

THE INFLUENCE OF THE POLARIZING FIELD ON THE PYROELECTRIC COEFFICIENT IN TGS

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The results from measurements made for single-crystal tryglycine sulphate of pyroelectric coefficient versus temperature in the vicinity of the Curie point ($T_0 \pm 0.5$ K) at a constant polarizing field are reported. It was found that the peak value of the pyroelectric coefficient appears at the same temperature ($T_0 \pm 0.03$ K) independently of the polarizing field in the range of the applied field ($0-10^5$ V/m). The constant β determined from these experiments equals 3.9×10^{11} Vm⁵C⁻³.

1. Introduction

An externally applied constant electric field to a ferroelectric crystal may influence the pyroelectric coefficient in two ways. It changes both its value and the location of the peak value on the temperature scale. This problem has been studied previously [1-6], but conflicting results have been reported. The displacement of the peak value of the pyroelectric coefficient toward a higher temperature with an increasing electric field strength has been observed by Glass [1] in SBN crystal, Kremenchugskii and Samoilov [2] (0.2-0.3 K) in TGS and Bye et al. [3] in LATGS at a high field ($E = 80$ kV/cm). However, Fatuzzo [4] and also Shaulov and Simhony [5], based on the thermodynamic theory, have shown that the electric field does not influence the peak position of the pyroelectric coefficient. In every case, however, the diminishing of the pyroelectric coefficient and its flattening with the increasing electric field was observed.

In this investigation an attempt was made to determine accurately the influence of the electric field on the position and value of the pyroelectric coefficient in a single-crystal TGS.

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2. Experiment

The TGS crystal sample, cut out perpendicularly to the ferroelectric axes with gold electrodes, was annealed in the paraelectric phase before the measurements. During cooling at a constant rate equal to 4.36×10^{-3} K/s, the pyroelectric current of the sample flowing through a load resistance, having a value approximately 10^6 times lower than that of the sample, produced a voltage drop. This voltage drop versus temperature was recorded with an x - y recorder. During the measurements a constant polarizing electric field was applied to the sample. Its sense agreed with the natural unipolarity of the crystal. EMF from a copper-constantan thermocouple compensated by a constant voltage of $1970.0 \mu\text{V}$ was, after amplification, connected to the x -terminal of the recorder. Measurements of signal voltages with a digital voltmeter (MERATRONIX 534 and KEITHLEY 180) were also made. The accuracy of the temperature readout was ± 0.01 K, whereas the dispersion of indications connected with long time instability of the circuit was ± 0.03 K.

3. Results and discussion

Results of the registered pyroelectric coefficient (p) and its dependence on temperature for three values of the polarizing electric field are shown in Fig. 1. Using the value of the pyroelectric coefficient determined for curve b , the function ($p^{-1/2} = f(T - T_0)$) was calculated and is shown as curve d . For low values of p this function is linear. Several

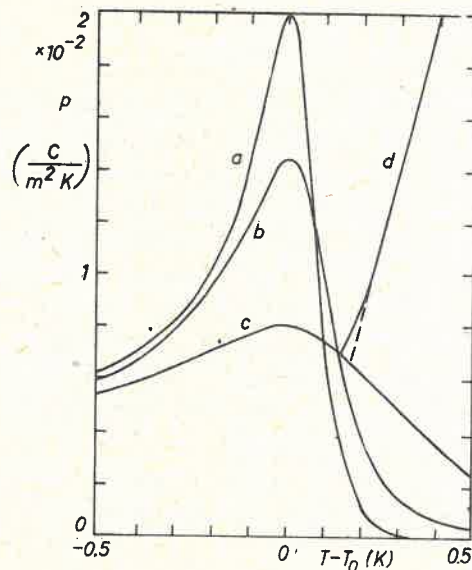


Fig. 1. Temperature dependence of the pyroelectric coefficient at a constant polarizing electric field (E). $a - E = 300$ V/m, $b - E = 3 \times 10^3$ V/m, $c - E = 2 \times 10^4$ V/m, d — quantity $p^{-1/2}$ calculated from curve b , in arbitrary units

measurements of the pyroelectric coefficient were made for different electric field values. In the range of the fields applied, i. e., from 0 to 10^5 V/m, the peak value of the pyroelectric coefficient appeared at the same temperature with an accuracy of 0.03 K. The peak value of the pyroelectric coefficient was recorded for each value of the electric field and these results are shown in Fig. 2. It can be seen that the dependence of $\log p = f(\log E)$ in the

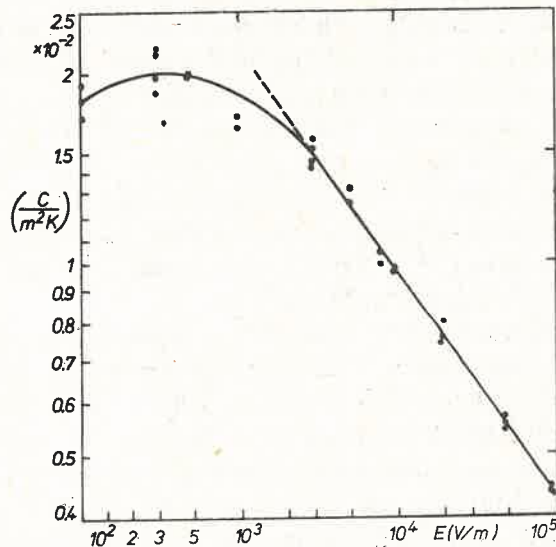


Fig. 2. Peak value of pyroelectric coefficient versus externally applied polarizing field

range of fields greater than 3×10^3 V/m is linear. The maximal pyroelectric coefficient was obtained for a field not equal to zero but approximately 300 V/m.

In the thermodynamic theory of ferroelectrics a relation between an external polarizing field (E) and polarization (P) is given by the formula:

$$E = \alpha(T - T_0)P + \beta P^3, \quad (1)$$

where α , β are constant. It is known [4, 5] that the field (E) influences the peak value of the pyroelectric coefficient and does not influence its temperature position. Close to the phase transition temperature (T_0) the pyroelectric coefficient is:

$$p = -\alpha/(3\beta^{2/3}E^{1/3}) + \alpha^3(T - T_0)^2/(3^3\beta^{4/3}E^{5/3}) \quad (2)$$

and in the paraelectric phase at the temperature satisfying the condition $T - T_0 > (3\beta^{1/3}E^{2/3})/(2^{2/3}\alpha)$, this coefficient equals

$$p = -E/\alpha(T - T_0)^2 + 5\beta E^3/4\alpha^4(T - T_0)^5. \quad (3)$$

These experimental results agree with the given formulas (2), (3) derived from Eq. (1). As seen from the experiment and the second term of Eq. (2), the pyroelectric coefficient diminishes parabolically with increasing $|T - T_0|$. In the paraelectric phase at a higher temperature, the decisive term in Eq. (3) is the first. This term may be presented as

$|p|^{-1/2} = (T - T_0)(\alpha/E)^{1/2}$. However, at the temperature approaching T_0 the second term must be included. The experimental result presented in Fig. 1 (curve *d*) is in agreement with Eq. (3). From the linear part of this curve the constant α can be calculated. The accuracy of such a measurement is, however, lower than that obtained from the measurement of $\varepsilon(T)$. At the phase transition temperature $T = T_0$, which is in accord with the first term of Eq. (2), $\log p$ is a linear function of $\log E$. From the linear part of the graph shown in Fig. 2, it is possible to calculate both the power index $E^{1/\delta}$ as well as the constant $\alpha/3\beta^{2/3}$. The critical index $\delta = 3$ calculated in this way is in agreement with the theoretical value as seen from Eq. (2). The constant α calculated from the permittivity measurement $\varepsilon(T)$ agrees with the literature data and most frequently is found to be equal to $(3.3 - 3.5) \times 10^7$ Vm/CK. For the assumed value $\alpha = 3.5 \times 10^7$, the constant β , calculated from the curve, equals $(3.9 \pm 0.5) \times 10^{11}$ Vm⁵C⁻³. According to the available literature, the constant β , measured by different methods for TGS, takes different values from the lowest, i. e., 3.79×10^{11} Vm⁵C⁻³ in [7] and (isothermal value) 4.2×10^{11} in [8] to the maximum which is approximately 10.37×10^{11} [9].

Note that the calculated β may have some systematic error. For example, if the applied electric field is not uniform in the sample, that is, if it is greater than the mean value in surface layer and lower than the mean value in the bulk, then the value of β decreases. Another factor which influences the value of β is the internal bias field. This suggests that this field is responsible for the non-typical course of the peak value of the pyroelectric coefficient (Fig. 2) at low fields $E \lesssim 3 \times 10^3$ V/m. It is also responsible for the finite value of p and for the value of the unipolarity coefficient smaller than unity at $E = 0$, ($T = T_0$). The experiment itself must be done carefully because the external electric field when applied to the sample for a long time increases its internal bias field.

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