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REGULAR ARTICLE

A Piezo-Optical Effect in Lead Nitrate

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A piezo-optical effect has been discovered in Lead nitrate crystals. A possible mechanism of the piezo-optical effect in divalent nitrates is discussed. It's assumed that the cause of the piezo-optical effect in the crystals is the displacement of the Nitrogen atoms relatively to the planes of Oxygen atoms in NO_3^- groups under application of mechanical stress. Due to this shift, the planar NO_3^- groups become pyramidal, with the Nitrogen atoms at the apexes of the pyramids. It is noted that it's precisely such a displacement that can cause a deformation of the optical indicatrix. In the absence of mechanical stress, the optical indicatrix has the shape of a sphere, since light birefringence is not observed in this case. Under mechanical stress the optical indicatrix becomes an ellipsoid. To observe the piezo-optical effect, single crystals of Lead nitrate of equilibrium shape, which is cuboctahedron, were used. These single crystals were grown from a saturated seed solution of $Pb(NO_3)_2$ by a slow temperature decreasing method. The paper describes a setup for piezo-optical coefficients of Lead nitrate measurements, which was specially created for research. It is noted that for crystals of the T_h group the following components of the matrix of piezo-optical coefficients are independent: π_{11}, π_{12}, π_{13}, π_{44}. It where these components or their combinations that we measured. The measurements have shown that the piezo-optical optical coefficients of Lead nitrate have rather large values, so the crystals can be used as a material for piezo-optical transducers.

Keywords: Lead nitrate, Nitrate group, Piezo-optical effect, Piezo-optical coefficients, Optical indicatrix, Birefringence

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1. INTRODUCTION

In a series of recent articles (e.g. [1]) we discussed a mechanism of phase transitions in divalent nitrates $Ba(NO_3)_2$, $Sr(NO_3)_2$ and $Pb(NO_3)_2$. It was noted that a number of phase transitions Pm3 ↔ Pa3 ↔ $P2_13$ occurs in these materials. According to our assumption, phase transitions in these crystals take place due to the special mobility of NO_3^- groups inside unit cell. Pm3 → Pa3 phase transition realizes because of the "freezing" of NO_3^- group rotation around Nitrogen atom that takes place in Pm3 phase. This rotation turns to oscillation of NO_3^- groups around Nitrogen atoms in Pa3 phase. Pm3 → $P2_13$ phase transition occurs due to the displacement of a Nitrogen atom relatively to a plane of Oxygen atoms and such an arrangement of nitrate groups inside the unit cell that it losses an inversion center.

We have made an assumption that the special mobility of NO_3^- groups in divalent nitrates may also lead to existence of piezo-optical effect in these materials.

The linear piezo-optical effect is described by the equation:

$$\Delta a_{ij} = \pi_{ijkl}\sigma_{kl}, \quad (1)$$

where Δa_{ij} is the change in polarization constants (second-rank tensor); π_{ijkl} is the tensor of piezo-optical coefficients (fourth-rank tensor); σ_{kl} is the tensor of applied mechanical stress (second-rank tensor). In short, the fourth-rank tensor π_{ijkl} can be written as a 6 × 6 matrix. Taking into account the Fumi principle, for the T_h of the cubic system, to which divalent nitrates belong, only four components of the matrix of piezo-optical coefficients are not equal to zero: π_{11}, π_{12}, π_{13}, π_{44}. These four components are independent. That is, the matrix of piezo-optical coefficients for divalent nitrates looks as follows:

$$\begin{pmatrix} \pi_{11} & \pi_{12} & \pi_{13} & 0 & 0 & 0 \\ \pi_{12} & \pi_{11} & \pi_{12} & 0 & 0 & 0 \\ \pi_{13} & \pi_{12} & \pi_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & \pi_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & \pi_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & \pi_{44} \end{pmatrix} \quad (2)$$

If a mechanical stress is applied to the sample along [001] direction and light is transmitted along [100] direction, then the induced birefringence is proportional to the difference π_{11}-π_{12}. Similarly, if a mechanical stress is applied to the sample along [100] direction and light is transmitted along [110] direction, the birefringence is found to be proportional to the difference π_{11}-π_{13}. Finally, if light is transmitted along [$\underline{1}$11] direction and a mechanical load is applied along [211] direction, the birefringence is proportional to π_{44}.

2. INVESTIGATION METHODOLOGY

2.1 Methods of Samples Preparation

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For the study, single crystals of Lead nitrate of a high optical quality were grown. The linear dimensions of the crystals reached 45 millimeters. To grow the crystals a special crystallizer was constructed. The crystals were grown on seeds from a saturated aqueous solution by the method of a slow decreasing of temperature. The initial temperature of the solution was 318 K, the final temperature was the ambient one. The rate of temperature decreasing was 0.02 degrees per hour.

The form of the crystals grown in this way was a cuboctahedron – a combination of a cube, or hexahedron (faces (100), less denoted square faces) and an octahedron (faces (111), more denoted hexagonal faces) (Fig. 1).

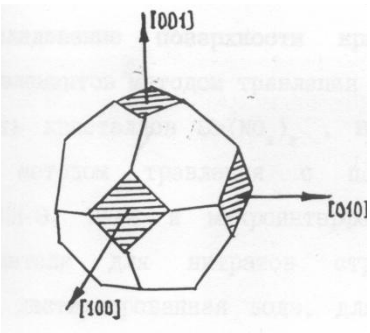

Fig. 1 – Cuboctahedron – an equilibrium facet of divalent nitrate crystals. Shaded rectangles are faces of (100) type; light hexagons are faces of (111) type

$Pb(NO_3)_2$ samples for piezo-optical coefficients measurements were cut from the single crystals obtained in this way. The samples were rectangular plates of high optical quality with (100), (110) and (111) developed facets. The samples were ground with abrasive powder No 5 with addition of machine oil, after that they were sequentially polished with diamond pastes ASM 40/28-NOM, ASM 20/14-NOM, ASM 10/7-NOM, ASM 2/1 NOM until maximum transparency was achieved.

2.2 Methodology of Experiments

The measurements of piezo-optical coefficients were carried out using a polarization-optical method. The basic scheme of the setup for piezo-optical coefficients measurements is shown in Fig. 2.

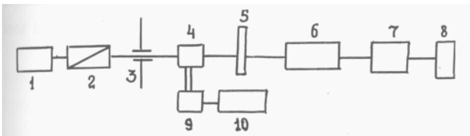

Fig. 2 – Basic scheme of the setup for measuring of piezo-optical coefficients

In Fig. 2: 1 – laser radiation source; 2 – polarizer; 3 – diaphragm; 4 – test sample; 5 – mica plate $\lambda/4$; 6 – analyzer; 7 – photoelectric multiplier; 8 – indicator; 9 – electromagnetic holder for creating a pressure on the sample; 10 – current source.

The crystal plate under study was installed so that the principal axes of the optical indicatrix section, perpendicular to the direction of light propagation, formed angles of 45° with the direction of oscillations in the polarizer. The $\lambda/4$ plate in this circuit is located so that one of it coincides with the direction of oscillations in the polarizer. The analyzer can rotate freely relatively the axis of the system. The transmission of such optical system can be expressed as follows:

$$I = \cos^2[(\pi/\lambda)\cdot d\cdot \delta\Delta n - \varphi] \quad (3)$$

where λ – wavelength of light; d – the optical path length of light in the sample; $\delta\Delta n$ – induced birefringence; φ is an analyzer rotation angle. It is obvious that by changing the angle φ, it is possible to achieve a situation where the induced phase difference will be compensated and the transmission of the system I will be equal to zero, which is achieved under the condition:

$$\delta\Delta n = [\lambda/(d\cdot\pi)]\cdot(\varphi + \pi/4) \quad (4)$$

The load on the sample placed in the electromagnetic holder was set using the source of a current. By turning the analyzer, we achieved the minimum throughput of the system, which corresponded to the minimum on the indicator. After this the rotation angle of the analyzer was fixed. The values of the piezo-optical coefficients were found using the formula:

$$\pi_{pq} = 14{,}1\cdot[a/(n_0^3\cdot F)]\cdot\gamma[brewster] \quad (5)$$

where a – sample thickness (in millimeters), n_0 – refractive index of the sample at zero load, F – the load on the sample (in Newtons), γ – analyzer rotation angle (in degrees). The absolute measurement error of piezo – optical coefficients using the described setup was 0, 125 brewster.

3. RESULTS OF THE EXPERIMENTS AND THEIR DISCUSSION

For a sample of [100] orientation, with a mechanical stress applied along [001] direction, the measurements results are presented in a Table 1. The sample thickness was 4 millimeters.

Table 1 – Dependence of rotation angle γ of analyzer on load F, applied to the sample of [100] orientation

F (10^{-2}N)	γ_1 (degrees)	γ_2 (degrees)	γ_3 (degrees)
0	98.0	94.0	101.0
20	102.5	100.0	98.0
40	100.5	102.25	103.5
60	105.5	101.0	104.5
80	103.5	101.0	107.0
100	106.0	100.0	105.25
120	103.5	105.5	101.5
140	103.5	103.0	106.0
160	105.5	104.5	106.5
180	104.5	104.5	106.5
200	107.0	106.5	107.5
220	106.0	105.0	108.0

According to the data presented in Table 1, the following values were used for calculations in formula (5): $\Delta F = 2$ N; $\Delta\gamma = 8$ degrees; $a = 4$ mm.

For a sample of [110] orientation, with a load applied along [001] direction, the measurements results are presented in a Table 2. The sample thickness was 5 millimeters.

Table 2 – Dependence of rotation angle γ of analyzer on load F, applied to the sample of [110] orientation

F (10^{-2}N)	γ_1 (degrees)	γ_2 (degrees)	γ_3 (degrees)
0	98.5	95.5	92.75
20	98.5	96.5	96.0
40	95.5	96.75	98.5
60	100.0	99.5	98.0
80	102.0	101.0	99.0
100	99.0	101.25	104.0
120	104.5	102.0	103.5
140	103.5	104.5	104.5
160	102.5	103.5	104.0
180	105.5	105.5	104.5
200	106.5	105.0	105.5
220	105.0	107.0	106.0

According to Table 2, the following values were used for calculation using formula (5): $\Delta F = 2$ N; $\Delta\gamma = 10$ degrees; $a = 5$ mm.

For a sample of [111] orientation, with a load applied in $[\bar{2}11]$ direction, the measurements results are presented in Table 3. The sample thickness was 3 millimeters.

Table 3 – Dependence of rotation angle γ of analyzer on load F, applied to the sample of [111] orientation

F (10^{-2}N)	γ_1 (degrees)	γ_2 (degrees)	γ_3 (degrees)
0	93.7	94.3	92.8
50	94.8	95.0	96.0
100	96.0	95.8	96.2
150	97.0	99.6	98.5
200	99.2	101.4	100.2
250	101.0	103.5	103.5

The following values were used for the calculations in formula (5): $\Delta F = 2$ N; $\Delta\gamma = 8$ degrees; $a = 3$ mm.

Based on the data presented in Tables 1-3, the following values were obtained:

$$\tfrac{1}{2}\, n_0^3(\pi_{11} - \pi_{12}) = -\,42{,}1 \text{ Brewster};$$
$$\tfrac{1}{2}\, n_0^3(\pi_{11} - \pi_{13}) = -\,65{,}8 \text{ Brewster};$$
$$n_0^3\pi_{44} = -\,16{,}0 \text{ Brewster}.$$

It should be noted that

$$1 \text{ Brewster} = n_0^3 \cdot 1 \text{ brewster}.$$

The values of piezo-optical coefficients that we obtained in the study of Lead nitrate single crystals, exceed similar indicators for recognized piezo-optical materials such as KDP and ADP (tetragonal syngony) [2, 3], potassium alum (cubic syngony) [4], some Perovskites (cubic syngony) [5].

In our opinion, the large values of piezo-optical coefficients in divalent nitrates crystals are associated with the high mobility of Nitrogen atom in the NO_3^- groups. Due to such mobility, when mechanical stress is applied to the samples of the studied crystals, NO_3^- group changes from planar to pyramidal. Apparently, the height of the pyramid increases with increasing of mechanical stress. This circumstance leads to a strong dependence of the form of the optical indicatrix on the magnitude of the applied mechanical stress, that is, to large values of the piezo-optical coefficients.

4. CONCLUSIONS

Crystals in which the photoelasticity is observed have found a wide application. Firstly, they have long been used to study mechanical stresses in construction and mechanical engineering. Secondly, after the invention of lasers, interest in photoelasticity increased greatly, since this effect can be used to control laser beams in optical communication lines and signal processing devices. Currently, the photoelastic effect is widely used for modulating and deflecting laser beams, Q-modulating of lasers and obtaining short and powerful radiation pulses, recording of holograms. The large piezo-optical coefficients of Lead nitrate make these crystals promising for use as a material for photoelastic transducers. In the case of possible use of Lead nitrate, as well as Barium and Strontium nitrates, as materials for piezo-optical transducers, it becomes important to study mechanical characteristics, such microhardness and elastic constants in order to determine whether the studied crystals meet the requirements for materials, proper for the manufacture of piezo-optical transducers.

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П’єзооптичний ефект в нітраті свинцю

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В кристалах нітрату свинцю виявлений п’єзооптичний ефект. Обговорюється можливий механізм п’єзооптичного ефекту в нітратах двовалентних елементів. Зроблене припущення, що причиною п’єзооптичного ефекту в кристалах є зміщення атому азоту відносно площини атомів кисню в групі NO_3^- під час прикладання механічної напруги. Завдяки такому зміщенню планарні NO_3^- – групи стають пірамідальними, з атомами азоту при вершинах цих пірамід. Відмічається, що саме таке зміщення може викликати деформацію оптичної індикатриси. У відсутності механічної напруги оптична індикатриса має форму сфери, оскільки у цьому випадку двозаламлення світла не спостерігається. Під впливом механічної напруги оптична індикатриса стає еліпсоїдом. Для спостереження п’езооптичного ефекту були використані монокристали нітрату свинцю рівноважної форми, якою є кубооктаедр. Ці монокристали були вирощені з насиченого розчину $Pb(NO_3)_2$ на затравку методом повільного зниження температури. В роботі описана установка для вимірювання п’єзооптичних коефіцієнтів нітрату свинцю, яка була спеціально створена для цих досліджень. Відмічається, що для кристалів групи T_h незалежними є наступні компоненти матриці п’єзооптичних коефіцієнтів: π_{11}, π_{12}, π_{13}, π_{44}. Саме ці компоненти або їхні комбінації були виміряні. Вимірювання показали, що п’єзооптичні коефіцієнти нітрату свинцю мають доволі великі значення. Таким чином, кристали можуть бути використані в якості матеріалу для п’єзооптичних перетворювачів.

Ключові слова: Нітрат свинцю, Нітратна група, П’єзооптичний ефект, П’єзооптичні коефіцієнти, Оптична індикатриса, Механічна напруга, Двозаламлення.