Photo-induced currents in the sandwich metal-ferroelectric-metal structure

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Abstract. It was described the photoelectric effect in thin sandwich metal-ferroelectric-metal system. The effect was observed in doped lithium niobate crystals with two electrodes of different metals. The effect is observed only in doped lithium niobate crystals and has a maximum for concentrations of impurities of iron around 0.3 % weight. This paper proposed thermal model of the investigated phenomena resulting from field contact potential difference on the borders of section of metal-ferroelectric material. The results obtained can be used to develop radiation receivers, as well as in the interpretation of experimental results on studying the properties of sandwiched metal-ferroelectric-metal structure.

1. Introduction

It is well known the photovoltaic effect (PVE) in lithium niobate with iron and copper impurities. It has been established that the density of steady-state current $J$ is proportional to the laser intensity $\Phi(t)$: $J_{ph} = \alpha K_G \Phi(t)$, where $K_G$ is the Glass constant dependent on the nature of absorbing centers and wavelength of radiation and independent of the crystal geometry, electrode configuration and impurity concentration, $\alpha$ is the absorption factor [1]. In paper [2] the thermovoltaic effect consisting in initiation of the electric voltage between the opposite faces of the semiconductive sample of samarsucilphide (SmS) on heating to 400–500K is described. The effect has appeared in the presence of the donor impurity concentration gradient directed towards these faces. The electric voltage has initiated in the absence of temperature gradient along the direction of the electrodes’ arrangement on the sample on heating and, therefore, cannot be explained by the classic Seebeck effect. There are a number of such nonequilibrium phenomena in the ferroelectrics which have no unambiguous theoretical description. In paper [3] the thermally-stimulated EMF arising only in the highly-alloyed with iron crystal of lithium niobate with the evaporated electrodes from a pair of different metals was described. A sign of the thermo-EMF is determined by location of electrodes applied by vacuum evaporation on the opposite faces of crystal and does not depend on orientation of the crystallographic axes of the sample in reference to electrodes.

In this paper, the results obtained in experimental studies of the light induced current in the thin-layer metal-lithium niobate-metal structure are presented. This effect is similar to photovoltaic effect, but it is characterized by thermal nature.
2. Thermovoltaic effect in the sandwich-like metal-lithium niobate-metal structure

In contrast with another thermal classic pyroelectric effect (classic pyroelectric effect), the value of observed thermal response is proportional to the crystal temperature increase and does not depend on the rate of its change. Formula for the thermally-induced voltage across the load resistance $R_n$ is, in general case, as follows:

$$U_n = \gamma SR_n \frac{dT}{dt} + U_{n0} + P_{el} SR_n \Delta T e^{-\frac{t}{\tau}},$$  \hspace{1cm} (1)

where $S$ is the area of crystal surface covered with sputtered (evaporated) electrode; $P_{el} = (R_n S)^{-1} \Delta U_n / \Delta T$ is the coefficient characterizing the thermo-EMF value in the mode of voltage $U_n$ measurement (A/K·cm$^2$); $\Delta U_n$ is the change of voltage across $R_n$ when the sample temperature changes by $\Delta T$; $\Delta T$ is the difference between the initial and final temperatures of sample; $U_{n0}$ is the initial voltage at $T_0$ ($T_0$ is the initial temperature of crystal); $\tau$ is the time of thermal response relaxation; $\gamma$ is the pyroelectric coefficient; $t$ is the time.

The first term in (1) corresponds to the classic pyroeffect while the second one describes the non-equilibrium thermovoltaic effect proper outstanding substantially of the general signal owing to large relaxation time ($\tau \geq 10^5$ s).

The study of the effect was performed using the slow modulation of the crystal temperature under the conditions of thermostat and the thermo-EMF was recorded, inter alios, under the stationary conditions (for observation time $\tau \sim 10^5$ s, relaxation was not observed).

The measurements showed that the thermo-voltaic effect has approximately equivalent values in the $Y$- and $Z$-cut crystals. Because the sufficiently high rates of heating (about 0.1 K/s) were used in the installation, the pyroelectric current for the polar $Z$-cut crystals reached the considerable value. Therefore, the principal measurements were performed for the $Y$-cut lithium niobate crystals where the thermal response could be observed in pure form.

The metal electrodes were applied by vacuum evaporation with thickness varying from 0.1 to 1 $\mu$m. The samples with different pairs of electrodes (aluminum (Al) – chromium (Cr), indium (In) – chromium (Cr), aluminum (Al) – copper (Cu), silver (Ag) – aluminum (Al)) were studied.

For studying the dependence of the thermo-EMF value on the concentration of impurity, the lithium niobate crystals with the same areas and thicknesses [$S = 0.5$ cm$^2$, $d = 1$ mm] were used. The iron concentration varied in the range of $1.3 \cdot 10^2$ wt% to 0.6 wt%. The results are given in Fig. 1. For the impurity concentration of less than 0.25 wt%, the thermo-voltaic effect is not observed. Beginning with the impurity concentration of 0.25 wt%, the coefficient $P_{el}$ rises sharply and reaches maximum ($P_{el} = 5 \cdot 10^{-12}$ A/cm$^2$K) at concentration of $0.3 \div 0.4$ wt% of Fe.

The known contact phenomena in the sandwiched structures with ferroelectrics do not explain the observed quasi-stationary currents [4, 5]. The presence of initial voltage $U_{n0}$ across the sample allows us to suggest the non-equilibrium nature of the thermally stimulated current.
When investigating the dependence of the coefficient $P_{el}$ value on the sample geometry, it was identified that $P_{el}$ value increases sharply with the crystal thickness decrease (Fig. 2).

When increasing the heating range, it was found that $P_{el}$ depends nonlinearly on the sample temperature (Fig. 3).
A sharp increase in the thermally induced current for small thicknesses of crystal (less than 100 μm) points to the crucial role of the near-contact zones with thicknesses of less than 100 μm. In this paper, the thermo-chemical model of the studied phenomenon caused by the field of contact potential difference at the metal-ferroelectric interface was proposed [4]. In case of the same electrode materials, the contact potential differences are equal and oppositely directed. Therefore, the resulting EMF is equal to zero. The model assumes that the electrochemical oxidative reaction proceeds in the area of metal-ferroelectric contact which leads to the appearance of electrochemical potential gradient [5].

3. Conclusions
Therefore, it is evident from the research results that in the thin-layer metal-ferroelectric-metal system the thermo-voltaic response exists which increases essentially with decreasing the crystal thickness. This effect can be comparable to photovoltaic effect in light experiments.

The results obtained can be used to develop the radiation detectors as well as to interpret the experimental results of investigating the properties of the sandwiched pyroelectric structures [5, 6].

References
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