Investigation of the phase delay of radiation by a transparent ferroelectric polymer film

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Abstract. In this paper, we study samples of an optically transparent ferroelectric polymer film with deposited nanoscale electrically conductive coatings designed to modulate the transmitted electromagnetic radiation of the visible and near-infrared wavelengths. Such films can be used, for example, in interference devices for phase delay compensation or for the implementation of the Phase Shifting Interferometry, in adaptive optics, etc. To measure the phase delay of the radiation passing through the samples under study, an installation based on the Mach-Zehnder interferometer was used. In the illumination branch of the installation, a broadband radiation source and an acousto-optic tunable filter are installed; in one of the arms of the interferometer, the test sample is installed. The interference pattern was recorded on a matrix radiation receiver; the phase information was decoded by digital holography methods. The report presents the results of measurements and shows that a modulation of the passed optical radiation occurs under the influence of the electric field as a result of changes in the geometrical dimensions of the film.

1. Introduction

Transparent piezoelectric films are promising materials for use in various fields of science and technology. On their basis, it is possible to manufacture flexible transparent biocompatible nanogenerators of electricity, pressure, temperature and detection sensors with autonomous power, including for medical purposes, thin-film transparent acoustic devices for flexible electronics of a new generation [1–8].

Promising materials for the manufacture of such piezoelectric films are polyvinylidene fluoride copolymers with tetrafluoroethylene or trifluoroethylene [9] or trifluoroethylene [10], which belong to the class of crystalline polymers in which crystalline and amorphous phases with significantly different elastic and electrical characteristics can exist. Such heterogeneity leads to the manifestation of piezoelectric properties in these materials [11, 12]. These materials are flexible (unlike piezoelectrics on a ceramic basis), broadband in the acoustic range, have a relatively high piezoelectric coefficient (d33 to 30 pC/N on commercially available samples) and high transparency (up to 99% in the visible wavelength range). Also it’s possible to manufacture films of a large area.

For practical application, films based on PVDF copolymers are subjected to texturing, for example, by their uniaxial drawing, as well as polarization by means of exposing of a high-intensity field. After that, the matrix of piezoelectric coefficients acquires 5 nonzero components due to symmetry effects [11, 12].

Such films can also find applications, for example, in interference devices for phase delay compensation or for implementing the phase step method, in adaptive optics, etc. The supply of an AC voltage of an audio frequency with an amplitude of several volts to the transparent electrodes causes the...
stretching-contraction of the polymer film due to the piezoelectric effect and, as a consequence, a change in the optical path length of the transmitted radiation. In this case, an important characteristic of the film structure is the value of the phase delay of radiation at different values of the applied voltage. To determine it, a measurement scheme based on the Mach-Zender interferometer was developed. The technique was tested to determine the phase delay of transmitted radiation by transparent piezoelectric films based on PVDF copolymer.

2. Methods
The studied sample (figure 1) is an optically transparent 16 µm thick ferroelectric film made of vinylidene fluoride and tetrafluoroethylene copolymer (VDF/TFE) F2m grades manufactured by JSC Plastpolymer with the chemical formula \([\text{CH}=\text{CF}_2]_n[\text{CF}_2=\text{CF}_2]_m\), where \(m/n = 0.05\), on which optically transparent electrically conductive ITO coatings 98 nm thick with a specific surface resistivity of 190 Ohm/□ were deposited on both sides by magnetron sputtering. The obtained samples were polarized by applying an alternating sine or triangular voltage with an electric field strength of about 1 V/µm.

![Figure 1. Appearance of the experimental sample.](image)

To experimentally evaluate the phase modulation when the electromagnetic signal passes through the sample, the scheme shown in figure 2 was used.

The collimated beam of radiation produced by a supercontinuum generator (Rock 400 2, Leukos) after expander (2) (with a magnification of 5x) was fed to the input of the Mach-Zender interferometer, in one arm of which sample (O) was installed. The plane of the sample was optically conjugated with the camera sensor plane (8) (DMK 37BUX273, The Imaging Source) via a confocal system consisting of objective (6) \((f = 100 \text{ mm})\) and objective (7) \((f = 75 \text{ mm})\). A light filter (3) was used to create monochromatic radiation.

Next, the matrix photodetector recorded an interference pattern (digital hologram) from two wavefronts, one of which carries information about the phase delay introduced by the sample. Thereafter the registered image was processed using the methods of digital holography [13].

The reflection coefficient of the samples under study was determined using a Shimadzu UV-3600i Plus spectrophotometer in the wavelength range from 380 nm to 1000 nm with a resolution of 1 nm at normal incidence of light on the sample.

3. Results and discussion
The main steps of reconstruction of the spatial distribution of the phase delay are shown in figure 3. At the first stage of processing the Fourier transform of registered digital hologram \(I(x, y)\) was performed. The result of the Fourier transform is a spectrum of the signal, in which 3 main components, i.e. 0-th, 1-th and -1-st orders, are distinguished (figure 3(b))
\[ \tilde{I}(\nu_x, \nu_y) = \tilde{A}(\nu_x, \nu_y) + \tilde{C}(\nu_x - \nu_{0x}, \nu_y - \nu_{0y}) + \tilde{C}^* (\nu_x + \nu_{0x}, \nu_y + \nu_{0y}), \]  

(1)

where the Fourier transformation and complex conjugation are marked by tilde and asterisk, correspondingly, 

\[ C(x, y) = \frac{1}{2} B(x, y) e^{i \Delta \phi(x, y)}, \]

\[ B(x, y) = 2 \sqrt{I_R(x, y) I_S(x, y)} \]

is the amplitude of the interference fringes, \( I_R(x, y) \) and \( I_S(x, y) \) are the intensities of the reference and sample waves, correspondingly, and \( \Delta \phi(x, y) \) is the phase delay to be determined.

Figure 2. Scheme (a) and photo (b) of the setup for measuring phase modulation: O – object, 1 – laser (or broadband source + filter), 2 – beam expander, 3 – light filter, 4 – beam splitting cubes, 5 – mirrors, 6, 7 – lenses, 8 – matrix photodetector.

After centering of the -1 order in the spectrum (figure 3(c)) the inverse Fourier transform was performed, and the phase \( \Delta \phi(x, y) \) of the signal was reconstructed (figure 3(d)). The value of phase delay was calculated by digital holography methods and averaged over the sample area of 2.5x2.5 sq mm.

Since the operating frequencies of the voltages applied to the samples are unity kilohertz, and accordingly the value of phase delay varies with the same frequencies, in each mode a series of several images (from 10 to 15) was registered, each with an exposure time of 1/10000 sec, which is significantly less than the period of variation of the introduced phase delay. Images in each series were recorded at arbitrary moments of time in order to obtain images recorded at moments of voltages close to the maximum and close to the minimum with high probability.

The phase delay was measured for each image, and from the set of measured phase delays the magnitude of its change \( \Delta \phi \) was established as the difference between the maximum \( \phi_{\text{max}} \) and the minimum \( \phi_{\text{min}} \) phase delay in the series of measurements. The same measurements were also performed.
in the absence of voltage in order to estimate the random errors of the results. The results of phase delay measurements are shown in table 1.

![Figure 3](image1.png)  
(a)  
![Figure 3](image2.png)  
(b)  
![Figure 3](image3.png)  
(c)  
![Figure 3](image4.png)  
(d)  

**Figure 3.** Stages of digital hologram processing: a – registered image $I(x, y)$, b – Fourier transform of the original image; c – selected and centered -1 order in the Fourier image $\hat{C}(\nu_x, \nu_y)$; d – phase delay $\Delta \phi(x, y)$.

| Measurement series number | Switching on the sample | Measurement results |
|---------------------------|-------------------------|---------------------|
|                           | $U$, V                  | $f$, kHz            | $\phi_{\text{max}}$, rad | $\phi_{\text{min}}$, rad | $\Delta \phi$, rad |
| 1                         | 0                       | 0                   | 1.54                      | 1.37                      | 0.17                |
| 2                         | 5                       | 2                   | 1.73                      | 1.40                      | 0.33                |
| 3                         | 5                       | 3.7                 | 1.89                      | 1.25                      | 0.64                |
| 4                         | 9                       | 3.7                 | 1.90                      | 1.27                      | 0.63                |

From table 1 we can conclude that the value of $\Delta \phi$ in the absence of voltage is significantly lower than when voltage is applied in either mode, indicating the presence of phase modulation when AC voltage is applied to the sample.

The transmission spectrum of the test sample is shown in figure 4. The transmittance coefficient calculated for the wavelength range of 380–780 nm in accordance with GOST R 54164-2010, is 74.8%.
4. Conclusion

Thus, it was found experimentally that when an alternating voltage is applied to the studied sample, phase modulation of the optical radiation passing through it occurs. At the same time, the sample has a sufficiently high light transmission (~75%).

For more detailed conclusions, for example, concerning the dependence of the depth of phase modulation on the parameters of the applied voltage (shape, amplitude and frequency) it is necessary to perform more detailed studies, for example, to increase the sensitivity of the installation by improving the optical system, reducing the exposure time or increasing the number of images to be registered.

Acknowledgments
This work was supported financially by the Ministry of Science and Higher Education of the Russian Federation within the state task № 0705-2020-0032. The results of phase delay measurements were obtained using the equipment of the Scientific and Technological Center of Unique Instrumentation of the Russian Academy of Sciences (STC UI RAS) [http://ckp.ntcup.ru].

References
[1] Park H K, Lee K Y, Seo J S, Jeong J A, Kim H K, Choi D and Kim S W 2011 Advanced Functional Materials 21 1187
[2] Kim K B, Jang W, Cho J Y, Woo S B, Jeon D H, Ahn J H and Sung T H 2018 Nano Energy 54 91
[3] Sun J G, Yang T N, Wang C Y and Chen L J 2018 Nano Energy 48 383
[4] Jeong C K, Hyeon D Y, Hwang G T, Lee G J, Lee M K, Park J J and Park K I 2019 Journal of Materials Chemistry A 7 25481
[5] Kang J H, Jeong D K and Ryu S W 2017 ACS Applied Materials & Interfaces 9 10637
[6] Lin L, Hu Y, Xu C, Zhang Y, Zhang R, Wen X and Wang Z L 2013 Nano Energy 2 75
[7] Lee K Y, Gupta M K and Kim S W 2015 Nano Energy 14 139
[8] Do Y H, Jung W S, Kang M G, Kang C Y and Yoon S J 2013 Sensors and Actuators A: Physical 200 51
[9] Kochervinskii V V, Kiselev D A, Malinkovich M D, Pavlov A S and Malyshkina I A 2015 Colloid and Polymer Science 293 533 https://doi.org/10.1007/s00396-014-3435-1
[10] Kochervinskii V V, Kiselev D A, Malinkovich M D, Korlyukov A A, Lokshin B V, Volkov V V, Kirakosyan G A and Pavlov A S 2017 Crystallography Reports (Russia) 62 324
[11] Kochervinskii V V 2003 Crystallography Reports (Russia) 48 649

Figure 4. Transmission spectrum of the test sample.
[12] Kochervinskii V V 2003 *Polymer Science Series B* **45** 326

[13] Machikhin A S, Polschikova O V, Ramazanova A G and Pozhar V E 2017 *Journal of Optics* **19** 075301