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Switching current shape analysis in LBGO single crystals

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Abstract. The switching current shape during polarization reversal at room temperature by rectangular field pulses was studied. The activation-type external field dependence of switching time was revealed. The domain structure was imaged after partial polarization reversal by piezoresponse force microscopy with high-spatial resolution and the main geometrical parameters were extracted. The switching currents for complete polarization reversal in field range from 15 to 20 kV/mm were fitted by modified Kolmogorov-Avrami formula taking into account the geometrical parameters of the domain structure. The obtained excellent fitting of the switching current confirmed the proposed model of the switching process.

1. Introduction

The borate-based nonlinear-optical crystals have emerged as the most promising candidates for creation of efficient laser sources in UV range based on frequency conversion of coherent light from visible and IR sources. The $LaBGeO_5$ (LBGO) single crystals grown for the first time in 1990s [1,2] with high nonlinear optical coefficients and ferroelectric properties allow realizing quasi-phase matched second harmonic generation (SHG) with the highest nonlinear optical coefficient (d_{33}=1.3 pm/V [3]) and without the light wave walk-off effect. Moreover, the LBGO crystals are non-hygroscopic in contrast to other borate-based crystals (β-BaB_2O_4 and $CsLiB_6O_{10}$) applied for UV light generation. The LBGO single crystals possessing the stillwellite structure have the ferroelectric phase transition at 532°C [4,5]. The outstanding crystal properties have stimulated the strong scientific interest resulting in detail study of thermal, dielectric [4], nonlinear optical [3], structural [5,6], and main ferroelectric properties [7,8].

The first polarization reversal in LBGO at room temperature was realized in Ref. [9]. The frequency and activation-type temperature dependences of coercive field during polarization reversal by triangular field pulses were studied [9,10]. It was shown that the switching time obtained during polarization reversal by rectangular field pulses followed the activation-type temperature dependence with activation energy 0.86 eV [10]. The attempt to analyse and describe the shape of switching currents obtained during polarization reversal at 120°C using the Kolmogorov-Avrami (K-A) model was made [11] with poor fitting of experimental data.

The domain structure imaging in LBGO single crystals was realized by the optical microscopy after selective chemical etching [11] and by SHG interference microscopy [10] with low spatial resolution. It is necessary to point out that the modern high-resolution domain imaging methods were not applied.

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The obtained results in studying the polarization reversal process in LBGO single crystals allowed developing the domain engineering methods for creation of periodically poled samples for the first-order SHG at 365 nm [12] with period 5.5 μm. However, the attempts to create the period 2.2 μm for generation at 266 nm ended with creation of period 4.4 μm for the second-order SHG with lower efficiency [13]. Thus, the deeper study of domain structure evolution is required for further development of domain engineering methods in LBGO to obtain the periods below 3 μm.

In the present paper, we provide the detail study of the switching current shapes in LBGO single crystals obtained during polarization reversal at room temperature accompanied by visualization of static domain structure with high spatial resolution.

2. Experimental

The studied c-cut plates of LBGO single crystal grown by Czochralski pulling method (Oxide corp., Japan) had dimensions 5x15x0.5 mm^3 along a, b, and c axes, correspondingly. The liquid electrodes (LiCl saturated aqueous solution) were used for polarization reversal. The sample was glued over the 1-mm-in-diameter round aperture in the supporting glass and sandwiched between two glass plates covered by transparent ITO electrodes [14]. The liquid electrodes provided the contacts between the ITO electrodes and polar surfaces of the sample.

The rectangular voltage pulses were generated by ADC/DAC board (NI PCI 6251, National Instruments, USA) and amplified by high-voltage amplifier (TREK 20/20C, Trek Inc., USA). The applied voltage and switching current were measured by feedback ammeter. The rectangular field pulse with rise-time from 0.1 to 0.01 s, amplitude E_{ex} from 15 to 20 kV/mm and duration long enough to obtain full polarization reversal under the electrode area was used for forward switching. The obtained switching currents were smooth enough, so the measurement rate of 5 KHz could be used. The unipolar pulse of reverse polarity with E_{ex} = 20 kV/mm and duration 3 s was applied after each forward switching. The 3 min interval was maintained before application of the next forward field pulse. The switching charge was calculated by integration of the switching current. The switching time was obtained as the time interval corresponding to 0.95 of the total switching charge.

3. Results and discussion

The typical shape of the switching current in LBGO is presented in Figure 1a. The smooth shape of the switching current was obtained for the whole studied field range. The switching current consisted of two peaks: the first one – at the rise of external field, and the second one – at the constant field. It was demonstrated by applying two subsequent field pulses of the same polarity that the first peak was not related only to the capacitive input of the switching current (charging of the linear sample capacitance) as this input was much smaller (Fig. 1b). In previous works this peak was attributed to

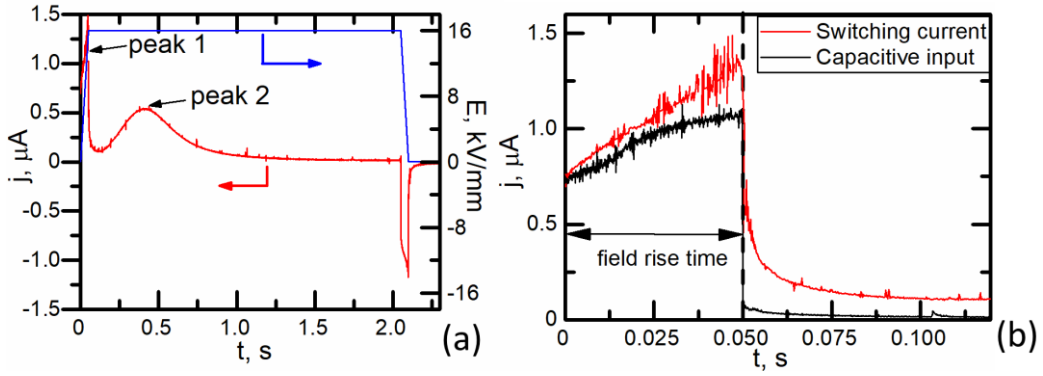

Figure 1. Typical switching current obtained in LBGO during switching with liquid electrodes: (a) switching pulse and applied electric field, (b) the magnified first peak of the switching pulse (red line) and capacitive input to measured switching current (black line) measured during application of the second subsequent forward field pulse. Amplitude of the field pulse 16 kV/mm, rise time 0.05 s.

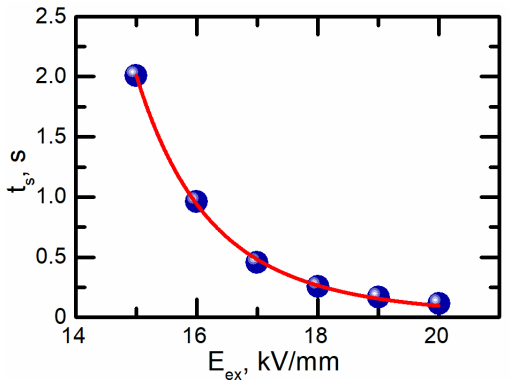

Figure 2. Field dependence of the switching time fitted by Eq. 1.

the nucleation of new domains [11,15]. In our experiments, this peak can be related to the domain nucleation at the electrode edges, while the second peak – to formation and growth of domains under electrode area.

The switching time t_s was measured by analysis of the time dependence of the switching charge corresponding to the second peak only. The activation-type dependence was used for fitting t_s (E_{ex}) data (Fig. 2):

$$t_s(E_{ex}) = t_0 \exp\left(-\frac{E_{ac}}{E_{ex}}\right) \quad (1)$$

where E_{ac} is activation field and t_0 is switching time in the limit of large switching fields. The best-fit parameter values: E_{ac} = (180 ± 5) kV/mm, t_0 = (11 ± 3) ms.

The detail study of the switching current requires the preliminary information about the domain structure geometry. The sample was switched partially by electric field pulse with E_{ex} = 15 kV/mm and duration 0.7 s (Fig. 3). The chosen pulse parameters allowed switching about the half of the area covered by electrode.

The imaging of static domain structure was realized using the scanning probe microscope (SPM) Asylum MFP-3D SA (Asylum Research, USA) in Piezoresponse Force Microscopy (PFM) mode. Commercial probes NSC18 with titanium-platinum conductive coating (MikroMash, Estonia) with a radius of curvature R = 35 nm, resonance frequency f = 70 kHz, and spring constant k = 3 N/m were used. Piezoresponse measurements were realized by the application of 5 V AC voltage close to contact resonance frequency to the SPM tip. The static domain structure (Fig. 4) represented the irregularly shaped domains nucleated with density of about $4 \cdot 10^4$ mm^{-2}. Such high values of domain nucleation densities can be attributed to defects of the sample surface. It can be seen from the Figure 4b that new domains nucleate mostly along the linear imperfections of the surface.

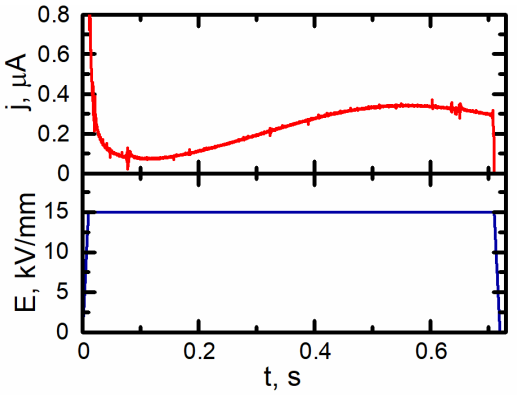

Figure 3. External field pulse and switching current obtained during partial polarization reversal.

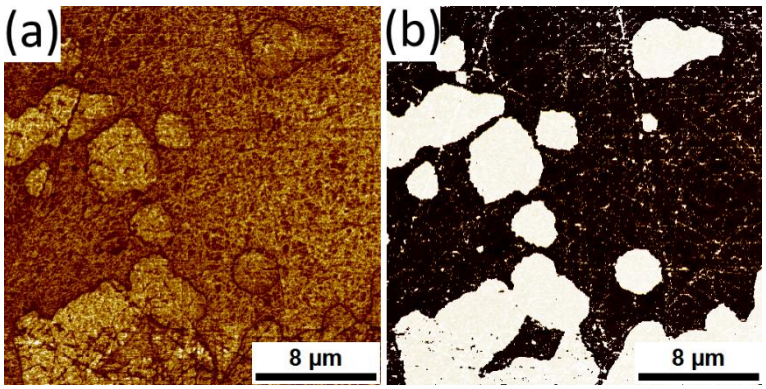

Figure 4. SPM visualization of static domain structure after partial polarization reversal: (a) amplitude signal, (b) phase signal. White contrast at (b) corresponds to switched domains.

The visualized large number of domains with wide variety of sizes ranged from 100 nm to 10 μm at the middle of the polarization reversal process allows to suppose that the main part of the switching process occurs by α(2D) process within the K-A model, while at the end of the switching the β(1D) process is expected. It should be noted that the proposed model is applicable for describing the second (main) peak of the switching current only. Additional study of the domain structure is required for full description of switching current.

The switching currents obtained for complete polarization reversal in field range from 15 to 20 kV/mm were fitted by K-A formula modified for switching in finite volume taking into account the transition from α(2D) to β(1D) processes (geometrical catastrophe) at t_{cat} [16,17]:

$$j(t) = \begin{cases} 2P_S A \frac{3}{t_{0\alpha}} \left(\frac{t-t_{st}}{t_{0\alpha}}\right)^2 exp\left(-\left(\frac{t-t_{st}}{t_{0\alpha}}\right)^3 \left(1-\frac{t}{t_m}\right)\right), & \text{for } t \le t_{cat} \\ 2P_S A \frac{1}{t_{0\beta}} exp\left(\frac{-(t-t_{st})}{t_{0\beta}}\right), & \text{for } t \ge t_{cat} \end{cases} \quad (2)$$

where P_S = 4 μC/cm^2 is the spontaneous polarization, $t_{0\beta}$ and $t_{0\alpha}$ are characteristic times of corresponding processes, t_{st} is the time of switching process start, t_m accounts for impingement of growing domains on the electrode edges (boundaries of switching media) [16,17], and A is the switched area.

The obtained excellent fitting of the switching current (Fig. 5) confirmed the proposed model of the switching process. The fit parameter A was significantly higher than the real electrode area indicating that the switching outside the electrode area occurred.

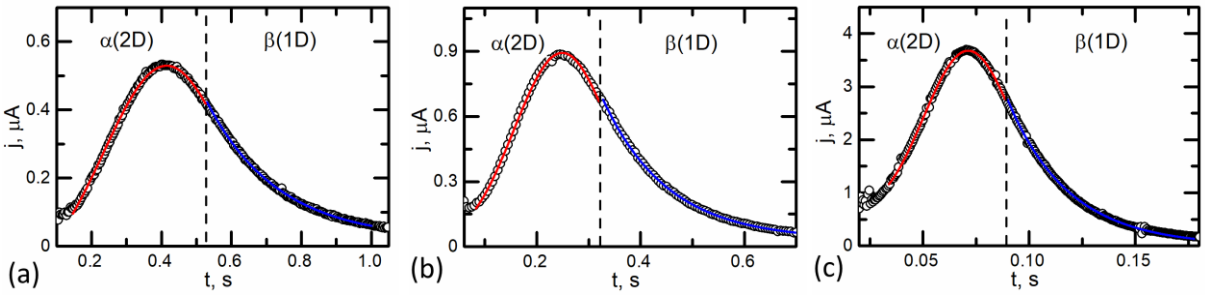

Figure 5. Switching current fitted by K-A formula modified for switching in finite volume taking into account the change of the growth model from α(2D) to β(1D) (Eq. 2). Amplitude of field pulse (a)16 kV/mm, (b) 17 kV/mm, (c) 20 kV/mm. The experimental points number was reduced to improve the visibility.

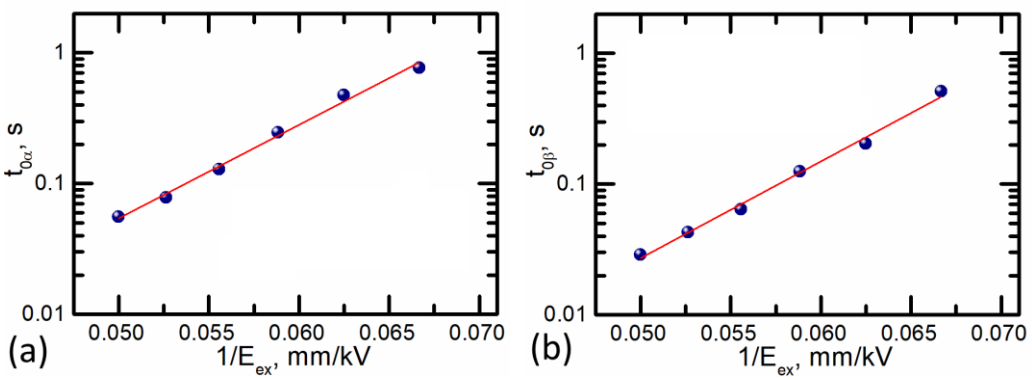

Figure 6. Field dependence of characteristic process times: (a) α(2D) process, (b) β(1D) process.

It was shown that the characteristic times $t_{0\alpha}$ and $t_{0\beta}$ followed the activation type field dependences (see Eq. (1)) with activation fields (165±5) kV/mm and (170±7) kV/mm, respectively. The equal (within the experimental error) activation fields indicate that both processes are governed by the same field-activated mechanism of domain growth.

4. Conclusion

The switching current shape obtained during polarization reversal at room temperature by rectangular field pulses was studied in LBGO single crystals. It was shown that the switching time followed the activation-type field dependence with activation field (180 ± 5) kV/mm. The visualization of static domain structure after partial polarization reversal using the scanning probe microscope in piezoresponse force microscopy mode demonstrated that domain structure represented the irregularly shaped domains nucleated with density of about $4 \cdot 10^4$ mm^{-2}. The switching currents obtained for complete polarization reversal in the field range from 15 to 20 kV/mm were fitted by Kolmogorov-Avrami formula modified for switching in finite volume taking into account the transition from α(2D) to β(1D) processes (geometrical catastrophe). The model choice was based on the results of domain structure imaging. The obtained excellent fitting of the switching current confirmed the proposed model of the switching process. It was shown that the characteristic times of α(2D) and β(1D) processes followed the activation type field dependences with activation fields (165±5) and (170±7) kV/mm, respectively. The equal (within the experimental error) activation fields indicated that both processes were governed by the same field-activated mechanism of domain growth. Obtained results improved understanding of domain structure evolution in LBGO single crystals and will be used for development of domain engineering for creation of compact and effective laser light sources in UV range.

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