





MEMRISTIVE SWITCHING BEHAVIOR OF SOL–GEL DERIVED Ga<sub>2</sub>O<sub>3</sub> THIN FILMS

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Gallium oxide (Ga<sub>2</sub>O<sub>3</sub>) is an ultrawide-bandgap semiconductor (~4.8–5.0 eV) that has recently gained considerable attention for next-generation nanoelectronic and memory devices owing to its superior breakdown field, chemical durability, and thermal robustness. In this study, Ga<sub>2</sub>O<sub>3</sub> thin films were fabricated through a sol-gel spin-coating route and subsequently annealed at 1000 °C. X-ray diffraction revealed that annealing led to structural evolution from an amorphous state to the stable monoclinic β-Ga<sub>2</sub>O<sub>3</sub> phase. Electrical measurements exhibited reproducible bipolar resistive switching with an ON/OFF resistance ratio exceeding 10<sup>2</sup> and relatively low set/reset voltages. The observed switching is interpreted within the framework of conductive filament formation and rupture, predominantly governed by oxygen vacancy dynamics. The combination of low-cost synthesis, scalable processing, and robust memristive performance highlights sol-gel derived Ga<sub>2</sub>O<sub>3</sub> thin films as strong contenders for future resistive random-access memory (RRAM) architectures and neuromorphic computing technologies.

**Keywords:** Gallium oxide; Sol-gel method; Thin films; Memristor

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## INTRODUCTION

Wide-bandgap (WBG) semiconductors such as SiC, GaN, and Ga<sub>2</sub>O<sub>3</sub> have attracted significant attention due to their potential applications in high-power electronics, high-frequency devices, and deep-ultraviolet optoelectronics [1,2]. Among these, gallium oxide (Ga<sub>2</sub>O<sub>3</sub>) is particularly attractive because of its ultrawide bandgap (~4.8–5.0 eV), exceptionally high breakdown electric field (~8 MV/cm), excellent thermal stability, and chemical robustness [3]. Furthermore, Ga<sub>2</sub>O<sub>3</sub> possesses five polymorphs (α, β, γ, δ, ε), with the monoclinic β-Ga<sub>2</sub>O<sub>3</sub> phase being the most thermodynamically stable, making it highly suitable for device applications [4].

In addition to its role in power and optoelectronic devices, Ga<sub>2</sub>O<sub>3</sub> has recently been investigated for resistive random-access memory (RRAM) devices, where resistive switching (RS) behavior plays a central role [5–7]. RRAM is considered a strong candidate for next-generation non-volatile memory due to its high switching speed, scalability, low power consumption, and compatibility with neuromorphic computing architectures [8–11]. The resistive switching mechanism in oxide memristors is typically associated with the formation and rupture of conductive filaments, primarily governed by the migration of oxygen vacancies.

Various fabrication methods, including pulsed laser deposition (PLD), molecular beam epitaxy (MBE), and RF sputtering, have been employed to grow Ga<sub>2</sub>O<sub>3</sub> thin films [12–14]. Although these techniques produce high-quality films, they require expensive equipment and are not ideal for large-area or low-cost processing. In contrast, the sol-gel spin-coating technique provides a simple, low-cost, and highly scalable approach, making it attractive for industrial applications [15,16]. Nevertheless, systematic studies of memristive switching in sol-gel derived Ga<sub>2</sub>O<sub>3</sub> thin films remain limited, with most prior works focusing primarily on structural and optical characteristics.

Furthermore, research on other oxide-based memristors has demonstrated how doping and defect engineering can introduce new functionalities. For example, Murodov *et al.* recently reported tunable negative differential resistance (NDR) in SnO<sub>2</sub>:Co memristors fabricated on p-Si substrates, showing that controlled nanocluster formation can significantly influence switching behavior [17]. In addition to single-oxide memristors, bilayer and heterojunction systems have also been investigated to enhance switching performance. For instance, our recent study on SnO<sub>2</sub>/ZnO heterojunction thin films demonstrated stable bipolar resistive switching with an ON/OFF ratio above 10<sup>2</sup> and forming-free operation, highlighting the role of interface engineering and oxygen vacancy dynamics in modulating device characteristics [18]. Such works emphasize the importance of material engineering in tailoring memristive responses and inspire further exploration of wide-bandgap oxides, such as Ga<sub>2</sub>O<sub>3</sub>.

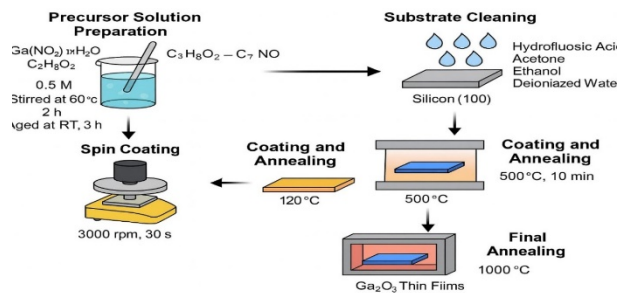
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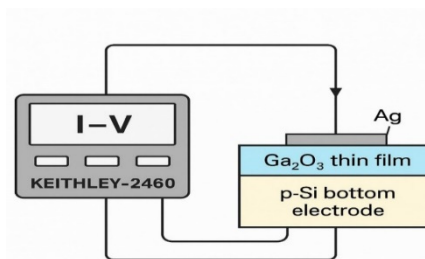
In this context, the present study investigates sol–gel derived  $\text{Ga}_2\text{O}_3$  thin films annealed at  $1000^\circ\text{C}$ . Structural analysis revealed evolution from an amorphous to  $\beta\text{-Ga}_2\text{O}_3$  phase, and electrical measurements confirmed reproducible bipolar resistive switching with a high ON/OFF ratio ( $>10^2$ ) and low set/reset voltages. The switching behavior is interpreted in terms of oxygen vacancy-mediated conductive filament formation. These findings establish sol–gel derived  $\text{Ga}_2\text{O}_3$  thin films as promising candidates for cost-effective, scalable, and high-performance RRAM and neuromorphic computing applications.

## METHODS

Gallium oxide ( $\text{Ga}_2\text{O}_3$ ) thin films were prepared using a sol–gel spin-coating method. The overall preparation process is schematically illustrated in Figure 1, which depicts the main steps of solution preparation, coating, drying, and high-temperature annealing. Gallium nitrate hydrate [ $\text{Ga}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$ ] served as the precursor, dissolved in 2-methoxyethanol ( $\text{C}_3\text{H}_8\text{O}_2$ ), while monoethanolamine (MEA,  $\text{C}_2\text{H}_7\text{NO}$ ) was employed as a stabilizing agent at a 1:1 molar ratio with respect to gallium nitrate. The precursor concentration was adjusted to 0.5 M. The solution was continuously stirred at  $60^\circ\text{C}$  for 2 hours, then aged at room temperature for 36 hours to achieve uniformity and stability. Silicon (100) substrates were thoroughly cleaned in hydrofluoric acid, acetone, ethanol, and deionized water to eliminate native oxides and organic residues. The sol–gel solution was spin-coated onto the substrates at 3000 rpm for 30 seconds. After each deposition, the coated layers were dried at  $120^\circ\text{C}$  for 10 minutes on a hot plate and subsequently pre-annealed in air at  $500^\circ\text{C}$  for 15 minutes. This deposition and treatment sequence was repeated six times, yielding films with thicknesses ranging from 100 to 150 nm. Finally, the films were annealed at  $1000^\circ\text{C}$  for 30 minutes in air to induce crystallization.



**Figure 1.** Schematic of the  $\text{Ga}_2\text{O}_3$  thin films preparation method



**Figure 2.** Electrical measurement scheme for  $\text{Ga}_2\text{O}_3$  films with Keithley-2460

The structural properties of the films were examined using X-ray diffraction (XRD) ( $\text{Cu K}\alpha$  radiation,  $\lambda = 1.5406 \text{ \AA}$ ). The optical absorption spectra were measured with a UV–Vis spectrophotometer in the range of 200–800 nm. For electrical characterization, the measurement setup is shown schematically in Figure 2, where current–voltage (I–V) curves were obtained using a Keithley-2460 source meter in a two-probe configuration with Ag top electrodes on the  $\text{Ga}_2\text{O}_3$  thin films and a p-Si bottom electrode. For electrical measurements, Ag top electrodes ( $\sim 100 \text{ nm}$ ) were deposited onto the  $\text{Ga}_2\text{O}_3$  thin films through thermal evaporation using a shadow mask, forming circular pads with a diameter of  $200 \mu\text{m}$ . The bottom electrode was p-type Si (100) substrate, which served simultaneously as a mechanical support and as the bottom contact. This configuration resulted in a vertical Ag/ $\text{Ga}_2\text{O}_3$ /p-Si memristor structure. The use of Ag top electrodes facilitates the formation of conductive filaments during resistive switching, while the p-Si bottom electrode provides good ohmic contact with the  $\text{Ga}_2\text{O}_3$  layer.

## RESULTS

**Structural properties:** XRD analysis revealed that the as-prepared  $\text{Ga}_2\text{O}_3$  thin films were predominantly amorphous, with only substrate-related diffraction peaks corresponding to Si (100) being observed. This indicates that the deposited layers did not form a well-ordered crystalline lattice in the as-grown state. However, after thermal annealing at  $1000^\circ\text{C}$ , distinct diffraction peaks assigned to the  $\beta\text{-Ga}_2\text{O}_3$  phase appeared, confirming crystallization into the thermodynamically stable monoclinic structure [19,20].

The transition from the amorphous state to the crystalline  $\beta$ -phase demonstrates that high-temperature annealing plays a crucial role in improving the structural ordering of sol–gel derived  $\text{Ga}_2\text{O}_3$  films. This observation is consistent with previous reports, which have shown that metastable phases of  $\text{Ga}_2\text{O}_3$  ( $\alpha$ ,  $\gamma$ ,  $\delta$ ,  $\epsilon$ ) tend to transform into the stable  $\beta$ -phase upon sufficient thermal treatment. Thus, annealing at  $1000^\circ\text{C}$  is confirmed to be effective in enhancing crystallinity and stabilizing the  $\beta\text{-Ga}_2\text{O}_3$  phase in sol–gel processed thin films (Figure 3).

**Optical properties:** The optical absorption spectra of the  $\text{Ga}_2\text{O}_3$  thin films showed a pronounced absorption edge, which sharpened after annealing at  $1000^\circ\text{C}$ , indicating improved crystallinity and a reduction in defect states. The optical bandgap was estimated using Tauc plots derived from the absorption data. The calculated bandgap for the annealed films was approximately 4.9 eV, which is in good agreement with reported values for  $\beta\text{-Ga}_2\text{O}_3$  [21–23]. The corresponding Tauc plot for the annealed films is shown in Figure 4.

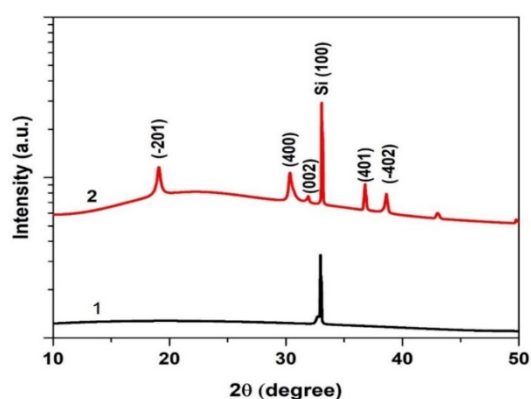


Figure 3. XRD patterns of sol–gel derived  $\text{Ga}_2\text{O}_3$  thin films

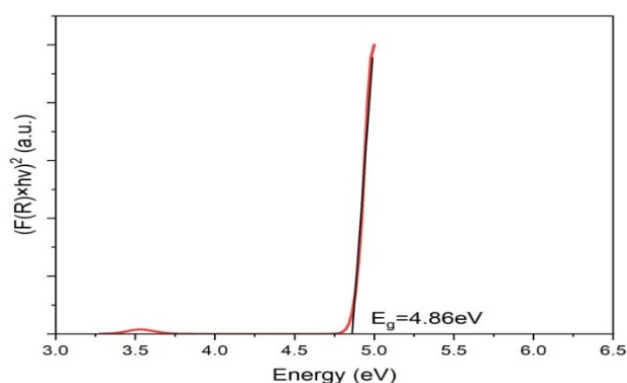


Figure 4. Tauc plot of  $\text{Ga}_2\text{O}_3$  thin films annealed at  $1000\text{ }^\circ\text{C}$ , indicating an optical bandgap of  $\sim 4.86\text{ eV}$

A noticeable blue shift in the absorption edge was observed as the annealing temperature increased. This shift can be attributed to the improved structural ordering of the films, which reduces sub-bandgap defect states, thereby widening the effective bandgap. Such behavior is commonly observed in oxide thin films and is typically attributed to the reduction of localized defect levels and the improvement of stoichiometry upon thermal treatment.

These results confirm that high-temperature annealing not only stabilizes the  $\beta\text{-Ga}_2\text{O}_3$  phase but also enhances its optical quality, making sol–gel–derived  $\text{Ga}_2\text{O}_3$  thin films suitable for optoelectronic and memory device applications that require a wide bandgap and high thermal stability.

**Electrical properties:** The current–voltage ( $I$ – $V$ ) characteristics of the  $\text{Ga}_2\text{O}_3$  thin films demonstrated reproducible bipolar resistive switching behavior. During the voltage sweep, the devices switched from a high-resistance state (HRS) to a low-resistance state (LRS) at a relatively low set voltage, and returned to the HRS at the reset voltage. The observed ON/OFF resistance ratio exceeded  $10^2$ , ensuring a clear distinction between the two resistance states and making the films suitable for non-volatile memory applications (Figure 5). The fabricated device exhibited stable bipolar resistive switching in the  $\text{Ag}/\text{Ga}_2\text{O}_3/\text{p-Si}$  configuration. The Ag top electrode played a crucial role in filament formation, as silver cations can drift under bias, assisting the creation of localized conductive paths in addition to oxygen vacancy migration. This hybrid filamentary mechanism contributed to the low set/reset voltages and stable endurance observed in our device.

Logarithmic  $I$ – $V$  plots further confirmed the presence of well-defined HRS and LRS states across an extended voltage range (Figure 6). The resistive switching mechanism is attributed to the formation and rupture of conductive filaments, predominantly governed by oxygen vacancy migration within the  $\text{Ga}_2\text{O}_3$  layer. In the SET process, oxygen vacancies drift under an electric field, forming localized conductive paths that reduce the resistance. Conversely, in the RESET process, these conductive filaments rupture, restoring the device to its high-resistance state.

These findings are consistent with previously reported  $\text{Ga}_2\text{O}_3$ -based memristors fabricated using physical vapor deposition techniques, such as RF sputtering and pulsed laser deposition [5–8]. However, the present results highlight that the sol–gel approach provides a cost-effective, solution-processable, and scalable alternative, while still achieving competitive switching performance parameters.

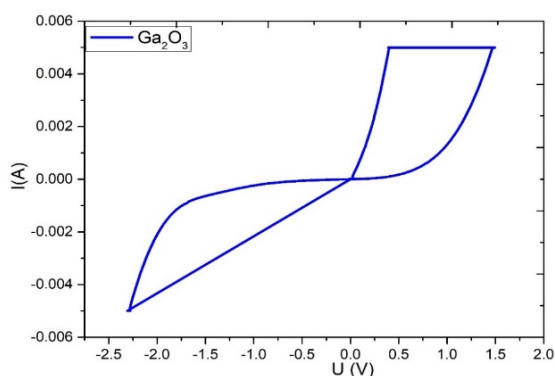


Figure 5. Current–voltage ( $I$ – $V$ ) characteristics of  $\text{Ga}_2\text{O}_3$  thin film demonstrating memristive switching behavior

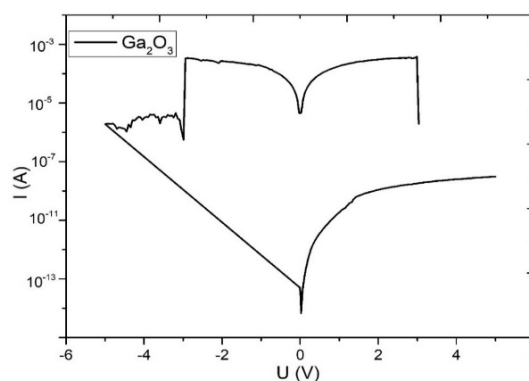
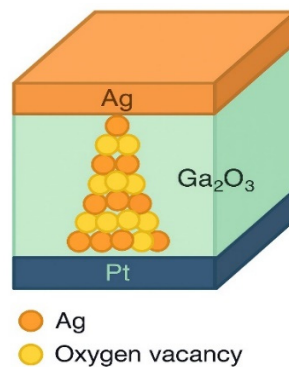


Figure 6. Log–scale  $I$ – $V$  characteristics of  $\text{Ga}_2\text{O}_3$  thin film showing high–resistance and low–resistance states over extended voltage range

The resistive switching behavior in  $\text{Ag}/\text{Ga}_2\text{O}_3$  memristors can be explained by the formation and rupture of conductive filaments, mediated by both oxygen vacancies and Ag cations (Figure 7). Under a positive bias, oxygen vacancies migrate and Ag ions drift from the top electrode into the  $\text{Ga}_2\text{O}_3$  layer, combining to form a localized filamentary

path that reduces the device resistance (SET process). When the polarity is reversed, Joule heating and electrochemical dissolution lead to the rupture of these filaments, restoring the high-resistance state (RESET process). This filamentary model is consistent with the observed bipolar switching and stable ON/OFF ratio in our devices.



**Figure 7.** Schematic of filament formation and rupture in Ag/Ga<sub>2</sub>O<sub>3</sub>/Pt memristor (orange: Ag ions, yellow: oxygen vacancies)

### CONCLUSION

In this work, Ga<sub>2</sub>O<sub>3</sub> thin films were successfully synthesized by a low-cost sol-gel spin-coating technique followed by annealing at 1000°C. Structural analysis confirmed the phase transformation from an amorphous state to the thermodynamically stable β-Ga<sub>2</sub>O<sub>3</sub> phase. Optical characterization revealed a sharp absorption edge with an estimated bandgap of ~4.9 eV, accompanied by a blue shift that can be attributed to improved crystallinity and reduced defect states. Electrical measurements demonstrated reliable bipolar resistive switching with a high ON/OFF resistance ratio (>10<sup>2</sup>) and low set/reset voltages. The switching mechanism was explained by the formation and rupture of oxygen-vacancy-mediated conductive filaments.

These results highlight the potential of sol-gel derived Ga<sub>2</sub>O<sub>3</sub> thin films as scalable, cost-effective, and high-performance candidates for resistive random-access memory (RRAM) and neuromorphic computing applications. Furthermore, this study demonstrates that solution-processed Ga<sub>2</sub>O<sub>3</sub> can achieve memristive characteristics comparable to those obtained by expensive physical vapor deposition techniques, thus opening new avenues for the development of next-generation memory technologies. Unlike many previous works that primarily emphasized Ga<sub>2</sub>O<sub>3</sub> film growth, our study highlights the memristive behavior of a complete device structure (Ag/Ga<sub>2</sub>O<sub>3</sub>/p-Si). The explicit consideration of contact engineering is essential, since both the choice of top electrode (Ag) and bottom electrode (p-Si) strongly influence resistive switching performance.

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**МЕМРИСТИВНА ПОВЕДІНКА ПЕРЕМИКАННЯ ТОНКИХ ПЛІВОК Ga<sub>2</sub>O<sub>3</sub>,  
ОТРИМАНИХ ЗОЛЬ-ГЕЛЬ МЕТОДОМ**

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Оксид галію (Ga<sub>2</sub>O<sub>3</sub>) – це напівпровідник з надширокою забороненою зоною (~4,8–5,0 eV), який нещодавно привернув значну увагу для наноелектронних пристроїв та пристроїв пам'яті наступного покоління завдяки своєму чудовому полю пробою, хімічній стійкості та термостійкості. У цьому дослідженні тонкі плівки Ga<sub>2</sub>O<sub>3</sub> були виготовлені методом золь-гель спінінгу та потім відпалені при 1000°C. Рентгенівська дифракція виявила структурну еволюцію від аморфного стану до стабільної моноклінної фази β-Ga<sub>2</sub>O<sub>3</sub> після відпалу. Електричні вимірювання показали відтворюване біполярне резистивне перемикання з коефіцієнтом опору ввімкнення/вимкнення, що перевищує 10<sup>2</sup>, та відносно низькими напругами встановлення/скидання. Спостережуване перемикання інтерпретується в рамках формування та розриву провідних ниток, переважно керованих динамікою вакансій кисню. Поєднання недорогого синтезу, масштабованої обробки та надійної мемристивної продуктивності підкреслює тонкі плівки Ga<sub>2</sub>O<sub>3</sub>, отримані методом золь-гель, як сильних претендентів на майбутні архітектури резистивної пам'яті з довільним доступом (RRAM) та нейроморфні обчислювальні технології.

**Ключові слова:** оксид галію; золь-гель метод; тонкі плівки; мемристор