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Brant E. Kananen

Larry E. Halliburton

Elizabeth M. Scherrer

K. T. Stevens

G. K. Foundos

See next page for additional authors

Authors

Brant E. Kananen, Larry E. Halliburton, Elizabeth M. Scherrer, K. T. Stevens, G. K. Foundos, K. B. Chang, and Nancy C. Giles

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Electron paramagnetic resonance study of neutral Mg acceptors in β-Ga_2O_3 crystals

B. E. Kananen,[1] L. E. Halliburton,[2] E. M. Scherrer,[1] K. T. Stevens,[3] G. K. Foundos,[3] K. B. Chang,[3] and N. C. Giles[1,a)]
[1]*Department of Engineering Physics, Air Force Institute of Technology, Wright-Patterson Air Force Base, Ohio 45433, USA*
[2]*Department of Physics and Astronomy, West Virginia University, Morgantown, West Virginia 26505, USA*
[3]*Northrop Grumman Synoptics, 1201 Continental Blvd, Charlotte, North Carolina 28273, USA*

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Electron paramagnetic resonance (EPR) is used to directly observe and characterize neutral Mg acceptors (Mg_{Ga}^{0}) in a β-Ga_2O_3 crystal. These acceptors, best considered as small polarons, are produced when the Mg-doped crystal is irradiated at or near 77 K with x rays. During the irradiation, neutral acceptors are formed when holes are trapped at singly ionized Mg acceptors (Mg_{Ga}^{-}). Unintentionally present Fe^{3+} ($3d^5$) and Cr^{3+} ($3d^3$) transition-metal ions serve as the corresponding electron traps. The hole is localized in a nonbonding p orbital on a threefold-coordinated oxygen ion adjacent to an Mg ion at a sixfold-coordinated Ga site. These Mg_{Ga}^{0} acceptors ($S = 1/2$) have a slightly anisotropic g matrix (principal values are 2.0038, 2.0153, and 2.0371). There is also partially resolved ^{69}Ga and ^{71}Ga hyperfine structure resulting from unequal interactions with the two Ga ions adjacent to the hole. With the magnetic field along the a direction, hyperfine parameters are 2.61 and 1.18 mT for the ^{69}Ga nuclei at the two inequivalent neighboring Ga sites. The Mg_{Ga}^{0} acceptors thermally convert back to their nonparamagnetic Mg_{Ga}^{-} charge state when the temperature of the crystal is raised above approximately 250 K. *Published by AIP Publishing.*
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At present, there is considerable interest in identifying suitable acceptors that will allow p-type gallium oxide (β-Ga_2O_3) to be readily produced. It is easy to make n-type material by doping with silicon or other Group IV ions, but making p-type materials is more challenging.[1,2] Because of its success in GaN,[3,4] magnesium has been considered a possible acceptor dopant in β-Ga_2O_3. According to several recent reports,[5–7] doping β-Ga_2O_3 bulk crystals or thin films with Mg produces semi-insulating material, thus demonstrating that these ions serve as compensating acceptors for donor impurities (e.g., the ubiquitous Si ions). To date, there have been no reports that Mg doping leads to effective hole conduction, possibly because the concentration of unintentionally present donors has always exceeded the concentration of isolated Mg acceptors. It is also possible that Mg acceptors have a sufficiently deep 0/– level that prevents the thermal generation of measurable concentrations of holes in the valence band even when donor concentrations are small.

In this letter, we use electron paramagnetic resonance (EPR) to identify and characterize the neutral charge state of Mg acceptors in a β-Ga_2O_3 crystal (the only previous EPR study[8] of acceptors in this material was focused on gallium vacancies). Singly ionized acceptors (Mg_{Ga}^{-}) are present in the as-grown Mg-doped crystal. They are converted into neutral acceptors (Mg_{Ga}^{0}) when the crystal is exposed to ionizing radiation (in our case, x rays) while being held at 77 K. The g matrix of these $S = 1/2$ defects is obtained from the angular dependence of their spectra. Partially resolved hyperfine structure from interactions with ^{69}Ga and ^{71}Ga nuclei are seen for all orientations of magnetic field. The model we assign to these Mg_{Ga}^{0} acceptors has an unpaired spin (i.e., the hole) localized in a "nonbonding" p orbital on an oxygen ion with one Mg neighbor and two Ga neighbors. In the future, advanced density-functional-theory (DFT) studies by other research groups are expected to further refine this model for the neutral Mg_{Ga}^{0} acceptor by including lattice relaxation and charge delocalization effects. These DFT investigations are proving to be useful in understanding and predicting the fundamental properties of point defects in β-Ga_2O_3 and similar materials.[9–12]

The localization of the hole on one oxygen ion suggests that the Mg_{Ga}^{0} acceptor is best viewed as a small polaron of the type described in detail by Schirmer[13,14] and Stoneham *et al.*[15] As demonstrated in these review papers,[13–15] acceptor-bound small polarons are widely observed in oxide crystals, with a particularly relevant example being the hole trapped adjacent to an Mg impurity in Al_2O_3 crystals.[16,17] In contrast, EPR shows that the neutral Mg acceptor in GaN behaves as a conventional shallow acceptor with a strongly anisotropic g matrix ($g_{\parallel} = 2.2$ and $g_{\perp} < 0.2$) when minimal strain is present.[18,19]

The Mg-doped β-Ga_2O_3 crystal used in this study was grown by the Czochralski method at Northrop Grumman Synoptics (Charlotte, NC). Approximately 0.20 mol.% of MgO was included in the starting material. An iridium crucible was used, and the growth atmospheres consisted of varying mixtures of Ar, CO_2, and O_2. The growth rate was 1.5 mm/h. This process routinely yields large boules of 35 mm in diameter and 30 mm in length. An EPR sample, with dimensions of $5.0 \times 1.4 \times 3.0\,mm^3$, was cut from the larger boule after the crystal directions were precisely determined using the x-ray Laue technique. The structure of β-Ga_2O_3 is monoclinic, and

a)Author to whom correspondence should be addressed: Nancy.Giles@afit.edu

the space group is C2/m (C_{2h}^3). Lattice constants[20,21] are $a = 12.214$ Å, $b = 3.0371$ Å, $c = 5.7981$ Å, and $\beta = 103.83°$. There are two inequivalent gallium sites and three inequivalent oxygen sites in this crystal. The Ga(I) ions are at tetrahedral sites, and the Ga(II) ions are at octahedral sites. The O(I) and O(II) ions have three gallium neighbors, and the O(III) ions have four gallium neighbors. A ball-and-stick model of the β-Ga_2O_3 crystal is shown in Fig. 1.[22]

A Bruker EMX spectrometer operating near 9.40 GHz was used to acquire the EPR spectra. An Oxford helium-gas flow system controlled the temperature of the sample, and a Bruker nuclear magnetic resonance (NMR) teslameter was used to measure the static magnetic field. The teslameter probe was placed alongside the microwave cavity in the magnet gap, and a small Cr-doped MgO crystal, replacing the β-Ga_2O_3 sample, was used to correct for differences in the magnetic field between the sample position and the tip of the teslameter probe (the isotropic g value for Cr^{3+} in MgO is 1.9800). An OEG-76H x-ray tube from Varian (operating at 60 kV and 30 mA) was used to produce the neutral Mg_{Ga}^0 acceptors. The irradiation times were typically 2–3 min. Because β-Ga_2O_3 has a 4.9 eV band gap, x rays were an efficient way to generate the "free" electrons and holes needed to form the Mg_{Ga}^0 acceptors at low temperature. An intense light beam near 250 nm, from either a laser or a lamp, should also produce the Mg_{Ga}^0 acceptors.

The as-grown Mg-doped β-Ga_2O_3 sample was semi-insulating (at room temperature, there was no infrared absorption due to free carriers and no nonresonant absorption of microwaves). Before an x-ray irradiation, the only observable EPR signals were from Fe^{3+} and Cr^{3+} ions. With the magnetic field along the b axis and a microwave frequency of 9.397 GHz, the Fe^{3+} ions have lines near 165, 304, and 606 mT and Cr^{3+} ions have lines near 131 and 1300 mT.[23,24] Then, as shown in Fig. 2(a), an irradiation at 77 K with x rays produced the EPR spectrum from neutral magnesium acceptors (Mg_{Ga}^0). (Note: This x-ray-induced spectrum could not be produced in β-Ga_2O_3 crystals that were not intentionally doped with Mg ions.) The spectrum in Fig. 2(a) was taken at 40 K with the magnetic field parallel to the a direction. During the irradiation, the sample was immersed in liquid nitrogen. Following the irradiation, the sample was inserted into a slot in a cylindrical Teflon holder while it remained in liquid nitrogen. The holder, with the sample, was then quickly transferred from the liquid nitrogen to the stream of cold helium gas that was flowing through the vertical quartz glassware (i.e., the cryostat) inside the microwave cavity. Negligible warming of the sample occurred during this transfer.

In addition to forming the neutral acceptors, the x rays caused the EPR signals from the Fe^{3+} and Cr^{3+} ions to decrease to approximately 35% of their initial intensity, as they convert to Fe^{2+} and Cr^{2+} ions by trapping an electron. The Fe^{3+} EPR line near 165 mT is shown in Fig. 3 before and after the x-ray irradiation. These spectra were taken at 40 K with the magnetic field along the b direction. Although it is difficult to determine absolute spin concentrations, the increase in the concentration of the neutral Mg acceptor appears to be approximately the same as the decrease in the concentration of the Fe^{3+} and Cr^{3+} ions.

Similar to previous observations[8] for the doubly ionized gallium vacancy (V_{Ga}^{2-}), the EPR spectrum of the neutral magnesium Mg_{Ga}^0 acceptor in β-Ga_2O_3 has partially resolved hyperfine structure from interactions with ^{69}Ga and ^{71}Ga nuclei. The ^{69}Ga nuclei are 60.1% abundant, and the ^{71}Ga nuclei are 39.9% abundant. Both isotopes have I = 3/2 nuclear spins, and their magnetic moments are $^{69}\mu = +2.0166\beta_n$ and

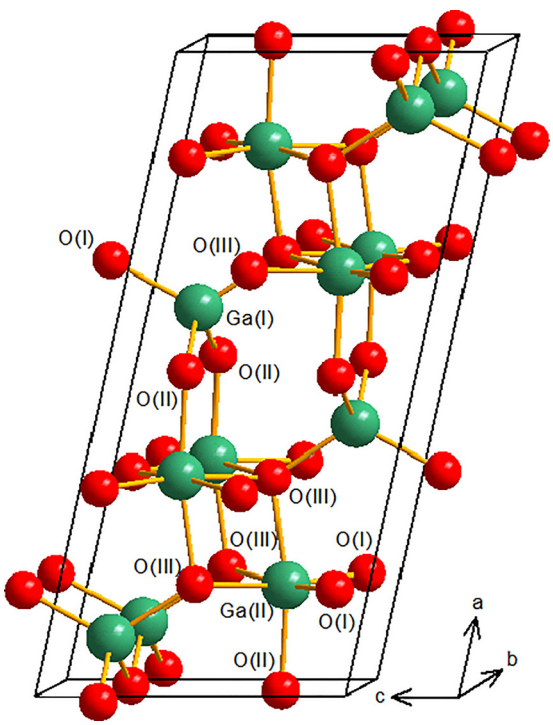

FIG. 1. Crystal structure of β-Ga_2O_3. Gallium ions are shown in green, and oxygen ions are shown in red. The two inequivalent gallium sites, Ga(I) and Ga(II), and the three inequivalent oxygen sites, O(I), O(II), and O(III), are labeled.

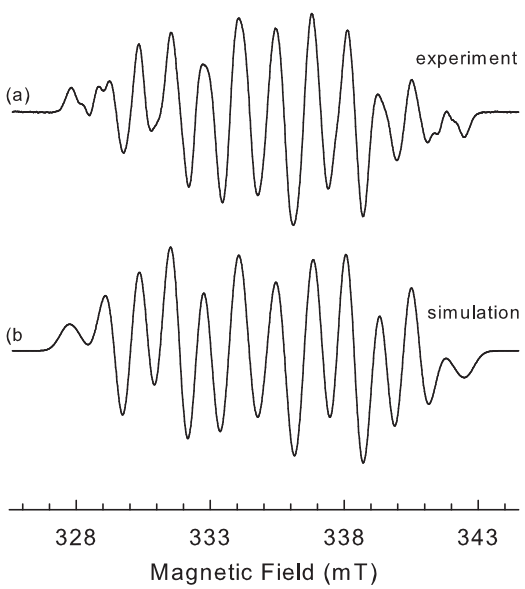

FIG. 2. (a) EPR spectrum from the S = 1/2 neutral magnesium acceptor (Mg_{Ga}^0) in Mg-doped β-Ga_2O_3. The spectrum was taken at 40 K after an irradiation at 77 K with x rays. The magnetic field is along the a direction, and the microwave frequency is 9.3979 GHz. (b) Simulated EPR spectrum produced using EasySpin (see Ref. 25).

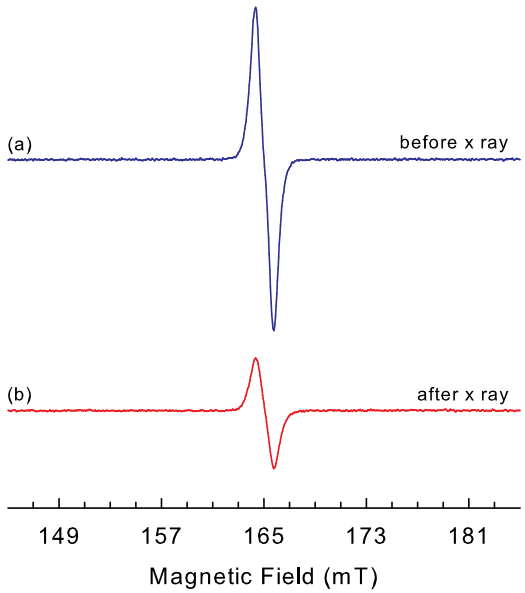

FIG. 3. Radiation-induced change in the EPR spectrum of Fe^{3+} ions in the Mg-doped β-Ga_2O_3 crystal. (a) Spectrum (blue curve) taken before an irradiation at 77 K with x rays. (b) Spectrum (red curve) taken after the irradiation. The spectra were acquired at 40 K, with the magnetic field along the *b* direction and a microwave frequency of 9.397 GHz.

$^{71}\mu = +2.5623\beta_n$, where β_n is the nuclear magneton. The complex hyperfine pattern in Fig. 2(a) is caused by unequal interactions of the unpaired spin with the gallium nuclei at two neighboring cation sites. Direct evidence that the interactions are unequal is the absence of a center line in the experimentally measured spectrum (equal interactions would require the most intense line to be at the center of a symmetrical hyperfine pattern, as was seen for the gallium vacancy[8]). The parameters describing the hyperfine interactions at one gallium neighbor are approximately a factor of 2.2 larger than the parameters describing the interactions at the other gallium neighbor. Since each of the neighboring gallium sites can have either a ^{69}Ga or a ^{71}Ga nucleus, the resulting EPR spectrum consists of 64 lines that are strongly overlapping because of their large linewidths, with only the "envelopes" of groups of these lines being well resolved.

The simulated spectrum in Fig. 2(b) was created with the EasySpin computer program.[25] Isotropic hyperfine matrices were used, with principal values of 2.61 and 3.32 mT for the ^{69}Ga and ^{71}Ga nuclei at the site with the larger hyperfine interactions and values of 1.18 and 1.49 mT for the ^{69}Ga and ^{71}Ga nuclei at the site with the smaller hyperfine interactions. In the simulation, the linewidth was set at 0.77 mT. The two spectra in Fig. 2 (experiment and simulation for the magnetic field along the *a* direction) are in good agreement, thus confirming our explanation that two gallium sites with unequal interactions are responsible for the experimental hyperfine pattern. Slight differences in the two spectra in Fig. 2 may be due to (1) the presence of one or more less intense lines from other defects underlying the experimental Mg^0_{Ga} acceptor spectrum and (2) the disregard of nuclear electric quadrupole terms in the simulation. The hyperfine parameters used to describe the EPR spectra are little changed when the magnetic field is along the *b* and *c* directions. These Ga hyperfine parameters are listed in Table I for the three crystallographic directions. The isotropic nature of these hyperfine matrices suggests that the unpaired spin density on the two neighboring ions is primarily in Ga 4*s* orbitals. As expected, hyperfine features from ^{25}Mg nuclei were not observed in our experimental spectra, primarily because of their natural abundance of only 10%, their larger I = 5/2 nuclear spin, and their most likely smaller hyperfine parameters.

TABLE I. Spin-Hamiltonian parameters describing the EPR spectra from the neutral magnesium acceptor (Mg^0_{Ga}) for different orientations of the magnetic field. The ^{69}Ga and ^{71}Ga parameters refer to interactions of the hole with the neighboring Ga(I) and Ga(II) ions (see Fig. 5). The estimated error is ±0.0004 for the *g* values and ±0.03 mT for the hyperfine values.

		Hyperfine parameters (mT)			
		Ga(I)		Ga(II)	
Direction of the magnetic field	*g* value	^{69}Ga	^{71}Ga	^{69}Ga	^{71}Ga
a crystal axis	2.0038	2.61	3.32	1.18	1.49
b crystal axis	2.0153	2.56	3.25	1.19	1.52
c crystal axis	2.0371	2.55	3.24	1.13	1.44

Figure 4 shows the angular dependence associated with the *g* matrix of the Mg^0_{Ga} acceptor. The data points in this figure represent the magnetic field at the center of the hyperfine patterns as the direction of the field is rotated from *a* to *b*, *b* to *c*, and finally *c* to *a** to –*c* in 15° steps. The *a** direction in Fig. 4 is 90° from *b* and *c*. Although small (less than 0.10 mT) and not shown in Fig. 4, site splitting due to the magnetic inequivalence of the two crystallographically equivalent orientations of the Mg^0_{Ga} acceptor was observed during rotation in the *b*–*c* plane. Site splitting was not observed for rotation in the *a*–*b* plane. The angular dependence in Fig. 4 is described by the electron Zeeman term in the spin Hamiltonian ($H = \beta \mathbf{S}\cdot\mathbf{g}\cdot\mathbf{B} + \sum(\mathbf{I_i}\cdot\mathbf{A_i}\cdot\mathbf{S})$). Turning points are near the *a* direction and near the *c* direction. Thus, principal axes of the *g* matrix are approximately along the

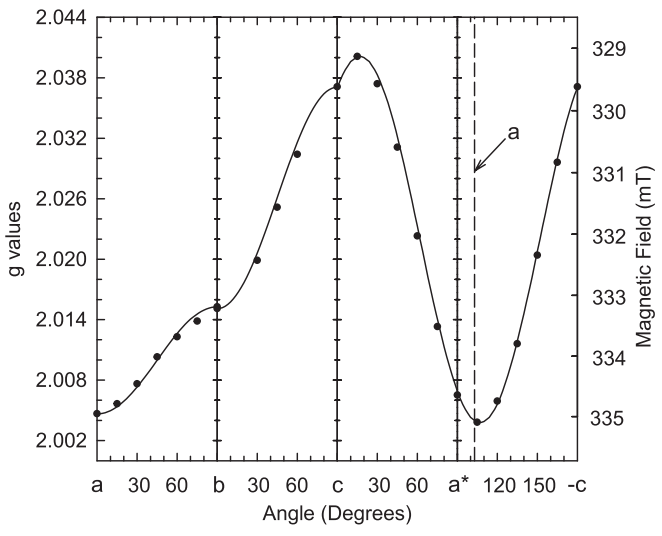

FIG. 4. Angular dependence of the *g* matrix of the Mg^0_{Ga} acceptor. The center of the hyperfine pattern is plotted versus the direction of the magnetic field for rotations in three planes. The discrete points represent the experimental results, and the solid curves represent computer-generated results using the *g* values in Table I. Magnetic field values along the right vertical axis correspond to a microwave frequency of 9.398 GHz.

crystallographic *a*, *b*, and *c* directions. The principal values corresponding to these principal-axis directions are listed in Table I.

As described in the analysis[8] of the doubly ionized gallium vacancy (V_{Ga}^{2-}) in β-Ga_2O_3, the small and positive *g* shifts observed for the neutral Mg_{Ga}^{0} acceptor indicate that the hole is located in a *p* orbital on an oxygen ion adjacent to the Mg ion. The resulting O^- ion has the $2p^5$ configuration, with the threefold orbital degeneracy of this ^2P state ($L = 1$, $S = 1/2$) removed by the local crystalline electric field. A spin-orbit interaction mixes these three states and gives rise to the observed *g* shifts. The value of 2.0038 for g_a in Table I is close to the 2.0023 "free" spin value, which implies that the p_z orbital containing the unpaired spin is aligned nearly along the *a* direction in the crystal.

The model for the neutral magnesium acceptor (Mg_{Ga}^{0}) is shown in Fig. 5. As expected for an acceptor-bound small polaron in the partially ionic β-Ga_2O_3 crystal, the Mg^{2+} ion on the Ga^{3+} site attracts a hole, with the hole residing on an adjacent oxygen ion instead of either forming an Mg^{3+} ion or having the hole delocalized but centered on the Mg ion. Our observation of resolved hyperfine structure with two, not three, gallium neighbors requires the hole to be on a threefold-coordinated oxygen ion with the Mg ion at one of the three nearest-neighbor gallium positions. This oxygen must be an O(I) ion, instead of an O(II) ion, because the two gallium hyperfine interactions are not equivalent. We place the Mg ion at the sixfold-coordinated gallium site, in agreement with computational studies[26,27] that suggest that Mg ions occupy the sixfold-coordinated Ga(II) sites in β-Ga_2O_3 [these sites provide more space for the slightly larger impurity ion than the fourfold Ga(I) sites]. An EPR study has also placed Cr impurities on the sixfold Ga(II) site.[24] Specifically, the model in Fig. 5 has the hole localized on a threefold oxygen ion at an O(I) site with the Mg ion at a neighboring sixfold Ga(II) site. The unpaired spin is in a nonbonding oxygen *p* orbital and thus avoids its positive Mg and Ga neighbors as much as possible. In a first approximation, the oxygen ion with the hole is near the center of a plane defined by its three cation neighbors (one Mg ion and two Ga ions) and the nonbonding *p* orbital is oriented perpendicular to this plane (nearly along the *a* direction in the crystal). Similar nonbonding orientations of the *p* orbital have been seen in holelike small polarons in SiO_2 and GeO_2 crystals.[28–32] In contrast, the doubly ionized gallium vacancy[8] in β-Ga_2O_3 has the hole localized in a *p* orbital that points toward the vacancy. This latter behavior is a result of the dominating influence of the large negative "effective" charge of the gallium vacancy (which attracts the positive hole). In Fig. 5, the inequivalent hyperfine interactions are with a gallium nucleus at a Ga(I) site and a gallium nucleus at a Ga(II) site. In the unrelaxed lattice near room temperature, the Ga(I) ion is 1.835 Å from the oxygen ion with the hole, whereas the Ga(II) ion is 1.937 Å from the oxygen ion.[21] In Table I, the larger hyperfine interaction is tentatively assigned to the Ga(I) ion because it is closer to the oxygen ion.

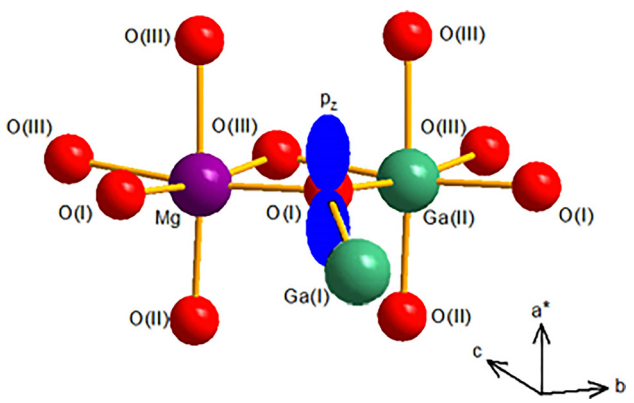

FIG. 5. Model of the neutral magnesium acceptor (Mg_{Ga}^{0}) in a β-Ga_2O_3 crystal. The unpaired spin (the hole shown in blue) is localized in a nonbonding *p* orbital on a threefold oxygen ion, O(I), adjacent to the Mg ion at a sixfold Ga(II) site. The primary hyperfine interactions are with the Ga(I) and Ga(II) ions adjacent to the hole.

The thermal stability of the neutral charge state of the magnesium acceptor (Mg_{Ga}^{0}) was determined by monitoring its EPR spectrum after the crystal was warmed briefly to several higher temperatures. Following an irradiation at 77 K with x rays, the Mg_{Ga}^{0} spectrum was acquired at 40 K. The crystal was then held at 225 K for 2 min and returned to 40 K where the spectrum was recorded. Next, after holding the crystal at 250 K for 2 min, the spectrum was again recorded at 40 K. The 225 K anneal did not significantly reduce the concentration of Mg_{Ga}^{0} acceptors, while the 250 K anneal destroyed nearly all of the neutral acceptors. These Mg_{Ga}^{0} acceptors thermally decay either because a trapped electron (from the Fe^{2+} and/or Cr^{2+} ions) returns to the acceptor or the hole "moves" away from the Mg ion via a small-polaron hopping process. Although it is not known which of these two mechanisms dominates, the thermal decay of the Mg_{Ga}^{0} acceptors near 250 K suggests that the associated activation energy is about 550 meV. We use the approximation $E \approx 25kT_m$ where T_m corresponds to the temperature where half of the acceptors have disappeared (in our case, T_m is ~250 K).[33–35] A possible relationship between this activation energy and the 0/– level of the magnesium acceptors needs to be explored. Also, the role of Fe^{2+} and Cr^{2+} ions in the thermal recovery of the Mg_{Ga}^{-} charge state of the acceptors is an area for further investigation.

In summary, an EPR spectrum from neutral magnesium acceptors in β-Ga_2O_3 is identified and characterized. These Mg_{Ga}^{0} acceptors are produced when an Mg-doped crystal is exposed to ionizing radiation (i.e., x rays) while being held at or near 77 K. Inadvertently present Fe^{3+} and Cr^{3+} impurities serve as the electron traps. Our model for the neutral magnesium acceptor has the unpaired spin (i.e., the hole) localized in a nonbonding *p* orbital on a threefold-coordinated oxygen ion adjacent to a magnesium ion at a sixfold-coordinated gallium site. Hyperfine interactions with the ^{69}Ga and ^{71}Ga nuclei at two neighboring gallium sites are partially resolved. The radiation-induced Mg_{Ga}^{0} acceptors become thermally unstable above 250 K, with an activation energy near 550 meV.

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