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Electromagnetic Radiation of Ferroelectric PZT Ceramics under Pulsed Compressive Stress

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Wideband electromagnetic pulses were received using indoor television antenna after a pulsed compressive stress of $Pb(Zr_{0.52}Ti_{0.48})O_3$ cylindrical samples. An investigation using a detector shows that it is generated a series of the electromagnetic pulses in the time range of about 1 μs. The electromagnetic pulses appear randomly and have approximately equal amplitude. Number of pulses in the series can vary from 2 to 12. It was also found that after the compressive stress it is generated a high voltage response at the sample. The response was registered by (0 – 100) MHz bandwidth oscilloscope using 1 : 470 voltage divider.

It is assumed that series of electrical pulses of ns time duration is formed just before and at the first μs of the response at the sample. A fast rising leading edge of these pulses causes the electromagnetic radiation. The electric pulses are generated because of competition between newly, stress-induced polarized state, and older state maintaining by opposing electric fields forming at grain boundaries.

The observed phenomenon can be useful for investigation of electric parameters and quality of ferroelectric crystals and ceramics. Possible practical applications of mechanically induced electromagnetic radiation are discussed.

Keywords: PZT ferroelectrics, compressive stress, sign-inverted high-voltage response, domain switching, wideband electromagnetic pulses.

INTRODUCTION

Generation of electromagnetic radiation (EM) is often caused by changes in a material structure. Radio and optical radiation were registered during crystallization of liquids [1]. X-rays were registered during breakdown of solids [2]. The EM radiation of radio frequencies was observed during the breakdown of very thin dielectric fibres [3], where it was found that essentially different in diameter fibres radiate signals of almost the same amplitudes. Recently it was found that the EM radiation of radio frequencies can be also caused by mechanical loading of solid materials. Amongst them are piezoelectric crystals [4], layered and composite dielectric materials [5], and composite materials on the base of concretes [6]. The results show that duration of the EM pulses can vary from microseconds down to nanoseconds, hence indicating that the nature of generation is different. Thus, the generated EM radiation can be a valuable tool to investigate internal structure of composite materials.

As it is known, ferroelectrics are the materials that have spontaneous polarization below Curie temperature T_C. If the applied voltage is increased, it may cause saturation of the polarization or, if the direction of electric field is changed to opposite, it can reduce the polarization to zero value (so called coercive electric field). Ferroelectric and ferroelastic properties are closely entwined in ferroelectrics with perovskite structure. Solid solutions of lead zirconate–titanate $Pb(Zr, Ti)O_3$, or just PZT, are materials of such kind.

Difficulties in understanding of dynamic polarization and repolarization in ferroelectrics were noted by Kurchatov [7]. Modern high resolution technique has disclosed the hierarchy of their internal microstructure.

Papers [8,9] showed that physical properties of PZT ceramics strongly depend on grain boundaries, domain walls and porosity. Paper [10] showed that at pressure levels up to 10^8 Pa the electrical properties of PZT could be modified by elastic change in the distances between ions in the crystal structure, by the changes in domain structure and by creation or annihilation of microdefects.

An electromechanical phenomenon – domain switching – was found during the investigation of repolarization dynamics in ferroelectrics [11 – 13]. There was noted that pressure level up to 10^8 Pa was large enough to induce domain switching and movement or clamping of domain walls. Stress influence on electronic conditions of grain boundaries was shown to play important role in the repolarization and in the domain switching [14].

PZT ceramics have found wide practical applications in mechanical sensor and actuator devices. A promising but still not realized application of PZT ceramics is wireless networks with sensors operating under low power or even without power supply [15]. However up to now the devices containing PZT ceramics elements suffer from limited reliability. Investigations of microscopic processes responsible for the fatigue of the ceramics caused by mechanical loading would help to improve mechanical and electrical stability of the material [16,17].

In this work we report on the registered EM radiation of television frequency range (50 MHz – 850 MHz) generated after pulsed compressive stress of polycrystalline PZT ceramics samples and analyse physical reasons causing the radiation.

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SAMPLES

A commercial PZT composition was used. The samples were cylinders of 4 mm high and 2 mm in diameter. Both circular faces were sputter coated with 200 nm thick Ag electrodes. The crystalline structure of the samples was examined by X-ray diffractometer Bruker AXS D8 using Cu K_{α} radiation with a 2θ step size of 0.02° and a scan rate of one step every 5 s. It was found that all the investigated samples were $Pb(Zr_{0.52}Ti_{0.48})O_3$ polycrystalline alloys and had the tetragonal symmetry ($a = 0.408$ nm, $c = 0.404$ nm). Typical X-ray diffraction (XRD) pattern of the samples is shown in Fig. 1. Some of the samples in the investigated direction had more developed (111) preferred orientation. Statically compressively stressed samples did not show any changes in the XRD patterns at pressures up to 10^8 Pa.

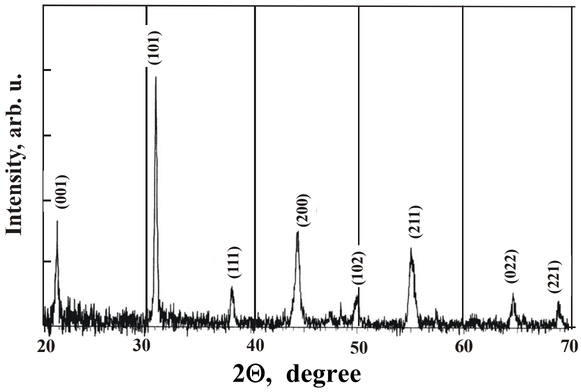

Fig. 1. Typical XRD pattern of the samples. The presented spectrum corresponds to $Pb(Zr_{0.52}Ti_{0.48})O_3$ alloy with tetragonal lattice symmetry

EXPERIMENTAL

Static stress of the samples did not cause a voltage signal rise between the contacts. The force of pressure threshold was determined by applying a set of weights. The pressure of about $0.5{\cdot}10^8$ Pa was found as a ratio of the force and the sample base area. Pulsed mechanical loading was realized by quick compressing of the samples placed into stress limiting device. To avoid a surface arc discharge the samples were coated with 2.5 mm-thick dielectric case. A high voltage response generated under pulsed compressive stress was registered by real time oscilloscope of (0 – 100) MHz bandwidth using 1 : 470 high voltage probe. The resistance of the probe had value of 470 MΩ. The high-voltage response is steadily generated up to 20 loadings, and then, after 10 minutes of rest, properties of the ceramics are regenerated.

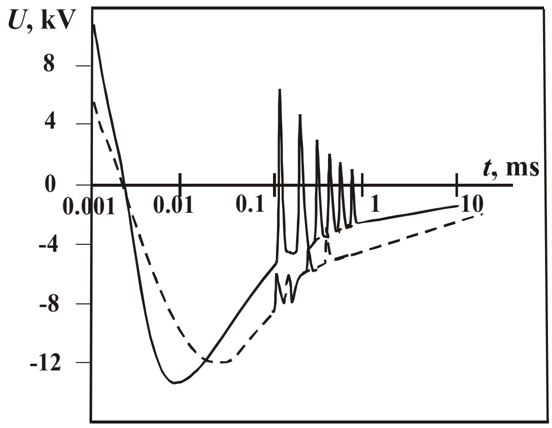

Fig. 2. Typical dynamics of the high voltage response after loading of PZT sample. Dashed curve corresponds to the non-resonant case during relaxation

Dynamics of the high voltage response is presented in Fig. 2. Typically the response changes polarity and relaxes in tens of milliseconds. The sign inversion point appears in the time range from 1 μs to 3 μs. The negative part of the signal reaches maximum at the moment from 10 μs to 15 μs. Series of relatively high voltage peaks of μs duration and of positive polarity appear during the relaxation (after 100 μs). As a rule, the peaks demonstrate periodicity and the series contain up to 6 – 8 peaks (solid curve); however, sometimes they used to appear randomly in the time range from 100 μs to 2 ms (dashed curve).

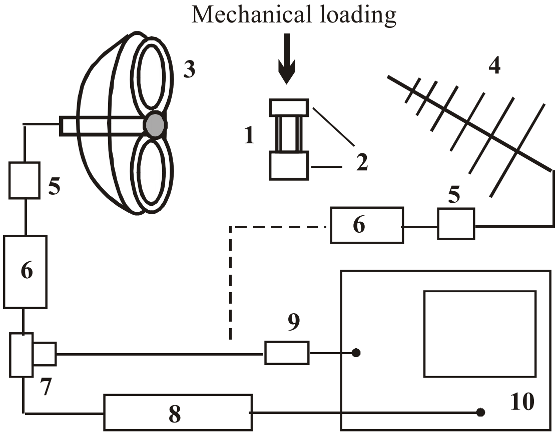

Fig. 3. Schematic of the experimental setup for receiving of EM pulses: 1 – PZT sample, 2 – stress limiting device, 3 – broadband TV antenna, 4 – log periodic antenna, 5 – (75 – 50) Ohm transition, 6 – broad band amplifier, 7 – matched tee, 8 – delay line, 9 – attenuator, 10 – real time broadband oscilloscope

The scheme of experimental setup to receive EM pulses in (50 – 800) MHz frequency range is shown in Fig. 3. The sample (1) was placed near the commercially available broad band TV antenna (3) at a distance of about 20 cm – 30 cm. The signal radiated after the loading was received by TV antenna (3) and transmitted through the (75 – 50) Ohm transition (5), a broadband amplifier (6) (amplification was 25 dB at 500 MHz), and the delay line (8) to the input of the broadband (0 – 5) GHz oscilloscope (10). Matched tee (7) was used to split the synchronizing signal. Attenuator (9) was used to reduce the existing interference signal. The measurements were improved using another synchronization channel (dashed line in Fig. 3) using a log periodic antenna (4).

A broadband (50 – 3000) MHz detector was placed between the tee (7) and the delay line (8) to control the periodicity of the series of wideband pulses shown in Fig. 4. Experiments indicated that the series can contain from 2 and up to 12 pulses in a time window of about 1 μs. As a rule, the pulses demonstrated non-periodicity, but sometimes they used to appear periodically.

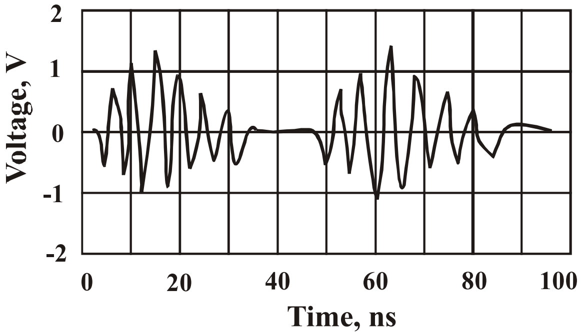

Fig. 4. Typical oscillograms of the wideband pulses from the series radiated by PZT samples

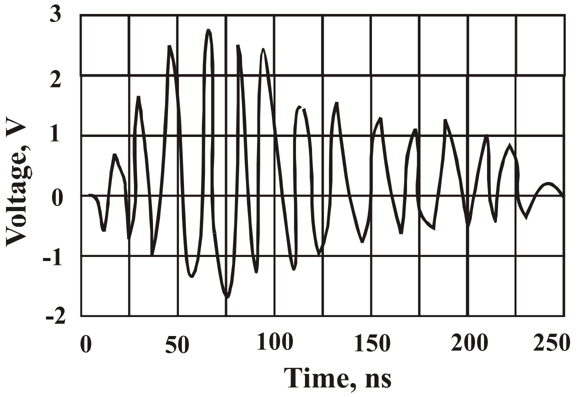

Fig. 5. Single pulse of longer duration and higher amplitude radiated by some of PZT samples

Typical oscillograms of the wideband serial pulses are shown in Fig. 4, while Fig. 5 demonstrates single wideband pulse having higher amplitude. It is worth noting that mostly of samples radiate only serial pulses, while theothers sometimes radiated single pulses. The delay line (8) enabled precisely to identify the moment of EM radiation relatively of the start of the high voltage response.

DISCUSSION

The cylindrical samples were loaded by mechanical pulses. Broadband TV antenna was used to register radiation of wideband EM pulses generated during the loading pulse. Oscillograms of 40 ns and longer pulses are shown in Figs. 4 and 5. Radiation of the EM pulses indicates that there exist short electrical processes during the loading. These could be either subnanosecond electrical pulses or longer electrical pulses having subnanosecond rise time. As a rule, inertial forces resisting to quick changes in the sample act after the end of the loading. The mentioned short electrical pulses as well as high voltage response (see Fig. 2) appear due to polarization and repolarization processes induced by these particular forces.

Barkhausen current pulses [18, 19] are known to result from repolarization processes and appear as transient current pulses accompanying replacement currents. But these are not so fast-rising and are coupled with repolarisation of volumes larger than the grain size. As recent investigations show, repolarisation in thick PZT films can last nanoseconds [20]. Concept of “dynamic coercive electric field” was introduced to explain the fast repolarization processes. It was shown [21] that nanosecond electrical pulses appeared after the removal of external repolarization voltage. The authors pointed out that reaction time of ferroelectric (ionic) subsystem to applied strong electric field was of the order of 10^{-12} s, but it lasted less than 10^{-7} s for electronic subsystem in the volume of the grain and on the grain boundaries and depended on PZT ceramic parameters. Since ferroelastic and ferroelectric properties of PZT ceramics are closely entwined, the similar effect can occur in our case after the removal of mechanical loading.

Polycrystalline ferroelectrics are known to damp external electric field by strong internal electric field existing in Schottky regions at the grain boundaries [14, 21]. Competition between newly polarized states and former states is sustained by electric fields of Schottky barrier-like regions, and after the removal of external voltage the return of the system to initial state is accompanied with generation of electrical pulses of single polarity.

In our case we have similar situation. Dynamic repolarization induces metastability and return of the system to the initial state after the removal of mechanical loading. Most probably this metastability causes fast switching of polarised regions near the grain boundaries inducing fast-rising electrical pulses at the sample contacts; thus EM pulses are generated (see Figs. 4 and 5). Non-periodicity of radiated EM pulse series results from different duration of the induced electrical pulses having the same amplitudes. These EM pulses do not have perfect sinusoidal forms. This means that TV antenna possibly limits higher harmonics of the pulses.

High voltage response having polarity inversion (see Fig. 2) is a well-known time dependence of surface charge decrease for ceramic ferroelectrics and other electrets. The measurements show that EM radiation is generated just before and at the first μs the rise of the positive part of the electrical response. High voltage growth could stop the generation of ns electric pulses. On the other hand, of high voltage pulses are applied to PZT ceramics to induce fast interactions between spontaneous and space-charge polarization [22] which also leads to possible ns pulse generation. This generation depends on the properties of PZT ceramics. EM radiation also could be generated with high voltage peaks appearing after 100 μs (see Fig. 2). Further experiments are needed to clarify these considerations.

High voltage peaks appear with a time of periodicity about 100 μs. This generation can be understood as a result of elastic oscillations of cylindrical sample as a whole, after loading during relaxation, thus confirming the strong coupling between ferroelectric and ferroelastic properties in PZT.

Differences between Fig. 4 and Fig. 5 indicate that there exist differences or different processes in the investigated samples. Hence it seems to be possible to distinguish internal parameters of the samples. For example, the EM pulse amplitude should depend on density of surface states of grain boundaries, which can be controlled during the manufacturing.

The investigated phenomenon of EM generation in PZT materials can find application in sensors where mechanical energy is transformed into the electromagnetic one.

CONCLUSIONS

Cylindrical $Pb(Zr_{0.52}Ti_{0.48})O_3$ polycrystalline samples were axially compressively stressed with abrupt stress cut off. Using a wideband (50 MHz – 850 MHz) indoor television antenna it was registered the series of wideband EM pulses of ns duration in a time window of about 1 μs. The pulses are randomly distributed in the time window, number of pulses in series can reach up to 12 and they are of approximately the same amplitude. Investigations, using 0 MHz – 100 MHz bandwidth oscilloscope and high voltage divider representing 470 MΩ load, show appearance of a sign-inverted high voltage response at the samples. The processes of the series generation and of the high voltage growth are both of the 1 μs duration.

It is assumed generation of series of short (tens of ns) electrical pulses of different durations after the end of the loading with fast rising leading edges that causes generation of the EM pulses.

The observed phenomenon can be useful for investigation of electric parameters and quality of ferroelectric crystals and ceramics. It also can be used in ferroelectric sensors realizing mechanical–electro and mechanical–electromagnetic transformations.

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