

Effects of illumination on the dielectric response of barium-strontium niobate ceramics

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Abstract. A study of the effects of white light on the low and infra-low frequency relaxation of polarization in the barium-strontium niobate (SBN) ceramics is reported. The light is found considerably decreasing the contribution of space charge at temperatures corresponding to the range of the relaxor phase (it is, around the T_m).

1. Introduction

Presently the studies of photosensitive materials are of high importance, for instance, the photo-response of submicron capacitor structures such as $M/Pb(ZrTi)O_3(PZT)/M$ to visible or UV irradiation in relation to possible use of photovoltaic currents [1,2] for non-destructive reading of information. However, toxicity (evaporation of lead) during production of the PZT materials used in a variety of devices is urging to look for alternative materials without lead one of which being barium-strontium niobate (SBN) [3]. Since long the ferroelectric (FE) $Sr_{0.75}Ba_{0.25}Nb_2O_6$ (SBN-75) attracts attention of researchers as a photosensitive ceramic material [1,4,5].

The present study is focused on the effect the illumination by light at low intensity has on the dielectric response at low and infra-low frequencies in the SBN-75 ceramics.

2. Samples and methods

The sample of barium-strontium niobate $Sr_{0.75}Ba_{0.25}Nb_2O_6$ (SBN-75) used for the study was obtained by conventional ceramics technology. Dielectric measurements were performed on a plate of the size of $6mm \times 5mm \times 0.88mm$ supplied with silver paste electrodes. One electrode had several series of perforations 0.5 mm in diameter. A 5034W2C-DSA-A LED providing “cold” visible radiation at the intensity of $0,15 \text{ mW/cm}^2$ was used as the source of light directed at right angle to the perforated electrode parallel to the measuring field E . Since depth of penetration by visible light of that intensity into opaque ceramics is negligible, the effects were assumed to take place mainly in the layer adjacent to the perforated electrode, and surface area of the electrode was used in calculations of the dielectric response.

Measurements of polarization loops in dark and under light were performed sequentially heating up the sample beforehand cooled below the room temperature to a series of selected fixed thermostatic temperatures. Polarization loops were obtained under continuous illumination at different field amplitudes and frequencies of 0.1, 1, and 10 Hz. After that light was switched off and the next temperature set up to repeat the cycle of measurements.



Effective dielectric permittivity $\varepsilon'_{eff} = P_E / \varepsilon_0 E_m$ and dielectric loss ($\varepsilon''_{eff} = tg\delta \cdot \varepsilon'_{eff}$, $tg\delta = \frac{S}{\pi E_m P_E}$) where E_m is the amplitude of the sinusoidal field at measuring polarization loops, P_E – polarization at E_m , ε_0 – the dielectric constant, S – the polarization loop area in the P - E coordinates, were calculated from data obtained on a Sawyer-Tower circuit modified to be used connected to a computer.

3. Results and discussion

The shapes of polarization loops (PL) at frequency 0.1 Hz and temperatures of 14 °C, 20 °C, and 62 °C of dark (a) and illuminated (b) sample are shown in figure 1. The selected temperatures cover the range of the broad phase transition in SBN-75 ceramics as found [6] from $\varepsilon'(T)$ and $\varepsilon''(T)$ measurements at low field intensities.

As seen from figure 1, a considerable relaxation of polarization is displayed by the rounded PL at 62 °C in agreement with the conclusion [6] of an essential effect of conductivity around T_m of the material. The same general pattern of PL is maintained at 23 °C; only at $T < T_{room}$ (14 °C) a typical response of a FE material is displayed by the Rayleigh pattern of PL at amplitude $E < E_C$ (E_C – coercive field) [6]. Comparing the loops in dark and illuminated sample at 62 °C reveals a decrease of the loop area – narrowing of the loops caused by illumination. Effective parameters – effective dielectric permittivity ε'_{eff} , effective dielectric loss ε''_{eff} and $tg\delta_{eff}$, obtained from PL are used hereafter for quantitative assessment of the effects of illumination on the polarization process.

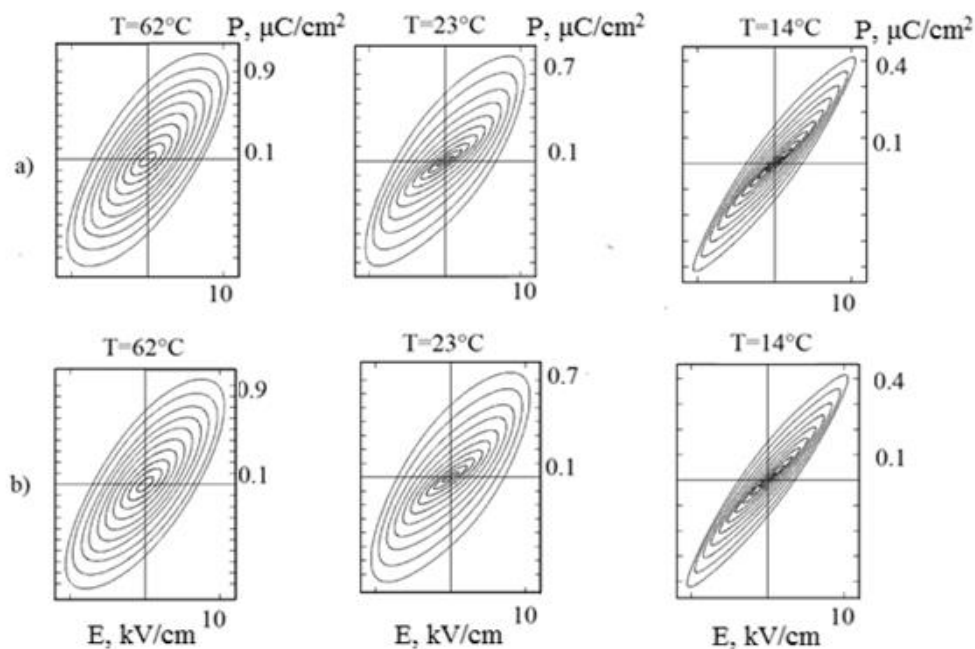


Figure 1. Dependence of polarization loops in SBN-75 ceramics on field intensity at frequency 0.1 Hz: a – in dark, b – at illumination.

Dependence on field intensity of $\varepsilon'_{eff}(E)$ and $\varepsilon''_{eff}(E)$ at frequency of 0.1 Hz at different temperatures in dark and illuminated sample (figure 2) shows anomalies on both curves at 62 °C. In the first case (a) – as a local maximum on $\varepsilon'_{eff}(E)$ at $E \approx 8$ –8.5 kV/cm and a flattening on the $\varepsilon''_{eff}(E)$ curve in the 6–8 kV/cm range (b). In the other case – as flattening of $\varepsilon'_{eff}(E)$ at $E \approx 8$ –10 kV/cm (c) and as a bending point on the $\varepsilon''_{eff}(E)$ curve in the 7–8 kV/cm amplitude range (d). Behaviour of the parameters of SBN-75 at 62 °C, which is below $T_m \approx 110$ °C [6], might point to saturation of polarization $P(E)$ as the cause of the additional maximum of $\varepsilon'_{eff}(E)$. The field intensity at which the broad maximum appears on $\varepsilon''_{eff}(E)$ would correspond to coercive field E_c when the material in the FE

state starts “switching” under applied field exceeding E_c . However, the oval pattern of PL at 62 °C (figure 1) remaining the same at all field intensities does not allow relating the anomalies to the FE nature of switching polarization.

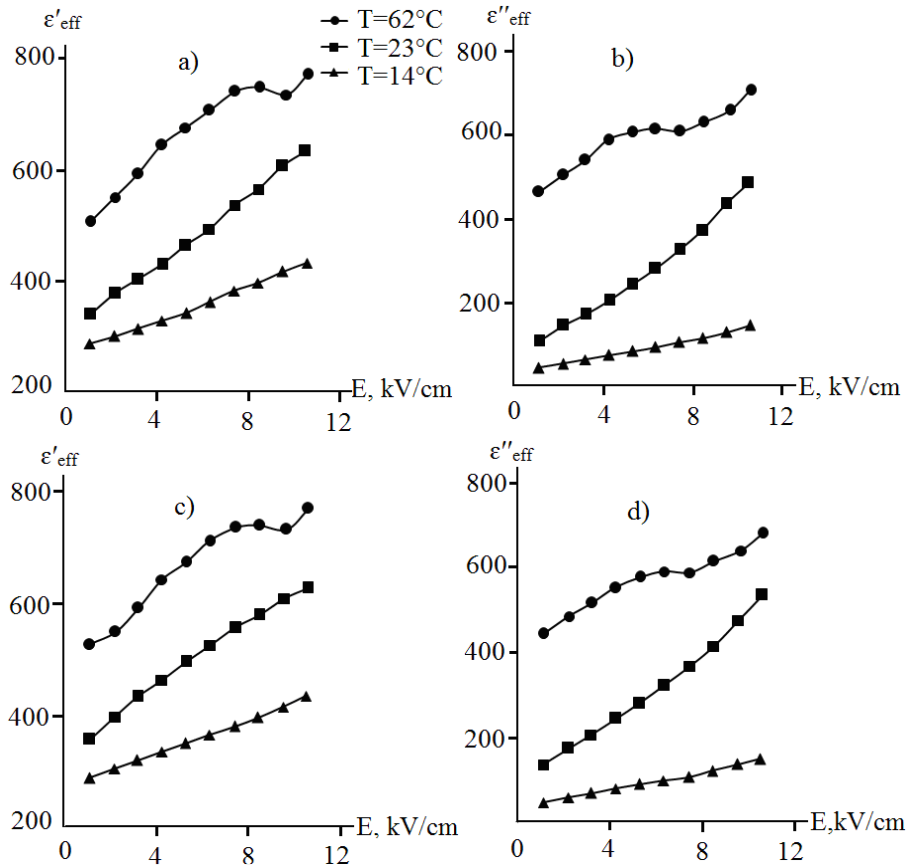


Figure 2. Dependence of $\epsilon'_{eff}(E)$ and $\epsilon''_{eff}(E)$ on field intensity at 0.1 Hz frequency at temperatures $T = 14^\circ\text{C}$, $T = 23^\circ\text{C}$ and $T = 62^\circ\text{C}$ in dark (a, b) and illuminated (c, d) sample.

Most likely the anomalies are due to a substantial relaxation of space charge at 62°C and frequency 0.1 Hz the maximum contribution made by field intensity amplitudes between 4 and 8 kV/cm. An evidence of it can be provided by $\text{tg}\delta_{eff}$ (figure 3) the magnitude of which is proportional to the area of the polarization loop.

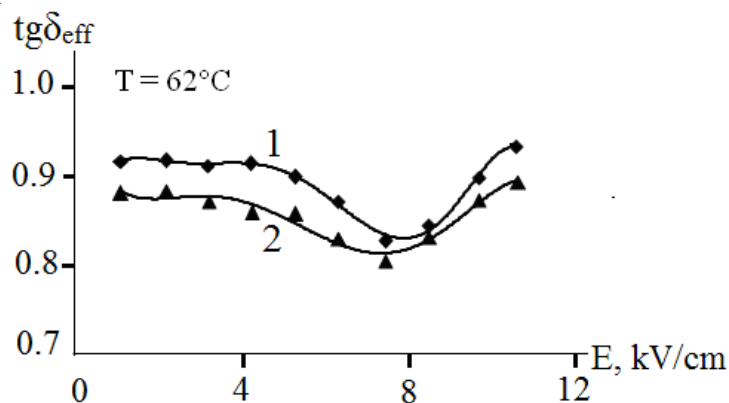


Figure 3. Field dependence of $\text{tg}\delta_{eff}(E)$ at frequency 0.1 Hz in dark (curve 1) and illuminated (curve 2) sample.

As follows from comparison of effective parameter values of dark and illuminated sample, the light mainly affects the behaviour of $tg\delta_{eff}$ and ε''_{eff} . Illumination decreases the values of $tg\delta_{eff}$ and ε''_{eff} while shifting the anomalies of $\varepsilon_{eff}(E)$ and $\varepsilon''_{eff}(E)$ to a stronger field intensity E .

Since the main contribution to relaxation of polarization at infra-low frequencies and $T > T_{room}$ is provided by space charge, the decrease of dielectric loss observed at illumination of the sample might be related to the effect of it on conductivity of the material. In the present case, unbalanced carriers emerging in ferroelectrics under illumination [5] would reduce the space charge and, consequently, reduce the effect of it on the relaxation of polarization. Under conditions of through-flow conductivity starting at higher field amplitudes (the sharp increase of $tg\delta_{eff}$ at $E > 8$ kV/cm, figure 3) the light-induced charge carriers have no effect on the features of the dielectric response of the material. Behaviour of $\varepsilon_{eff}(E)$ and $\varepsilon''_{eff}(E)$ at lower temperatures and higher frequencies when the contribution of conductivity to dielectric response is substantially smaller point to conductivity of the ceramics being mainly affected by illumination. As it follows from figure 1, the dielectric response at 14 °C and 0.1 Hz is typical to FE ceramics the effect of illumination being around 1 % – practically of the order of accuracy of the PL measurements.

The values of ε_{eff} and ε''_{eff} measured at frequency of 10 Hz in dark and illuminated sample are practically the same at 14 °C and 23 °C, only at temperatures closer to T_m the light-induced increase of ε_{eff} is detectable. In this case the increment of ε_{eff} can be related to the effect of unbalanced carriers on the intrinsic field of defects as shown by the study of dielectric response in SBN-75 single crystal [7]. The increase of ε_{eff} is achieved by partial compensation of the intrinsic field facilitating mobility of domain (phase) boundaries under applied field. Decrease of the coercive field E_c under illumination has been reported in studies of light-induced effects on polarization switching in SBN-61 crystals [8].

4. Conclusions

By comparing the values of effective parameters $tg\delta_{eff}$, ε_{eff} and ε''_{eff} obtained from polarization loops of dark and illuminated SBN-75 ceramic samples within the range of temperatures of the relaxor phase at frequencies below 10 Hz and characterising relaxation of polarization the effect of light is found to present a decrease of the $tg\delta_{eff}$ and ε''_{eff} values.

Unbalanced carriers induced by light in the SBN-75 ceramics decreasing the space charge and its contribution to relaxation of polarization most likely produce the decrease of the effective dielectric loss in illuminated samples.

5. References

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