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# PLZT ceramic shutters: properties and application

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The voltage dependence of relative light intensity transmitted through a 9/67/33 PLZT ceramic plate at room temperature has been investigated. The static and dynamic electrooptic properties of PLZT modulator with Kerr effect have been described. The possible applications of those new electrooptical elements in different techniques and scientific disciplines have been discussed.

## Introduction

PLZT* ceramic is a new electrooptical material. It was made for the first time in USA in 1969 [1]. The transparent electrooptic ceramics is a solid homogeneous solution of the La-modified lead zirconate — titanate material. These ceramics come from the $PbTiO_3$ —rich side of the $PbZrO_3$–$PbTiO_3$ phase diagram and contain more than 5% of La substituted for Pb. Lanthanium contents and Zr:Ti ratio in solid solution of Pb, $La(ZrTi)O_3$ determine a fundamental phase of the stuff in the room temperature. We can distinguish three basic phases: ferroelectric and antiferroelectric. The characteristic properties of PLZT stuffs are their phase transitions influenced by temperature or applied electric field.

The elements of electrooptic ceramics are made by hot-pressing techniques. The range of hot-pressing varies within 800–1300°C temperature and depends on the remaining conditions and parameters of hot-pressing, such as pressure, time of hot-pressing, etc.

The production technology of transparent PLZT ceramics with the required properties is much complicated, involving the purest raw materials and a very accurate processing. The composition of primary material should approach the stoichiometric one, while their crystallities must have adequate dimensions. The size of the latters has a real influence [2] on electrooptic properties and should be carefully selected to individual applications. The PLZT ceramics with the Zr:Ti ratio equal to 65:35 and the content of atomic lanthanium ranging from 6 to 12% seems to be the most frequently investigated material in the literature. Compositions, in which Zr:Ti ratio is high as 65:35 are the optimum ones with respect to ferroelectric, electrooptic, and other properties.

Owing their electrooptical properties this ceramics are used in light modulation and for the construction of electrooptical measuring systems. The ceramic light modulator has many advantages when compared with mono-crystalline light modulators. The light modulators usually contain mono-crystal, the birefringence of which

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* Pb(lead) — La(lanhanium) — Zr(zirconium) — Ti(titanium).

is controlled by means of the electric field created in the crystal. To this end, the linear electrooptic effect (Pockles effect) or quadratic electrooptical effect (Kerr effect) [3] can be used. In such cases the voltage of the full light modulation (halfwave voltage) may reaches even several kilovolts.

The application of crystals to the light modulation involves great difficulties, related to the preparation of good electrooptical crystals [4, 5], which are often hygroscopic and sometimes are rapidly getting aged and frosted. These crystals need suitable cutting, axis orientation, grinding and polishing of walls. The electrooptical properties of crystals are strongly temperature dependent. Therefore the applications of the crystalline light modulators to measuring systems are limited.

The ceramic light modulators are free of these shortcomings. In free state all electric domains in PLZT sample are arranged chaotically and accidentally. The material possesses isotropic optical properties. Under the influence of electric field within the sample the domains pass into the ordered state, and the material in this area acquires features gains the properties of the unaxial optically negative crystal. The ceramic PLZT samples used as light modulators need not be oriented optically and the required degree of modulation is obtained by a suitable shaping and their localization of electrodes.

## Properties of PLZT ceramic modulator

The ceramic plates in this country were for the first time made in Research Work Laboratory of Elpod-Unitra in Warsaw [2]. Then were prepared by mixing up the raw materials and sintering pressing techniques in oxygen atmosphere with simultaneous control of PbO vapour pressure [3]. The obtained product has a homogeneous structure, i.e. a structure of a solid homogeneous solution of Pb, La (Zr, Ti) $O_3$ crystallized in a regular crystall lattice with a lattice coefficient $a = 4.0839$ Å. The crystals are formed as dodecahedron with predominat pentagonal polyhedron.

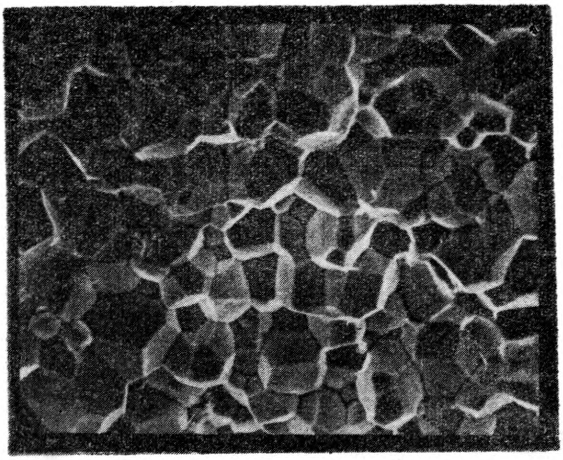

Fig. 1. The structure of the PLZT ceramics examined by means of a scanning microscope, 1 cm–3 μm

The dimensions of crystallites of the obtained materials depend on atmosphere sintering parameters. In our case the size of the crystallites is c. 6 μm.

The grain structure of material examined is shown in fig. 1 and fig. 2.

The remaining electric and piezoelectric properties are shown in the table.

The elements of the ceramic PLZT material are mostly formed in form of circular plates of diameter ranging from 10 to 18 mm and thicknes 100–200 μm. The 9/67/33 PLZT samples 200 μm thick are characteristic of the transparency 35% at Ne–He laser radiation (wavelength $\lambda = 0.63$ μm), and transparency about 26% at unpolarized white light [6]. The element prepared to quadratic light modulation is made as shown in fig. 3. Several metallic electrodes situated on the plate may be placed on one or both its surfaces. The space between electrodes situated on one side of the ceramic plate is a working space of the modulator. To this working space a linear polarized light beam is directed. The proper work of the modulator is conditioned by radiation perpendicular to the surface of the ceramic element. The angle

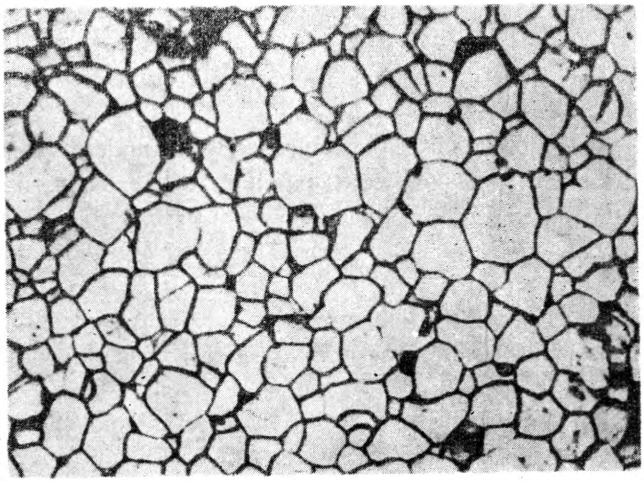

Fig. 2. The structure of the PLZT sample investigated in reflected light by using television microscope, 1 cm–3 μm

Some electric and piezoelectric properties of investigated PLZT ceramics*

| Tan of loss angle tan $\delta$ | Relative dielectric constant $E^T_{33}/E_0$ | Differential resistance $R_d$ [Ω] | Resistivity [Mm] | Poisson coefficient $\sigma$ | Electromechanical factor $Q$ | Electromechanical coupling coefficient $k_p$ | Coercive field $E_c$ [kV/cm] | Residual polarization $P_s$ [C/m²] |
|---|---|---|---|---|---|---|---|---|
| 0.038–0.049 | 3660–4623 | 110–310 | $8.1\cdot10^9$–<br>$-3.1\cdot10^{10}$ | 0.321 | 69–103 | <0.1 | 1.9–2.2 | $(6\text{–}18)^{1}10^{-2}$ |

* All dielectric and piezoelectric measurements were made by the resonanse method, according to the IRE Standard using two-terminal-pair $\pi$ type systems. Measurement of hysteresis loop was made by the Sowyer–Tower method in room temperature at frequency 0.1 Hz. All measurements refer to polarized samples.

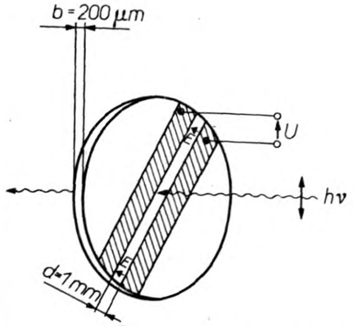

Fig. 3. View of PLZT ceramic modulating plate

between polarization plane of the light beam and direction of electric field formed between the electrodes must be 45°.

The optical axis which arises in the ceramic element under influence of the voltage applied to the electrodes is directed parallelly to the direction of the electric field formed in the material. The voltage applied to the electrodes causes the phasic modulation of the light. The transformation of the latter into the amplitude modulation is done by placing a polarizer behind the plate. Polarization surface of polarizer and the light incident onto the modulator are perpendicular constituting amplitude light modulator. Its properties are examined in the measuring circuit, shown in fig. 4.

The presented measuring system makes possible to measure the static and dynamic characteristic curves, as well as to observe the electrooptic hysteresis phenomenon.

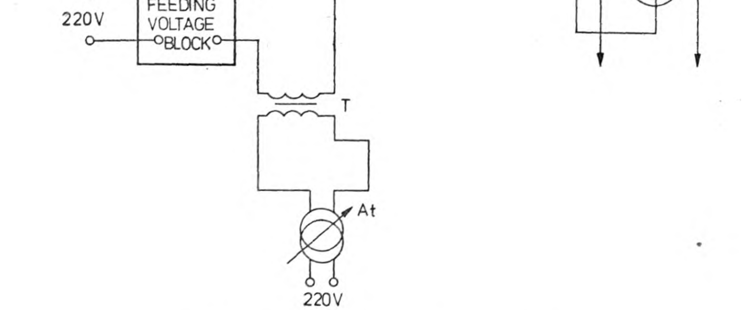

Fig. 4. Scheme of measuring system

*P* — polarizer, *F* — photoelement, *M* — analogue meter of light intensity, *Os* — oscilloscope, *T* — transformer, *At* — feeding voltage regulator

## Static properties

The studies of the static properties depended on the measurement of the intensity of light coming from the ceramic modulator to which direct voltage had been applied. The intensity of light was measured by an analogue meter (fig. 4). As a source of light, Ne–He laser and/or electric bulb were used. To apply the bulb the additional polarizer had to be placed in front of the modulator. The measured light signal is shown in fig. 5 as a function of applied voltage coming from linear photooptic converter.

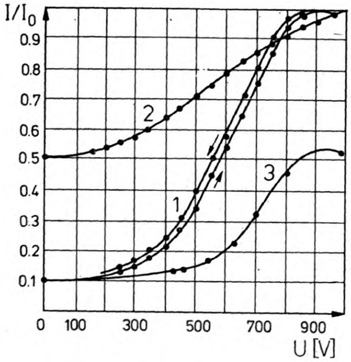

Fig. 5. Static electrooptic characteristics of PLZT modulator at room temperature ($t = 22^{0}$C): $I_0$ — intensity of light coming into the photodetector $F$ (fig. 4) when the polarizer $P$ (fig. 4) is parallel to the polarization plane of the laser beam

$I/I_0$ — relative intensity of light, *1* — $\lambda = 0.63$ μm, 2 — for unpolarized white light, *3* — mean value of light intensity, $\lambda = 0.63$ μm vs. the amplitude of alternation voltage of frequency $f = 50$ Hz

Curve *1*, obtained by using of the Ne–He laser, is non-linear. It is, however, linear from 450 to 800 V in the voltage range. This range of linearity amounts to 40% of the whole range of full light modulation voltage. It is clear that the said modulator shows an optic hysteresis, connected with a residual polarization of ferroelectric material.

Similar measurements were made at unpolarized white light (curve *2*). In this case higher intensity of light observed at $U = 0$ seems to be related — in our opinion — to scattering of unpolarized white light passing through the tested sample which is stronger than that of monochromatic laser light. Hence, a depth of modulation defined as

$$m = \frac{I_{\max}-I_{\min}}{I_{\max}+I_{\min}}$$

is equal to 82% for laser light and only to 30% for the white light.

When the alternating voltage of frequency $f$ is applied to the electrodes of modulator, the modulated light beam with frequency $2f$ is obtained. In this case it is possible to measure the mean value of the modulated intensity of light for different values of amplitudes of the applied voltage (curve *3*). Such measurements were made for laser light only. This method makes it possible to achieve 70% of the depth of the modulation and can be successfully used in the electrooptic measuring systems.

The white light beam might be similarly modulated. Its modulation allows to achieve the characteristic curve with a small gradient and a little (23%) depth of modulation.

## Dynamic properties

The dynamic electrooptic properties of the said modulator have been measured in the circuit shown in fig. 4. The alternating voltage was set by voltage regulator, the time and voltage processes of the modulated light intensity being observed on the oscilloscope.

When the sinusoidal voltage alone is applied to the electrodes (fig. 6), the light beam modulated with duble-frequency is obtained behind the modulator. The time process of the modulated light is deformed. After an additional polarization of the sample with direct voltage 625 V the working point of the modulator move to the centre of the linear section of the electrooptical characteristics (fig. 5). In such a case the alternating electrical signal evokes a much greater alternating optical signal This signal is non formed and its frequency is in agreement with that of the modulating voltage.

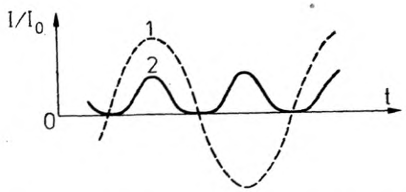

Fig. 6. Timing of modulating voltage (curve *1*) and the corresponding range of light intensity timing (curve *2*). Laser beam $\lambda = 0.63$ μm

The shape of curve shown in fig. 6 was not deformed in the frequency range of 20–4000 Hz. In higher frequency range the measurements were not performed.

If the voltage in a form of the rectangular unipolar impulse is applied to the electrodes of the PLZT ceramic modulator, then the time delay of the sample reaction may be noticed (fig. 7). At the moment of switching on the voltage the intensity of the transmitted light reaches the maximum value very speedily, and after 60 s consolidates on the level of 80% of maximum value. A sudden switch-off of the voltage does not cause any speedy regaining of the initial state of the sample. The initial state of the sample becomes established within about 2 min. along the exponential curve.

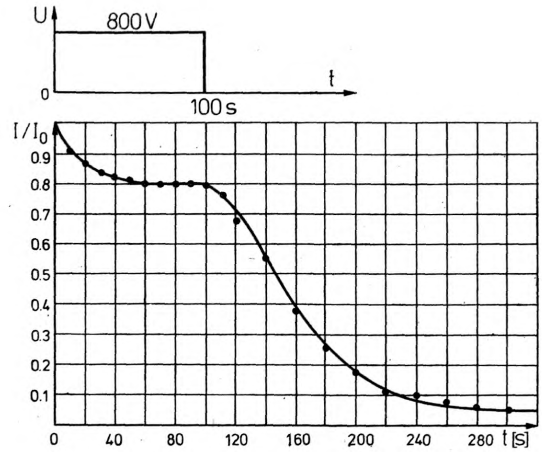

Fig. 7. Response of 9/67/33 PLZT ceramic modulator to a unipolar rectangular impulse of voltage, $t = 22°C$. Laser beam $\lambda = 0.63$ μm

Similar phenomena may be observed if the voltage applied to the electrodes of PLZT modulator has the form of rectangular bipolar impulse (fig. 8). At the moment when the polarity of the sample is changed, the light intensity is reduced to the minimum very speedily, and after 2 min. it reaches again its consolidated value.

The delay time of the modulator reaction upon the rectangular electrical impulse, is explained by relaxation of ferroelectric domain and the influence of the sample capacity. The capacity of the sample is particulary great when the electrodes are placed on the both surfaces of the plate.

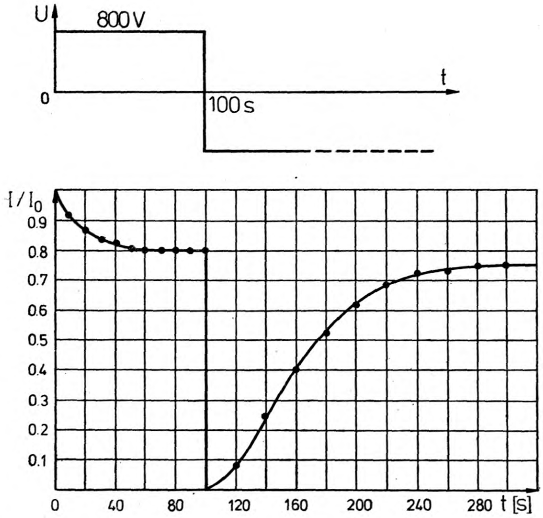

Fig. 8. Response of 9/67/33 ceramic modulator to a bipolar rectangular impulse of voltage, $t = 22°C$. Laser beam $\lambda = 0.63$ μm

The investigated sample displayed also an electrooptic hysteresis, which is shown in fig. 9. The hysteresis loop was determined in the measuring circuit (fig. 4), but the alternating voltage applied to the modulator electrodes was also applied to the *X*-plates of oscilloscope. With the gradually increasing frequency the hysteresis loop was getting narrower and moved along *Y*-axis.

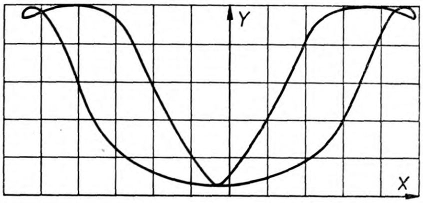

Fig. 9. Electrooptical hysteresis loop of 9/67/33 ceramic modulator, $t = 22^{0}\mathrm{C}$. Laser beam $\lambda = 0.63\ \mu\mathrm{m}$, $X = 200$ V/div, $Y = 0.2 \frac{I}{I_0}$ /div, $f = 50$ Hz

The above picture of the electrooptical hysteresis loop of the ceramic modulator was observed when the direct polarizing voltage was 0. At polarizing voltage $U \neq 0$ the hysteresis loop is getting asymmetric, like the typical electric hysteresis loop.

## Applications

Electrooptic properties of the PLZT ceramics result from chemical composition of the relative phase of PLZT, domain structure and size of crystallites. These properties may be modified by the influence of electrical field radiation, for the given wavelength, tension, temperature, etc.

The PLZT materials, due to its different properties, can be used in devices transmitting information in form of the pictures or $\alpha$-numeric signs, in laser technique as light modulator and in holographic and digital memory systems. They can also find application in the protective eyepieces and windows, filters, shutters, optical voltage sensors, electrooptic converters, logical systems, etc.

The ceramic material which differ from one another by electrooptical properties differ also by other parameters such as coercive field, dielectric constant, electromechanical coupling coefficient, as well as form and gradient of the electric hysteresis loop. Depending on the composition and technology of the PLZT ceramic various effects (Kerr effect, Pockles effect, linear electrooptical light modulation, dynamic scattering of light and optical memory effect) may be observed. The optical memory effect depends on the permanent birefringence which is forced by presence of residual polarization.

The ceramic memory elements can be substitute the ferroelectric monocrystals and other memory elements used up to now. In particular, the ceramic memory cells can replace the liquid crystal memory cells [7].

The capacity of monocrystals and other memory elements is limited because of their small storage capacity, low accessibility, small recording-velocity and obliteration of information, etc. For example, the monocrystals of unrestricted shapes and relatively large sizes cannot be obtained. Besides the preparation of homogeneous crystals is faced with difficulties.

The advantage of electrooptic ceramic PLZT materials lies mainly in their ability of selective memorization. They are also characterized by a high spatial resolving power, high density of bits, high contrast, short switching time, and low feeding voltage.

Lastly, the firm Unitra-Elpod has produced the PLZT of 9/67/33 composition, and 50% transmittance for the white unpolarized light and of transmittance 30% for the laser light.

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## Керамический оптический вентиль PLZT: свойства и применение

Описаны электрооптические свойства оптоэлектронных элементов — керамических пластинок PLZT со складом 9/67/33 — работающих подобно модуляторам света с эффектом Керра. Описаны также статические и динамические свойства названных модуляторов, а также применение их в различных областях науки и техники.