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THE EFFECT OF ELECTRIC FIELD SWITCHING MODES ON POLARIZATION PROPERTIES OF THE LEAD ZIRCONATE-TITANATE-BASED EPITAXIAL HETEROSTRUCTURES

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Abstract. In order to extend the serviceability of ferroelectric devices, this paper proposes an experimental approach (for the first time) to clearing up the mechanism of polarization fatigue development in a lead zirconate titanate-based ferroelectric thin film. For this purpose, the influence of the recording pulse sequence configuration on the polarization of a selected object has been analyzed. The coercive field (CF) and remanent polarization of ferroelectric hysteresis loops were compared under two electric field application modes (they differed in the order of positive and negative pulses). The change in CF components was found to be insignificant during cyclic switching, whereas the difference between CF values reached 10% or more after pauses between the measurements. Experiments showed that the polarity of the final pulse of the switching signal set favorable conditions for stabilizing domains in the film whose dipole moment was aligned with the pulse polarity, and after resuming the measurements, a field of higher intensity was required to reverse the polarization.

Keywords: ferroelectric, epitaxial thin film, polarization fatigue, remanent polarization

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ВЛИЯНИЕ РЕЖИМОВ ПЕРЕКЛЮЧЕНИЯ ЭЛЕКТРИЧЕСКОГО ПОЛЯ НА ПОЛЯРИЗАЦИОННЫЕ СВОЙСТВА ЭПИТАКСИАЛЬНЫХ ГЕТЕРОСТРУКТУР НА ОСНОВЕ ЦИРКОНАТА-ТИТАНАТА СВИНЦА

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Аннотация. С целью продления работоспособности устройств на основе сегнетоэлектриков, в работе впервые предложен экспериментальный подход к выяснению механизма развития поляризационной усталости в тонкой сегнетоэлектрической пленке на основе цирконата-титаната свинца. Для этого проанализировано влияние

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конфигурации последовательности записывающих импульсов на поляризацию выбранного объекта. При двух режимах приложения электрического поля (они различались порядком следования положительных и отрицательных импульсов) было проведено сравнение значений коэрцитивного поля (КП) и остаточной поляризации сегнетоэлектрических петель гистерезиса. Установлено, что в моменты циклических переключений изменение компонент КП незначительно, тогда как после пауз между измерениями разница между значениями КП достигала %10 и более. Эксперименты показали, что полярность завершающего импульса переключающего сигнала задает благоприятные условия для стабилизации в пленке доменов, дипольный момент которых сонаправлен с полярностью импульса, и после возобновления измерений требуется поле большей величины для изменения поляризации на противоположную.

Ключевые слова: сегнетоэлектрик, эпитаксиальная гетероструктура, поляризационная усталость, остаточная поляризация, цирконат-титанат свинца

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Introduction

Thin-film ferroelectric capacitors attract considerable interest due to their wide range of applications. One such application is ferroelectric random access non-volatile memory (FeRAM), combining the high speed of random-access memory with the ability to retain data when power is switched off [1, 2]. In such devices, information is written by applying pulses of a certain polarity: a negative pulse sets the memory cell to logic “0”, while a positive pulse sets it to logic “1”. The key advantage of these devices lies in their material: ferroelectrics provide fast polarization switching and non-volatile data storage without a constant power supply.

However, one of the main obstacles to commercialization of ferroelectric devices is polarization fatigue, i.e., a decrease in remanent polarization after a large number of write cycles. As a result of fatigue, the states corresponding to logic “0” and “1” become less distinguishable.

This phenomenon has been the focus of much research, and several possible causes of polarization fatigue have been proposed. According to [3], the main mechanisms involve inhibition of domain nucleation and freezing of domain walls in the ferroelectric crystal. Beyond identifying these mechanisms, many studies have analyzed such contributing factors as capacitor and electrode materials [4–6], operating temperature [7], size effects [8], applied field parameters [9], and others. Nevertheless, many fundamental and applied questions remain unresolved.

The goal of this work is to find an effective approach to extending the service life of ferroelectric devices that would additionally provide insight into the mechanism of polarization fatigue.

Exploratory approach to the problem

The proposed approach to identifying the mechanism of polarization fatigue in the object of study, a thin ferroelectric PZT film, is discussed here for the first time. Our objective was to experimentally assess how the configuration of the writing pulse sequence affects the polarization properties of this film. To this end, a voltage was applied to the film in each write cycle as a sequence consisting of two pairs of triangular pulses with positive and negative polarity. We needed to determine how the coercive field and remanent polarization evolve over time under two different pulse sequence configurations.

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In the first case, a pair of positive pulses was applied to the film first, followed by a pair of negative pulses.

In the second case, the order of the pulse pairs was reversed.

Two parameters of the ferroelectric hysteresis loops recorded in each write cycle were monitored: the magnitudes of the remanent polarization $|\mathbf{P}_r|$ and the coercive field $|\mathbf{E}_c|$.

The remanent polarization $\mathbf{P}_r$ is a vector quantity that retains its value in a ferroelectric capacitor after the electric field is removed; its change under repeated field application characterizes the polarization fatigue of the material.

The coercive field $\mathbf{E}_c$ corresponds to the electric field that must be applied to the crystal to reverse the sign of the polarization.

Based on the measurement results, we plotted the positive and negative components of $\mathbf{E}_c$ and $\mathbf{P}_r$ as functions of the number of write cycles for the different sequence configurations.

Analysis of the obtained dependences makes it possible to assess the effect of the pulse sequence configuration on the processes occurring in the ferroelectric film.

Methods

Sample preparation. The object of study was an epitaxial $PbZr_{0.3}Ti_{0.7}O_3/LaSr_{0.7}Mn_{0.3}O_3$ (PZT/LSMO) heterostructure grown on a $SrTiO_3$ (STO) substrate by pulsed laser deposition. The PZT and LSMO layers (the latter serving as the bottom electrode) were 60 and 50 nm thick, respectively. The fabrication procedure is described in more detail in [10], where it is given for a similar sample. Gold electrodes 60 nm thick were deposited onto the film (Fig. 1,*a*). To improve adhesion of the contact, a 4.8-nm chromium interlayer was deposited between the film and the electrodes.

Measurement details. The triangular pulses of the control field had an amplitude of ±8 V and a duration of 0.02 s. The choice of the triangular shape was dictated by the results in [11], where such writing pulses were used for slow cycling at a very low frequency.

The measurements were performed in series of 50 write cycles with 2-minute pauses between the series. The objective in the first stage of the study was to examine the polarization properties of the ferroelectric film over a small number of switching cycles (on the order of 10^3 write cycles), so as to focus on the film behavior in the absence of fatigue signs [3].

A ferroelectric hysteresis loop was recorded in each write cycle using the Sawyer–Tower method [12]. This method consists in measuring the charge of a reference capacitor series-connected to the sample. The measurement circuit was modified within the framework of the double-wave method [13, 14] to eliminate the dielectric and conductive contributions, which would otherwise distort the results. Fig. 1,*b* shows an example of a hysteresis loop recorded in each cycle, along with the approach used to determine the parameters of the $\mathbf{P}_r$ and $\mathbf{E}_c$ vectors.

The positive (E_c^+) and negative (E_c^-) components of the coercive field $\mathbf{E}_c$ are defined as the electric field at which the remanent polarization reaches half the value of the corresponding $\mathbf{P}_r$ components (P_r^+ and P_r^-).

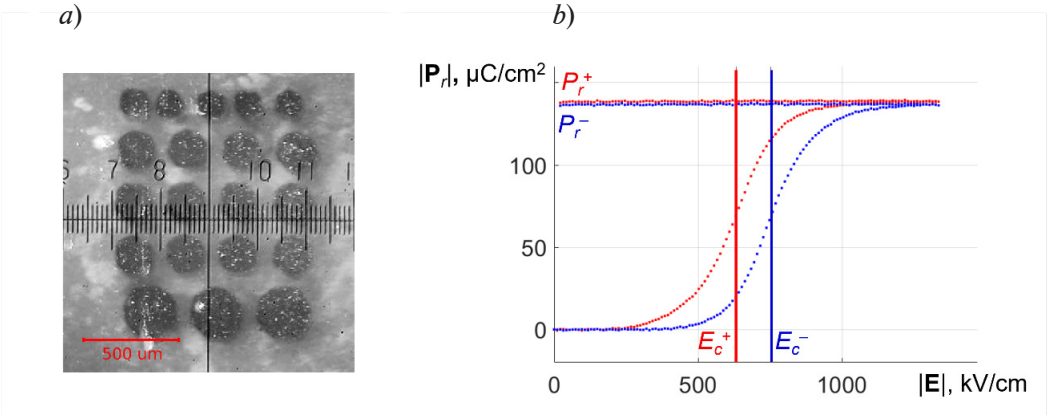


Fig. 1. Photograph of electrodes deposited on the film (*a*);
example of ferroelectric hysteresis loop measured in each write cycle (*b*);
P_r^+, P_r^- are the remanent polarization components; E_c^+, E_c^- are the coercive field components

Results and discussion

Fig. 2 shows the coercive field components as functions of the number of write cycles N for the two pulse sequence configurations. The vertical dashed line marks the point at which the pulse sequence configuration was changed. The differences between the measurement series, each consisting of 50 write cycles, are apparent from the plots. The coercive field components E_c^+ and E_c^- change only slightly for both pulse sequence configurations during cyclic switching, whereas jumps in $|\mathbf{E}|$ are observed after pauses between measurements. The amplitude of these jumps can reach 10% or more of the initial values.

Analyzing the dependence $|\mathbf{E}_c|(N)$, we can conclude that the effect of the pulse sequence configuration on E_c manifests itself as a correlation between the polarity of the last applied pulse and the amplitude of the jump in E_c after a pause between measurements.

If the preceding measurement series ended with pulses of negative polarity, then the amplitude of E_c^+ increased noticeably after the pause, while that of E_c^- decreased, and vice versa.

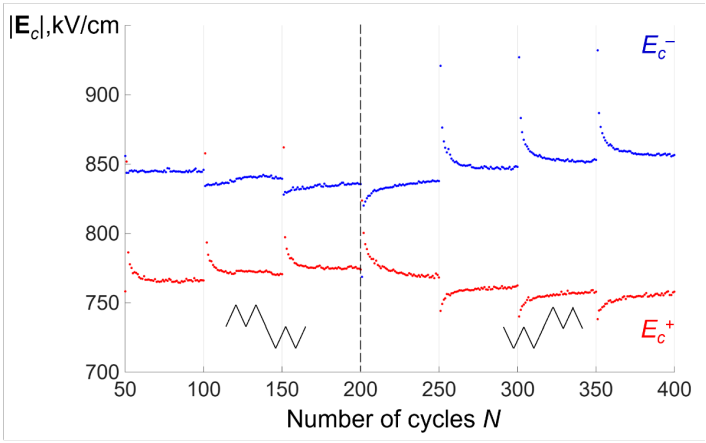


Fig. 2. Coercive field components $\mathbf{E}_c$ as functions of the number of write cycles for the two pulse sequence configurations
(the vertical dashed line marks the point at which the configuration was switched)

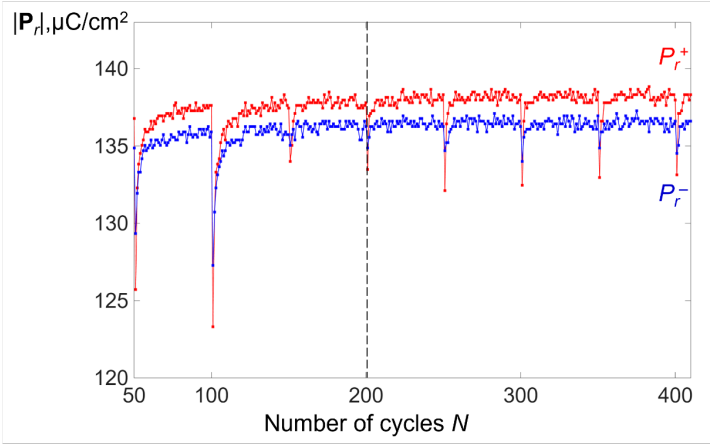


Fig. 3. Remanent polarization components $\mathbf{P}_r$ as functions of the number of write cycles for the two pulse sequence configurations

Taking into account the physical meaning of the coercive field E_c, this observation can be interpreted as follows: the polarity of the final writing pulse creates conditions under which domains whose dipole moments are oriented along the pulse direction become stabilized in the ferroelectric crystal.

Fig. 3 shows the remanent polarization components $\mathbf{P}_r$ as functions of the number of write cycles. As in Fig. 2, the experimental points belonging to individual measurement series are well distinguishable, and the pauses between the series are accompanied mainly by minor drops in $|\mathbf{P}_r|$. However, unlike the dependence $|\mathbf{E}_c|(N)$, changing the pulse sequence configuration from "two positive–two negative" to "two negative–two positive" has no qualitative effect on the behavior of $|\mathbf{P}_r|(N)$.

Conclusion

This study was conducted to evaluate how the configuration of switching pulses affects the remanent polarization and coercive field in a thin epitaxial lead zirconate titanate film. The analysis showed that the polarity of the final pulse applied to the film in a series of write cycles influences the jump in $|\mathbf{E}_c|$, creating favorable conditions for stabilizing domains whose dipole moments are aligned with the pulse polarity. No correlations were found between the polarity of this pulse and the sign of remanent polarization after the measurements were resumed.

We plan to further develop the approach proposed in this paper. One direction for future work is to gradually bring the writing parameters closer to those used in practical data storage devices, namely, by increasing the number of write cycles and reducing pulse duration.

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