Thermopolarization currents in dielectrics

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Pyroelectric currents have been observed in a temperature gradient in crystals of lead magnoniobate, $PBMg_{1/3}Nb_{2/3}O_3$, which have a center of inversion. These pyroelectric currents are accompanied by the onset of a dielectric polarization (this is a thermopolarization effect). The thermopolarization coefficient B_{11} has been evaluated.

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Gurevich[1] recently showed theoretically that a dielectric polarization P could theoretically arise in a temperature gradient ∇T ($P_i = B_{ij} \nabla_j T$) in dielectrics of arbi-

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trary symmetry (this is an "electrothermal" effect or a "thermopolarization" effect, as we will refer to it below). In piezoelectric materials this effect is accompanied by a third-order pyroelectric effect, which also contributes to the polarization.[2-4]

In the present experiments we attempted to observe the thermopolarization effect in crystals having a center of inversion. Above the Debye temperature the thermopolarization coefficient B is described by[5]

$$|B| \cong \frac{\chi k_B}{\sqrt{q_0 M w^2}}, \tag{1}$$

where χ is the dielectric susceptibility, a_0 is the lattice constant, M is the average mass of the atoms, w is the average sound velocity, and k_B is the Boltzmann constant. The largest values of P can thus be expected in compounds having a large dielectric susceptibility.

The simplest way to observe the thermopolarization effect is to measure the pyroelectric current (the thermopolarization current) which flows in the external circuit when one end of a sample is heated and ∇T varies over time. For a dielectric in the absence of an external electric field, the current density may be written

$$j \sim a \frac{dP'}{dt} + B \frac{d}{dt}(\nabla T), \tag{2}$$

where P' is the polarization associated with the random unipolarity of the sample, and t is the time. The pyroelectric current described by the first term on the right side of (2) can be taken into account easily, since—in contrast with the thermopolarization current—it does not change direction when the sign of ∇T changes.

A convenient material for studying the thermopolarization effect is lead magnoniobate, $PbMg_{1/3}Nb_{2/3}O_3$ (PMN). The resistivity of PMN at 300 K is $\sim 10^{14}$ $\Omega \cdot$cm, and the Seebeck effect which accompanies the thermopolarization effect is very small. Above 273 K, PMN is in a paraelectric state, and it has the perovskite cubic structure with a center of inversion (class O_h); because of the proximity to T_c, however, and because of the diffuse nature of the phase transition, this material retains high values of the dielectric function over a broad temperature range, including room temperature.[6]

In the present experiments we used samples of pure (deliberately not doped) PMN single crystals which were elongated parallelepipeds with $\{100\}$ faces and dimensions of $3 \times 3 \times 8$ mm. Electrodes of Degus paste were deposited on the longest faces. A temperature difference ΔT was imposed between these faces: One end of the sample was held at room temperature while the other was heated. All the measurements were carried out in a vacuum of $\approx 10^{-3}$ Torr.

Figure 1 shows the time dependence of the density of current flowing in the external circuit (curve 1), the temperature difference which excites this current (2), and the rate of change of the temperature difference (3). When the sign of ∇T is changed (i.e., when the opposite end of the sample is heated), the magnitude of the current changes insignificantly, indicating that the sample is slightly unipolar. It can be seen

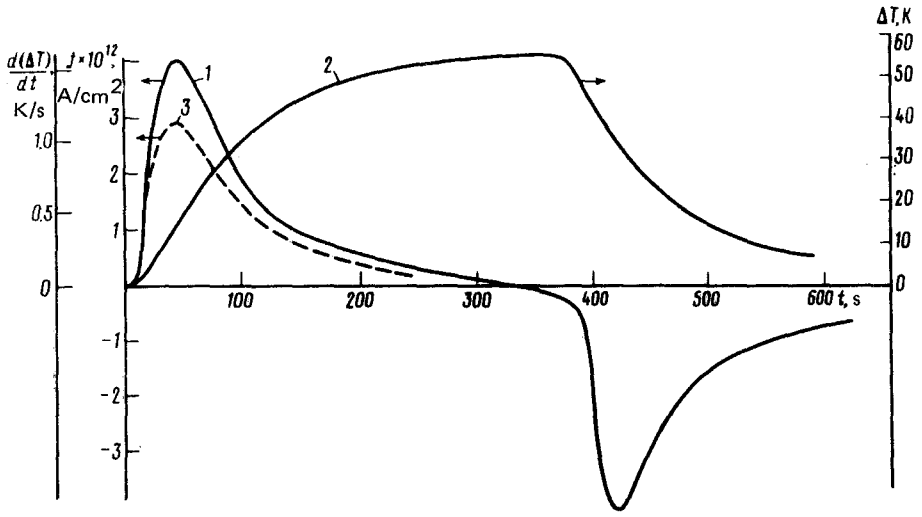

FIG. 1. Time dependence of several quantities. 1–Thermopolarization current density j; 2–difference between the temperatures at the ends of the sample, ΔT; 3–the derivative $d(\Delta T)/dt$ in PMN.

from Fig. 1 that the magnitude of the observed current is proportional to the rate of change of the temperature difference; the current also changes direction at $d(\Delta T)/dt < 0$. Figure 2 is a plot of the maximum current density against the quantity $[d(\Delta T)/dt]_{\max}$ at various heating rates. It is not difficult to see that this de-

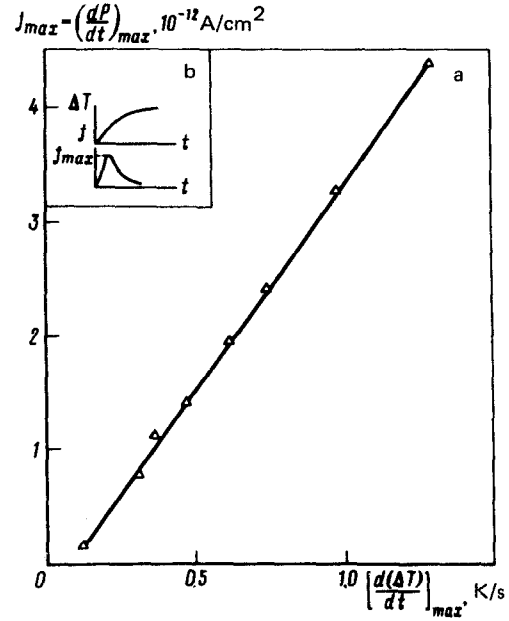

FIG. 2. a–Dependence of the maximum current density $j_{\max}$ on $[d(\Delta T)/dt]_{\max}$ for various heating rates; b (inset)–definition of $j_{\max}$.

pendence is linear, and the slope gives us the thermopolarization coefficient, $B_{11} \approx 3 \times 10^{-12}$ C/(cm · K).

The observed current in PMN thus arises because of the appearance of a polarization associated with the thermopolarization effect and a change in this polarization. The magnitude of this polarization is $\sim 10^{-10}$ C/(cm^2 · K) ($T = 310$ K, $\nabla T = 30$ K/cm).

Evaluating B_{11} from (1) for $\chi = 10^3$, $a_0 = 4 \times 10^{-8}$ cm, and $k_B T/Mw^2 = 10^{-2}$, we find $B_{11} \approx 10^{-13}$ C/(cm · K), which is close to the experimental value in order of magnitude. We should add that the lattice-anharmonicity parameter $k_B T/Mw^2$ can reach $\sim 10^{-1}$ or even more in real crystal lattices,[1] causing the coefficient B to increase.

The thermopolarization effect opens up some new opportunities for studying crystals with a center of inversion.

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