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► **To cite this version:**

Z. Málek, J. Janta, G. Chanussot. THE INFLUENCE OF SURFACE EFFECTS ON THE PYROELECTRIC BEHAVIOUR OF T. G. S. CLOSE TO THE PHASE TRANSITION. *Journal de Physique Colloques*, 1972, 33 (C2), pp.C2-233-C2-234. 10.1051/jphyscol:1972280 . jpa-00215016

HAL Id: jpa-00215016

<https://hal.science/jpa-00215016>

Submitted on 4 Feb 2008

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THE INFLUENCE OF SURFACE EFFECTS ON THE PYROELECTRIC BEHAVIOUR OF T. G. S. CLOSE TO THE PHASE TRANSITION

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Résumé. — Une étude du coefficient pyroélectrique p^* a été réalisée pendant une élévation de température à taux constant compris entre 0,03 et 1,5 °C.s⁻¹. On a trouvé que p^* varie avec l'épaisseur (quand celle-ci décroît de 3 mm à 0,4 mm, p^* décroît 8 fois).

Ces variations sont expliquées par l'existence de couches de surface ferroélectriquement inactives. Leur influence doit se manifester dans toute méthode de mesure.

Abstract. — From the study of pyroelectric coefficient p^* during constant-rate-temperature-increase (0,03-1,5 °C.s⁻¹) its thickness dependence was found (for the thickness decrease from 3 mm to 0,4 mm p^* decreases eight times) and explained by the influence of ferroelectrically inactive surface layers. The effect is to be expected at any measuring method.

Introduction. — During the investigation of pyroelectric behaviour of ferroelectric triglycine sulfate crystals it has been found out that various physical mechanisms strongly influence the pyroelectric response of this material. This concerns in particular :

a) the compensation charges at the surface [1] which are supposed to be responsible for the pyroelectric response found regularly also in absence of any external field close above T_c [1], [2] ;

b) the internal domains [1] which are partially responsible for the asymmetry of the response ;

c) the lattice defects which influence particularly the magnitude and symmetry of the residual pyroelectric effect in the paraelectric phase [2] ;

d) the back-switching phenomena which modify strongly the pyroelectric response at low rates of temperature increase, especially in the region close below T_c [3] ;

This contribution presents experimental results, showing that sample thickness also strongly influences the pyroelectric behaviour.

Experimental procedure. — From T. G. S. single crystal cylindrical rods (the diameter of which was 2 mm) by successive cleaving samples of different thickness were prepared (the rotational axes of the rods were parallel to the b -axes of the crystals). The fresh plane surfaces were consequently treated in an identical way and covered with vacuum deposited gold electrodes.

The pyroelectric coefficient was determined by the method which had been described in previous paper [3],

i. e. the pyroelectric current i and its integrated value q were directly recorded while increasing the temperature of the sample from 46 °C across the Curie point. The range of rates of temperature increase b covered the interval from 0.03 to 1.5 °C.s⁻¹. Before each measurement the investigated sample was annealed at 90 °C, until the released charge, measured by the same method as quoted above, drops practically to zero ; then the sample was cooled down, always with the same rate, to the temperature 46 °C. Afterwards for about 5 s the dc field 1 kV.cm⁻¹ was applied in the direction, given by the polarity of the pyroelectric current delivered by the virgin sample, afterwards short-connected for a few seconds and finally connected to the input of the measuring apparatus and submitted to heating at a given rate $b = dT/dt$ through T_c . Under these conditions the reproducibility of the temperature dependences of the pyroelectric signal, measured at any given rate higher than 0.1 °C.s⁻¹ was better than 5 %.

Results. — The temperature dependences of pyroelectric coefficient $p^* = (1/S) dq/dT = i/bS$ (where S is the electrode area) evaluated from the current i measured by the method described above for three samples of the same rod are presented in figure 1. It is evident that the increase of the sample thickness in the range at least to 3 mm causes in the ferroelectric region an increase of the p^* value. Nevertheless, the total charge released in the paraelectric region is not dependent of the thickness. Analogous investigations performed in the interval of rates b given above with various groups of samples cleaved from several rods

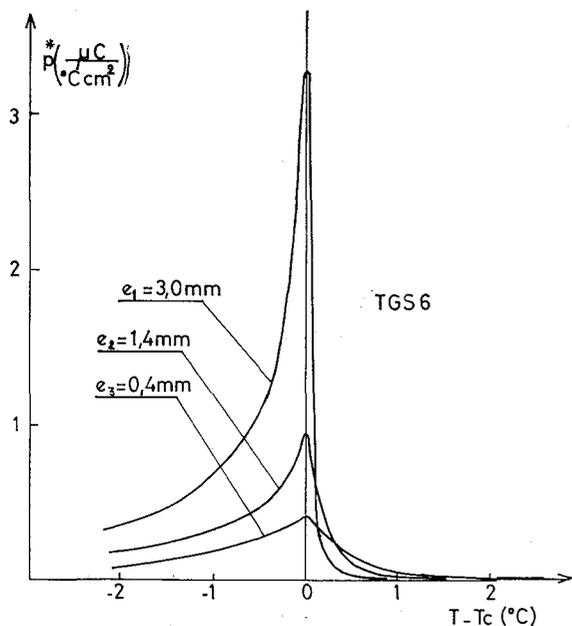


FIG. 1. — Temperature dependences of the pyroelectric coefficient p^* at the rate of temperature increase $b = 0,15 \text{ }^\circ\text{C}\cdot\text{s}^{-1}$ for three samples of the same rod, differing in their thickness e .

cut from different crystals provided qualitatively the same results.

Discussion. — The thickness dependence of p^* could be expected as a consequence of a depolarizing field inside the sample. This field, being inversely proportional to the thickness, would increase the probability of back-switching and thus decrease the total polarization of thinner samples. Nevertheless, as a result of the applied procedure the depolarizing field is compensated by the free charge from the poling source and cannot act in the way mentioned above.

Chinkolkar and Unruh [4], explained successfully the thickness dependence of permittivity of T. G. S. crystals, taking into account the role of nonferroelectric dielectric surface layers with lower value of permittivity. Let us consider their influence in our case. As the thickness of the surface layers of our samples is roughly constant (with respect to the technology mentioned above), their influence on the pyroelectric response would depend on the sample thickness.

The schematic diagram of the sample and the measuring circuit is given in figure 2. For the sake of simplicity let us consider such a small change of temperature that p and ϵ may be here taken as constant,

During the temperature increase at a rate b the change of the charge q at the capacitor C_f in the time dt is given by the difference of the charge released due to pyroelectric effect and the charge transferred through the both C_s to the measuring circuit :

$$dq_f = pb \, dt - i \, dt.$$

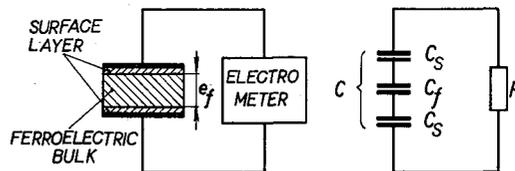


FIG. 2. — Schematic diagram characterizing the influence of surface layers capacitances in the measuring circuit.

The corresponding voltage across C_f equals to the sum of voltages across the remaining parts of the circuit :

$$du = \frac{1}{C_f} dq_f = R \, di + \frac{2}{C_s} i \, dt. \quad (1)$$

From (1) a differential equation follows immediately, the solution of which is

$$i = \frac{pb}{1 + 2C_f/C_s} \left[1 - \exp\left(-\frac{t}{RC}\right) \right].$$

With respect to the very small time constant RC in our case the steady state may be considered only and the measured pyroelectric coefficient p^* may be expressed as

$$p^* = \frac{i}{b} = \frac{p}{1 + k/e_f} \quad (2)$$

where e_f is the thickness of the ferroelectric bulk and $k = 2e_s \epsilon_s / \epsilon_f$ is the constant of proportionality.

From the relation (2) the thickness dependence of the measured pyroelectric coefficient p^* is obvious. As the surface layer parameters e_s and ϵ_s are not known, the quantitative evaluation is not possible.

Conclusion. — The thickness dependence of the pyroelectric coefficient evaluated from the direct measurement of the pyroelectric current during constant-rate-temperature-increase was explained by the influence of nonferroelectric surface layers which separate the pyroelectrically active part of the sample from the measuring apparatus. Obviously, this influence of surface layers plays a corresponding role in any method of investigation of pyroelectric properties. Thus it should be also considered when projecting pyroelectric detectors.

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