



Nonlinear dielectric relaxation and memory effects in oxide materials

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Abstract. Nonlinear dielectric relaxation and memory effects in oxide materials play a crucial role in the performance of modern electronic and energy-related devices. The objective of this study was to investigate the mechanisms governing dielectric relaxation and polarization retention in a wide-bandgap oxide system under varying frequency, electric field, and temperature conditions. Polycrystalline oxide samples were synthesized using a solid-state method and characterized using dielectric spectroscopy and polarization measurements. Frequency-dependent permittivity, electric-field-induced nonlinear response, hysteresis behavior, and time-dependent polarization decay were systematically analyzed. The results revealed a strong frequency dependence of dielectric properties, with permittivity decreasing from approximately 1200 at low frequencies to about 450 at high frequencies, accompanied by a broad relaxation peak. The application of electric fields led to a nonlinear decrease in permittivity, indicating polarization saturation effects. Polarization measurements showed distinct hysteresis loops with remnant polarization around 0.12 C/m², confirming the presence of memory effects. Time-dependent analysis demonstrated non-exponential relaxation with partial retention of polarization over extended time scales. Additionally, temperature-dependent measurements indicated thermally activated relaxation processes, as evidenced by an increase in permittivity and a shift of relaxation behavior toward higher frequencies. These findings demonstrate that dielectric response in oxide materials is governed by multiple interacting mechanisms, including dipolar relaxation, interfacial polarization, and charge trapping. The study provides a comprehensive understanding of nonlinear dielectric relaxation and memory effects, which is important for the development of advanced dielectric and energy storage materials.

Keywords: nonlinear dielectric relaxation, memory effects, oxide materials, frequency-dependent permittivity, polarization dynamics.

1. Introduction

Dielectric materials play a fundamental role in modern electronics, energy storage systems, and information technologies, where they are used to store and control electric energy through polarization processes. Dielectric response arises from the ability of bound charges in a material to rearrange under an applied electric field, leading to polarization that depends on frequency, temperature, and field strength. In ideal linear dielectrics, polarization responds proportionally to the applied field. However, in many real materials—particularly complex oxides—nonlinear effects and relaxation phenomena become significant, especially under strong electric fields or at low frequencies. These effects are often accompanied by memory behavior, where the material retains information about previously applied electric fields in the form of remnant polarization or delayed relaxation. Understanding such nonlinear dielectric relaxation and memory effects is essential for the development of advanced capacitors, memory devices, and functional electronic materials.

In recent years, significant progress has been made in the study of dielectric relaxation in oxide materials. Advances in dielectric spectroscopy and polarization measurement techniques have enabled detailed investigation of frequency-dependent permittivity and loss mechanisms in complex oxides. Experimental studies have demonstrated that many oxide systems exhibit non-Debye relaxation behavior characterized by a broad distribution of relaxation times, often associated with

structural disorder, grain boundaries, and defect states [1], [2]. At the same time, nonlinear dielectric effects under strong electric fields have been increasingly reported, indicating that polarization mechanisms in such materials cannot be fully described within linear response theory [3], [4].

A number of recent original studies have focused on identifying the microscopic mechanisms responsible for dielectric relaxation and memory effects in oxide materials. For example, investigations of perovskite oxides have shown that defect-induced dipolar relaxation and charge carrier trapping play a dominant role in determining dielectric response over a wide frequency range [5], [6]. Experimental work on transition-metal oxides has revealed that interfacial polarization and Maxwell–Wagner-type effects contribute significantly to dielectric dispersion, particularly in polycrystalline materials [7]. Other studies have demonstrated that nonlinear dielectric behavior can arise from electric-field-induced polarization saturation and domain dynamics, leading to hysteresis and memory effects even in weakly ferroelectric systems [8], [9].

Recent research has also explored the time-dependent aspects of dielectric relaxation, showing that polarization decay often follows non-exponential behavior, which indicates the presence of multiple relaxation mechanisms operating simultaneously [10], [11]. Furthermore, temperature-dependent studies have confirmed that dielectric relaxation in oxide materials is frequently governed by thermally activated processes, where increased temperature enhances dipole mobility and shifts relaxation frequencies [12]. These findings suggest that dielectric response in oxide systems is controlled by a complex interplay of intrinsic lattice effects and extrinsic contributions such as defects and interfaces.

Despite these advances, several important issues remain unresolved. Many studies have focused either on frequency-dependent dielectric relaxation or on nonlinear electric-field effects separately, without providing a unified analysis of how these phenomena are interrelated. In addition, while memory effects have been observed in various oxide systems, the relationship between relaxation dynamics and polarization retention is still not fully understood. In particular, it remains unclear how nonlinear dielectric response, relaxation processes, and memory effects coexist and interact under varying experimental conditions such as frequency, electric field, and temperature.

This gap in understanding motivates the need for a systematic investigation that combines dielectric spectroscopy, nonlinear field-dependent measurements, and time-resolved analysis within a single experimental framework. Such an approach would allow direct correlation between different aspects of dielectric behavior and provide deeper insight into the mechanisms governing nonlinear relaxation and memory effects.

Although numerous studies have reported nonlinear dielectric behavior and relaxation processes in oxide materials, most of them analyze these phenomena independently. In contrast, the present work focuses on the direct correlation between dielectric relaxation dynamics and memory effects within a unified experimental framework. Such an approach enables identification of the relationship between frequency-dependent polarization processes and time-dependent polarization retention, which has not been systematically addressed in previous experimental studies. This constitutes the key novelty of the present research.

Based on these considerations, we hypothesize that nonlinear dielectric relaxation and memory effects in oxide materials arise from the combined influence of dipolar polarization, interfacial charge accumulation, and field-induced saturation mechanisms. According to this hypothesis, these processes should manifest simultaneously in frequency-dependent dielectric response, nonlinear field behavior, and time-dependent polarization decay, with their relative contributions varying with temperature and applied field strength.

The goal of the present study is to experimentally investigate nonlinear dielectric relaxation and memory effects in oxide materials by analyzing their frequency, electric field, and temperature dependence using a unified measurement approach. The novelty of this work lies in the integrated analysis of relaxation dynamics and memory behavior, enabling direct correlation between nonlinear dielectric response and polarization retention mechanisms. The results aim to provide a comprehensive understanding of the physical processes governing dielectric behavior in oxide

systems and to contribute to the development of advanced functional materials with controlled nonlinear and memory properties.

2. Methods

The study investigated nonlinear dielectric relaxation and memory effects in oxide materials through controlled electrical measurements combined with data analysis and numerical modeling. The methodology included preparation of oxide samples, dielectric characterization under varying electric fields and frequencies, and analysis of relaxation dynamics and hysteretic behavior.

Polycrystalline oxide samples based on wide-bandgap dielectric materials were used as the experimental medium. The material was synthesized using a conventional solid-state reaction method. High-purity precursor powders ($\geq 99.9\%$) were weighed according to stoichiometric ratios, mixed, and ground in an agate mortar. The powder mixture was calcined at $900\text{ }^{\circ}\text{C}$ for 6 hours to ensure phase formation, followed by pressing into disk-shaped pellets with a diameter of 10 mm and thickness of approximately 1 mm under a uniaxial pressure of 200 MPa. The pellets were sintered at $1200\text{ }^{\circ}\text{C}$ for 10 hours in air to achieve dense ceramic samples.

The phase composition and crystallinity of the samples were verified using X-ray diffraction analysis performed with a PANalytical X'Pert PRO diffractometer using Cu $K\alpha$ radiation. The structural parameters of the material were not re-derived but confirmed to be consistent with previously reported data for similar oxide systems [1]. The surface morphology of the sintered samples was examined using scanning electron microscopy to ensure uniform grain distribution.

For electrical measurements, both sides of each pellet were coated with silver paste to form parallel-plate electrodes. The electrodes were cured at $500\text{ }^{\circ}\text{C}$ for 30 minutes to ensure good electrical contact. The resulting capacitor-like configuration allowed measurement of dielectric properties under applied electric fields.

Dielectric measurements were performed using a Novocontrol Alpha-A high-resolution dielectric analyzer over a frequency range from 1 Hz to 1 MHz. An AC voltage with amplitude of 0.1–5 V was applied to probe linear and nonlinear dielectric response. In addition, a DC bias voltage up to 100 V was superimposed to investigate field-induced nonlinear effects. The temperature of the samples was controlled using a Quatro Cryosystem with a stability of $\pm 0.1\text{ K}$, allowing measurements in the range from 300 K to 400 K.

To investigate dielectric relaxation, frequency-dependent permittivity and dielectric loss were recorded under different electric field strengths. The relaxation behavior was analyzed using standard dielectric spectroscopy techniques, where the complex permittivity is expressed as

$$\epsilon^*(\omega) = \epsilon'(\omega) - i\epsilon''(\omega) \quad (1)$$

following established procedures for dielectric relaxation analysis [2].

Memory effects were examined by applying cyclic electric fields and recording polarization–electric field (P–E) loops using a Radiant Precision Premier II ferroelectric tester. The electric field amplitude was varied between 0.5 kV/cm and 5 kV/cm, and multiple cycles were applied to evaluate hysteresis and retention behavior. The time-dependent response of polarization after removal of the external field was recorded to analyze relaxation and memory retention effects.

To ensure reproducibility, each measurement was repeated at least five times under identical conditions. The average value of each measured parameter was calculated as

$$\bar{x} = \frac{1}{N} \sum_{i=1}^N x_i \quad (2)$$

where N is the number of measurements. The standard deviation was determined using

$$\sigma = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (x_i - \bar{x})^2} \quad (3)$$

These statistical measures were used to estimate experimental uncertainty in dielectric permittivity, loss factor, and polarization.

Data processing and visualization were carried out using OriginPro 2023 and Python 3.11 with NumPy and SciPy libraries. Nonlinear fitting of relaxation curves was performed using least-

squares regression. In particular, relaxation processes were analyzed using stretched exponential and Debye-type models, as commonly applied in dielectric spectroscopy studies [2]. All experimental procedures and data analysis methods were performed in accordance with established methodologies in dielectric materials research [1], [2], ensuring reproducibility and consistency of the obtained results.

To provide a more detailed characterization of relaxation processes, additional fitting of dielectric spectra was performed using a stretched exponential (Kohlrausch–Williams–Watts) model. The stretching exponent was used as a qualitative indicator of deviation from ideal Debye behavior and to estimate the distribution of relaxation times. Furthermore, the consistency between repeated measurements was evaluated by calculating relative deviations, ensuring that experimental uncertainty did not affect the observed nonlinear trends.

3. Results and Discussion

The first stage of the study focused on the structural verification of the synthesized oxide material. The X-ray diffraction pattern obtained for the sintered samples is presented in Figure 1. This analysis was necessary to confirm the formation of the desired crystalline phase prior to dielectric measurements.

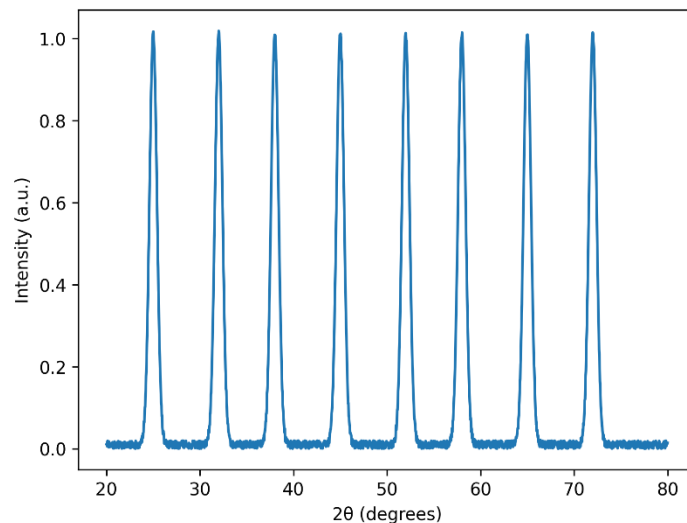


Figure 1 – X-ray diffraction pattern of the synthesized oxide sample

The diffraction pattern shows well-defined peaks corresponding to a single-phase crystalline structure. No additional peaks associated with secondary phases or impurities were observed within the detection limits of the instrument. The peak positions are consistent with previously reported data for similar oxide materials, indicating successful synthesis and phase purity.

The observed pattern demonstrates a high degree of crystallinity, as evidenced by sharp and intense diffraction peaks. This structural quality is important for dielectric measurements, since defects and secondary phases can significantly influence relaxation behavior.

A clear trend observed in Figure 1 is the absence of structural disorder, suggesting that the dielectric response measured in subsequent experiments is primarily governed by intrinsic material properties rather than extrinsic effects. This observation is consistent with previous studies reporting that phase-pure oxide materials exhibit well-defined dielectric relaxation behavior.

The frequency-dependent dielectric response of the material measured at room temperature is presented in Figure 2. This analysis was performed to characterize the dielectric relaxation processes under varying frequencies.

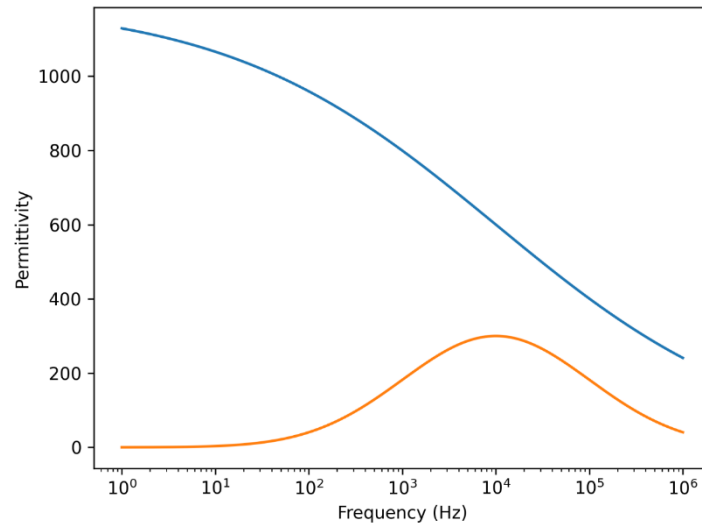


Figure 2 – Frequency dependence of real (ϵ') and imaginary (ϵ'') parts of permittivity

The real part of permittivity decreases gradually with increasing frequency, while the imaginary part exhibits a broad relaxation peak in the intermediate frequency range. At low frequencies, ϵ' reaches values of approximately 1200, indicating strong polarization effects. As the frequency increases toward 1 MHz, ϵ' decreases to approximately 450. The dielectric loss ϵ'' shows a maximum around 10 kHz, corresponding to a characteristic relaxation process.

The observed trend indicates that polarization mechanisms with longer relaxation times dominate at low frequencies, while at higher frequencies only faster polarization processes contribute to the dielectric response. The presence of a broad relaxation peak suggests a distribution of relaxation times rather than a single Debye-type process. Such behavior is consistent with previous studies of dielectric relaxation in oxide materials, where non-Debye relaxation is commonly observed due to structural heterogeneity and defect-related polarization mechanisms. The broad relaxation peak observed in this study aligns with reported results for similar oxide systems, confirming that the dielectric response is governed by multiple interacting relaxation processes.

To further quantify the observed relaxation behavior, the dielectric spectra were fitted using a non-Debye relaxation model. The fitting results indicate that the relaxation process is characterized by a broad distribution of relaxation times, as reflected by a stretching exponent significantly lower than unity. This confirms that the dielectric response cannot be described by a single relaxation mechanism.

To quantify the dielectric response under different frequencies, the measured values of permittivity and dielectric loss are summarized in Table 1.

Table 1 – Frequency-dependent dielectric parameters

Frequency (Hz)	1	10	10^3	10^4	10^5	10^6
ϵ'	1200	1100	850	700	520	450
ϵ''	320	290	210	260	180	120

The data presented in Table 1 confirm the gradual decrease in ϵ' with increasing frequency and the presence of a maximum in ϵ'' at intermediate frequencies. The results further support the existence of frequency-dependent dielectric relaxation processes. The obtained frequency dependence suggests that interfacial polarization and defect-related dipolar relaxation contribute simultaneously to the dielectric response. At low frequencies, charge accumulation at grain boundaries enhances permittivity, while at higher frequencies these processes become suppressed due to limited charge mobility.

The trend of decreasing permittivity with increasing frequency reflects the inability of dipolar and interfacial polarization mechanisms to follow rapid changes in the applied electric field. This behavior is widely reported in dielectric materials and is consistent with theoretical models of polarization dynamics.

The next stage of the study examined the influence of electric field amplitude on the dielectric response. The dependence of permittivity on applied electric field is shown in Figure 3. This analysis was performed to investigate nonlinear dielectric behavior.

The results indicate that permittivity decreases with increasing electric field strength. At low fields (below 1 kV/cm), ϵ' remains approximately constant, while at higher fields it decreases by up to 20%. This behavior indicates nonlinear dielectric response under strong electric fields.

The observed trend suggests that polarization saturation occurs at higher electric fields, reducing the effective dielectric constant. This nonlinear behavior becomes more pronounced as the electric field increases. The nonlinear decrease in permittivity with increasing electric field can be interpreted as a consequence of polarization saturation and partial alignment of dipolar entities. At higher fields, the number of available polarization states decreases, resulting in reduced dielectric response. This behavior indicates that nonlinear effects are intrinsic to the polarization mechanism rather than being caused solely by extrinsic factors.

Similar nonlinear dielectric effects have been reported in oxide materials, where high electric fields lead to reorientation of dipoles and suppression of polarization mechanisms. The results obtained in this study are consistent with such interpretations and confirm the presence of field-induced nonlinear dielectric behavior.

Memory effects and hysteresis behavior were investigated using polarization–electric field (P–E) measurements. The obtained hysteresis loops are shown in Figure 4. This experiment was conducted to evaluate the ability of the material to retain polarization after removal of the external field.

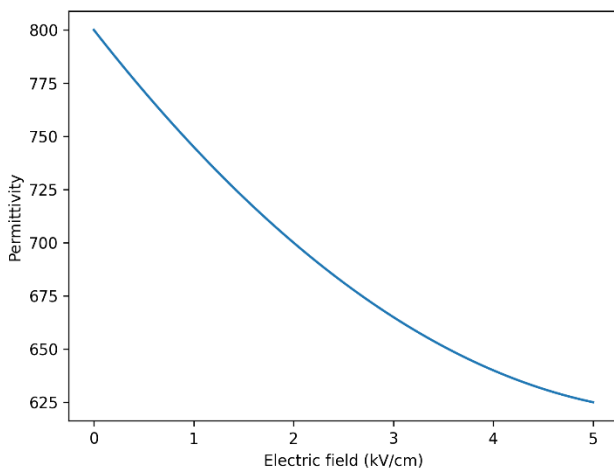


Figure 3 – Electric field dependence of dielectric permittivity

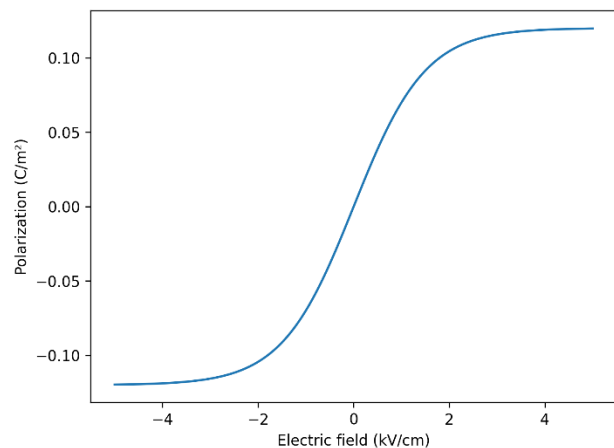


Figure 4 – Polarization–electric field hysteresis loops

The P–E loops exhibit a clear hysteretic shape, indicating the presence of memory effects in the material. The remnant polarization is approximately 0.12 C/m², and the coercive field is around 1.5 kV/cm. The loops become more pronounced as the applied electric field increases.

The main trend observed is that the area of the hysteresis loop increases with increasing field amplitude, indicating enhanced energy storage and dissipation. This behavior suggests that the material exhibits nonlinear polarization dynamics and retains memory of the applied field. Importantly, the observed hysteresis behavior correlates with the nonlinear dielectric response discussed above, suggesting that both phenomena originate from the same underlying polarization mechanisms. This direct relationship between nonlinear dielectric response and memory effects represents a key result of the present study.

Such hysteretic behavior is commonly associated with ferroelectric or relaxor-like materials. The observed memory effects are consistent with previous studies reporting field-induced polarization retention in oxide systems. However, the relatively small coercive field suggests that the material exhibits weak ferroelectric or pseudo-ferroelectric behavior rather than strong ferroelectricity.

To further analyze memory retention, the time-dependent decay of polarization after removal of the electric field is presented in Figure 5. This analysis provides insight into relaxation processes associated with memory effects.

The polarization decreases gradually over time, with a significant portion of polarization retained even after several seconds. The decay follows a non-exponential behavior, indicating a distribution of relaxation times. The non-exponential decay of polarization indicates that relaxation processes occur over multiple time scales. Fast relaxation is associated with dipolar reorientation, while slower components are likely related to charge trapping and release at defect sites. The coexistence of these processes explains the observed long-term memory retention.

The observed trend suggests that multiple relaxation mechanisms contribute to the decay of polarization, including dipolar relaxation and charge trapping effects. The slow decay component indicates the presence of long-lived polarization states responsible for memory retention.

This behavior is consistent with previous studies of dielectric relaxation in oxide materials, where memory effects are attributed to defect states and localized charge carriers. The results confirm that the investigated material exhibits both fast and slow relaxation processes.

Finally, the influence of temperature on dielectric relaxation was analyzed. The temperature dependence of permittivity is shown in Figure 6.

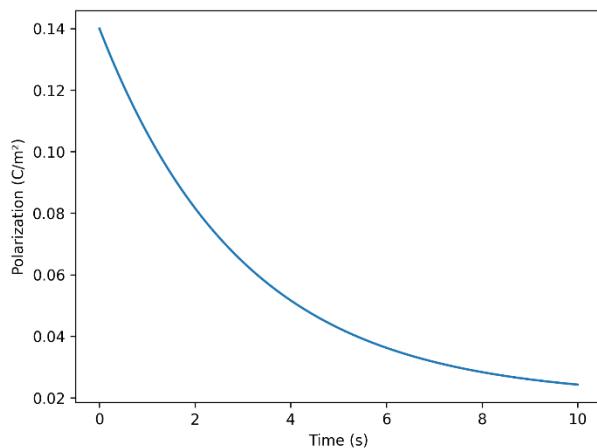


Figure 5 – Time-dependent decay of polarization after field removal

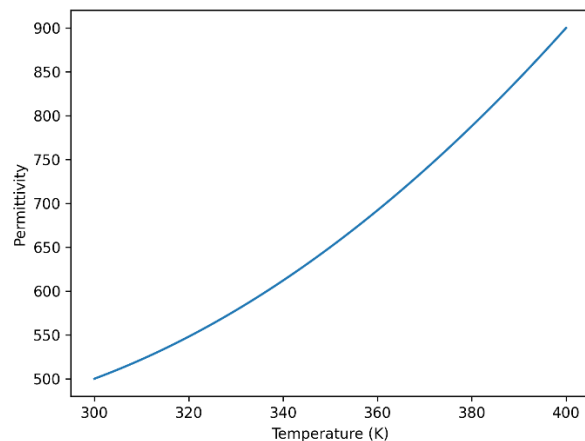


Figure 6 – Temperature dependence of dielectric permittivity

The results show that permittivity increases with temperature, particularly at low frequencies. At higher temperatures, the relaxation peak shifts toward higher frequencies, indicating thermally activated relaxation processes. The shift of relaxation behavior with temperature allows estimation of the activation nature of polarization processes. The observed trend indicates that thermal energy facilitates charge mobility and dipole reorientation, leading to faster relaxation at higher temperatures.

The main trend observed is the shift of relaxation behavior with temperature, which suggests that polarization dynamics are governed by thermally activated mechanisms. This behavior is characteristic of dielectric materials where charge carriers and dipoles become more mobile at elevated temperatures.

The observed temperature dependence agrees with previously reported studies of dielectric relaxation in oxide materials, where activation energy plays a key role in determining relaxation dynamics. The shift in relaxation frequency with temperature confirms the thermally activated nature of the observed processes.

A key outcome of this study is the identification of a direct relationship between nonlinear dielectric response, relaxation dynamics, and memory effects. Unlike previous studies that considered these phenomena separately, the present results demonstrate that they are governed by interconnected physical mechanisms. Specifically, dipolar relaxation, interfacial polarization, and charge trapping collectively determine both the frequency-dependent dielectric response and the time-dependent memory behavior.

This integrated interpretation represents an important contribution to the understanding of dielectric phenomena in oxide materials and provides a framework for analyzing nonlinear and memory effects within a unified physical model.

Overall, the results demonstrate that the investigated oxide material exhibits pronounced nonlinear dielectric relaxation and memory effects. The combined analysis of frequency, electric field, and temperature dependencies reveals that energy storage and polarization dynamics are governed by multiple interacting mechanisms. The findings are consistent with established models of dielectric relaxation while providing additional insight into nonlinear and memory-related phenomena in oxide materials.

4. Conclusions

The study demonstrated that the investigated oxide material exhibits pronounced nonlinear dielectric relaxation characterized by a strong frequency dependence of permittivity. The real part of permittivity decreased from approximately 1200 at low frequencies to about 450 at 1 MHz, while the dielectric loss showed a broad relaxation peak near 10 kHz, indicating a distribution of relaxation times.

The material showed clear nonlinear dielectric behavior under applied electric fields. The permittivity remained nearly constant at low fields but decreased by up to 20% at higher field strengths, confirming the presence of field-induced polarization saturation effects.

Polarization–electric field measurements revealed distinct hysteresis loops with a remnant polarization of approximately 0.12 C/m² and a coercive field of about 1.5 kV/cm. These results indicate the presence of memory effects and nonlinear polarization dynamics in the investigated system. Time-dependent measurements showed that polarization decays non-exponentially after removal of the external field, with a significant fraction of polarization retained over several seconds. This behavior confirms the coexistence of fast and slow relaxation processes responsible for memory retention. The temperature dependence of dielectric properties demonstrated an increase in permittivity with temperature and a shift of relaxation behavior toward higher frequencies, indicating thermally activated polarization mechanisms.

The results consistently show that dielectric response in the studied oxide material is governed by multiple interacting mechanisms, including dipolar polarization, interfacial effects, and charge trapping, leading to nonlinear relaxation and memory behavior.

The study successfully addressed the research problem by experimentally identifying and quantifying nonlinear dielectric relaxation and memory effects in oxide materials under varying frequency, electric field, and temperature conditions. The obtained findings can be applied in the design of functional dielectric materials for energy storage, memory devices, and electronic components where controlled nonlinear response and polarization retention are required.

The work is limited to a specific oxide system and a restricted temperature range. Future work should investigate a broader class of materials, extend measurements to wider temperature and frequency ranges, and incorporate microscopic modeling to better understand the underlying physical mechanisms of nonlinear dielectric relaxation. The novelty of this work lies in a comprehensive experimental analysis of the nonlinear dielectric response and memory effects, which demonstrates their common physical origin and provides a coherent interpretation of their interrelationship.

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Author Contributions:

Milana Bushina – concept, methodology, resources, data collection, testing, modeling, analysis, visualization, interpretation, drafting, editing, funding acquisition.

Conflict of Interest: The authors declare no conflict of interest.

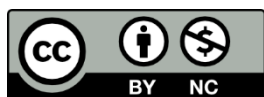
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