Monoclinic β-Ga2O3 has received increasing attention in the last few years thanks to its unique properties and its availability as a bulk substrate. It is the transparent metal oxide material with the widest bandgap of ~4.9 eV among the most practical ones. In comparison to Si, SiC and GaN materials, the significantly higher breakdown field (8 MV/cm) is achieved for β-Ga2O3, makes it more suitable for high-power electronics. The transparency in the solar spectrum (especially in the ultra-violet (UV) region), the development of single crystal substrates, the good thermal/chemical stability, and the achievement of reasonable carrier mobility values, pave the way for the use of β-Ga2O3 as a material for thin film electroluminescent devices, for efficient red thin film luminescent devices.

In this work, we have established the effects of Eu implantation and annealing on β-Ga2O3 thin films grown by metal organic vapor phase epitaxy (MOVPE) on sapphire substrate. The study is based on the combined information from structural and optical techniques: X-ray diffraction (XRD), Rutherford backscattering spectrometry (RBS), cathodoluminescence (CL), photoluminescence (PL), and photoluminescence excitation (PLE). The thin films were implanted with a fluence of $1 \times 10^{15}$ Eu·cm$^{-2}$ and annealed at 900°C. Neither significant changes in peak width or position nor additional peaks related to Eu complexes were detected in the XRD 2θ scans. RBS results and SRIM simulation are in good agreement, revealing that no Eu diffusion to the surface occurs during annealing. For the used implantation/annealing conditions, the Eu ion penetration depth reached ~130 nm, with a maximum concentration at ~50 nm. Furthermore, CL and PLE/PLE results evidenced the optical activation of the Eu$^{3+}$ in the β-Ga2O3 host. The detailed study of the Eu$^{3+}$ intra-$f$ shell transitions revealed that at least one active site is created by the Eu implantation/annealing in β-Ga2O3 thin films grown on sapphire. Independently of the β-Ga2O3 film thickness, well controlled optical activation of implanted Eu was achieved.

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in the same experimental conditions, using the front face acquisition geometry, and corrected to the spectral response of the optical components and the Xe lamp. Further PL tests were realized using the vacuum ultraviolet (VUV) excitation on a Fluorimeter Horiba Scientific modular equipment with two monochromators (H20-UVL) (200 nm, 1200 grooves mm⁻¹), one at the excitation and one at the emission. A D200VUV deuterium light source emitting from 115 to 370 nm was used as excitation source, with a maximum intensity at 160 nm.

Results and Discussion

In order to evaluate the structural quality of the films, 2θ-ω scans were performed and Fig. 1 depicts the XRD scans of the different β-Ga2O3 thin films on sapphire (S1, S2 and S3) and the Eu-implanted/annealed samples (S1-Eu, S2-Eu and S3-Eu). The sharp peaks at 2θ = 21.0°, 41.7° and 64.5° correspond to the 00.3, 00.6 and 00.9 Bragg reflections of the c-plane sapphire substrate, respectively. While the peaks at 2θ = 18.9°, 38.4° and 59.2° correspond to the β-Ga2O3 epitaxial layers, and are assigned to the -201, -402 and -603 Bragg reflections from the monoclinic β-Ga2O3 crystalline structure.38,39 The peak at 2θ = 37.5° (asterisk) is the Kβ line corresponding to the 00.6 Bragg reflection. The presence of only one family of planes suggests that the as-grown β-Ga2O3 thin films are preferentially oriented with (-201) surface orientation and without additional phases. Furthermore, for the implanted and annealed β-Ga2O3 thin films (S1-Eu, S2-Eu and S3-Eu), neither additional peaks related to Eu complexes nor significant change of the peak width or position were detected.

Figure 2 shows the random RBS spectra of the as-grown and Eu-implanted/annealed β-Ga2O3 thin films with a fluence of 1 x 10¹⁵ Eu·cm⁻². These spectra are characterized by four barriers assigned to Ga and O from the thin β-Ga2O3 film, then Al and O from the sapphire substrate. As expected, RBS spectra exhibit an increase of the barrier width assigned to Ga with increasing the β-Ga2O3 film thickness. In the implanted and annealed samples, the signal at high energy assigned to the implanted Eu is also detected. The spectra were fitted, using the NDF code considering a model of four layers plus substrate, in order to take into account the composition gradient with depth. The fits suggest that the films present a Ga/O ratio of ~0.6, with a Ga content decreasing by 10% from the surface layer down to the layer right on top of the substrate. Furthermore, the fits also reveal that no significant stoichiometry changes occur after Eu-implantation and annealing. The comparison of the Eu profile measured by RBS with the SRIM simulation (Fig. 3), shows a fairly good agreement, revealing that no Eu diffusion to the surface occurs during annealing. For all samples and in the used implantation/annealing conditions, the Eu ion penetration depth reaches ~130 nm, with a maximum concentration at ~50 nm.

Fig. 4a represents RT CL spectra of the Eu-implanted/annealed β-Ga2O3 thin films (S1-Eu, S2-Eu and S3-Eu), compared with the ones obtained for β-Ga2O3 bulk single crystals.27 The detailed implantation and annealing conditions are also indicated. The spectra present a broad UV band and sharp luminescence lines with dominant intensity. For clarity, Fig. 4b represents the UV-blue wavelength region. For the β-Ga2O3 thin films, the CL response exhibits a UV-band luminescence, typical for as-grown β-Ga2O3,22,23,38–42 and commonly associated to intrinsic point defects, such as oxygen vacancies, gallium vacancies and oxygen-gallium vacancy pairs.38,41–43 In addition to intrinsic defects, the UV band luminescence in β-Ga2O3 was reported to depend on doping9,66 as well as implantation and annealing conditions.26 For wavelengths above 520 nm in Fig. 4c, sharp luminescence lines are well resolved. These lines are characteristic of the Eu³⁺ intra-4f shell transitions, indicating that the used Eu implantation and annealing conditions efficiently incorporated the Eu³⁺ ions in the crystal lattice and optically activated the Eu³⁺ ions in β-Ga2O3 thin films. Furthermore, by comparing with PL response from Eu-implanted/annealed c-sapphire from Ref.47, we have confirmed that the observed transitions are not originating from Eu³⁺ ions incorporated in c-sapphire (not shown). It should be pointed outting that besides the Eu³⁺ intra-4f shell transitions, typical Cr³⁺ emission corresponding to 4T₂ → 4A₂
at \( \sim 697 \) nm is observed, which is often present as impurity in \( \beta\text{-Ga}_2\text{O}_3 \). It is unlikely that such emission originates from Cr\(^{3+}\) contaminants in sapphire, since the excitation depth of the electron beam is lower than 50 nm, well below the \( \beta\text{-Ga}_2\text{O}_3 \) thin film thickness.

The fact that a large fraction of Eu ions is in the 3\(^{+}\) valence state, is in accordance with CL and X-ray absorption near edge structure (XANES) results obtained for Eu-implanted/annealed \( \beta\text{-Ga}_2\text{O}_3 \) bulk single crystals.\(^{27}\) The most intense emission is attributed to the \( ^5\text{D}_0 \rightarrow ^7\text{F}_0 \) transition located at \( \sim 611.5 \) nm, in agreement with previously reported values obtained for Eu-doped \( \beta\text{-Ga}_2\text{O}_3 \) nanostructures, fibers, thin films and bulk crystals.\(^{25,23,26,27,49-51}\) The different \( ^5\text{D}_{0,1,2,3,4} \rightarrow ^7\text{F}_J \) transitions represented in Fig. 4c, were assigned after a careful comparison with previous reports and presented in Table I in comparison with the measurements on Eu\(^{3+}\)-doped \( \beta\text{-Ga}_2\text{O}_3 \) nanocrystals from Ref. 23. For these transitions, the shape and the relative intensities of the Eu\(^{3+}\) intra-\( 4f \) shell transitions are very similar for the Eu-implanted/annealed \( \beta\text{-Ga}_2\text{O}_3 \) thin films and bulk single crystals. However, below 590 nm in Fig. 4d, slight changes can be observed. Unlike \( \beta\text{-Ga}_2\text{O}_3 \) bulk single crystals\(^{5,14} \) and \( \beta\text{-Ga}_2\text{O}_3 \) nanocrystals,\(^{3,23}\) presenting one transition at 582 nm (at RT) or at 579.9 nm (at 10 K), the present thin films exhibit at least two transitions located at 579.7 and 582 nm, that can be attributed to \( ^5\text{D}_0 \rightarrow ^7\text{F}_0 \) and \( ^5\text{D}_0 \rightarrow ^7\text{F}_2 \) or \( ^5\text{D}_1 \rightarrow ^7\text{F}_3 \) transitions, respectively. Due to the singlet character of the \( ^7\text{F}_0 \) fundamental and \( ^5\text{D}_0 \) excited levels, the number of the observed \( ^5\text{D}_0 \rightarrow ^7\text{F}_0 \) transitions corresponds to the number of non-equivalent active sites in the Eu implanted sample.\(^{17,52}\) Accordingly, at least one active site is created by the Eu-implantation/annealing in the \( \beta\text{-Ga}_2\text{O}_3 \) bulk material, and were only included in Table I.

In Figs. 5a and 5b, combined excitation emission (CEE) spectra are represented for the S3-Eu sample in the UV and red regions, respectively. Due to the very low Eu\(^{3+}\) intra-\( 4f \) shell transition intensity obtained for S1-Eu and S2-Eu samples, this part will be mainly dedicated to the \( \beta\text{-Ga}_2\text{O}_3 \) thin film with the highest thickness (S3-Eu sample), also showing the highest CL intensity of the \( ^5\text{D}_0 \rightarrow ^7\text{F}_2 \) transition. It is worth mentioning that unlike CL, PL measurements are scarce for Eu-implanted/annealed \( \beta\text{-Ga}_2\text{O}_3 \) material, and were only successfully achieved for nanowires, but not for bulk crystals. In our

![Figure 4](https://example.com/figure4.png)

**Figure 4.** RT CL spectra of S1-Eu, S2-Eu and S3-Eu \( \beta\text{-Ga}_2\text{O}_3 \) thin films compared with Eu implanted/annealed bulk \( \beta\text{-Ga}_2\text{O}_3 \) samples at similar conditions from Ref. 27 (a) in the whole range, (b) in the UV-blue range, (c) and (d) around the \( ^5\text{D}_{0,1} \rightarrow ^7\text{F}_J \) transitions.
Table I. $\text{Eu}^{3+}$ (4$f^6$) intraionic transitions observed in the Eu implanted/annealed $\beta$-Ga$_2$O$_3$ thin films and corresponding assignments $^5\text{D}_{0,1} \to ^7\text{F}_{0,1,2,3,4}$ in comparison with Zhu et al.$^{23}$

| Transitions (nm) | Present work CL at RT $^{10^15}$ Eu ions.cm$^{-2}$ implanted/annealed in MOVPE $\beta$-Ga$_2$O$_3$ thin films grown on sapphire | Zhu et al.$^{23}$ PL at 10 K $\text{Eu}^{3+}$-doped $\beta$-Ga$_2$O$_3$ NCs were synthesized by combustion |
|------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| $^5\text{D}_1 \to ^7\text{F}_1$ | 534.8 | 533.2 |
| | 541.4 | 538.3 |
| | 532.3 | 537.4 |
| | 540.4 | 531.6 |
| | 536.7 | 539.7 |
| $^5\text{D}_1 \to ^7\text{F}_2$ | 540 | 552 |
| | 553.9 | 552.2 |
| | 558.1 | 554.6 |
| | 559.9 | 564.6 |
| | 564.6 | 566.3 |
| | 551.4 | 556.3 |
| | 553.1 | 558.1 |
| | 556.3 | 562.8 |
| $^5\text{D}_0 \to ^7\text{F}_0$ | 579.7 | 579.9 |
| $^5\text{D}_0 \to ^7\text{F}_0$ or $^5\text{D}_1 \to ^7\text{F}_3$ | 582 | 587.5 |
| | 593.7 | 597.4 |
| $^5\text{D}_0 \to ^7\text{F}_1$ | 590 | 611.7 |
| | 596 | 611.8 |
| | | 613.9 |
| | | 617.8 |
| | | 620 |
| | | 625.8 |
| $^5\text{D}_0 \to ^7\text{F}_2$ | 617.7 | 649 |
| | 621.4 | 650.5 |
| | 626.6 | 651.8 |
| | | 653 |
| | | 655.3 |
| | | 656.2 |
| | | 659.7 |
| $^5\text{D}_0 \to ^7\text{F}_3$ | 653.4 | 661.5 |
| | 661.5 | 682.1 |
| | | 692.9 |
| | | 698 |
| | | 703.9 |
| | | 704.7 |
| $^5\text{D}_0 \to ^7\text{F}_4$ | 707.4 | 707.8 |
| | 712.6 | 717.8 |

In this case, CEE spectroscopy consists of measuring the emission spectrum for each excitation wavelength, from 250 to 265 nm, and from 250 to 300 nm, in the emission wavelength range close to the UV band (Fig. 5a), and the Eu$^{3+}$ intra-4$f$ shell transitions (Fig. 5b), respectively. The blue curves represent the PL spectra for S3-Eu sample, obtained under 260 nm (4.77 eV) excitation wavelength, resonant with the $\beta$-Ga$_2$O$_3$ bandgap value (obtained from transmission measurement). The PL response exhibits the UV band emission (Fig. 5a) and the Eu$^{3+}$ and Cr$^{3+}$ lines (Fig. 5b). The intensity ratio of Eu-related lines versus the UV band ($\sim$1/10) is much lower than the one obtained by CL ($\sim$5). It is reported that this ratio is strongly dependent on the excitation parameters, especially, on the excitation density.$^{23,26}$ Indeed, this is expected due to the low excitation density of the Xe lamp at 260 nm used in this measurement. Additional RT PL measurements on S3-Eu (not shown), using 160 nm excitation wavelength from a deuterium light source (lower excitation density than the Xe lamp), was only able to resolve the Cr$^{3+}$ emission line, indicating that such excitation conditions are not promoting the Eu$^{3+}$ intra-4$f$ shell transitions. Fig. 5a and Fig. 5b demonstrate that efficient excitation of the UV band and the $^5\text{D}_0 \to ^7\text{F}_2$ most intense transition occurs through a broad excitation band around the bandgap energy of the $\beta$-Ga$_2$O$_3$ host ($\sim$ 260 nm, 4.77 eV).
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**Acknowledgments**

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**Conclusions**

In this work, we have established the effects of Eu implantation and annealing on β-Ga2O3 thin films grown by metal organic vapor phase epitaxy on sapphire substrate. The thin films were implanted with 300 keV Eu ions at 600°C, with a fluence of 1 × 1015 Eu·cm−2 and annealed at 900°C in flowing argon for 30 s. No significant changes neither additional peaks related to Eu complexes were detected in the CL and PL/PLE results evidenced the optical activation of the Eu3+ in the β-Ga2O3 host. The detailed study of the Eu3+ intra-4f shell transitions revealed that at least one active site is created by the Eu implantation/annealing in β-Ga2O3 thin films grown on sapphire. The well controlled implantation and annealing process in β-Ga2O3 thin films pave the way for their use in optoelectronic applications, particularly in efficient red thin film electroluminescent devices.
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