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Electrocaloric effect in (1-x)(0.8Na0.5Bi0.5TiO3-0.2BaTiO3)-xCaTiO3 solid solutions at high electric fields

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Abstract: Recently, many efforts have been made to find high values of reversible electrocaloric effect (ECE) induced temperature change DT, which is the most important parameter for creation of ECE-based cooling systems. The application of larger electric fields has shown promise as a way of increasing DT. However, there are only a small number of publications where ECE is directly measured at electric fields in the range above 20-30 kV/cm. The present work provides a detailed overview of ECE in (1-x)(0.8Na0.5Bi0.5TiO3-0.2BaTiO3)-xCaTiO3 (x=0.05-0.125) solid solutions. For these compositions, we have measured DT as a function of temperature and applied fields of up to 100 kV/cm using the direct measurement method. At lower concentrations of CaTiO3, values of DT above the electric field-induced first order phase transition reach 1ºC with a large contribution from an entropy jump. At higher CaTiO3 concentrations, the electric field-induced phase transition is suppressed. This causes an expressed reduction of DT, despite a moderate reduction of electric field-induced dielectric polarization. Furthermore, a comparison of the direct measurement method of ECE temperature change with the indirect one using Maxwell's relations is presented. Here, an inconsistency between the results obtained by both methods is demonstrated and interpreted.

Keywords: Electrocaloric effect; Large electric field; Polarization; Field-induced first order phase transition

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Results and Discussion

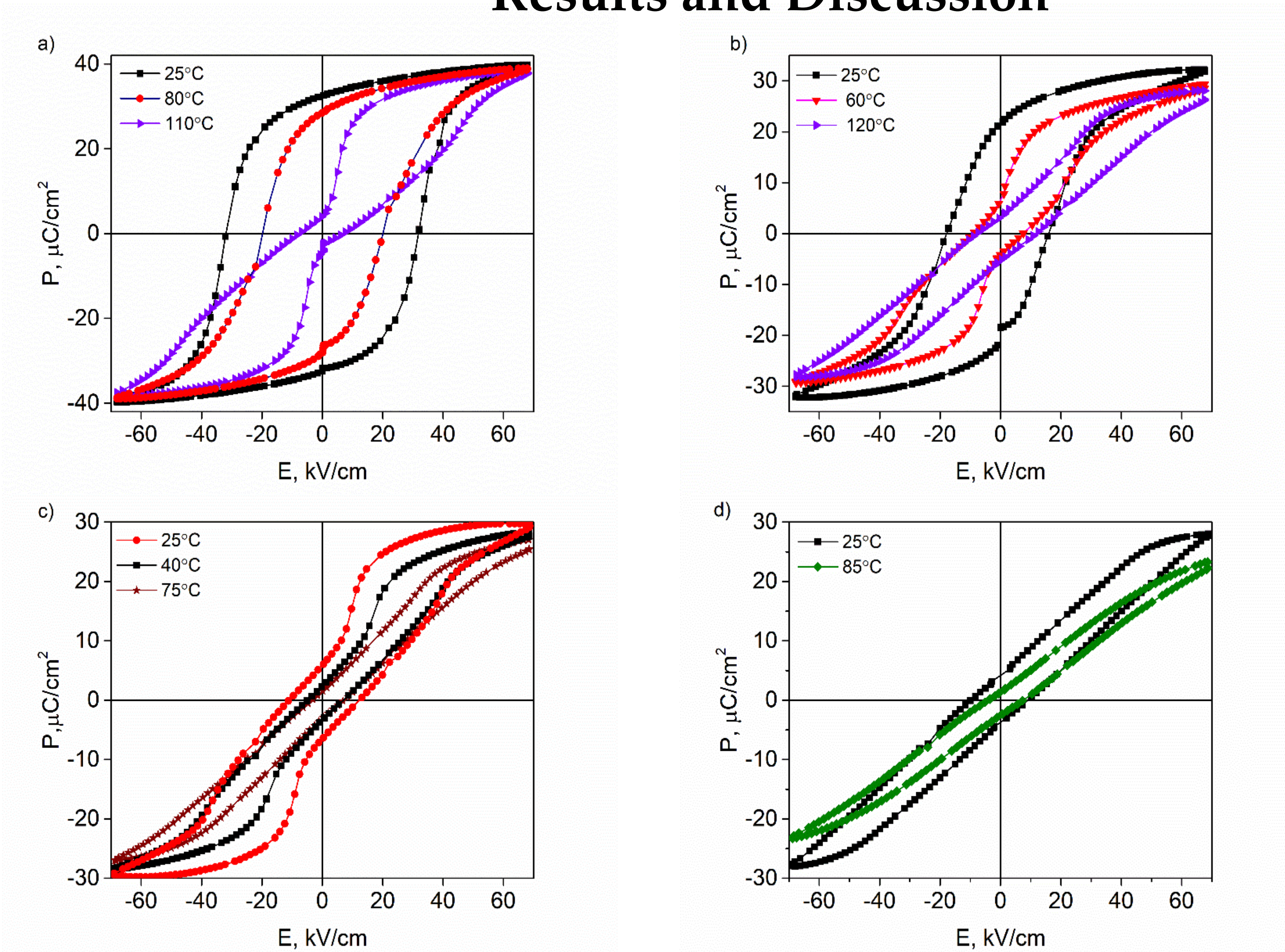

Figure 1. Polarization hysteresis loops at bipolar electric field pulses for (1-x)(0.8NBT-0.2BT)-xCT compositions x=0.050 (a), x=0.075 (b), x=0.100 (c) and x=0.125 (d) at various temperatures.

Results and Discussion

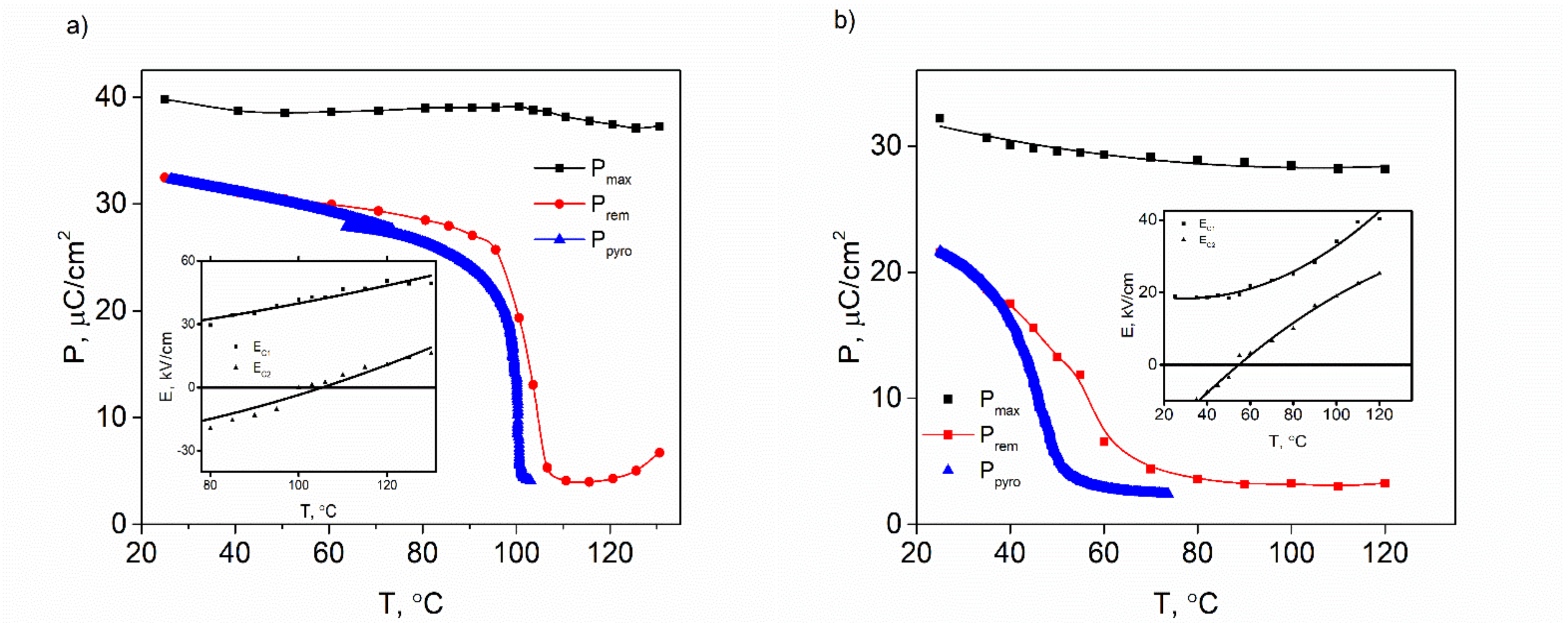

Figure 2. Temperature dependences of maximal and remnant polarization for (1-x)(0.8NBT-0.2BT)-xCT compositions with x=0.050 (a) and 0.075 (b), determined from polarization hysteresis loops, as well as from static pyroelectric effect measurements. Inset: Temperature dependence of the critical electric fields.

Results and Discussion

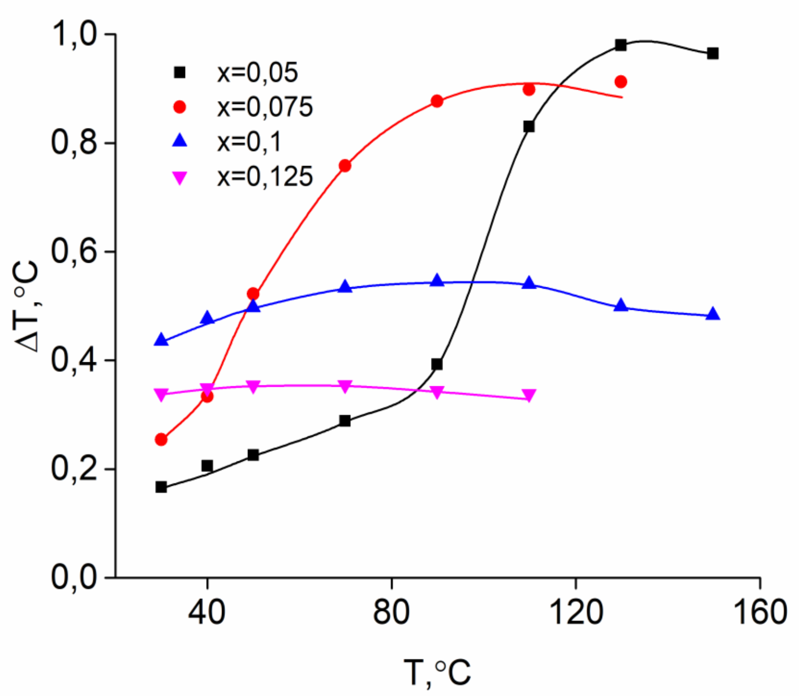

Figure 3. ΔT(T) for (1-x)(0.8NBT-0.2BT)-xCT with different concentrations, measured as a result of switching off electric field pulse E=100 kV/cm.

Results and Discussion

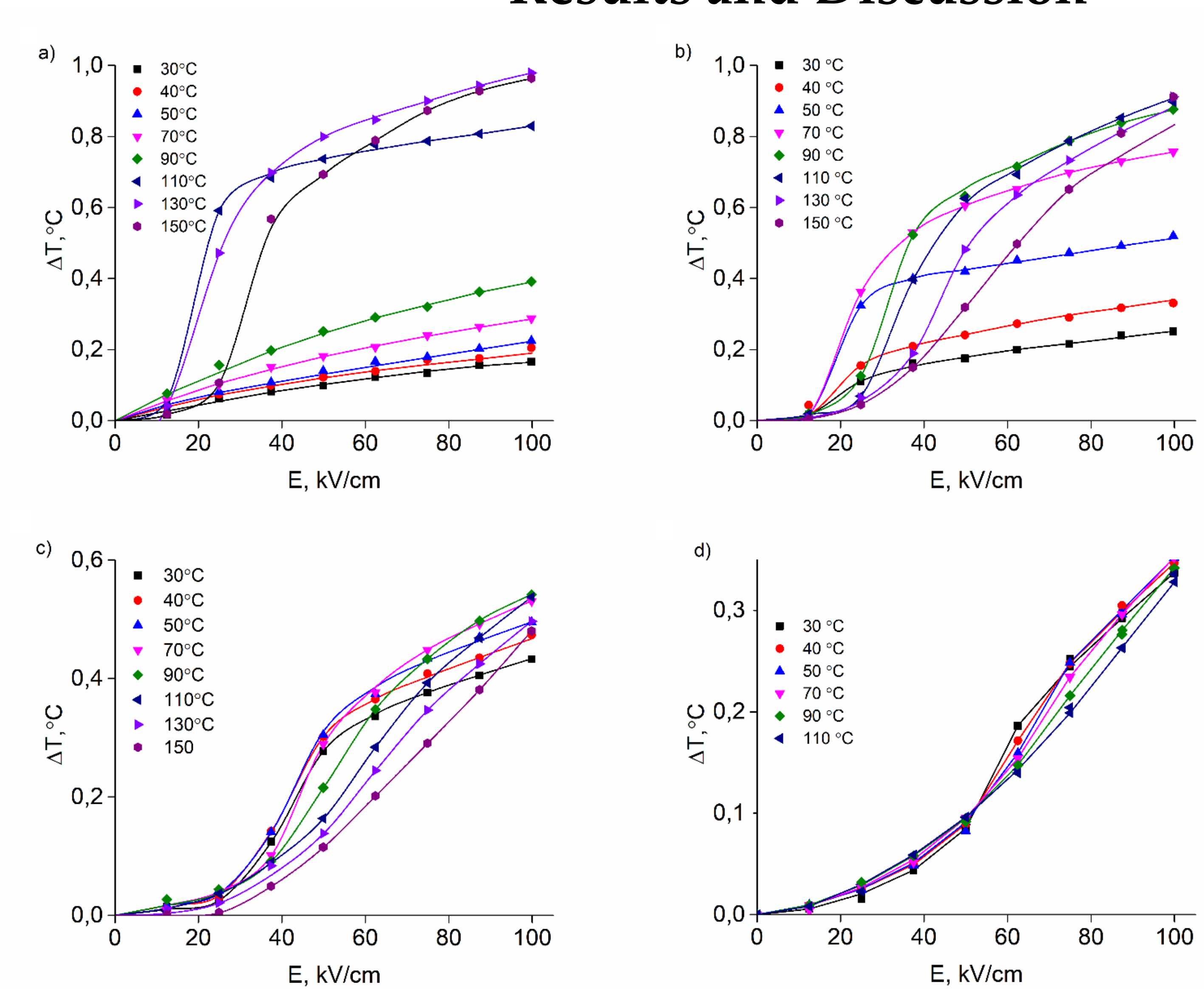

Figure 4. ΔT(E) for (1-x)(0.8NBT-0.2BT)-xCT compositions with x=0.050 (a), x=0.075 (b), x=0.100 (c) and x=0.125 (d), measured at different temperatures in the case when electric field pulse is switched off.

Results and Discussion

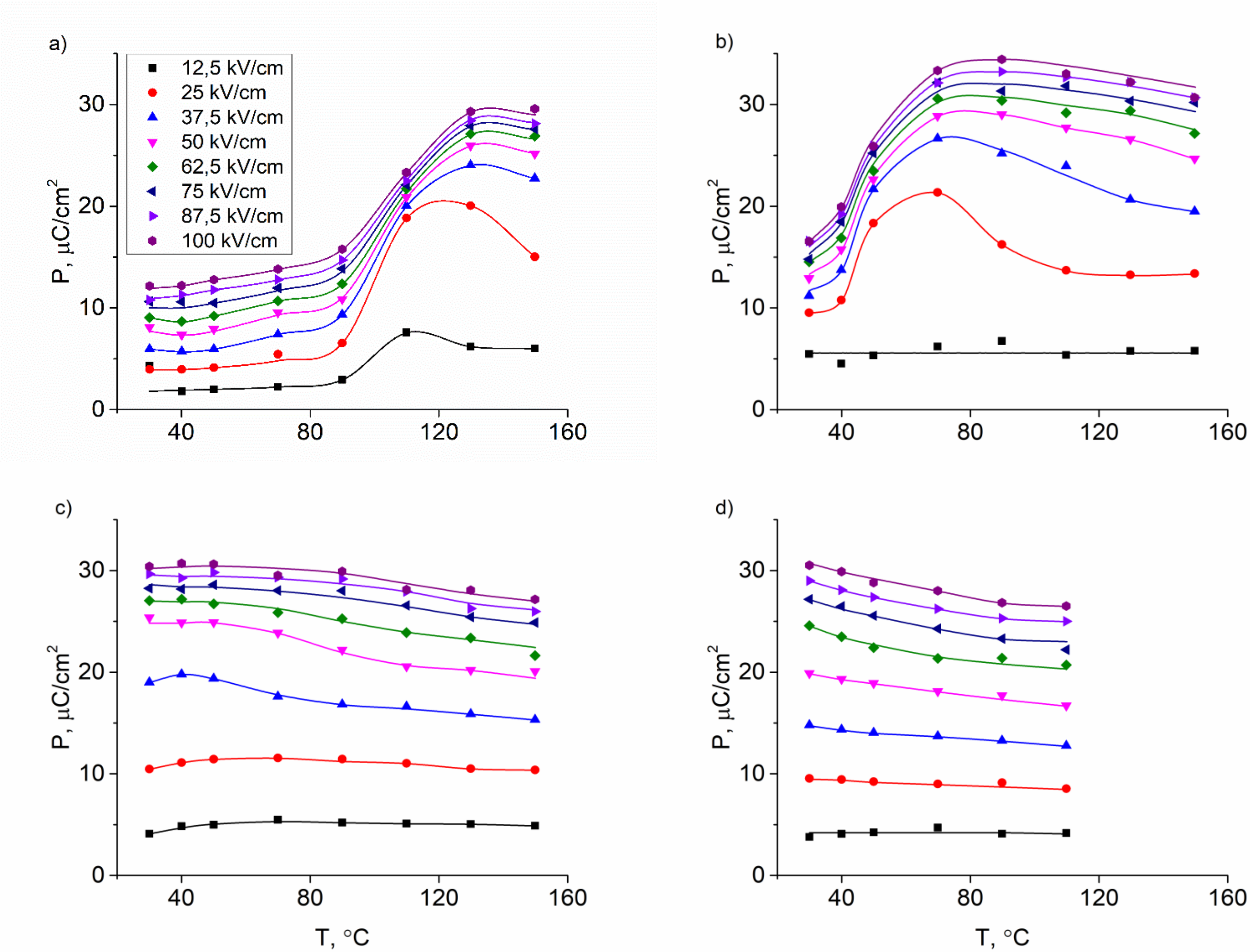

Figure 5. Temperature dependence of polarization at different electric fields for (1-x)(0.8NBT-0.2BT)-xCT compositions with x=0.050 (a), x=0.075 (b), x=0.100 (c) and x=0.125 (d), obtained from measurements of polarization current simultaneously with measurements of ΔT.

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Results and Discussion

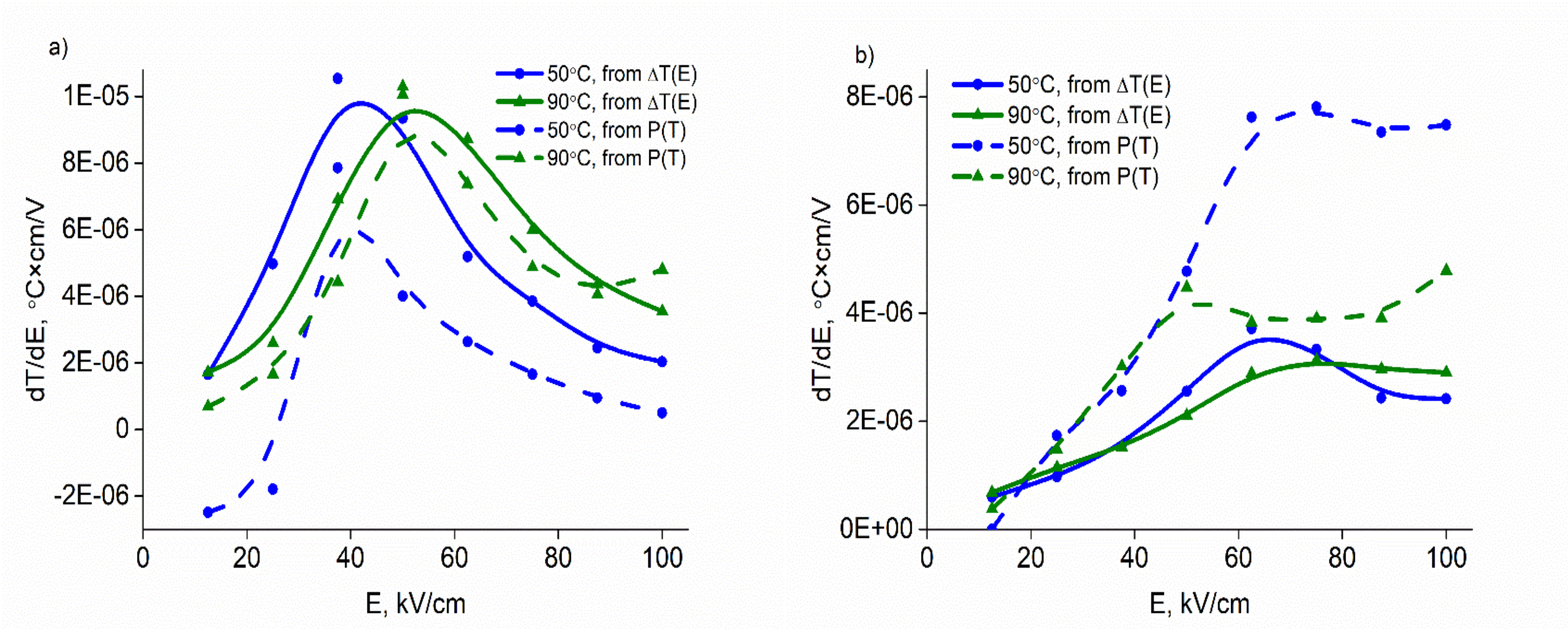

Figure 6. Comparison of dT/dE values obtained by the direct measurements of ΔT(E) and extracted from P(T,E) according to Eq. (3), using data from experimentally measured polarization current, for (1-x)(0.8NBT-0.2BT)-xCT compositions with x=0.100 (a) and x=0.125 (b).

Eq. (3).

$$\frac{dT}{dE} = -\frac{T}{c_E} \cdot \left(\frac{\partial P}{\partial T}\right)_E$$

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Conclusions

- The maximal values of ECE temperature change ΔT are found in the temperature region slightly above depolarization temperature. In the case of the composition with x=0.050, ΔT reaches 1.0 ºC at 100 ºC.
- In a wide temperature range around the depolarization temperature, values of ΔT measured upon switching off the electric field pulse are lower than the values obtained upon switching on the electric field pulse.
- Increasing of CaTiO3 concentration allows reducing of depolarization temperature, but simultaneously significantly decreases the attainable value of ΔT.
- In the temperature range, where electric field at least partly induces a ferroelectric state, the shape of the calculated electric field dependence of the derivative dT/dE corresponds to the directly measured dependence, although the absolute values can differ significantly.

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Conflicts of Interest

The authors declare no conflict of interest.