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Citation: *Appl. Phys. Lett.* **108**, 202601 (2016); doi: 10.1063/1.4949338

View online: <http://dx.doi.org/10.1063/1.4949338>

View Table of Contents: <http://aip.scitation.org/toc/apl/108/20>

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Al-doped MgB₂ materials studied using electron paramagnetic resonance and Raman spectroscopy

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(Received 27 January 2016; accepted 29 April 2016; published online 16 May 2016)

Undoped and aluminum (Al) doped magnesium diboride (MgB₂) samples were synthesized using a high-temperature solid-state synthesis method. The microscopic defect structures of Al-doped MgB₂ samples were systematically investigated using X-ray powder diffraction, Raman spectroscopy, and electron paramagnetic resonance. It was found that Mg-vacancies are responsible for defect-induced peculiarities in MgB₂. Above a certain level of Al doping, enhanced conductive properties of MgB₂ disappear due to filling of vacancies or trapping of Al in Mg-related vacancy sites. Published by AIP Publishing. [<http://dx.doi.org/10.1063/1.4949338>]

The effect of additives in MgB₂ materials has been investigated for many years after the discovery of the material's superconductive properties in 2001 by Nagamatsu *et al.*¹ It was reported that by introducing defects the coherence length decreases and accordingly H_{c2} increases, which is a limiting factor in MgB₂. Moreover, defects can introduce pinning centers to make the irreversibility line sharper.² It has also been reported that Al doping gradually reduces J_c ³ and T_c .^{4,5} By contrast, low-level Al doping⁶ and addition of carbon-nanotube-coated Al revealed the enhancement of J_c in MgB₂.⁷ This has been explained by first-principle studies where the loss of superconductivity was attributed to the filling of holes in the valance band.^{8,9} Superconductivity of Al-doped samples might disappear when around half of the Mg is replaced by Al due to electron doping of Al into the 2D σ -band of MgB₂.^{6,8,10,11} However, it is still not quite clear if doping enhances the superconducting behavior of MgB₂ or not. This difficult issue crucially depends on the type and concentration of the doping element. Carbonaceous materials (C and graphene oxide),¹² transition-metal (Ti¹³ and V¹⁴), and rare-earth metal (Ce, Sm, and Gd)¹⁵ ions have been used effectively as additives for many years to achieve a better superconducting response from the material. Very recently, adding magnetic nanoparticles, such as CuFe₂O₄¹⁶ and NiFe₂O₄,¹⁷ has yielded a significant enhancement of superconductivity of MgB₂.

In most of the aforementioned dopants, replacements in either B or Mg sites are not easily achieved. Among them, C and Al doping appears to be most convenient.¹⁸ The ease of replacement of Mg by Al is due to the similar crystal structures of MgB₂ and AlB₂. However, a high extension along the c direction is observed for MgB₂ with a c/a value of 1.143 as compared with 1.083 for AlB₂.¹⁰ This directly shows that due to the large difference in the ionic radii of the two elements ($r_i(\text{Mg}^{2+}) = 65$ pm; $r_i(\text{Al}^{3+}) = 50$ pm), substitution cannot simply generate a single-phase compound, and Al-doping chemistry remains to be a complicated issue.^{19,20} Many researchers have reported structural changes and

superconductivity of Al-doped MgB₂ materials (Mg_{1-x}Al_xB₂, $x = 0-1$) in four phase regions: (i) the region with Al doping up to $x = 0.1$ shows almost the same structure and a T_c similar to pure MgB₂;⁴ (ii) for $0.1 \leq x \leq 0.25$, the coexistence of Al-rich and Mg-rich phases was reported;^{8,21} (iii) for $0.25 \leq x \leq 0.6$, a super-structured phase (Mg_{0.5}Al_{0.5}B₂) occurs, in which hexagonal Mg layers are alternatively replaced by Al layers. This phase was reported to be superconductive but at very low T_c ;^{10,22} and (iv) for $x > 0.6$, the material is no longer superconductive and it exhibits an AlB₂-type structure.^{22,23}

To date, a number of experimental and theoretical studies dealt with the superconductive properties of both undoped and Al-doped MgB₂ materials.^{3-6,8,10,11,22-25} But the defect structure, which can be decisive for the material's superconductivity, remains largely unclear. The aim of this work is to monitor, by means of Raman and electron paramagnetic resonance (EPR) spectroscopy, the electronic property changes induced by different Al-doping levels into MgB₂. In particular, we strive for an understanding of the effect of Al on the possible intrinsic vacancies present in the crystal structure as so-called defects.

Five different Mg_{1-x}Al_xB₂ samples with $x = 0, 0.25, 0.5, 0.75$, and 1 were prepared using a high-temperature solid-state synthesis method. As starting powders, aluminum (Alfa Aesar, 99.97%, -325 mesh) and nanoboron (Pavezyum, 98.5%) were used. A stoichiometric mixture of the elements Al and B with a total weight of 5 g was placed into a rotating tumbler at a speed of 40 rpm for 4 h and pressed to pellets ($\varnothing = 25$ mm). The samples were placed into steel crucibles and heated under protective argon gas to 900 °C for Mg_{1-x}Al_xB₂ phases with $x = 0.25, 0.5, 0.75$, and 1 , and to 700 °C for pure MgB₂ ($x = 0$). All specimens were annealed for 4 h and cooled down in the furnace to room temperature. The title compounds were obtained as sintered grayish solids; they could easily be pestled to fine powders.

The products were analyzed using powder X-ray diffraction (PXRD) recorded with a Bruker D8 ADVANCE (Cu-K α radiation with VANTEC-1 detector). Raman spectroscopic measurements were performed using a Raman microscope RENISHAW inVia with a 532-nm excitation laser source. X-band (9.86 GHz) continuous-wave-EPR measurements were performed with a Bruker EMX spectrometer. A super

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FIG. 1. PXRD patterns of $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples with $x=0, 0.25, 0.5, 0.75$, and 1. (a) Full range of diffraction angles; (b) (002) and (110) reflections; (c) (101) reflections.

high-Q X-band resonator (Bruker) was used. The offset in the magnetic field and accurate g -values were determined with a polycrystalline DPPH (2-diphenyl-1-picrylhydrazyl) reference sample with well-known g -factor (2.0036). Additional EPR spectral analyses have been performed using the WINEPR software from Bruker. For cooling to liquid-helium temperatures, an Oxford CF-935 cryostat was used. The temperature was regulated to ± 0.5 K by an Oxford ITC-503 temperature controller.

PXRD data of $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples with $x=0, 0.25, 0.5, 0.75$, and 1 are plotted in Fig. 1(a). All samples contain some traces of MgO, which are originating from the starting materials.²⁶ The effect of MgO has been discussed elsewhere.²⁶ Unreacted Al is also detected in the samples with higher Al content. In addition, by increasing x , all $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ -related peaks are shifted toward higher diffraction angles; however, the extent of the shifts is not uniform for all reflections. Figure 1(b) illustrates more specifically the differences in the shifts of the AlB_2 -type structured materials as a function of x . Some of the added Al, which substitutes Mg, changes intra-plane boron layers (c axis) more than inter-plane boron layers (ab plane). Consequently, the shifts of (002) reflections are much higher than those of (110) reflections. Moreover, the (110) reflections reveal sharp patterns at $x=0$ and 1 but in $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples with $x=0.25, 0.5$, and 0.75 rather broad peaks are observed. This proves that at intermediate Al concentrations the samples are not single-phased but contain more than one AlB_2 -type phase along with some unreacted Al. The presence of mixed AlB_2 -type phases has also been reported previously for various x values.^{4,20}

Raman spectra are depicted in Fig. 2 for undoped MgB_2 , undoped AlB_2 , and Al-substituted $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples with $x=0.25, 0.5$, and 0.75 . As the Raman-active soft mode (E_{2g}) is related to inter-plane vibrations of boron atoms,²⁷ this mode in MgB_2 around 600 cm^{-1} can be easily affected either by changes in defect concentration or elemental doping.²⁸ In this study, a reduction in defect concentration by doping

with Al can be deduced from the spectra shown in Fig. 2. The intensity of the E_{2g} mode for MgB_2 is reduced in the sample with $x=0.25$, and it almost disappears in samples with higher doping concentrations. In addition, a new, broad peak emerges in samples with $x=0.25$ and 0.5 , which becomes sharper in samples with $x=0.75$ and 1. This fingerprint Raman E_{2g} mode at around 980 cm^{-1} for AlB_2 ²⁹ is strongly connected to the Al doping level, which shows a red shift with increasing Al concentration. With this finding we disentangle the effect of Al on the Raman spectrum, which has already been mentioned in various reports.^{24,30,31} In most of these studies,^{30,31} Al was investigated in combination with other co-doping ions so that the effect of Al, based solely on the Raman spectra, was not easy to interpret. Our Raman spectroscopic findings shed some light on previously reported Raman data^{11,24} regarding MgB_2 , AlB_2 , and Al-doped MgB_2 .

EPR spectroscopy is a useful method to study defect centers of superconductive materials.^{32–34} The method uses

FIG. 2. Raman spectra of $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples with $x=0, 0.25, 0.5, 0.75$, and 1.

FIG. 3. First-derivative EPR spectra of $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$ samples (a) with $x = 0, 0.25, 0.5, 0.75$, and 1. (b) Deconvolution of (integrated) EPR spectra of EPR-active samples. (c) Defect concentration with respect to Al doping values calculated by spin counting via double integration of the spectra plotted in (a), or integration over the area of the curves given in (b).

unpaired electron spins as messengers, whose signal responses provide precise information on their direct local environment. Therefore, EPR spectroscopy is well suited to help understanding the role of defect centers since it provides a sensitive means to monitor different paramagnetic states of vacancies³⁵ or paramagnetic metal ions;³⁶ the method thus complements other experimental techniques such as Raman spectroscopy.³⁷

EPR results reveal that the effect of Al substitution on the electronic properties of MgB_2 is closely related to not only its defect structure but also the crystal symmetry. Possible defect centers in MgB_2 have been described by the MgB_2 spin Hamiltonian concept in our previous studies.^{26,29} Paramagnetic defects have been observed and assigned to Mg vacancies. EPR results of Al-doped MgB_2 specimen also confirm that paramagnetically active structural defects mainly arise from Mg vacancies. Figure 3(a) shows that the systematic increase of the Al concentration reduces the defect concentration until pure AlB_2 is reached, which does not exhibit any paramagnetic centers. In Fig. 3(b), the integrated EPR spectra of Fig. 3(a) are presented that allow for an exact determination of the defect concentration. Via spin counting, we calculated the absolute amount of defects as a function of Al doping (see Fig. 3(c)). Each green fit curve in Fig. 3(b) indicates the anisotropic behavior of g -factors in the defect centers. The maximum of each green fit curve gives us the accurate g -factor component, g_x , g_y , and g_z . Strong g -anisotropy is observed for Al-doped samples of MgB_2 , thus indicating considerable crystal deformations (see Fig. 3(b)). Undoped MgB_2 has axial symmetry, and therefore, its EPR signal exhibits only two principal components, parallel and perpendicular, of the g -tensor. Our EPR data reveal that doping triggers the quenching of Mg vacancies via trapping of Al ions. In a previous study,²⁸ we have shown that in C-doped MgB_2 defects still play a significant role in conductivity. In Al-doped MgB_2 , however, the role of defects smears out above a certain level of Al content, and Al reduces the conductivity of MgB_2 . Moreover, it is obvious that by increasing the Al content both EPR (Fig. 3(b)) and the XRD (Fig. 1) peaks reveal broadened lines, which

indicates the close relation between symmetry distortion of the crystal and the defect concentration.

Fig. 4 shows continuous-wave X-band EPR spectra recorded in the temperature range between 8 and 50 K, which includes the critical temperature of MgB_2 . Therefore, it is quite meaningful to compare EPR results at low temperatures for MgB_2 that is doped with different concentration levels of Al. In all samples, the EPR signal is induced by Mg vacancies around $g \sim 2$. It is remarkable that for $x = 0.25$ (Fig. 4(a) and inset) the temperature dependence of $1/(I \times T)$ falls off drastically around 25–30 K. The EPR intensity is expected to be proportional to the dc susceptibility of the spins, as can be seen in the inset of Fig. 4(a). Clearly, only the sample with $x = 0.25$ follows the Curie–Weiss law ($\chi = C/(T - \theta)$, where C and θ are the Curie constant and the Curie–Weiss temperature, respectively), thus indicating pure paramagnetic behavior until the respective critical temperature is reached. Similar Curie–Weiss paramagnetism has been observed previously for MgB_2 not only indirectly by

FIG. 4. Low-temperature EPR measurements (a) for $x = 0.25$ and (b) for $x = 0.5$ and 0.75 Al-doped MgB_2 material. The inset in Fig. 4(a) shows the temperature-dependent EPR signal intensity for the sample with $x = 0.25$. Note, that the low-temperature EPR results of undoped MgB_2 have been published elsewhere and a decrease in T_c has been reported.²⁸

superconducting quantum interference device (SQUID) measurements³⁸ but also directly using EPR for undoped and C-doped MgB_2 .²⁸ However, it is not possible to determine quantitatively the dependency of the superconducting fraction from EPR.³⁹ Accordingly, at this point it is important to note that Kim *et al.*⁴⁰ and Tyagi, Gould, and Bhagat *et al.*^{41–43} studied non-resonant magnetically modulated microwave absorption (MMA) method below the critical transition temperature which has proved to be a useful method to probe the electrodynamic behavior of superconductors. Of course EPR is not a convenient technique to study the superconducting states in general. The magnetic field acts on the sample in the normal state would be expected to be uniform and the resonance line-width would be narrow compared with line-widths when the sample is in the superconducting state, depending on the temperature, the sample would have differing amounts of flux exclusion in different parts of the sample. This can substantially contribute to the line-width of EPR. Thus, the room temperature results presented in Fig. 3(b) should be also carefully interpreted in case of at low temperatures (near and below T_c) by considering such facts. In the present work, only paramagnetically active defects were investigated that reveal EPR signal above T_c ; thus, here only EPR were applied. Below the critical temperature, the susceptibility starts to strongly deviate from a linear temperature dependence, and therefore, deviates from the Curie–Weiss behavior. This shows that the spin-dynamic properties of the samples are highly sensitive to the temperature, here T_c . If the EPR intensity follows the Curie–Weiss law at low temperatures, this evidently allows us to assume that the Mg-vacancy defects were both uniformly distributed in MgB_2 samples. Below T_c , paramagnetic defects in MgB_2 completely smear out and the materials become superconductive. However, just above T_c , before the paramagnetic defects smear out, both paramagnetic defects (here, V_{Mg}) still interact with the magnetic field and phonons, and thus cause quite “instable” EPR spectra.²⁸ This can be clearly seen in Fig. 4(a) that by approaching the T_c value the signal-to-noise ratio of the EPR spectra strongly decreases as compared with higher temperatures. In particular, the sample with $x = 0.25$ shows rapidly decreasing signal-to-noise ratio of EPR spectra measured below 32 K (see Fig. 4(a)), predominantly caused by an inhomogeneous distribution of the magnetic field inside the material due to the appearance of vortices.^{44,45} Such an EPR signal at the superconducting state was described recently by the realization of vortex structure in MgB_2 samples.⁴⁴ This effect has been also reported for different impurity centers in MgB_2 .⁴⁴ The inset of Fig. 4(a) shows that the superconducting state starts in the temperature region 25–30 K, and the sample becomes totally superparamagnetic at 17 K. This means that defect-induced paramagnetism, quite interestingly, still holds even below the bulk T_c value (39 K). The decrease in T_c has been also reported for increasing levels of Al: recently Karpinski *et al.* studied the effect of Al substitution in MgB_2 , and they reported a moderate decrease of the superconducting transition temperature for the samples with low Al-doping levels (i.e., small x values).^{46–48} More specifically, they stated that very mild (1%–2%) Al doping causes a decrease in T_c by 2–3 K.⁴⁸ At high levels of Al ($x = 0.32$)

T_c drops by 12 K. Such a reduction in T_c can be rationalized in three ways: (i) MgB_2 synthesized by a nanoboron precursor²⁶ reduces T_c due to its unique properties, (ii) interaction defects between phonons or between each other alter the superconducting temperature, and/or (iii) defects or Al doping enhance the lattice distortion. As expected, Al-doping levels greater than 0.25 did not reveal any superconducting state even at 8 K (see Fig. 4(b)). This is quite consistent with magnetic measurements of highly Al-doped MgB_2 samples.^{9,21,46,48–50}

A series of Al-doped MgB_2 samples ($\text{Mg}_{1-x}\text{Al}_x\text{B}_2$, $x = 0, 0.25, 0.5, 0.75$, and 1) was synthesized using a high-temperature solid-state method. Systematic substitution of doped Al into Mg sites was followed by means of X-ray diffraction peaks that shift to higher 2 theta degrees. Raman spectroscopic results reveal the strong correlation between Al doping, defect concentration (Mg vacancies), and the intensity and position of the E_{2g} mode. The latter is nearly absent in the Al-doped MgB_2 samples, and instead, another E_{2g} mode related to Al appears. EPR results are in agreement with the findings from Raman spectroscopy; they prove that the paramagnetic defects are mainly related to Mg vacancies. Combining the results from Raman spectroscopy and EPR clearly demonstrates that a reduced Mg content decreases also the Mg-related defect sites in the samples. The effects of structural defects on the electronic properties are gradually quenched by increasing the Al or decreasing the Mg contents. This confirms that paramagnetism of MgB_2 at room temperature is strongly related to Mg vacancies and Al plays a role as vacancy-filling ion. It is also worth to mention that EPR is an effective tool to understand the critical temperature in superconducting materials. EPR data showed suppression of T_c , at low levels of Al doping. At high levels ($x > 0.25$) superconductivity does not exist. With this study we contribute to an understanding of the complex structure of defects and Al-doping effects in superconductive MgB_2 .

This work was financially supported by the *Deutsche Forschungsgemeinschaft* (DFG), Grant No. ER 662/1-2 and the Koç University Graduate Program. The authors would like to thank Dr. Barış Yağcı (Koç University Surface Science and Technology Center (KUYTAM)) and Lorenz Heidinger (University of Freiburg) for contributing Raman spectroscopy and low temperature EPR measurements, respectively.

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