	7.6×10^{-4}	5.9×10^{-5}	3.7×10^{-3}	3.0×10^{-3}
0.21	5.6	2.3	6.0×10^{-4}	1.0×10^{-4}	4.8×10^{-3}	1.4×10^{-3}

from the dirty-limit two-gap theory. The dotted lines are the theoretical curves of the one-gap GL model. The error bars in this data are comparable to or less than the symbol size. The two-gap theory describes the data better than the GL model for $x=0.12$. For $x=0.12$, a small difference between the two-gap theory and the GL model is apparent and as predicted, is most pronounced at the middle-angle regions. This is a strong indication of the two-gap nature of Al-doped MgB_2 single crystals. This behavior is very similar to that of MgB_2 single crystals, where a deviation from GL behavior was observed to be peaked at $T\approx0.8T_c$. For $x=0.21$, the difference between the two-gap theory and the anisotropic GL model is very tiny and is the case for all the temperatures we investigated. Despite the small difference in angular dependence predicted by the anisotropic GL model and the two-gap theory for this doping, the shapes of the $H_{c2}(T)$ curves and the values of the fitted diffusivities show the existence of two distinct gaps. If the sample for $x=0.21$ followed the one-gap GL model, the upward curvature would not have been observed.

Finally, $\gamma_H(T)$ for $x=0$, 0.12, and 0.21, extracted from the $H_{c2}(T,\theta)$ data, are plotted as functions of the reduced temperature T/T_c in the inset of Fig. 3(a). The values of γ_H are systematically reduced, and for $x=0.21$, γ_H is virtually temperature independent at high temperatures, slightly increasing at low temperatures. The γ_H at low temperatures significantly changes with Al doping and the γ_H's merge to 2–2.5 at $T=T_c$ for all doping levels. This behavior is thought to result from the isotropization of the σ bands. The decreasing tendency of γ_H with increasing temperature for $x=0.12$ and 0.21 is in good agreement with the case of dirty σ bands, predicted by using the dirty-limit two-gap theory.

If the effects of impurity scattering can be ignored in $x=0.12$ and 0.21 single crystals, H_{c2} will evolve according to changes in the electronic structure and the lattice constant. Among these, the main effect is due to changes in the electronic structure caused by doping with electrons, resulting in a shift of Fermi level E_F to higher energies. At moderate doping levels, where a rigid band model is valid, an increase in E_F modifies the band-averaged Fermi velocities, primarily in the σ bands and the $\gamma_H(0)$, which is $\gamma_{v_F}\equiv v_{F,\sigma}^{ab}/v_{F,\sigma}^{c}$ in the clean limit. Here, $v_{F,\sigma}^{ab(c)}$ is the in-plane (out-of-plane) Fermi velocity of the σ bands. According to the calculation by Putti *et al.*,[19] $v_{F,\sigma}^{c}$ remains approximately constant while $v_{F,\sigma}^{ab}$ substantially decreases with Al doping for $x<0.3$. At doping levels of $x=0.0$, 0.12, and 0.21, values of $\gamma_{v_F}=5.6$, 5, and 4.2, respectively, were obtained from their calculations. The value at $x=0.0$ is nearly consistent with $\gamma_H(0)$ estimated from the experimental data, as shown in the inset of Fig. 3(a). In contrast, the values at $x=0.12$ and 0.21 are significantly larger than the estimated $\gamma_H(0)$. In fact, the $\gamma_H(0)$'s at $x=0.12$ and 0.21 are better represented by the parameter $\gamma_\sigma\equiv\sqrt{D_\sigma^{ab}/D_\sigma^{c}}$, which contains information on not only the Fermi velocity but also impurity scattering. Therefore, as mentioned earlier, $Mg_{1-x}Al_xB_2$ single crystals ($x=0.12$ and 0.21) are in the dirty limit with anisotropic impurity scattering. This is in sharp contrast to the conclusions for Al-doped MgB_2 polycrystalline samples.[14,19] Those polycrystalline samples might have less impurities than single crystals, which is very improbable in normal situations. It is noted that while γ_σ decreases with Al doping, γ_π increases from 1.1 to 1.8.

The electron diffusivities are related to the value of resistivity by the relation $1/\rho\propto N_\sigma D_\sigma+N_\pi D_\pi$,[6] where N_σ and N_π are partial densities of state in σ and π bands, respectively. In the present case, since the electron diffusivities in the π bands are larger than those in the σ bands, the electron diffusivities in the π bands determine the resistivities of our samples and resistivity should increase with x. This tendency holds in our samples. We calculated the values of resistivities by using the diffusivity values of Table I and the partial densities of state calculated by Ummarino *et al.*[16] and obtained 10 and 12 $\mu\Omega$ cm for $x=0.12$ and $x=0.21$, respectively. The discrepancy of the absolute values, especially for $x=0.21$ might originate from a large error in calculating the resistivity of small-size samples.

To summarize, we investigated the effect of Al substitution on $H_{c2}(T,\theta)$ of MgB_2 single crystals. From an analysis of $H_{c2}(T,\theta)$ using the dirty-limit two-gap theory, we found that Al substitution influenced the electronic structure, increasing impurity scattering along the c direction in the π bands while making the σ bands less anisotropic. Accordingly, temperature dependence of γ_H was systematically reduced and for $x=0.21$, γ_H was virtually temperature independent. The isotropization, especially of the σ bands, originates not only from increased anisotropic impurity scattering but also from electron doping. In $H_{c2}(\theta)$, we also observed a strong indication of the dirty-limit two-gap nature of Al-doped MgB_2.

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