ISSN 1392–1320 MATERIALS SCIENCE (MEDŽIAGOTYRA). Vol. 19, No. 4. 2013

Broadband Electromagnetic Emission from PZT Ferroelectric Ceramics after Shock Loading

Fiodoras ANISIMOVAS *, **Steponas AŠMONTAS, Oleg KIPRIJANOVIČ, Andrius MANEIKIS, Bonifacas VENGALIS**

Center for Physical Sciences and Technology, A. Goštauto 11, LT-01108 Vilnius, Lithuania

crossref http://dx.doi.org/10.5755/j01.ms.19.4.3137

Received 10 December 2012; accepted 11 March 2013

It was experimentally registered pulsed electromagnetic (EM) radiation in frequency range higher than television one using wideband horns with coaxial and waveguide outputs. The EM radiation was received during shock loading of lead zirconate titanate (PZT) ceramics cylinders in conventional piezoelectric ignition mechanisms. Digital oscilloscope allows registering whole series of EM pulses and each pulse from the series transmitted from horn antenna of (1 – 18) GHz operating band frequencies. There is (1 – 4) μs delay between the shock and the first pulse of the series. Duration of the series is about (3 – 4) μs.

The PZT cylinders were cleaved along their axes and the surfaces formed in the process were investigated by scanning electron microscope. It was concluded that from electrical point of view PZT ceramics contain interacting subsystems. It was found that EM radiation spectrum of pulse detected by waveguide detector heads has harmonics reaching 80 GHz. Presence of harmonics higher than 20 GHz indicates on radiation due to deceleration of electrons emitted during the switching. The EM pulses in the series appear randomly and have different amplitudes which partly confirmed thesis on independent switching dynamics of small volumes governed by a local electric field.

Keywords: PZT ceramics, shock loading, pulsed EM radiation, fast polarization switching, electron emission, electron deceleration.

INTRODUCTION

Nowadays attention is paid to investigations of materials with complex internal structure. The investigations include analysis of their improved mechanical properties considering scale and nonlinear effects [1 – 3]. Sometimes these materials are expected to possess necessary properties in relation to certain electromagnetic (EM) and chemical phenomena. In ferroelectric ceramics and composite dielectrics mechanical-electrical and mechanical-EM transformations after shock loading are also investigated for these purposes [4, 5].

Pulsed mechanical loading of piezoelectric crystals [6], layered and composite dielectric materials [7, 8], induce the pulsed EM radiation. It was found that duration of the EM pulses varies up from microseconds down to nanoseconds, which hints at differences in origins of such generations. Generation of broad band signals and their application, including medical, is of great interest. Investigation of super wideband EM radiation using antennas is difficult, especially of single pulses. Wideband antennas and wideband oscilloscopes are needed for the investigation. Now creation of super wideband antennas and wideband digitized oscilloscopes makes the investigations easier [9]. Both theoretical studies [10] and experimental research [11] keep uncovering new types of super wideband signals.

Previously our experimental investigation of EM radiation after pulsed compressive stress of lead zirconate titanate (PZT) ferroelectric ceramic cylinders was carried out using wideband television antennas [12]. It was found series of short EM pulses (30 ns – 50 ns) spanning ~1 μs. The pulses in this series have no indication of period or constant value of amplitude. It was proposed that the stress leads to appearing of high voltage electric pulse series. The electrical pulses have ns duration and during their fast rising fronts the EM pulses are generated. The processes that linked the stress and the generation of electrical pulse series were not sufficiently explained.

Piezoelectric ignition mechanisms (PIM) with PZT ceramics are used for high voltage generation, which works on shock loading for example in piezoelectric cigarette lighters. The aim of this work is to register pulsed EM radiation during action of PIM in the range from 1 GHz up to 80 GHz using different horn antennas with coaxial or waveguide outputs. We also analyze cleaved PZT samples under SEM and try to clarify what causes the series of electrical pulses to appear.

EXPERIMENTAL

Conventional PIM with PZT cylindrical components (cylinders) of 4 mm high and 2 mm in diameter is used in experiments. Such cylinders are fabricated as follows. Firstly PZT powder is calcined at temperatures (1000 – 1300) °C for (1 – 2) h until the required PZT phase is acquired. Then the samples are shaped using pressing or hot-pressing methods. Different piezoelectric properties are imparted depending on how long the pressure is applied at maximal temperature [13]. It is worth noting that during growth of PZT crystallites impurities concentrate in crystallite boundaries whose thickness increases causing changes of their electrical properties. Key problem is in controlling the properties of the boundaries during the fabrication.

*Corresponding author. Tel.: +370-5-2619759; fax: +370-5-2627123.
E-mail address: *anis@pfi.lt* (F. Anisimovas)

Crystalline structure of the PZT cylinders was examined by X-ray diffractometer Bruker AXS D8 using CuK_α radiation. We should investigate the PZT ceramics internal structure to establish if it is linked with EM pulse series generation along with its intended function. In this work there were investigated surfaces formed after cleaving the cylinders along its axes. To investigate the surfaces we employed scanning electron microscope (SEM) Hitachi 3000. The backscattered electrons are detected to form an image of the surfaces. Such procedure also allows determining composition of surface material.

Schematic diagram of high voltage block of the PIM is presented in Fig. 1. There are two PZT cylinders (1) of 130 mg each with Ag metalized bases (2) and high voltage electrode (3) placed between them. They are rested against a metallic foundation of 800 mg (6). On another cylinder base there is a metallic disk with a bulge of 20 mg (5) which is being hit by metallic hammer of 300 mg (4). The hammer is accelerated by spring which has stiffness $2.75 \cdot 10^3$ N/m. Energy stored in the spring before the shock is equal to $34 \cdot 10^{-3}$ J. The purpose of the disk is to align shock force evenly on the cylinder base. Both cylinders are in two-layer insulated cases: inner layer is plastic (7) and outer layer is epoxy (8). Under the action of a spring (not shown) the hammer (4) hits the disk (5) and high voltage discharge through the air between (3) and (6) is initiated. The length of the discharge spark channel reaches 6 mm.

Voltage dynamics induced by the hammer are investigated inserting high voltage resistive divider 1 : 470 between the electrode (3) and foundation (6). Schematic diagram of this investigation including oscilloscope having 1 MΩ input and (0 – 100) MHz bandwidth corresponds to the one presented in [12]. Appearing voltage is divided and reduced signal is registered by the oscilloscope.

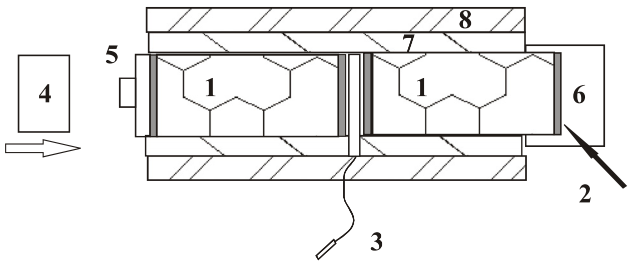

Fig. 1. Schematic diagram of high voltage block of the PIM. 1 – PZT cylinder, 2 – cilinder base metallization, 3 – high voltage electrode, 4 – hammer, 5 – disk, 6 – foundation, 7 – plastic insulator, 8 – epoxy insulator

The EM emission of PIM after the shock loading was investigated using wideband antennas and digital and real time oscilloscopes. Schematic diagram of the experimental arrangement for EM pulses registration is presented in Fig. 2. The EM emission registration occurs in a following way. The PIM (1) is placed at small distance from the horn antenna aperture (2) and is excited by pressure. We have used the pyramidal horn with coaxial output. The horn has aperture (34×26 cm^2) and (1 – 18) GHz operational frequency range. The EM emission is collected by the horn and electrical signal is then transmitted directly to wideband digital oscilloscope (4) by 50 Ω impedance cable. Since the signal was powerful enough, to protect sensitive, 1 V max, oscilloscope input we had to include the 10 dB attenuation (3).

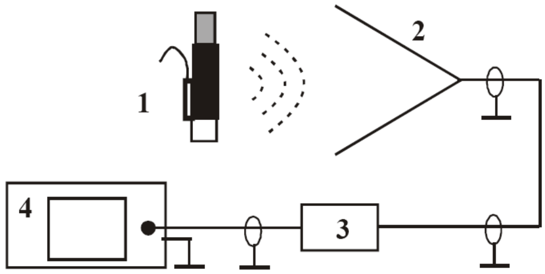

Fig. 2. Schematic diagram of experimental set-up for EM radiation registration using digital oscilloscope. 1 – PIM, 2 – horn antenna operating in (1 – 18) GHz frequency range, 3 – 10 dB attenuator, 4 – digital oscilloscope of (0 – 6) GHz bandwidth

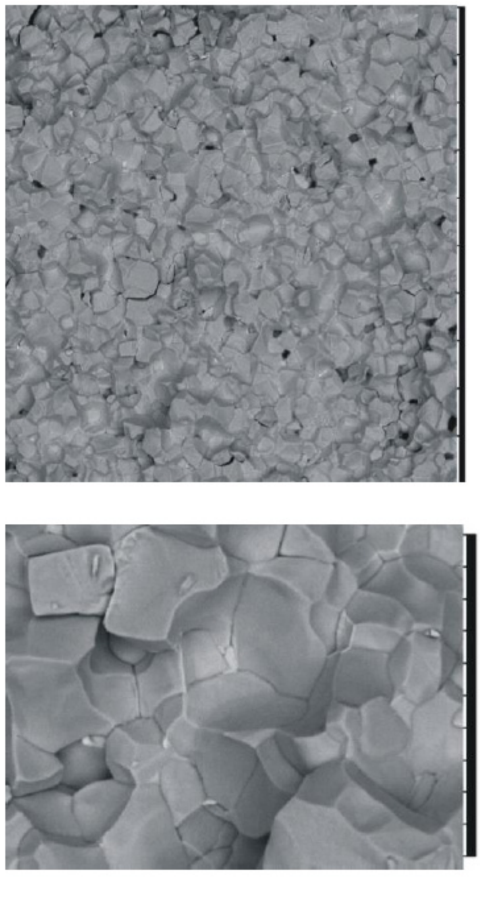

Fig. 3. Surface images of PZT cylinder cleaved along its axis obtained by SEM. Full vertical scales are 200 μm (above) and 30 μm (below)

Detection of the higher than 18 GHz frequency harmonics was realised using a number of different horn antennas with waveguide outputs. In these measurements it was used (0 – 7) GHz bandwidth real time oscilloscope. Schematic diagram of experimental set-up is structurally similar to that as in Fig. 2. Difference between the diagrams is that now synchronization channel is added. The signal is branched by a tee and a delay line is placed before the oscilloscope input (see in Ref. [12]). The horn waveguides are loaded by detector heads. Signals from the heads are amplified in (0.8 – 2.4) GHz range and transmitted to the oscilloscope input using short cables. The frequency range can be broadened employing waveguide-to-waveguide transition to one with smaller cross section.

There were employed following horns in the investigations. Horn with aperture (10×8 cm^2) with waveguide output of (11×5.5) mm^2 cross section corresponding to (17.44 – 25.95) GHz range; horn with aperture (5×4.5 cm^2) with output waveguide 7.2×3.4 mm^2 cross section corresponding to (25.95 – 37.5) GHz range; horn with aperture (3.5×3 cm^2) with waveguide output of 3.6×1.8 mm^2 cross section corresponding to (53.6 – 78.3) GHz range. We have used transition from 7.2×3.4 mm^2 to 5.2×2.6 mm^2 waveguide which corresponds to (37.5 – 53.6) GHz range.

EXPERIMENTAL RESULTS

Information obtained from X-ray diffraction patterns is essentially the same as presented in Ref. [12]. The cylinders are $PbZr_{0.52}Ti_{0.48}O_3$ compound with tetragonal symmetry and some of the samples had more developed (111) preferred orientation**.** The Zr : Ti proportion of this composition, near morfotropic transition Zr : Ti = 55 : 45, indicates on the PZT ceramics having high piezoelectric effect. Composition analysis using SEM shows somewhat distorted results because of the presence of carbon and additional oxygen at the surface of the cleaved samples.

The experiments on high voltage dynamics when discharge is initiated through high ohmic resistive divider show results, which mainly repeat those obtained in [12]. The experiments reveal that the shock induces voltage growth, which reaches its maximum after (1 – 3) μs. Discharge dynamics has zero sign inversion point, which appears in time range from 4 μs to 5 μs. Afterwards in span of (10 – 15) μs voltage decreases, and reaches negative minimum comparable by absolute value with positive one. However important part of the response at the first μs could not be observed mainly because of divider bandwidth. After reaching negative minimum the voltage has lengthy relaxation part of ms order.

Such behaviour of the voltage-time dependence curve with a sharp drop from positive to negative values is observed when investigating surface charge density changes in ferroelectrics. Except in our case at the beginning of the relaxation part there is series of relatively high voltage spikes of microsecond duration with decreasing amplitude (see Fig. 2 in Ref. 12).

The PZT cylinder was split along its axis and the split surface images made by SEM can be seen in Fig. 3. There is chaotic distribution of crystallites having different sizes and forms. This is clearly seen on both pictures despite them having different magnification. The distances between the crystallites boundaries differ from tightly merged to containing nanometer size air filled openings. Crystallites of large size, higher than 2 μm indicate on calcining at 1300 °C for 1 h following by non hot pressing.

Using broadband digital oscilloscope reveals additional results during registration of EM pulse series. Half of one of the registered series is presented in Fig. 4 where every pulse appears as dashed vertical line. There is a delay between the moment of shock, which is also accompanied by pulse or several pulses, and the first series pulse. The pulse induced by the shock is zero moment of the oscillograms and is not displayed in Fig. 4. The delay ranges from 1.2 μs to 4 μs depending on the sample. Series duration is longer than 1 μs as it was proposed in [12] and can be up to 4 μs. The delay is comparable with the time needed for voltage to reach the maximum.

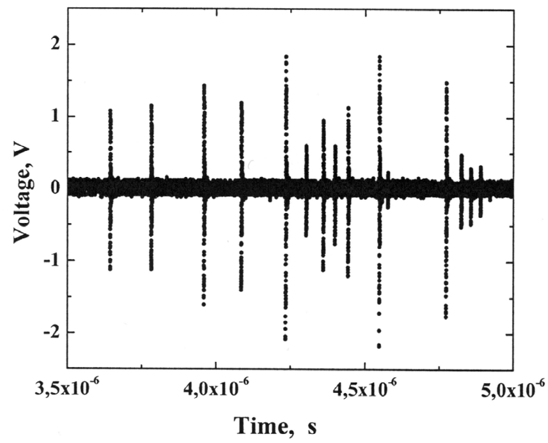

Fig. 4. Oscillogram of the half of EM pulse series obtained employing the (1 – 18) GHz horn and digital oscilloscope with (0 – 6) GHz bandwidth. Zero time corresponds to the shock of the hammer

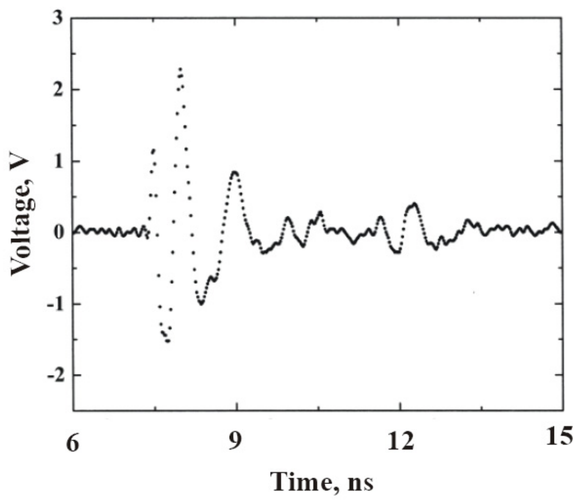

Fig. 5. Typical stretched EM pulse from the presented in Fig. 4 series. Higher harmonics are cut by (0 – 6) GHz bandwidth oscilloscope

The oscilloscope allows us to stretch every pulse of the series. They are short wideband pulses, which differ mainly in amplitude. The typical stretched pulse is presented in Fig. 5 and they have durations of (3 – 5) ns. However the form of this pulse is slightly distorted because higher than 6 GHz harmonics are cut off by both the (0 – 6) GHz bandwidth oscilloscope and the cable. Such pulse received on wideband TV antenna with upper frequency about 1 GHz has duration of (30 – 50) ns as shown in [12]. This is an example of oscillogram change depending on the receiving antenna operational bandwidth.

Oscillogram of the several pulses from the series detected by detector head of (37.5 – 53.6) GHz range is presented in Fig. 6. The forms of spikes on oscillogram differ depending on semiconductor type in the heads. They can be negative and stretched in time because of their sluggishness.

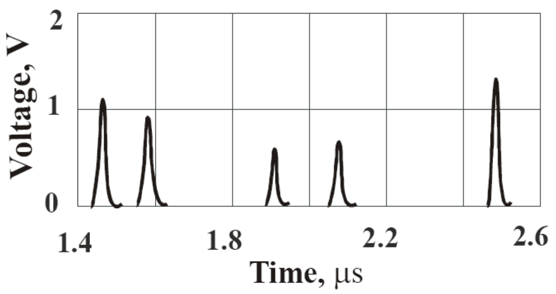

Fig. 6. Typical oscillogram of the part of EM pulse series obtained employing the waveguide horn antennas loaded by detector heads and amplification in (0.8 – 2.4) GHz range

DISCUSSION

So, usage of the broadband digital oscilloscope allows for more extensive measurements of the EM series. It is followed from Fig. 4 that there is 3.6 μs delay, which can be (1 – 4) μs in other cases, between the hammer shock and the first pulse of the EM series. The correct evaluation of the EM series duration was not possible in [12] because the fact of this delay was not known. The experiments reveal that series duration is longer and is at least (3 – 4) μs instead of ~1 μs.

The EM pulse series with its complex structure has certain features indicating on re-reflections of pulses. Sound speed in the crystallite structure with boundaries has relatively small value. This means that applied hammer shock with a pulse of 4.5 kg·m/s can induce deformation wave and even shock wave if the sound speed is low enough. Such pulses can be soliton-like deformation waves appearing after the hammer shock [14], and electrical pulse, which causes EM pulse, is formed during reflection of this wave from the cylinder base. Clarifying of the situation is complicated because overlapping EM pulses generated from two cylinders are undistinguishable. The waves should be formed after the shock from the two bases, one of them near the disk and another near the foundation. However the hammer shock is relatively long, comparing with ps laser pulse, for example, and interaction of the cylinder with the PIM foundation allows us to propose that cylinders were compressed as a whole rather than in a way which allows forming the soliton-like deformation waves.

It is worth to mention that the high voltage spikes can be understood as a result of elastic oscillations of the cylindrical sample during relaxation. It would mean that mechanical behaviour of the cylinders during the oscillations is as if they were solid.

As one can see from Fig. 3 the cylinder consists of varied form and size crystallites each having a surface and invisible internal domains with their own walls. Electrical properties of the crystallite content and its surface are different. There are Schottky barrier-like surface regions and surface levels, which play important role in polarization screening [15]. When we also take into account switching of the domains and movement of their walls it becomes clear how complex the cylinder structure is from electrical point of view. Processes and their response times and relaxation time constants in such media at applied electric field vary in duration from 10^{-12} s to $\sim 10^{-2}$ s. But high dielectric constant of PZT, which is changeable during repolarization, and entwined ferroelectric and piezoelectric properties could lead to fast, ps or ns, unforeseen processes.

Series of EM pulses can be generated due to generation of electrical pulses of ns durations with fast rising fronts during surface charge changes at the cylinder bases. Therefore it is possible sudden significant weakening of polarization and then returning to its initial state. Polarization switching and accompanying processes including electron emission and shielding are investigated in variety of works [16 – 19]. Investigators in the mentioned works used configuration with application of external electrical pulses to induce the repolarization, following by the polarization switching. In this work we met the problem of fast polarization switching while clarifying the EM radiation origin.

In our case self-repolarization process begins at the maximal voltage value during dynamic changes of cylinder polarity. The velocity of voltage inversion is of the order of 20 kV in 15 μs, which means that self-repolarization is quite fast indeed. The EM series beginning coincides with the start of self-repolarization process and a spark discharge in external circuit occurs after the start. We do not, however, exclude possibility that the series begins before the voltage maximum is reached, which indicates that fast reversible polarization switching occurs during the rapid voltage changes. Most probably we have experimental confirmation of the reversible metastable process including fast polarization switching and electron emission [20]. The polarization recovery to the initial state became possible due to differences in fast ferroelectric response times and slower adjusting times of surface state electrons. This process is also important in organizing an efficient emission process. We suppose that in our case fast reversible polarization switching occurs in regime of self-excitation and is accompanied by electrical pulse series. Our observed EM radiation occurred during these fast polarization changes.

Harmonics above 20 GHz point on the presence of electron emission during recovering and their deceleration, and therefore on the EM radiation. However we have no experimental confirmation whether the spikes in Fig. 6 correspond to the every pulse whose lower frequency harmonics registered and shown in Fig. 4 or to the every second.

Most of the classic models of polarization switching are based on the so called Kolmogorov-Avrami-Ishibashi approach, which considers the switching process to occur in two basic stages: nucleation and growth of reversed domains. This approach cannot accurately describe the fast switching in our experiment because of nonhomogeneous PZT ceramics with complex structure and entwined ferroelectric-piezoelectric properties. In modern theories such media is considered an ensemble of many regions with independent switching dynamics, governed exclusively by a local electric field [20, 21]. If it is true, the self generation of fast polarization switching mostly appear randomly and, possibly, could have different amplitude and durations of up to hundreds of nanoseconds. This is partially confirmed in our experiments as seen from Fig. 4 where the EM pulses of different amplitudes are spread randomly.

The mean value of amplitude of EM pulse series should depend on density of surface states in crystallites and on domain structure both of which can be regulated during manufacturing. Therefore using different PZT ceramics in devices with shock loading allows generating complex broadband signals which can be amplified and exploited in various fields including medical applications [22].

CONCLUSIONS

1) It is confirmed that after shock loading of PIM cylinders it is generated broadband EM pulse series. Usage of broadband digital oscilloscope uncovered that the first pulse of the series appears after a delay and the duration of the series reaches 4 μs.

2) The series pulse is extremely broadband and its form on the oscillogram depends on antenna operational frequency band and on bandwidth of oscilloscope.

3) Ferroelectric PZT cylinder has complex structure with interacting subsystems and therefore its reaction on external disturbances is characterized by different response times and by relaxation times constants of huge time range.

This facilitates processes with screening effects which damping fast electric field changes. Applying temporary mechanical structure displacements in such media induces not only piezoeffect but also can cause unforeseen fast, ps or ns, processes.

4) It is explained that unforeseen presence of such broadband EM emission suggests presence of self-exited generation of fast reversible polarization switching which is accompanied by generation of electrical pulse series of ns duration which begins at voltage maximum and lasts about 4 μs.

5) Presence of EM harmonics in the pulse up to 80 GHz indicates on deceleration processes of emitted electrons during fast polarization switching.

REFERENCES

1. **Mehta, A., Barker, G., Luck, J.-M.** Heterogeneities in Granular Materials *Physics Today* May 2009: pp. 40 – 44.
2. **Goriacheva, I. P., Morozov, N. F.** Mechanics – Science of XXI century *Vestnik of Russian Academy of Science* 82(7) 2012: pp. 638 – 647 (in Russian).
3. **Dimitrienko, Ju.**, **Sokolov, A.** Numerical Modeling of Composites with Multiscale Microstructure *Bulletin of the Russian Academy of Sciences. Physics* 75 (11) 2011: pp. 1457 – 1461.
4. **Rouquette, J., Hines, J., Bornard, V., Pintard, M., Papet, Ph., Bousquet, C., Konczewicz, L., Gorelli, F., Hull, S.** Pressure Tuning of the Morphotropic Phase Boundary in Piezoelectric Lead Zirconate Titanate *Physical Review B* 70 2004: 014108 p.
5. **Millett, J., Bourne, N., Deas, D.** The Response of Two Ferroectric Ceramics to One-Dimensional Shock Loading *Journal of Physics D: Applied Physics* 45 2012: 165203 p.
6. **Sud'enkov, Yu. V.** Electromagnetic Radiation Induced by the Failure of Piezoelectrics under the Action of Submicrosecond Stress Pulses *Technical Physics* 46 (12) 2001: pp. 1588 – 1590.
7. **Fursa, T., Surzhikov, A., Horsov, N., Osipov, K.** Interrelation between the Stressed State in Composites and the Parameters of the Electromagnetic Response to a Mechanical Impulse *Technical Physics* 51 (4) 2006: pp. 519 – 521.
8. **Surzhikov, A., Fursa. T.** Mechanoelectrical Transformations upon the Elastic Impact Excitation of Composite Dielectric Materials *Technical Physics* 53 (4) 2008: pp. 462 – 465.
9. **Reid, A.**, **Judd, M.** Ultra-wide Bandwidth Measurements of Partial Discharge Current Pulses in SF_6 *Journal of Physics D: Applied Physics* 40 2007: pp. 2948 – 2953.
10. **Kravchenko, V., Lazorenko, O., Pustovoit, V., Chernogor, L. F.** A New Class of Fractal Ultra-Wideband Signals *Doklady Physics* 52 (3) 2007: pp. 129 – 133. http://dx.doi.org/10.1134/S1028335807030019
11. **Anisimovas, F., Ašmontas, S., Kiprijanovič, O., Matuzas, J.** Radiation of Large Bandwidth and Long Duration Signals by Pulsed Excitation of Monopole Antennas *Proc. Intern. Conf. "Radiation Interaction with Material and its Use in Technologies" May 14 – 17, Kaunas, Lithuania* Kaunas, Technologija, 2012: pp. 574 – 577.
12. **Ašmontas, S., Anisimovas, F., Dapkus, L., Gradauskas, J., Kiprijanovič, O., Senulis, M.** Electromagnetic Radiation of Ferroelectric PZT Ceramics under Pulsed Compressive Stress *Materials Science* (*Medžiagotyra*) 16 (3) 2010: pp. 264 – 267.
13. **Okazaki, K.** Ceramics Engineering for Dielectrics. Energija, 1976 (in Russian).
14. **Kudriavtsev, E., Lotkova, E., Zotov, S., Autric, M., Emel'yanov, V., Rogacheva, A.** Temperature Dependence of the Velocity of a Slow Soliton Wave of Changes in Optical Reflection and Conductivity Excited by a Laser Pulse in Solids *Laser Physics* 10 (3) 2000: pp. 733 – 740.
15. **Fridkin, V.** Ferroelectrics-Semiconductors. Nauka, Moscow, 1976 (in Russian).
16. **Zhukov, S., Genenko, Yu., Hirsch, O., Glaum, J., Granzow, T., Seggern, H.** Dynamics of Polarization Reversal in Virgin and Fatigued Ferroelectric Ceramics by Inhomogeneous Field Mechanism *Physical Review B* 82 2010: 014109 p.
17. **Rabkin, L. M., Ivanov, V. N.** Electron Energy in Exoelectronic Emission from a Ferroelectric *Technical Physics Letters* 24 (7) 1998: pp. 565 – 566.
18. **Sadykov, S., Borodin, V., Agalarov, A.** Reversible Polarization of Ferroelectric Ceramics in a Rapid-Growing Electric Field *Technical Physics* 45 (6) 2000: pp. 776 – 779. http://dx.doi.org/10.1134/1.1259719
19. **Rezinkin, O., Axelsson, A., Vytrishko, V.** Test Bench for Investigating the Pulsed Polarization Dynamics of Nonlinear Dielectrics *Instruments and Experimental Techniques* 53 (5) 2010: pp.755 – 760.
20. **Pavlov, A. N., Raevski, I. P., Sakhnenko, V. P.** Electron Emission during Pulsed Polarization Switching of Ferroelectric Ceramics *Technical Physics* 44 (7) 1999: pp. 782 – 785.
21. **Tagantsev, A., Stolichnov, I., Setter, N.** Non-Kolmogorov-Avrami Switching Kinetics in Ferroelectric Thin Films *Physical Review B* 66 2002: 214109 p.
22. **Isogawa, T., Kumashiro, T., Song, Ho-Jin, Ajito, K., Kukutsu, N., Iwatsuki, K., Nagatsuma, T.** Tomographic Imaging Using Photonically Generated Low-Coherence Terahertz Noise Sources *IEEE Transactions on Terahertz Science and Technology* 2 (5) 2012: pp. 485 – 492.