The field-induced jump-like dynamics of a single planar domain wall in single crystalline uniaxial ferroelectric-ferroelastic Gd$_2$(MoO$_4$)$_3$ and needle domains in the perovskite BaTiO$_3$ are compared. Acoustic emission as well as ferroelectric Barkhausen pulse measurements were performed in order to investigate time coincidences with optically observed jumps. In both crystals significant amounts of acoustic emissions occur. The coincidences of both event types in Gd$_2$(MoO$_4$)$_3$ are determined by the particular character of extended defects in the crystal and the relative orientation of the acoustic sensor. Hardly any Barkhausen pulses but huge amounts of acoustic pulses are observed for the annihilation of needle domains. The underlying microscopic mechanisms are discussed.

**Keywords:** Ferroelectrics; BaTiO$_3$; Gd$_2$(MoO$_4$)$_3$; acoustic emissions; ferroelectric Barkhausen pulses

**PACS:** 77.80.-e, 77.80.Fm, 77.84.Dy, 61.72.Hh

### INTRODUCTION

The polarization reversal process in ferroelectrics is known to be a complicated process of domain nucleation and subsequent growth by domain wall motion [1–3]. The individual nucleation acts, forward domain growth, and the interaction of the domain walls with defects (pinning) are often accompanied by discontinuities in the switching current, the ferroelectric
Barkhausen pulses [2–4], and in the elastic response as acoustic emission events [5, 6]. The analysis of the Barkhausen pulse and acoustic emission data may constitute an effective way to study the elementary discontinuous switching processes [7].

Two materials are chosen for a comparative study, the improper ferroelectric-ferroelastic gadolinium molybdate Gd$_2$(MoO$_4$)$_3$ (GMO) [8–15] and BaTiO$_3$. Barkhausen pulses in GMO have been known to occur when a domain wall is moved across an artificial electric field inhomogeneity near the edges of a gadolinium molybdate crystal under a.c. driving [12–14]. Acoustic emissions in ferroelectrics on the other hand have been identified during domain wall nucleation [16, 17], domain wall annihilation, and the motion of domain walls across internal defects [18].

The interaction of a wall with larger defects generally stops its smooth motion [18]. When the driving force exceeds a particular value, the wall is released generating discontinuous jumps. A different source of acoustic emissions may arise, when the domain positions are stabilized due to space charges during aging. An illustrative study of such an effect was performed [19]. The acoustic emission pattern drastically changed after chemical etching away of a near surface layer on a BaTiO$_3$ crystal, even though the authors did not identify space charges to be responsible for the effect at the time.

In this study the particular role of needle domains in BaTiO$_3$ and coincidence of acoustic emission events and Barkhausen-pulses are discussed for 90$^\circ$ and 180$^\circ$ domain switching processes.

**EXPERIMENTAL**

Orthorhombic $\beta'$-Gd$_2$(MoO$_4$)$_3$ [20] crystals were grown by Czochralski method. The growth conditions allowed to introduce some bulk defects yielding pinning centers for the domain [15, 18]. The crystal surfaces were oriented along the [001] and [110] planes. A single 180$^\circ$ domain wall normal to the [110] face separates two domains of point inverted symmetry [21]. Transparent In$_2$O$_3$:Sn-electrodes were sputtered onto the single crystalline plate in form of two u-shaped electrodes [12, 13, 22]. The BaTiO$_3$ crystals were commercial high purity crystals cut along the crystal axes (dimensions 1 $\times$ 10 $\times$ 10 mm$^3$, [100] $\times$ [010] $\times$ [001]). Initially a striped domain structure of alternating c- and a-domains (with respect to the 10 $\times$ 10 surface [100]) had aged into the sample. The c- as well as a-stripes contained further 180$^\circ$ domain walls. Silver paint electrodes were applied to the 1 $\times$ 10 surfaces [00 $\pm$ 1] and a first set of measurements was performed removing the aged domain structure. In a second step a thin slice of the sample [010]
was taken out by a diamond wire saw inducing mechanically driven needle domains (Fig. 1(a)).

The commercial acoustic emission setup [6] (AMS3, Vallen Systeme, Icking, Germany) of analog bandwidth 200 kHz to 2 MHz records amplitude, energy (1 e.u. = \(10^{-18}\) J electrical), and the transients (5 MS/s) of each event using a resonant type microphone (150 kHz). The amplification was 54 dB (60 dB), threshold 25 \(\mu\)V (32 \(\mu\)V), for AE, and 43 dB (49 dB), threshold 400 \(\mu\)V (25 \(\mu\)V), for the Barkhausen pulse detection on a serial resistance (\(R = 100 \Omega \ (4700 \Omega)\)) for \(\text{Gd}_2(\text{MoO}_4)_3 \langle \text{BaTiO}_3 \rangle\). For GMO a somewhat wider gap between the sample and the microphone was introduced to avoid fracture. Acoustic contact in each case was provided by liquid Fluorinert® (FC-77, 3M corporation). The electrical circuit is depicted in Fig. 2. Bipolar triangular electric fields of rising amplitude were applied to the samples. Large wall jumps in GMO were visible in an polarized optical microscope and recorded onto video tape with frequency 25 Hz.

**Figure 1.** Illustration of the needle pattern in the sample.

**Figure 2.** Electrical circuitry for determining acoustic emissions and ferroelectric Barkhausen pulses. High voltage source (HV), microphone (MIC), sample (S), serial resistor (R), electronics (AE), electrometer (V).
RESULTS

Acoustic emissions and Barkhausen pulses are only partially coincident in GMO. As we previously demonstrated, the dynamics of single domain walls change according to the crystal defects encountered by the domain wall during its electrically driven motion [18]. Figure 3 depicts an extended view of a single bipolar cycle. The initial breakup of the

![Graph](a)

![Graph](b)

**Figure 3.** First acoustic emission. (a) Coincidence patterns for acoustic emissions and Barkhausen pulses for GMO crystal and (b) The microphone axis lies along the orthorhombic c-axis [001].
Figure 4. The acoustic emission pattern (a) for a sequence of external field cycles of rising field amplitude (b) along [001] applied to an aged BaTiO₃ crystal. Significant switching of c-domains ([100]) into the plane is observed. Amplitudes of acoustic emission (c) and polarization (d) versus the electric field.

single domain wall from its aged position only generates acoustic emissions (Fig. 3(a)). No Barkhausen pulse is found. Subsequent jumps generate coincident acoustic emissions and Barkhausen pulses (Fig. 3(b)). The ratio of acoustic emission amplitude to Barkhausen pulse amplitude varies from event to event and no clear correlation for the amplitudes is found.

In BaTiO₃ acoustic emissions occur for the breakup of the aged domain configuration (Fig. 4). The events only occur for rising electric fields and in field ranges, where a significant increase of the applied field is necessary to induce further switching of the crystal. This corresponds to the first hysteresis cycles, where fields well exceeding the standard coercive field are necessary (Fig. 4(d)). For maximum fields of 500 V/mm the textbook hysteresis loop is finally restored. Now acoustic emissions dominantly occur at high and low polarization values, while the major switching range at the coercive field $E_c$ exhibits significantly less acoustic emissions (Fig. 5). The entire set of experiments is performed on the aged initial c-/a-domain structure.

In the next experiment needle domains were introduced mechanically by inducing residual stresses from sawing. Figure 1 illustrates the domain pattern obtained. While an enormous number of acoustic emissions
occur, which corresponds to the annihilation of the needles, basically no Barkhausen pulse is found (Fig. 6(c)).

DISCUSSION

Acoustic emissions and Barkhausen pulses in previous literature on GMO were only observed when a domain wall is driven to an electrode edge [23]. At the edge high stresses and fields induce zigzag domain wall patterns
appearing and disappearing under emission of Barkhausen pulses due to the locally induced high mechanical stresses and electric fields [10]. This is a different mechanisms than encountered here. The stated one-to-one time coincidence between acoustic emissions and Barkhausen pulses has been shown by us to be invalid for GMO [18]. In this crystal two processes occur. First the breakup of the domain from its aged position (also termed bound internal field) [24] yields significant acoustic emissions. We were previously able to show, that the resonance spectrum of such an event is characteristic of the domain wall itself as a resonating membrane [18]. Under these circumstances, the domain wall obviously breaks away from its initial position sufficiently symmetrically with respect to the external electrodes that no effective Barkhausen pulse is observed. Only later, when the domain wall as a whole exhibits jumps, which do not excite the domain wall resonance, finite polarization jumps are observed. In this case the projection of polarization change onto the electrodes is sufficient to induce large electrical pulses.

It has to be clearly understood that in order to yield Barkhausen pulses as well as acoustic emissions, the microscopic sources have to be extremely fast. Slow continuous motion of domain walls is neither a source of acoustic emissions nor Barkhausen pulses. This is clearly seen in the fact that only few acoustic emissions occur during the major switching (Fig. 5). In the case of needle domains in BaTiO₃ it was particularly intriguing that essentially no Barkhausen pulses were observed. Barkhausen pulses observed in our setup occur as single channel transient recorder events (0.5 microseconds). All charges constituting the Barkhausen pulse are integrated to yield the respective voltage value for this channel. In order to estimate the detectable charge values the relative projection of the needle onto the external electrode surface is relevant. Needles extend over several millimeters. Typical jump distances as optically observed are at least 50–100 µm minimum up to complete annihilation of a domain in one step. The domains typically extend a few hundred µm into the depth of the sample (with respect to face [100]). A jump of a domain 100 µm deep and 400 µm jump distance would yield a pulse amplitude of 80 mV in our setup. Even smallest depths of a needle should yield in the range of 40 mV, which is 46 dB on the scale shown in Fig. 6 and thus far into the detectable range. If the propagation speed of the domain wall in a discontinuous jump of a ferroelectric-ferroelastic wall is the speed of sound, the pulse length is confined to about two channels maximum. The conclusion is that the annihilation of the domain wall is locally symmetric with respect to the electrodes, thus no effective charge jump occurs.
The elastic energy released during domain annihilation on the other hand is enormous. Figure 6(a) shows the acoustic emission amplitudes. The number of events by far outnumbers those encountered during one cycle of the deaging of the initial c-/a-domain structure. Very high amplitudes are observed corresponding to the large elastic energy stored in the needle [25, 26].

**SUMMARY**

Discontinuous acoustic pulses and ferroelectric Barkhausen pulses are not necessarily coincident, neither for 180° nor for 90° switching processes. In some cases large acoustic pulses reflect the liberation of elastic energy, while the polarization changes locally cancel to yield no macroscopic effect. This explains, why aside of the pure speed of the domain wall motion, polarization changes due to discontinuous pulses are comparatively rare with respect to the overall polarization switching of a crystal. The effect occurs for single planar domain walls in GMO, which for certain resonating modes do not yield finite polarization changes, as well as for needle domains in perovskite ferroelectrics exemplified by BaTiO$_3$.

**ACKNOWLEDGEMENT**

The support by the Deutsche Forschungsgemeinschaft (Lu729/5-1) and through “Basic Research in Russian Universities” (Grant UR. 06.01.031) and “Priority Research in High School. Electronics” (Grant No. 03-03-29), Grant No. 01-02-17443 of the Russian Foundation of Basic Research, by Grant No. 02-02-04006 of RFBR-DFG, by Award No. REC-005 of the U.S. CRDF are greatly acknowledged.

**REFERENCES**
