Dynamic secondary pyroelectric effect in lithium niobate

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It is predicted theoretically that a secondary pyroelectric effect will be observed in polar-neutral directions in noncentral crystals with an anisotropic limitation on thermal deformation. This prediction has been verified experimentally. This effect occurs in piezoelectric resonators in the frequency interval between the first and last acoustic resonances.

As the temperature of a polar crystal is changed by an amount $\delta T = T - T_0$, a the spontaneous polarization $\delta P_i = \gamma_i^{(1)}\delta T$ in the crystal changes. Here $\gamma_i^{(1)}$ is the pyroelectric coefficient corresponding to the primary pyroelectric effect. In addition, there is a thermal deformation $x_n = \alpha_n^E \delta T$ of the crystal, which causes a secondary pyroelectric effect, with a coefficient $\gamma_i^{(2)}$ (α_n^E is the thermal expansion coefficient). As a result, a uniform temperature change (grad $T = 0$, and there is no shear) causes a resultant pyroelectric effect

$$\gamma_i = \gamma_i^{(1)} + \gamma_i^{(2)} = \gamma_i^{(1)} + e_{in}^T \alpha_n^E, \tag{1}$$

where the sum $e_{in}^t \alpha_n^E$ determines the contribution of the secondary pyroelectric effect, and the e_{in}^T are the piezoelectric moduli. It has been assumed here that the sum in (1) is nonzero only in the polar directions in pyroelectrics, i.e., in the ten crystal classes which have a spontaneous polarization, while for nonpolar directions we have $\gamma_i^{(1)} = 0$ and $\gamma_i^{(2)} = e_{in}^T \alpha_n^E = 0$. The latter equality presupposes a free thermal deformation of a noncentral crystal, so the piezoelectric component of the thermal strain is completely canceled. In precisely the same way, we would have $\gamma_i^{(2)} = 0$ in a squeezed crystal, since deformation would be prevented.

As is shown below, a partial squeezing of a noncentral crystal (such a partial squeezing can be arranged with the help of, for example, an isometric piezoelectric resonators: a thin disk, a long rod, etc.) disrupts the cancellation of the dynamic component of the thermal deformation, with the result that there are effective values

$$\alpha_n^E e_{in}^T(\omega) \neq 0, \tag{2}$$

in the frequency interval $\omega_1 \leqslant \omega \leqslant \omega_2$, where ω_1 is the frequency of the first acoustic resonance (radial vibrations of a disk, longitudinal vibrations of a rod, etc.), and ω_2 is a higher frequency of an acoustic resonance (thickness vibrations of a disk or rod).

Inequality (2) holds for only so-called[1] polar-neutral directions in noncentral crystals: In such crystals, under certain boundary conditions (in particular, when there is a limitation on certain types of deformation by piezoelectric squeezing), the cancellation of the polarity may be disrupted, and a dynamic secondary pyroelectric effect may become possible. In crystals of group $3m$ (e.g., lithium niobate), for example, we have theoretically predicted and experimentally observed secondary pyroelectric effect in the [010] polar-neutral direction. This effect occurs in addition to the primary and secondary pyroelectric effects in the [001] polar direction. For the [100] direction, on the other hand, a secondary pyroelectric effect is theoretically impossible, and it is not seen experimentally. Significantly, the effect observed in lithium niobate is appreciable:

$$|\gamma_2^{(2)}/\gamma_3^{(2)}| \approx d_{22}^T/d_{31}^T \approx 15.$$

Analytic expressions for the dynamic pyroelectric effect were derived through a joint solution of the equations of the thermally induced piezoelectric effect and Maxwell's equations, by a method close to that of Ref. 2. The expressions derived describe the frequency dispersion of the components of the secondary-pyroelectric-effect vector for all 20 classes of noncentral crystals.

In particular, for disk-shaped piezoelectric and pyroelectric elements of crystals of the $3m$ group, the contributions of the secondary pyroelectric effect are $\gamma_1^{(2)}(\omega) = 0$,

$$\gamma_2^{(2)} = \frac{d_{22}^T(\alpha_1^E S_{33}^{E,T} - \alpha_3 S_{13}^{E,T})}{S_{11}^{E,T} S_{33}^{E,T} - (S_{13}^{E,T})^2} f(\omega), \qquad \gamma_3^{(2)} = \frac{2d_{31}^T \alpha_1^E}{S_{11}^{E,T} + S_{12}^{E,T}} f(\omega), \tag{3}$$

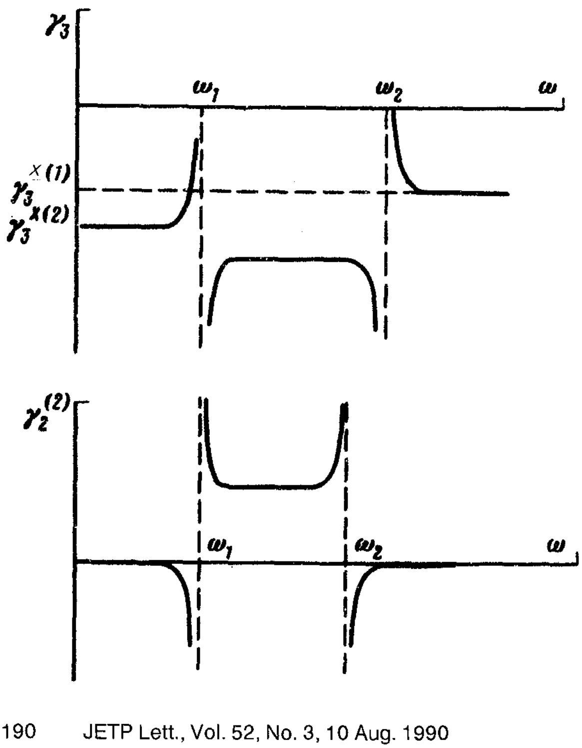

FIG. 1. Theoretical frequency dispersion of the components of the pyroelectric coefficient in lithium niobate (X is the voltage).

where $f(\omega) = 1 - 2\omega_0/\pi\omega\tan(\pi\omega/2\omega_0)$, ω_0 is the frequency of the acoustic resonance of radial vibrations of the disk, α^T_{in} are components of the piezoelectric modulus, and S^{ET}_{nm} are components of the elastic-compliance tensor. Corresponding expressions have been derived for rod-shaped resonators. Higher-index vibrational modes and damping were ignored in the calculations.

Figure 1 is a schematic diagram of the frequency spectrum γ_i of lithium niobate. In accordance with (3), we have $\gamma_1 = 0$, while the components of the primary and secondary effects add together in γ_3. It is assumed that $\gamma_3^{(1)}$ is independent of the frequency, while the contribution of the secondary effect, on the contrary, varies in the course of radial (ω_1) and thickness (ω_2) resonances (harmonic components are ignored). There is some new information regarding the frequency dependence of $\gamma_2^{(2)}$. In neither a free crystal ($\omega = 0$) nor a squeezed crystal ($\omega \to \infty$) is there a secondary pyroelectric effect $\gamma_2^{(2)}$. The frequency range in which the dynamic pyroelectric effect is manifested depends on the ratio of the diameter and thickness of the disk-shaped piezoelectric resonator.

The experimental apparatus used to study the dynamic pyroelectric effect includes a 50-mW LGN-215 laser, whose output is modulated by an ML-3 shutter over the frequency range 0–10^7 Hz. The pyroelectric response is measured by a V6-10 tuned voltmeter. The lithium niobate samples are disks 3.5 mm in diameter and 0.1 mm thick. Electrodes are deposited on the planes of the disk; the disk is suspended freely, and its end is exposed to the modulated laser beam.

Figure 2 shows the results of the measurements of the dynamic pyroelectric response. In agreement with the theory, there is no pyroelectric response in a disk oriented perpendicular to the [100] direction. This result is also of importance in the sense that this orientation of lithium niobate allows a tertiary pyroelectric effect.[3] The experimental conditions thus prevented a significant contribution from that effect.

In general, the frequency dependence of the pyroelectric coefficients γ_3 and $\gamma_2^{(2)}$ supports the theoretical predictions listed above. The secondary pyroelectric effect

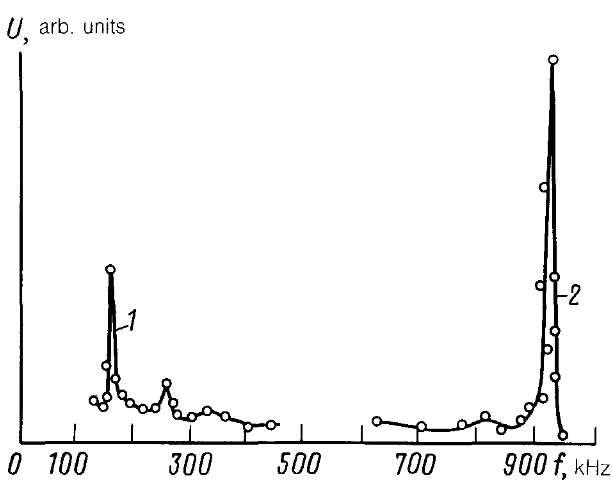

FIG. 2. Dynamic pyroelectric response of oriented lithium niobate disks. *1*—Crystallographic cut perpendicular to the [001] direction; *2*—crystallographic cut perpendicular to the [010] direction.

may be intensified significantly as a result of the piezoelectric resonance in (3). Significantly, the polar-neutral cut of the lithium niobate crystal leads to a dynamic secondary pyroelectric response much larger than that in the case of a polar cut. Calculations show that a smaller difference would be expected in lithium tantalate: $\gamma_2^{(2)}/\gamma_3^{(2)} \approx 2$.

Again in the case of nonpolar but noncentral crystals, there would evidently be conditions which would satisfy dynamic inequality (2) in the polar-neutral directions. In crystals of ammonium dihydrophosphate, for example, which are of group $42m$, a dynamic secondary pyroelectric effect would be manifested [according to calculations similar to those leading to (3)] in a long rectangular rod cut in the [110] direction with electrodes on (001) planes:

$$\gamma_3^{(2)} = 2d_{36}^T \alpha_1^E / 2S_{11}^{E,T} + 2S_{12}^{E,T} + S_{66}^{E,T}. \tag{4}$$

A calculation yields a pyroelectric coefficient $\gamma_3^{(2)} = 17 f(\omega) \mu C/(m^2 \cdot K)$. A dynamic pyroelectric response has been observed experimentally in a resonator consisting of along rod of ammonium dihydrophosphate.

[1] I. S. Zheludev, *Physics of Crystalline Dielectrics*, Nauka, Moscow, 1968.
[2] A. M. Glass and R. L. Abrams, J. Appl. Phys. **41**, 4455 (1970).
[3] V. F. Kosorotov, L. S. Kremenchugskiĭ, V. B. Samoĭlov, and L. V. Shchedrina, *The Pyroelectric Effect and Its Practical Applications*, Nauk. Dumka, Kiev, 1980.

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