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Experimental investigation of the electrocaloric response and simulation of solid-state ferroelectric cooler

A V Es'kov[1,2], P Yu Belyavskiy[1,2], A S Anokhin[2], A A Semenov[1], O V Pakhomov[2], I L Mylnikov[1,2], A A Nikitin[1,2], V A Krylov[2]

[1] St. Petersburg Electrotechnical University, St. Petersburg, 197376, Russia
[2] ITMO University, St. Petersburg, 197101, Russia

E-mail: pbeliavskiy@gmail.com

Abstract. The electrocaloric response in ferroelectrics was experimentally investigated in nonequilibrium thermal conditions. The electrocaloric response reached 6 mK per one cycle polarization-depolarization of the ferroelectric sample. Computer modeling based on experimental data demonstrated the cooling capacity of multilayered structure at 25 W/cm^3.

The finding of new cooling principles for the development of compact, ecologically friendly, solid-state heat pumps and coolers with low power consumption, working around room and cryogenic temperatures, are extremely important for modern society. The effective heat transformation could be implemented in the solid state structures using electrocaloric effect [1, 2]. The main technical difficulty standing in the way of developing a small-sized high-performance solid-state coolers on electrocaloric effect, is the need to use heat switches for heat removal process from the cooling object [3-5]. Earlier, in works [6-9], principle and thermodynamic cycle of the electrocaloric cooler without the use of thermal switches have been proposed. The principle of operation is based on the effect of the difference values of electrocaloric effect in ferroelectric materials with polarization and depolarization of the sample [10].

In this paper electrocaloric response in bulk ferroelectric samples based on a solid solution of barium strontium titanate ($Ba_{0.65}Sr_{0.35}TiO_3$) was experimentally investigated. The temperature dependence of the electrocaloric response was investigated for ferroelectric samples placed in special thermal conditions. The plane-parallel ceramic capacitors with a diameter of 10 mm and a thickness of 1 mm were used as a measured samples. The copper electrodes were created by vacuum evaporation. A thickness of electrodes was about 1 μm.

Figure. 1 shows the view of the electric field E time dependence in the plane-parallel ferroelectric capacitor and temperature variation due to the electrocaloric effect. At the initial time the sample is in contact with an environment at the temperature T_0. At time t_1 occurs polarization and temperature of the sample changes from T_0 to T_1 due to electrocaloric effect. The time interval from t_1 to t_2 corresponds to a temperature relaxation due to heat exchange with the environment. The time of changing of electric field in the sample from E_1 to E_2 and from E_2 to E_1 must be much less than the time of thermal relaxation. At time t_2 occurs depolarization of ferroelectric sample and its temperature decrees below T_0. In the time interval from t_2 to t_3 the sample temperature increase occurs due to heat exchange with the environment. Then, the cycle is repeated with a different initial temperatures. The measurement of electrocaloric response was carried out in the thermostatic bath of FP40-MA Refrigerated/Heating Circulator with using of arbitrary waveform generator Agilent 33522A, a high-voltage amplifier Trek 609E-6 and nanovoltmeter Agilent 34420A. Direct measurement of the temperature change due to polarization and depolarization of the ferroelectric sample produced with calibrated miniature platinum thermistor. On the sample were applied voltage pulses with amplitude of 2 kV which led in the sample the electric field 2 V/μm. Duration of periods t_1 - t_2 and t_2 - t_3 were 100 s. The durations of the leading and trailing edge of the pulse were 200 μs. Figure. 2 shows the dependence of the electrocaloric response due to polarization and depolarization of the ceramic ferroelectric sample based on $Ba_{0.65}Sr_{0.35}TiO_3$.

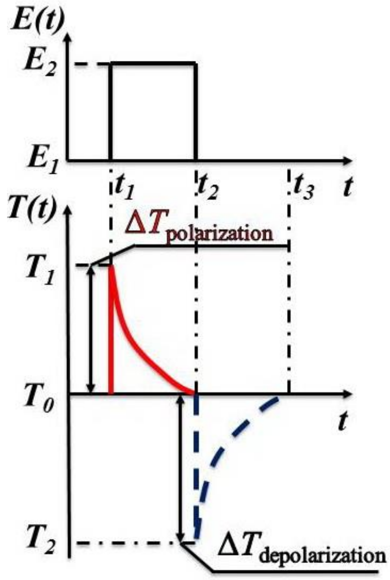

Figure 1. The time dependence of the electric field E in the plane parallel ferroelectric capacitor and temperature changes due electrocaloric effect.

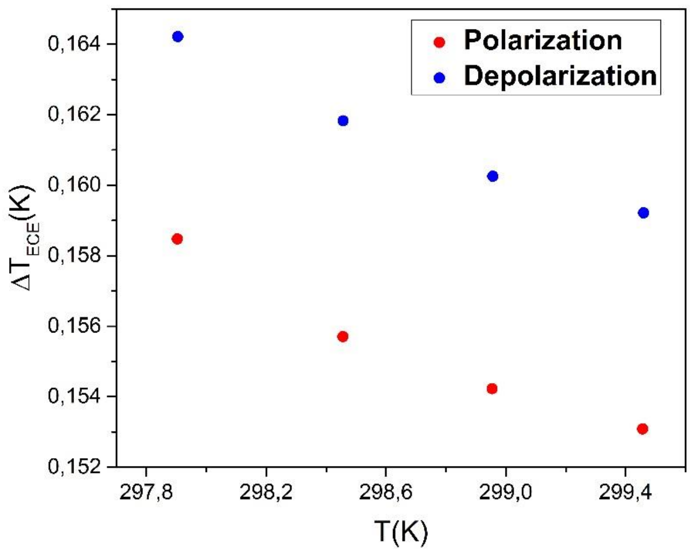

Figure 2. The dependence of the electrocaloric response due to polarization and depolarization of the ceramic ferroelectric sample based on $Ba_{0.65}Sr_{0.35}TiO_3$.

The results of the electrocaloric response experimental measurements have been used for computer simulation of active element of electrocaloric cooler without using of heat switches. The active element of cooler is a MLC capacitor with ferroelectric layers based on $Ba_{0.65}Sr_{0.35}TiO_3$. Figure. 3 shows a simple model of the cooling capacitor. It consists of ten ferroelectric layers separated from each other by nickel electrodes. The thickness of ferroelectric layer is 10 μm, thickness of nickel electrode is 1 μm. We will investigate a one-dimensional model under the assumption that the temperature changes only along the x axis. One boundary (x = 0) is thermally insulated, whereas the other boundary (x = l) is held under slow convection by air with temperature T_0, with equal to the initial temperature T_0. The heat capacity $C(x)$ and thermal conductivity $\lambda(x)$ are assumed to be constant for each segment of the line according to the materials of layers. In this case, the temperature distribution $T(x, t)$ along the line can be found by solving the heat conduction equation:

$$C(x)\frac{\partial T}{\partial x} = \frac{\partial}{\partial x}\lambda(x)\frac{\partial T}{\partial x} + Q(x,t,T) \tag{1}$$

which satisfies the initial and boundary conditions.

$$T(x,\ 0) = T_0; -\lambda\left.\frac{\partial T}{\partial x}\right|_{x=0} = 0\,, \lambda\left.\frac{\partial T}{\partial x}\right|_{x=L} = h(T - T_0) \tag{2}$$

where h is the heat transfer coefficient. The function $Q(x, t, T)$ determines the quantity of heat released (or absorbed) by a thermal electrocaloric source and is expressed in the following form:

$$Q(x,t,T) = -T\frac{\partial P}{\partial T}\frac{dE}{dt} \tag{3}$$

where E is the electric field strength of the electrocaloric element and P is the polarization of the ferroelectric. The function $Q(x, t, T)$ is nonzero only within the ferroelectric layers.

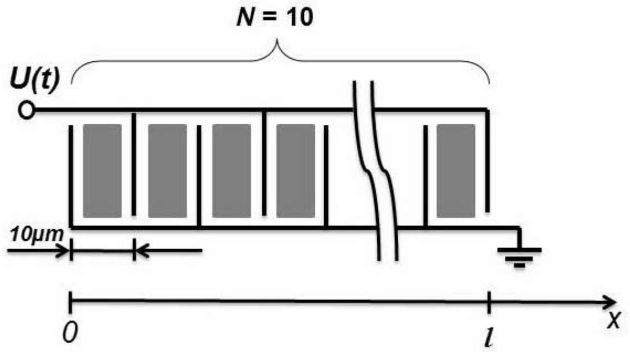

Figure 3. Schematic model of multilayered capacitor with 10 ferroelectric layers.

The initial temperature 298.5 K has been selected for the simulation procedure, in this case the temperature variation due to polarization and depolarization was about 0.006 K. Periodic square pulses of the electric field were supplied to the ferroelectric elements with different frequencies. The pulses were cause periodic heating (cooling) of the electrocaloric elements and a redistribution of the temperature field along the structure. After a series of cycles of the polarization and depolarization of the solid—state structure, we determined the temperature distribution on the free end of the cooling line (x = 0). We consider two different situation. In the first case, we have considered a simulation with in-phase pulses supplied to the ferroelectric elements. Figure 4 presents the results of the computer simulation of the temperature variation for in-phase pulses for all layers with frequency 1 kHz. In the second case, we have considered a simulation with pulses shifted by a half period for odd and even layers. Figure. 5 shows the schematic diagram of a such connection. Figure 6 presents the results of the computer simulation of the temperature variation on the free end of the cooling line (x = 0) for out-of-phase pulses with frequencies 500 Hz, 1k Hz and 2 kHz. Volumetric cooling capacity in case of phase shift was about 6 W/cm^3 for frequency 500 Hz, 9 W/cm^3 for 1 kHz and and correspondingly 25 W/cm^3 for 2 kHz. Thus, the cooler may show considerable cooling capacity when used the multilayer elements with thin layers and high-frequency out-of-phase pulses, even if there is a small difference between temperature changes due to polarization and depolarization.

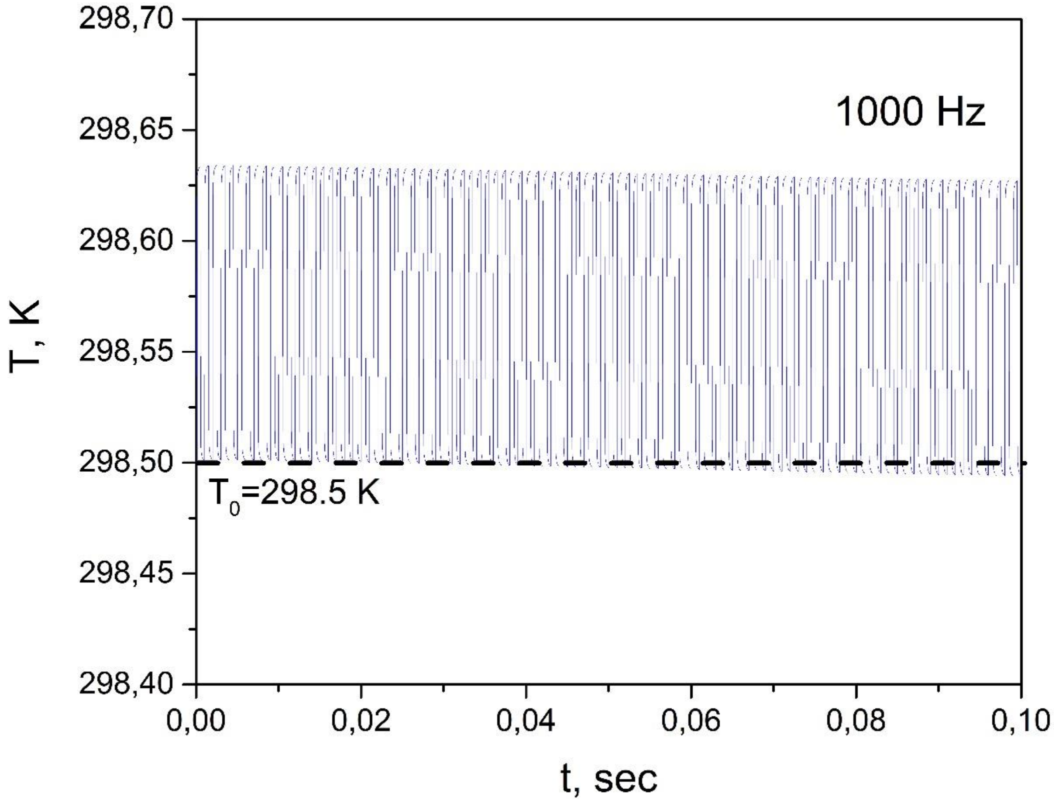

Figure 4. Dynamics of change in the temperature at the left end (x = 0) of the cooling elementin the case of in-phase square pulses.

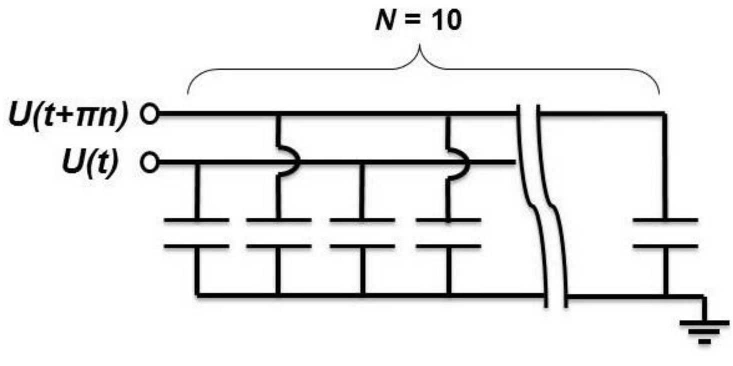

Figure 5. Schematic diagram of electric connection of layers in the case of out of phase pulses.

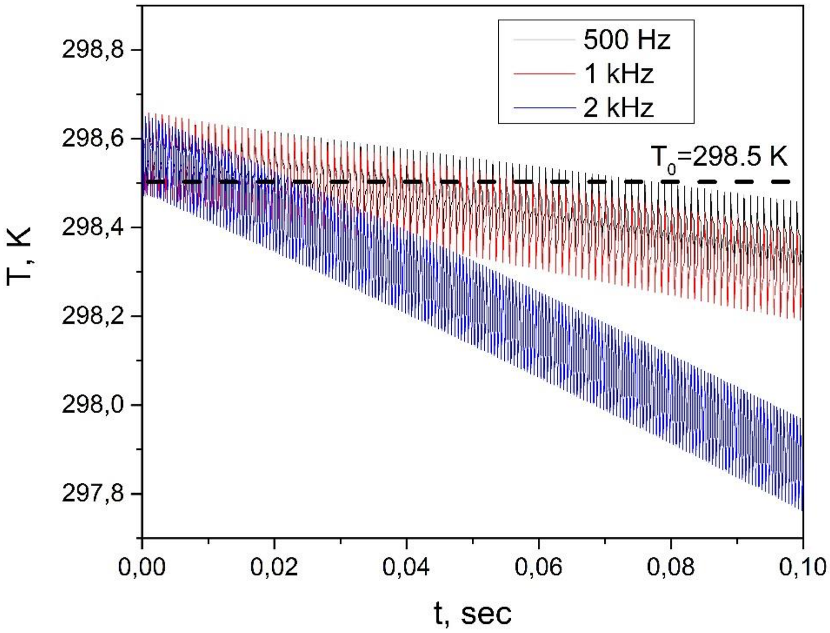

Figure 6. Dynamics of change in the temperature at the left end (x = 0) of the cooling element in the case of out of phase square pulses.

Acknowledgements
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