Available online at www.sciencedirect.com

SCIENCE DIRECT®

Journal of the European Ceramic Society 24 (2004) 1709–1714

www.elsevier.com/locate/jeurceramsoc

Electrical properties of photovoltaic lead lanthanum zirconate titanate in an electrostatic-optical motor application

M. Ichiki[a,*], Y. Morikawa[b], Y. Mabune[c], T. Nakada[c]

[a]*Institute of Mechanical System Engineering, National Institute of Advanced Industrial Science and Technology, 1-2 Namiki, Tsukuba, Ibaraki 305-8564, Japan*

[b]*Intelligent System Institute, National Institute of Advanced Industrial Science and Technology, 1-2 Namiki, Tsukuba, Ibaraki 305-8564, Japan*

[c]*School of Information Environment, Tokyo Denki University, 2-1200 Muzai-Gakuendai, Inzai, Chiba 270-1382, Japan*

Abstract

An electrostatic-optical motor is proposed as a new application device that uses photovoltaic lead lanthanum zirconate titanate (PLZT) as an energy transducer from optical energy to electrical energy. The photovoltaic electrical power output of the PLZT is transformed into electrostatic force to rotate the optical motor. Movement of the motor was observed but the response was second order. A number of problems must be solved in order to realize a high-performance electrostatic-optical motor. One is the improvement of the photovoltaic properties for high rotation power and fast response speed. Another problem is justification of the mechanical construction of the motor. The last issue is clarification of the mechanisms of the photovoltaic effect of lead lanthanum zirconate titanate to obtain a suitable sample size and electrical circuit. The poling conditions on samples were investigated to improve PLZT for the electrostatic-optical motor. Since electrical conductivity, including dark conductivity and photovoltaic conductivity, plays an essential role in the mechanisms of the photovoltaic effect, the electrical properties of the photovoltaic PLZT were examined quantitatively. A linear relationship between electrical conductivity and light intensity was confirmed in the authors' experiments.

Keywords: Electrical conductivity; Electrical properties; Functional applications; Perovskite; PLZT

1. Introduction

Various kinds of micro-fabrication technologies have been developed including device design, simulation, lithography, film formation, and structure and system integration. Ferroelectrics are useful materials since they have both piezoelectric and pyroelectric properties. These characteristics are applicable to many micro-electrical components, e.g., sensors, actuators, controllers and transducers. It is necessary to develop and establish a new energy transfer method to supply energy to micro-electromechanical systems (MEMS) [1–3] and micro-opt-electromechanical systems (MOEMS). In particular, wireless energy transfer has received much attention[4] in recent years. It is useful to apply photovoltaic materials, e.g., a lead zirconate titanate (PZT) system such as lead lanthanum zirconate titanate. Lead lanthanum zirconate titanate, i.e., $Pb_{1-x}La_x(Zr_yTi_z)_{1-x/4}O_3$ (abbreviated PLZT or PLZT($X/Y/Z$), where $X=100x$, $Y=100y$ and $Z=100z$), is a ferroelectric solid solution with wide-ranging material properties that depend on its composition.[5] PLZT(3/52/48) has photostrictive properties that are the superposition of photovoltaic[6] and inverse piezoelectric effects. The photostrictive effect is caused by illumination in the near-ultraviolet region. These materials can be used in MEMS devices to directly convert optical energy to mechanical energy, and the first of such devices proposed were opt-mechanical actuators.[7] This property is useful for energy conversion in MEMS and photo-acoustic devices.[8] Some reports have been published dealing with both the application[8,9] and material[10,11] aspects.

The objective of this research is to improve the photovoltaic effect of PLZT to realize a high-performance electrostatic-optical motor.[12] There are a number of problems to be solved. One is the improvement of the

* Corresponding author. Tel.: +81-298-61-7140; fax: +81-298-61-7169.

E-mail address: ichiki-m@aist.go.jp (M. Ichiki).

0955-2219/$ - see front matter © 2003 Elsevier Ltd. All rights reserved.
doi:10.1016/S0955-2219(03)00474-6

photovoltaic properties for high rotation power and fast response speed. Another problem is justification of the mechanical mechanisms in device construction. The last issue is clarification of the mechanisms of the photovoltaic effect of lead lanthanum zirconate titanate to obtain a suitable sample size and electrical circuit. It is also necessary to quantitatively clarify the electrical properties to investigate the phenomenological mechanisms of the photovoltaic effect.[13–15]

The poling conditions and estimation of photoconductivity were investigated to improve the characteristics of the electrostatic-optical motor. This paper describes the actuation principle of the electrostatic-optical motor and the estimation of the photovoltaic effect, and some experimental results of the photovoltaic effect will be presented in the following sections.

2. Electrostatic-optical motor and experimental set-up

A new mechanical rotation device, called an ''optical motor[12]'', was realized through the use of photovoltaic PLZT. Electrostatic force per unit mass is, generally speaking, inversely proportional to volume. It is, therefore, effective to use a small-scale electrostatic force for efficiency as well as in accordance with the driven device's smaller size. The driving principle of an optical motor using PLZT is shown in Fig. 1. The PLZT electrode is connected electrically with the stator pads of the optical motor which are arranged in parallel with each other. A moving pad is positioned between the stator pads with only some overlap. When near-ultraviolet light is illuminated onto the PLZT, electrical voltage is induced between the stators which draw the moving pad to the left side in Fig. 1(a).

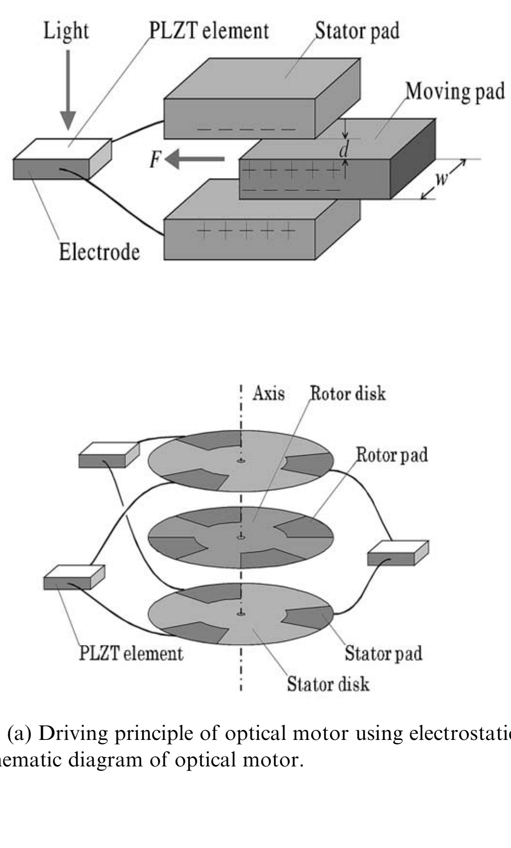

Fig. 1. (a) Driving principle of optical motor using electrostatic force. (b) Schematic diagram of optical motor.

Fig. 1(b) is a schematic of an optical motor in its simplest form. The system consists of three stators and four moving pads, the fewest number of components possible for this system. A pair consisting of a stator and a pad is connected electrically to the PLZT. A rotor disk is fixed to the motor axis which is held in place using a ball bearing. When the PLZT is illuminated, high electric voltage in the kilovolt order is induced and introduced into the connected stator electrically. The rotator is pulled toward the stator in the clockwise direction when viewed from the top.

The conventional solid-state reaction method was used[10,11] to prepare the PLZT(3/52/48) samples. The fired samples were cut into 10×10 mm on the sides and 1 mm in thickness. Archimedes method is used to obtain the density. The density of the samples was 7.7 g/cm^3, which corresponds to a value above 96% compared to the theoretical density. LCR meter was used to obtain

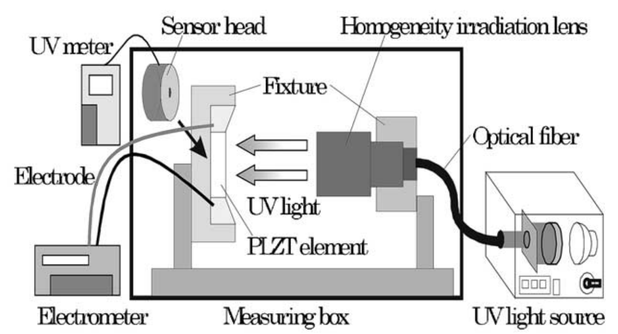

Fig. 2. Experimental apparatus for estimating photovoltaic properties.

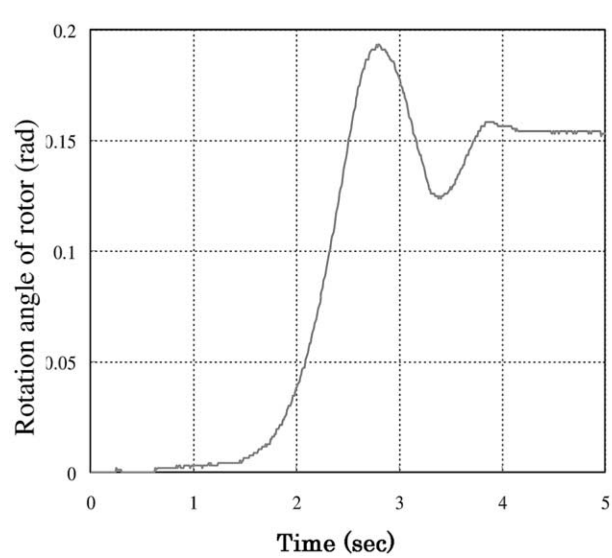

Fig. 3. Rotation angle of optical motor.

the dielectric constant. The value of the dielectrics was around 1000 for all samples, which is an average value for a ferroelectric PLZT.

Light intensity supplies an input energy which directly affects the response of the photovoltaic current. Generally speaking, the stronger the light intensity, the faster the photovoltaic response; however, the nonuniformity of the light intensity distribution also becomes larger at the same time. Therefore a homogeneous illumination lens was employed in our experimental system to maintain uniform light intensity even in the strong light intensity region. The experimental setup for the estimation apparatus is shown in Fig. 2. This apparatus consisted of an optical light source (UV light), a mount for the sample and an electrometer. The light was introduced into the measuring box through an optical fiber after being filtered to monochromatic light around 365 nm. The light was illuminated onto the PLZT through the homogeneous irradiation lens. A homogeneous irradiation lens is effective for accurate

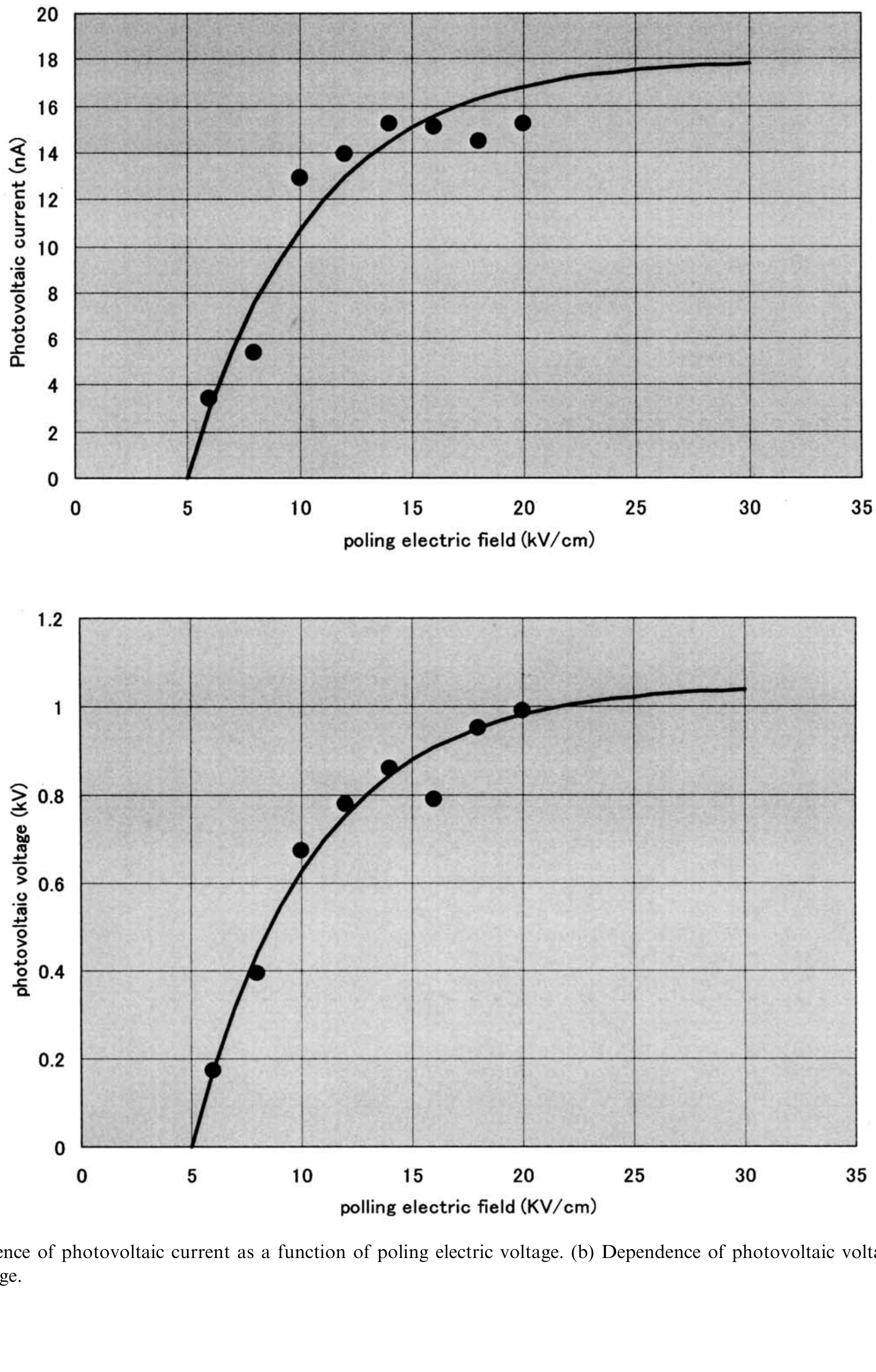

Fig. 4. (a) Dependence of photovoltaic current as a function of poling electric voltage. (b) Dependence of photovoltaic voltage as a function of poling electric voltage.

measurement of electrical properties. Photovoltaic voltages were measured using the electrometer in voltage application mode, in which we could measure the photovoltaic voltage indirectly, i.e., the photovoltaic current could be measured for an application voltage between-1 kV. The photovoltaic voltage is equal to the cross-point value of the I–V line and the V axis.[15] The distribution of the light intensity is about 10% in a 1 cm^2 region, which is equal to the size of the PLZT.[16] This means that uniform illumination was realized in the area of the sample.

An optical motor was tested for actuation using PLZT and the actuation has been confirmed.[12] In that study, an optical motor with five stators was used for the experiment. Fig. 3 shows the experimental result of dynamic observation of the electrostatic optical motor. The optical motor could be rotated but its rotation speed was slow, around second order. The motor started to rotate around 0.6 s after illumination and stopped at 2.5 s and 0.157 rad (9 deg.), at which point the rotor and stator were overlapped completely. This slow response is due to the high resistance of PLZT[13,17] in that it takes a long time to store up the electrostatic force with photovoltaic voltage over the maximum static friction force. More detailed discussions in view of form mechanics are now underway and will be presented in a separate paper.

There are, therefore, a number of problems to be overcome to achieve a high-performance electrostatic-optical motor; improvement of the PLZT characteristics, justification of the mechanical construction of the optical motor, and the clarification of the mechanisms of the photovoltaic effect. In the following sections some experimental results will be presented on the poling conditions of PLZT for material improvement and the estimation of photo-electrical conductivity for the photovoltaic effect. There are various process factors in the improvement of the PLZT characteristics, e.g., material composition, firing condition, poling, surface roughness and light intensity. Among these factors, poling conditions and photovoltaic effect have not yet been examined fully. These issues will be addressed in the next section.

3. Experimental results and discussions

Fig. 4 shows the relationships between (a) the photovoltaic current and the poling electric field, and (b) the photovoltaic voltage and the poling electric field. Both the photovoltaic current and voltage increase together with the increasing of the poling electric field and they approach the constant value gradually. The asymptotic value of photovoltaic voltage is 1.05 kV. To utilize the maximum power of PLZT it is therefore necessary to do the poling procedure at over 20 kV/cm.

Fig. 5 shows the relationship between the photovoltaic electrical current and the induced voltage (i.e., I–V line). The photovoltaic current and applied voltage have a linear relationship. The induced photocurrent was about 20 nA with 178 mW/cm^2 light intensity without applied voltage. The photovoltage of PLZT is around 2000 V, and this could be obtained at the point at which it crosses the voltage line. The slope of the tangent corresponds to electrical conductivity. The specific electrical conductivity was calculated from the

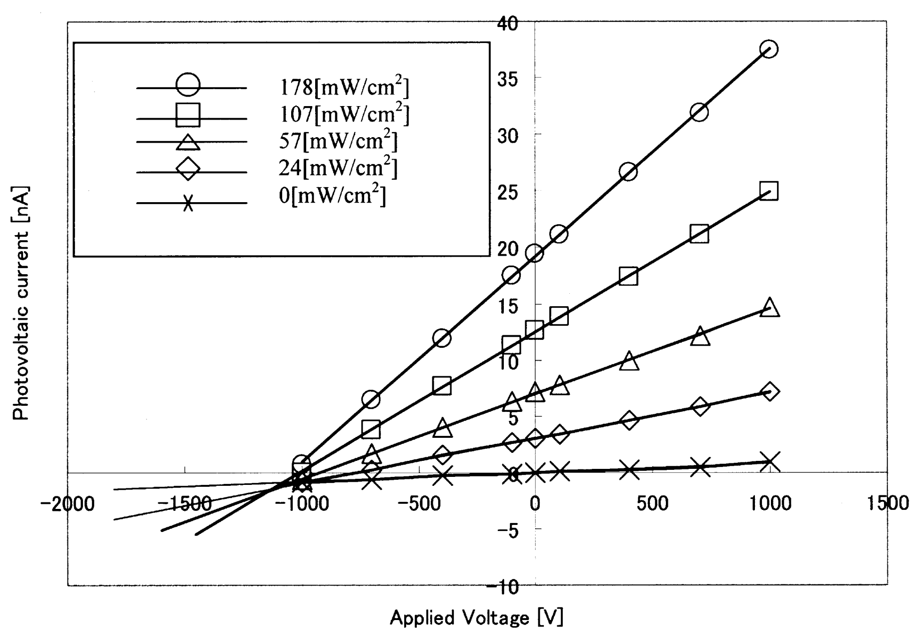

Fig. 5. Dependence of photovoltaic current as a function of applied voltage in several conditions of light intensity.

electrical conductivity and the sample size. Electrical conductivity σ_ is expressed in relation to light intensity $\boldsymbol{I}$ up to first term approximation as follows;[6]

$$\sigma = \sigma_d + \sigma_{ph} I \quad (1)$$

where σ_d is dark conductivity and σ_{ph} is photoconductivity.

Fig. 6 shows the relationship between light intensity and electrical conductivity. Conductivity was calculated from the slope of the linear line in Fig. 5 by the least-squares method. The relationship in Fig. 6 is almost linear up to the maximum light intensity region. Previous reports on this relationship are not in agreement; some report it to be linear[13, 14] and others nonlinear.[18] Our experiments were performed with less than 50 mW/cm^2 illumination. In Fig. 5 the lowest line indicates no illumination and the others are different intensities of illumination. The photo- and dark conductivities were 6.3×10^{-12} mΩ^{-1}W^{-1} and 8.8×10^{-10} Ω^{-1}m. These values were more accurate than those previously reported[13,14] at weaker intensity. Table 1 gives a comparison of electrical conductivity in previous studies and the present study. It was concluded that the above-described estimation method with the homogeneous lens was useful for obtaining the material properties of the photovoltaic effect. The light intensity was adequate for estimating the characteristics. The induced photovoltaic voltage was also better than that in previously reported values.[16]

Table 1
Comparison of PLZT electrical conductivity

Dark conductivity Ω^{-1}m^{-1}	Photoconductivity Ω m W^{-1}	Reference
2.0×10^{-12}		11
2.7×10^{-11}	1.8×10^{-10}	14
1.7×10^{-10}	1.2×10^{-11}	13
8.8×10^{-10}	6.3×10^{-12}	This study

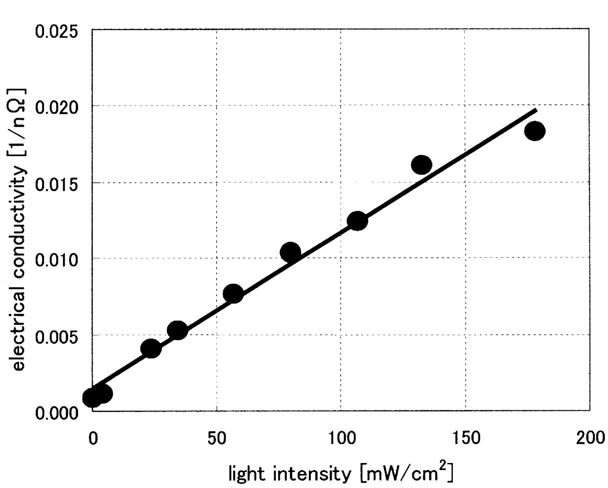

Fig. 6. Dependence of electrical conductivity as a function of light intensity.

We have confirmed that there is a linear relationship between light intensity and electrical conductivity at more than 100 mW/cm^2. Our results are consistent with some previously reported findings at less than 50 mW/cm^2 .[13,14] A nonlinear relationship has also been reported[18] but it was not observed in the light intensity region of our study. The estimation method may be sufficient for obtaining over 100 mW light intensity. Nonaka et al. recently reported that electrical conductivity could be controlled by the material composition.[11] The relationship between composition and response must, therefore, be clarified. Although there might exist a nonlinear term in the high-intensity light region, such nonlinearity was not observed in our experiments. Therefore, we have concluded that our experimental results were within the limitation of the durability of linearity in Eq. (1). Accurate photoconductive properties were obtained in this study.

4. Summary

The electrical properties of photovoltaic PLZT were examined accurately using an electrometer. The use of a homogeneous irradiation lens was advantageous for achieving uniform illumination on the PLZT surface within 10%. An optical motor rotated by electrostatic force was proposed as an application of photovoltaic PLZT. It was shown that a poling condition over 20 kV/cm is necessary to obtain the electrostatic force of PLZT. The obtained photo and dark conductivities were 6.3×10^{-12} mΩ^{-1}W^{-1} and 8.8×10^{-10} Ω^{-1}m. These values were estimated more accurately in the higher light intensity region than those in a previous report.

Acknowledgements

The authors thank Dr. R. Maeda and Dr. K. Nonaka (National Institute of Advanced Industrial Science and Technology, AIST) for their useful discussions, Ms. K. Tanaka (AIST) for the sample preparation, and Ms. T. Imai (AIST) for the manuscript preparation. The authors also thank Dr. Y. Tsutsui and Dr. K. Tanie (AIST) for their encouragement during this study. This study was partially supported by Grant-in-Aid for Young Scientists, 14750044 (KAKENHI).

References

1. Schroth, A., Lee, C., Matsumoto, S. and Maeda, R., Application of sol-gel deposited thin PZT film for actuation of 1D and 2D scanners. *Sensors and Actuators*, 1999, **73**, 144–152.

2. Matsumoto, S., Klein, A. and Maeda, R., Development of bi-directional valve-less micropump for liquids. *Proc. of IEEE Micro Electro Mechanical System*, 1998, 141–146.
3. Chu, J., Wang, Z. J., Maeda, R., Kataoka, K., Itoh, T. and Suga, T., Novel multibridge-structured piezoelectric microdevices for scanning force microscopy. *J. Vac. Sci. Tech.*, 2000, **B18**, 3604–3607.
4. Iddan, G., Merson, G., Glukhohvsky, A. and Swain, A., Wireless capsule endoscopy. *Nature*, 2000, **405**, 417.
5. Haertling, G. H. and Land, C. E., Hot-pressed (Pb, La)(Zr, Ti)O_3 ferroelectric ceramics for electro-optic applications. *J. Am. Ceram. Soc.*, 1971, **54**, 1–11.
6. Stuman, B. I. and Fridkin, V. M., *The Photovoltaic and Photorefective Effects in Noncentrosymmetric Materials*. Gordon and Breach Science Publishers, Amsterdam, 1992.
7. Brody, P. S., Optomechanical bimorph actuators. *Ferroelectrics*, 1983, **50**, 27–32.
8. Chu, S. Y. and Uchino, K., Photoacoustic devices using (Pb, La)(Zr, Ti)O_3 ceramics. *Proc.9th Int. Sym. Appl. Ferroelectrics*, 1994, 743–745.
9. Morikawa, Y. and Nakada, T., Bimorph type optical actuator using PLZT elements. *Jpn. Soc. Mech. Eng. Int. J.*, 1998, **C41**, 860–866.
10. Nonaka, K., Akiyama, M., Hagio, T. and Takase, A., Effect of multiple impurity doping on the photovoltaic properties of lead zirconate titanate ceramics. *Ferroelectrics*, 1999, **223**, 357–364.
11. Nonaka, K., Akiyama, M., Hagio, T. and Takase, A., Effect of Pb/(Zr + Ti) molar ratio on the photovoltaic properties of lead zirconate titanate ceramics. *J. Eur. Ceram. Soc.*, 1999, **19**, 1143–1148.
12. Morikawa, Y., Ichiki, M., Kawaguchi, K. and Nakada, T., Mechanisms of an electrostatic optical motor using PLZT elements. *Ext. Abstr. the 13th Symp. Electromagnetics and Dynamics*, 2001, 693–696 (in Japanese).
13. Nakada, T., Cao, D. H., Kimura, M. and Hui, C. Y., Study on optical servo system (Modeling for photovoltaic effect in PLZT elements). *J. Mech. Soc. Eng.*, 1992, **58**, 2513–2518.
14. Nonaka, K., Akiyama, M., Hagio, T. and Takase, A., Bulk photovoltaic effect in reduced/oxidized lead lanthanum titanate zirconate ceramics. *Jpn. J. Appl. Phys.*, 1995, **34**, 2344–2349.
15. Ichiki, M., Morikawa, Y. and Tanaka, M., Electrical properties of lead lanthanum zirconate titanate. *Ferroelectrics*, 2001, **262**, 149–154.
16. Kawaguchi, K., Ichiki, M., Morikawa, Y. and Nakada, T., Electrical properties of photovoltaic PLZT and its application to optical motors. *Ferroelectrics*, 2002.
17. Poosanass, P., Dogan, A., Thakoor, S. and Uchino, K., Influence of sample thickness on performance of photostrictive ceramics. *J. Appl. Phys*, 1998, **84**, 1508–1512.
18. Tonooka, K., Poossanaas, P. and Uchino, K., Mechanisms of the bulk photovoltaic effect in ferroelectrics. *Proc. SPIE*, 1998, **3324**, 224–232.