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► To cite this version:

V.V. Demjanov, S.P. Solov'Ev. Dielectric dispersion of irradiated BaTio3 near the phase transition. *Revue de Physique Appliquée*, 1972, 7 (2), pp.81-82. 10.1051/rphysap:019720070208100 . jpa-00243608

HAL Id: jpa-00243608

<https://hal.science/jpa-00243608>

Submitted on 1 Jan 1972

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DIELECTRIC DISPERSION OF IRRADIATED BaTiO₃ NEAR THE PHASE TRANSITION

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(Reçu le 16 novembre 1971, révisé le 10 février 1972)

Résumé. — Jusqu'à la fréquence de $1,5 \times 10^{10}$ Hz la perméabilité diélectrique et les pertes diélectriques ont été étudiées dans BaTiO₃ polycristallin irradié dans la pile jusqu'au flux intégré de neutrons rapides $\varphi \simeq 5 \times 10^{19}$ cm⁻². Lors de $\varphi_c \simeq 5 \times 10^{18}$ cm⁻² on a découvert l'accroissement brusque de la fréquence de dispersion f_0 et la transformation d'état induite par irradiation qui correspondent au maximum de la dépendance de ε avec φ . Les changements qualitatifs observés de ε et f_0 sont analogues à ceux des mêmes paramètres au voisinage du point de Curie pour BaTiO₃ non irradié. Lors de $\varphi < \varphi_c$ la valeur mesurée du déplacement de la constante de Curie dans la direction des températures inférieures dépend de la fréquence selon la loi $\Delta T_c(f) = \Delta T_0 \ln(f'_0/f)$ ou $\Delta T_0 = 4-5$ °C et $f'_0 \simeq 10^{11}$ Hz.

Abstract. — Frequency dependence of the dielectric constant (ε') and $\text{tg } \delta$ up to 1.5×10^{10} c/s have been measured for polycrystalline BaTiO₃ irradiated with fast neutron integrated fluxes up to $\varphi \simeq 5 \times 10^{19}$ cm⁻². The radiation induced phase transition takes place at $\varphi_c \simeq 5 \times 10^{18}$ cm⁻² corresponding to a maximum in the ε vs φ dependence and a fairly sharp increase in the dispersion frequency f_0 . Qualitatively, changes of ε' and f_0 are analogous to these near the Curie point for non-irradiated BaTiO₃. At $\varphi < \varphi_c$ the Curie point shift to lower temperatures depends upon the frequency according to the law $\Delta T_c(f) = \Delta T_0 \ln(f'_0/f)$, where $\Delta T_0 = 4-5$ °C and $f'_0 \simeq 10^{11}$ c/s.

It is known that irradiation of BaTiO₃ by fast neutrons causes the Curie point to shift to lower temperatures including a radiation induced phase transition into the paraelectric state at integrated neutron fluxes $\varphi > 10^{18}$ cm⁻² [1]-[4]. One qualitatively observed similar changes in BaTiO₃ crystal lattice parameters as a function of both T and φ .

For a more detailed study of the radiation induced phase transformation in BaTiO₃ the dielectric properties (ε' and $\text{tg } \delta$) of in-pile irradiated polycrystalline samples of BaTiO₃ were measured at radio and microwave frequencies using the method described in [5].

Low signal dielectric constant ε' vs φ dependences

FIG. 1. — Dielectric constant ε' (curves 1, 2) and frequency f_0 of microwave dispersion (curve 3) vs integrated fast neutron flux for polycrystal BaTiO₃. Curves 1 and 2 measured at 50 °C and at the frequencies 3×10^5 and 1.5×10^{10} c/s respectively.

are shown in figure 1 for the range of frequencies 3×10^5 c/s and 1.5×10^{10} c/s. It follows from figure 1 that at $\varphi \equiv \varphi_c \simeq (3 - 6) \times 10^{18}$ cm⁻², corresponding to the radiation induced phase transition [1]-[4], one observes in the $\varepsilon'(\varphi)$ dependence an anomalous increase of ε' similar to that in the $\varepsilon'(T)$ dependence at $T = T_c$ for non-irradiated BaTiO₃.

In the $\varphi < \varphi_c$ range ($< 10^{18}$ cm⁻²) one observes an increase in frequency corresponding to the start

FIG. 2. — Temperature dependences of ε' (curves 1-4) $\text{tg } \delta$ (curves 1'-4') and a factor $\eta = \varepsilon'_{15 \text{ Hz}} / \varepsilon'_{300 \text{ Kc/s}}$ (curves 5, 6). 1, 1', 2, 2', 5 — before irradiation ($\varphi = 0$). 3, 3', 4, 4', 6 — after irradiation $\varphi = 6 \times 10^{18}$ cm⁻². Curves 1, 1', 3, 3' and 2, 2', 4, 4' measured at frequencies 3×10^5 and 1.5×10^{10} c/s respectively.

of microwave dispersion, and one also observes an increase in the frequency f_0 corresponding to the midpoint of the dispersion which is also determined from the maximum of the $\varepsilon''(f)$ curve. Herewith the value of the dielectric constant, corresponding to the end of the dispersion, remains practically unchanged. These changes of the BaTiO₃ dielectric spectrum (not shown in Fig. 1-3) are in agreement with the assumption of domain fractionation as in the case of electron irradiation [5].

The data shown in figures 2 and 3 also testify to the fact that ferroelectric BaTiO₃ displays the properties of the paraelectric phase more and more

FIG. 3. — Frequency dependences of ε' (curves 1-3) and $\text{tg } \sigma$ (1'-3') for neutron irradiated BaTiO₃.

- 1, 1' — before irradiation ($\varphi = 0$).
 2, 2' — $\varphi = 6 \times 10^{18} \text{ cm}^{-2}$.
 3, 3' — $\varphi = 2 \times 10^{19} \text{ cm}^{-2}$.

intensively with increasing φ . In particular, the frequency f_0 increases and at $\varphi > \varphi_c$ gets into the frequency range characteristic of infrared dispersion

of BaTiO₃ (Fig. 1). At $\varphi \simeq \varphi_c$ in parallel with the f_0 increase, parameters of the Curie-Weiss law irreversibly change; T_c shifts to lower temperatures and the value of ε'_0 and ε'_∞ increase (Fig. 1).

At the same time one observes the broadening of the $\varepsilon'(T)$ and $\text{tg } \delta(T)$ curves measured both at low and microwave frequencies (Fig. 2). Herewith within the temperature range $T < T_c$, $\text{tg } \delta$ decreases and a factor η , which is called the depth of dispersion (see Fig. 2), increases. Within the range $T > T_c$ the opposite occurs. Upon irradiating simultaneously with the phase transition broadening, some details of the $\varepsilon'(T)$, $\text{tg } \delta(T)$ and $\eta(T)$ curves observed for non-irradiated BaTiO₃ [6], [7], vanish.

It is noteworthy that the $f_0(\varphi)$ behaviour near φ_c appears to be qualitatively similar to the $f_0(T)$ dependence near T_c for non-irradiated BaTiO₃ [6], [7] and must be related to the radical changes of domain structure at the radiation induced phase transition, namely to the transformation of domain structure into a dynamically stable system of nuclei of spontaneous polarization (polarization fluctuations) in the paraelectric phase [8]. The accumulation of high barrier radiation defects as a reason for the T_c shift [2], [3] must cause simultaneously, the decrease of the correlation radius of atomic displacements in the polarization nuclei and hence result in the observed increase of the dispersion frequency f_0 (Fig. 1).

It is of interest that the phase transition temperature shift ΔT_c , obtained from the data shown in figure 2 at $\varphi \gtrsim \varphi_c$, depends upon frequency according to the law $\Delta T_c(f) = \Delta T_0 \ln(f'_0/f)$ where $\Delta T_0 = 4 - 5^\circ\text{C}$ and $f'_0 \simeq 10^{11} \text{ c/s}$. The $\Delta T_c(f)$ behaviour for neutron irradiated BaTiO₃ is evidence of relaxation processes whose threshold energy is about 3 eV per defect at $\varphi = 8 \times 10^{18} \text{ cm}^{-2}$.

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