PHYSICAL MODEL FOR FERROELECTRICS BASED ON PYRROCURRENT CONSIDERATION

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Investigations of the ferroelectric properties carried out during last years have shown presence of the temperature hysteresis: the polarization [1], the dielectric permeability [2] and the thermal capacity [3] of ferroelectrics depend not only on temperature but also on whether temperature increased or decreased. Moreover, the dependence of measurable ferroelectric characteristics on the heating (cooling) rate was revealed experimentally (Fig.1), see [4]. Such a phenomenon cannot be described within the framework of the classical Landau-Ginzburg-Devonshire (LGD) theory [5]. The deficiency of these theory is explained as follows. If the polarization P, intensity E and temperature T are linked by any algebraic expression, then a certain set of polarization values P should correspond to any given values of E and T defined by this relation. Note that each value in this set does not depend on the prehistory of temperature variations. For the description of the temperature hysteresis phenomenon, the algebraic equations have to be changed to the differential ones.

For the solution of the problem discussed above we suggest a new approach based on the LGD model striving to describe experimental data more precisely. As the starting point we use Landau-Khalatnikov (LK) equation (also known as Time Domain Ginzburg-Landau-Devonshire) [6]

$$\alpha \frac{\mathrm{d}P}{\mathrm{d}t} = E - a_0(T - T_c) - bP^3. \tag{1}$$

where a_0 is the Curie-Weiss constant, T_c the Curie temperature and b is the coefficient of nonlinearity. The coefficient α has the sense of the “internal” resistance. Equation (2) means that a change of polarization is equivalent to the presence of the current defined as

$$j = \frac{\mathrm{d}P}{\mathrm{d}t} \equiv \frac{\partial P}{\partial E}\frac{\mathrm{d}E}{\mathrm{d}t} + \frac{\partial P}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}t} = j_{el} + j_{pyr}. \tag{2}$$

The impact of the “electrical“ current component j_{el} on the polarization is investigated in [6], where it has been shown that the account of j_{el} allows us to explain the distinction in values obtained by measurements of the sample temperature and calculated through the polarization [7]. To consider the impact j_{pyr} on the polarization we assume a linearity in temperature evolution with time: $T = V{\cdot}t + T_0$, where V is the rate of temperature change and T_0 is the initial temperature. In order to find the polarization one should use the equation

$$\beta \frac{\mathrm{d}P}{\mathrm{d}T} + a_0(T - T_c)P + bP^3 = E, \quad \beta = \alpha V. \tag{3}$$

It is important to note that we chose linear dependence between β factor and the rate of heating because it was predicted by the LK theory. Assuming a more complex dependence between these two values, we can easily eliminate the aforementioned deficiency. Detailed analysis of (3) makes it possible to write out the following statements: First, the pyroelectric coefficient $\partial P/\partial T$ is limited at $T = T_c$, i.e. the phase transition of the second kind disappears. In addition, the dielectric permittivity is limited also and the peak sharpness of $\varepsilon(T)$ is defined by the parameter β. Second, the spontaneous polarization at the Curie point does not turn into zero as well as it deviates from zero in paraphase in a small vicinity of the order of $\sqrt{\beta/a_0}$ at the Curie point. Third, equation (3) allows us to describe the temperature hysteresis. The difference of physical values at heating and cooling cycles appears proportional to the parameter β. Finally, the maximum of the electrocaloric effect is displaced towards higher temperatures at the Curie point. As an example for the model verification, we attracted the commonly used typical ferroelectric ceramics $Pb(Mg_{1/3}Nb_{2/3})O_3 - PbTiO_3$. Corresponding calculations performed using the LGD model and our refined version are compared against the experiment in Fig.2. The developed approach demonstrates rather better agreement between experiment and theory as compared with the LGD model.

Our work is derived from the conventional Landau-Khalatnikov model for ferroelectric materials, which is known to describe a wide range of experimental data. Comparing experiment results with findings from classical LGD theory and with those obtained by means of our refined model we showed that the latter one better represents the matter. In other words, we have proved that if a ferroelectric behavior could be described within the framework of the LK theory, then temperature dependence of polarization will be adequately described by the theoretical model we have proposed.

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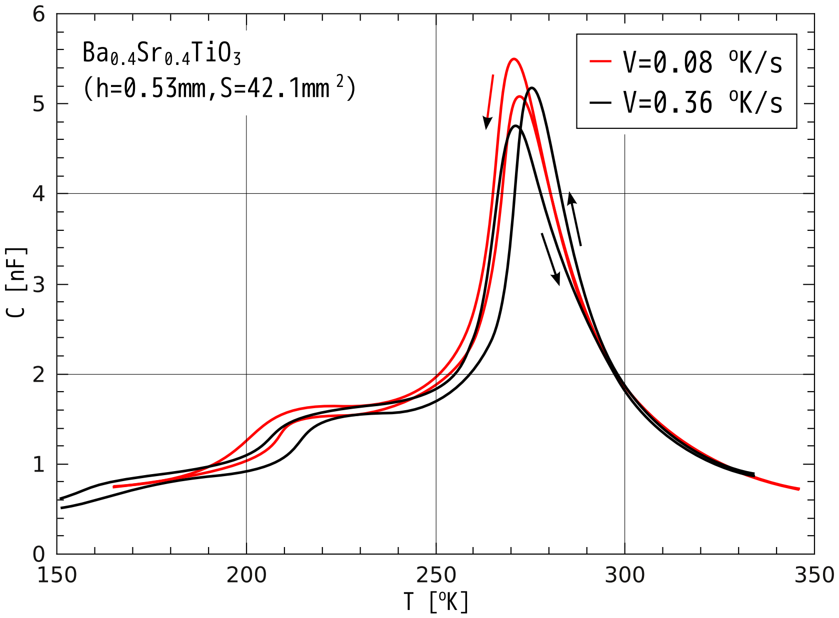

Figure 1: The temperature dependence of the capacitance for the ceramics $Ba_{0.4}Sr_{0.4}TiO_3$ at different heating rates.

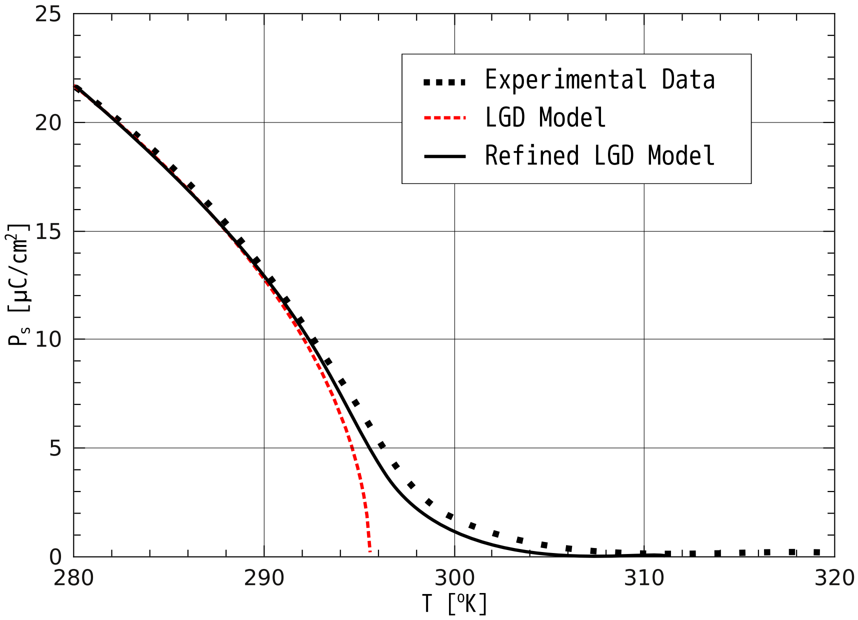

Figure 2: The temperature dependence of the spontaneous polarization for the ceramics $Pb(Mg_{1/3}Nb_{2/3})O_3 - PbTiO_3$: comparison between experiment, standard LGD model and our refined approach.