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STUDY OF THE SPONTANEOUS POLARIZATION AND COERCIVE FIELD IN TGS SINGLE CRYSTAL FROM 30 TO 1000 Hz*

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Sponataneous polarization P_s and coercive field E_c are studied at room temperature *vs.* the measuring field frequency f from 30 to 1000 Hz in TGS samples of various thickness. As f increases, P_s decreases, whereas E_c increases attaining a maximum and then decreasing. Samples aged, rejuvenated with electrodes, and rejuvenated without electrodes, were studied. Ones rejuvenated without electrodes exhibited, at a given f-value, time-dependent changes in E_c and slight changes in P_s.

The E_c-maximum is interpreted as related with the course of the polarization switching process; this approach permits the determination of the polarization switching time, leading to times of the order of 10^{-3} s.

1. Introduction

Few papers deal with measurements of the dielectric hysteresis loop parameters versus the measuring field frequency in TGS single crystals [1].

For low frequencies up to 0.1 Hz such studies have been carried out by Gurevich, Zheludiev and Rez [2], and by Gurevich and Zheludiev [3], who report an increase in coercive field with increasing frequency and strength of the measuring field.

Pawlaczyk and Hilczerowa [4] studied the polarization vs. frequency dependence for irradiated TGS samples from 30 to 3000 Hz, and report a decrease in spontaneous polarization with increasing frequency.

The present paper contains results for the spontaneous polarization and coercive field as functions of the measuring field frequency in TGS single crystals at various stages of the aging process.

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2. Experimental

2.1. Preparation of the samples

Samples for measurement were cut, perpendicularly to the ferroelaectric axis, from the growth pyramid (110) of TGS single crystals, grown in the paraelectric phase (at 51°C). Seven samples, with an area of $S = 0.59$ cm^2 and thickness ranging from 0.07 to 0,366 cm, were thus prepared.

The dielectric hysteresis loop was studied with a Diamant–Drenck–Pepinsky circuit [5], constructed by "Radiopan". The parameters of the loop were read from a digital voltmeter.

2.2 Experimental results

a) Thickness effect. Measurements of the spontaneous polarization P_s and coercive field E_c as functions of the measuring field frequency f were performed. Fig. 1 shows P_s vs f for samples of various thickness. P_s is found to decrease with increasing frequency,

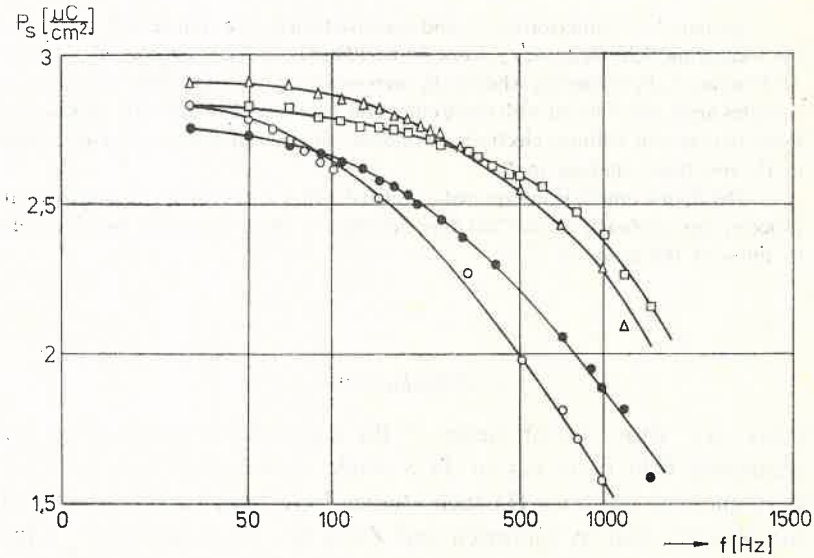

Fig. 1. Sponataneous polarization P_s vs measuring field frequency f for TGS samples of various thickness: □ — 0.70 mm, △ — 1.18 mm, ○ — 1.40 mm, ● — 3.66 mm

the more steeply the thicker is the sample. Fig. 2 shows E_c vs f for variously thick samples. Here, with increasing frequency, E_c at first tends to a maximum and then decreases. With increasing thickness of the sample, the maximum of E_c shifts towards lower frequencies. Table I gives the coercive field maximum E_c^{max} as well as the frequencies f of the measuring field corresponding to E_c^{max} for different values of the sample thickness.

b) Aging effect. Measurements of P_s and E_c vs f were carried out for aged samples, the ones rejuvenated with electrodes, and the ones rejuvenated without electrodes.

On rejuvenation with electrodes, E_c was found to have decreased. On electrode-less rejuvenation, E_c decreases considerably as soon as a 50 Hz AC field is applied; when the sample is maintained in the field, E_c increases. In Fig. 3, the successive numbers 4, 6, 8, ...

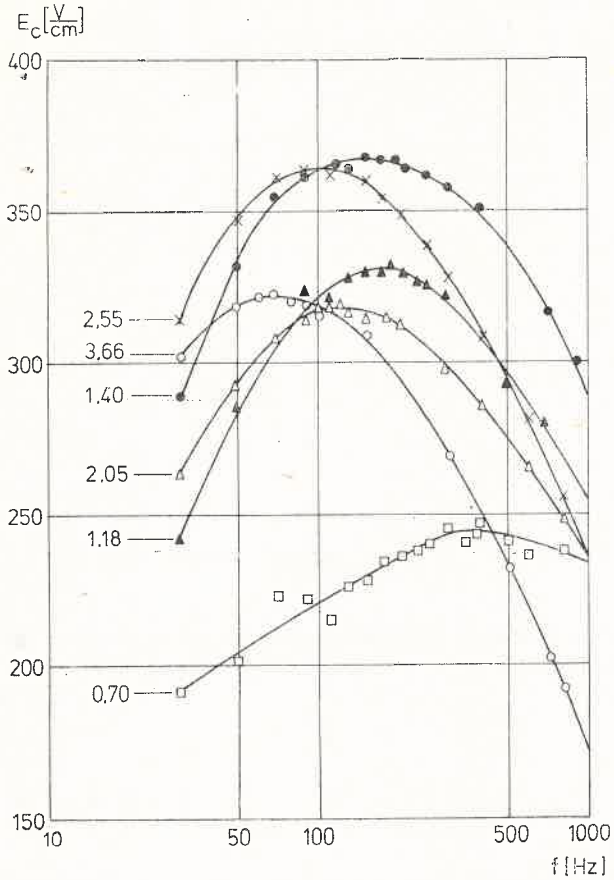

Fig. 2. Coercive field E_c vs measuring field frequency f for TGS samples of various thickness

TABLE I

Coercive field maximum E_c^{max} and corresponding measuring field frequency f for TGS samples of various thickness

d [cm]	f [Hz]	$E_c^{max}\left[\frac{V}{cm}\right]$
0.070	350	245
0.118	190	332
0.140	170	368
0.205	120	318
0.255	100	365
0.366	70	322

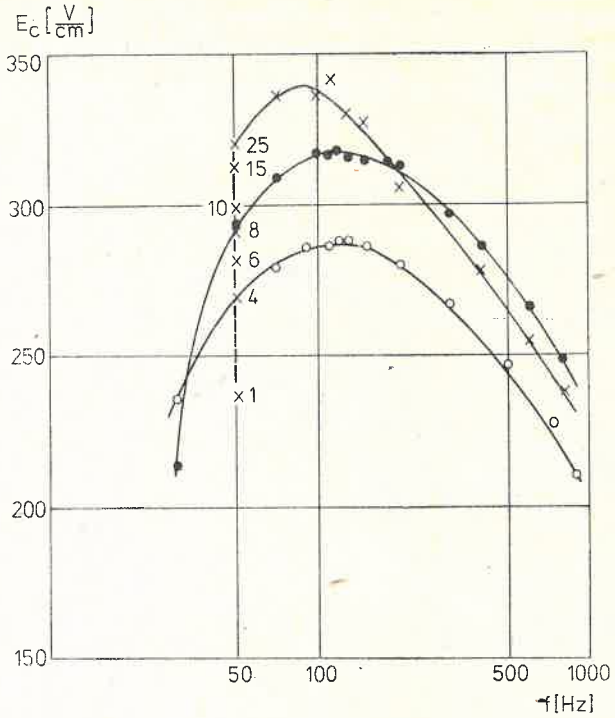

Fig. 3. Coercive field E_c vs measuring field frequency f for: aged TGS sample — ●; sample rejuvenated with electrodes — ○; sample rejuvenated without electrodes — ×

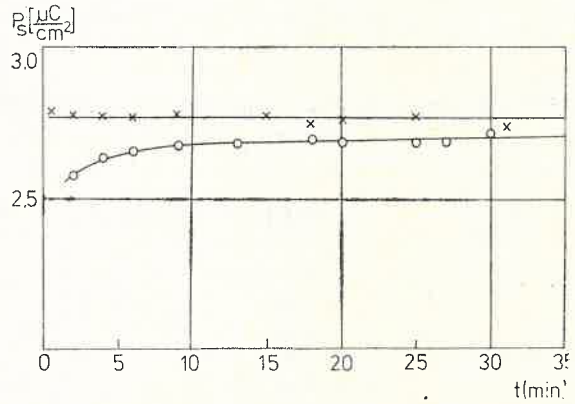

Fig. 4. Sponataneous polarization P_s vs time, at the frequency 50 Hz, for TGS sample rejuvenated with — × and without — ○ electrode

denote the time elapsed since the moment of application of the field. After about 25 min., E_c settles at a value which is higher than that of the aged sample. As examples, Figs 4 and 5 show graphs of the time-evalution of P_s and E_c for a sample rejuvenated with and without electrodes.

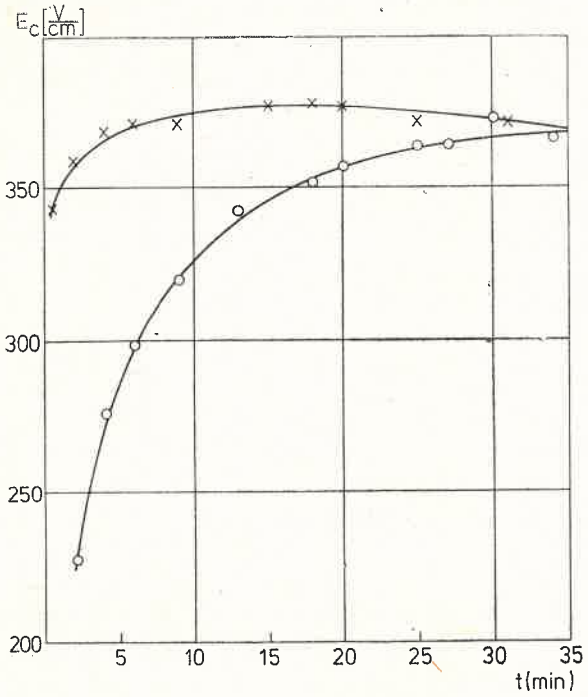

Fig. 5. Coercive field E_c vs time, at the frequency 50 Hz, for TGS sample rejuvenated with — × and without — ○ electrode

3. Discussion of the results

The coercive field maximum, dependent on the frequency of the measuring field, is accessible to explanation on the basis of domain structure mobility. At low frequencies, the domains keep pace with the variations of the measuring field vector, so that one observes low values of the coercive field. As the measuring field frequency is raised, some domains become more difficult to switch owing to their high inertia, and one observes a maximum $E_c^{\max}$ of the coercive field. At still higher frequencies of the measuring field some domains no longer keep pace with the AC measuring field and thus take no part in the process of polarization switching, so that one now observes a decrease in value of the coercive field. Once the frequency corresponding to $E_c^{\max}$ is known, one can evaluate the polarization switching time from Debye's formula:

$$f \cdot \tau = 1,$$

where f is the frequency, and τ the relaxation time.

Table I gives the relaxation time, calculated for samples of various thicknesses. The thicker the sample, the longer is the time of polarization switching. The time obtained is of the same order of magnitude as that determined from the polarization switching process [6]. The polarization switching studies [6] of Hilczerowa have led to a time of the order of 10^{-3} s in rejuvenated TGS samples in static electric fields of 1.5 kV/cm.

Our interpretation is furthermore supported by the fact that the product of the thickness and frequency at which the coercive field becomes maximal is, within the limits or error, constant (Table II).

TABLE II

Polarization switching time, frequency corresponding to E_c^{max}, and product of the thickness and above frequency f value, for TGS samples of various thickness

d [cm]	τ [s]	f [Hz]	$d \cdot f$ [cm · Hz]
0.070	$2.8 \cdot 10^{-3}$	350	24.5
0.118	$5.3 \cdot 10^{-3}$	190	22.4
0.140	$5.9 \cdot 10^{-3}$	170	23.8
0.205	$8.3 \cdot 10^{-3}$	120	24.6
0.255	$10.0 \cdot 10^{-3}$	100	25.5
0.366	$14.3 \cdot 10^{-3}$	70	25.6

The influence of the degree of ageing on the value of E_c, measured at 50 Hz, can be interpreted in terms of the domain structure of the various samples [7, 8]. In aged TGS samples, the domains are large and partially clamped, leading to large coercive field values (294 V/cm). On rejuvenation with electrodes, the number of domains remains unchanged but the structure is loosened, so that E_c decreases (270 V/cm). On electrode-less rejuvenation of the same sample, the structure undergoes a frittering (to 100 domains per mm in the growth pyramid (110)), the distribution of oppositely directed domains being equal. Immediately on application of the electric field, E_c is found to be low (230 V/cm), since the energy losses involved by polarization switching are small. As the sample is maintained in the field, it tends to the single-domain state, and E_c increases.

Resuming the preceding reasoning, the following statements can be made:

(1) When measuring the coercive field E_c as a function of the frequency f, a maximum of E_c is observed;

(2) the thicker the sample, the lower is the frequency f at which E_c becomes maximal;

(3) by having recourse to the preceding measurements and Debye's formula, it is feasible to calculate the polarization switching time of the sample;

(4) measurements of spontaneous polarization P_s vs f show that P_s decreases as f increases and that this process occurs the more steeply the thicker is the sample;

(5) a sample having a fine domain structure (subsequent to electrode less rejuvenation) exhibits a ow E_c-value which, after an electric field is applied, increases in time (for about 25 min.) up to the value exhibited by the sample on rejuvenation with electrodes.

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