High-accuracy determination for optical indicatrix rotation in ferroelectric DTGS

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Received 26.08.1999

Abstract

Optical indicatrix rotation in deuterated ferroelectric triglycine sulphate is studied with the high-accuracy null-polarimetric technique. The behaviour of the effect in ferroelectric phase is referred to quadratic spontaneous electrooptics.

Key words: ferroelectrics, triglycine sulphate, deuteration, optical indicatrix, null-polarimetry

PACS: 07.60; 64.70.Hz; 77.84.Fa; 78.20Jq

Introduction

Among ferroelectric materials, the crystals of triglycine sulphate (TGS) group are known as modeling objects, of which physical properties have been thoroughly studied [1]. TGS has the monoclinic point symmetry *2/m* in its paraelectric phase and undergoes a phase transition into ferroelectric state (point group *2*) at T_C = 49 °C. Because of the low symmetry, different crystal optical effects (birefringence, optical indicatrix rotation, optical activity) overlaped in TGS, making their correct experimental measurements difficult. This is why the optical parameters of TGS have been determined with a only relative success, and there still exists discrepancies in the results by different authors (see [2-5]). The situation is even worse for the deuterated isomorph of TGS (DTGS) which has attracted some attention of researchers in relation with the possible applications of its pulse repolarization [6].

The aim of this work is to measure, with highly accurate polarimetric methods, the indicatrix rotation in both paraelectric and ferroelectric phases, the effect which, to our knowledge, has not yet been investigated in DTGS.

Experimental Methods and Results

A 2.53 mm thick *y*-cut DTGS sample (the deuteration degree x = 0.7 and T_C = 56.5 °C) was prepared for the experiments. A thermostat enabled us to control the sample temperature with the tolerance better than 0.01 °C. The temperature range under test was from 27 to 68 °C and the working wavelength λ = 632.8 nm. No dc voltage had been applied, so that we dealt with a multidomain sample.

On contrary to most of the previous works where the method of synchronous polarizers' rotation has been employed for studying the indicatrix rotation, we have used the universal null-polarimetric technique described elsewhere [7]. The measured quantities are as follows:

$$dY/d\theta = \cos\Delta, \quad \varepsilon_0 = 2k - p_0 + \delta Y \cot(\Delta/2), \qquad (1)$$

$$\theta'_0 = (k-p)\cot(\Delta/2) + \delta Y/(1-\cos\Delta) + \theta'_{orig} + \Delta\theta, \qquad (2)$$

where Δ is the phase retardation defined by birefringence, k the eigenwave ellipticity related to optical activity, $\Delta\theta$ the indicatrix rotation, θ_{orig} the apparatus constant, and p, p_0 and δY the imperfection parameters of the optical equipment [7].

For the sake of conciseness we do not present here the temperature dependences of the $\cos\Delta$ and ε_0 parameters but only that of the characteristic azimuth θ'_0 (fig. 1).

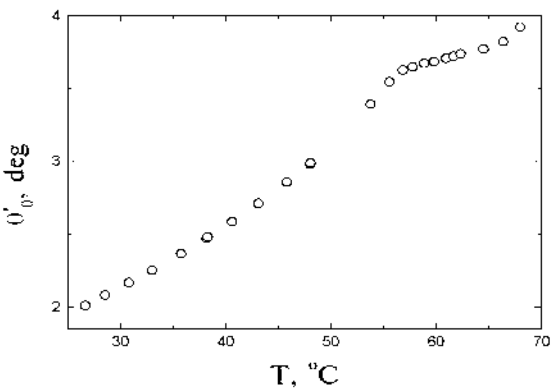

Fig.1. Temperature dependence of the polarizer reading θ'_0 for DTGS.

Identifying those data with the true indicatrix rotation, one can see that the latter varies nonlinearly with temperature in the paraelectric phase, the fact which is difficult to justify. On the second view, one realizes that this is associated with the terms p and δY in formula (2) rather than the optical behaviour of DTGS itself.

More accurate determination of $\Delta\theta$ thus requires elimination of those parasitic contributions and may be done in the following way. Since the plot of ε_0 vs $\cot(\Delta/2)$ proves to be linear with a low enough mean square deviation (4.7×10^{-3}deg), we see that appreciable unipolarity in the sample and the optical activity effect are absent ($k = 0$ - see formula (1)). This is also consistent with simple symmetry considerations. We obtain on this basis the values $p_0 = 0.283$ and $\delta Y = 2.1\times10^{-2}$deg. Let us now introduce the parameter

$$Y = \theta'_0 - \delta Y/(1-\cos\Delta) = -p\cot(\Delta/2) + \theta_{orig} + \Delta\theta. \qquad (3)$$

$\Delta\theta$ should behave linearly in paraphase ($\Delta\theta = \alpha T + \beta$), so one has from (3)

$$dY/dT = \alpha - p\, d[\cot(\Delta/2)]/dT. \qquad (4)$$

Differentiating graphically the $Y_0(T)$ and $\cot(\Delta(T)/2)$ dependences with formula (4) yields $p = -8.4\times10^{-3}$deg, with the mean square deviation 1.1×10^{-3} deg. The final refined indicatrix rotation data are depicted in fig. 2.

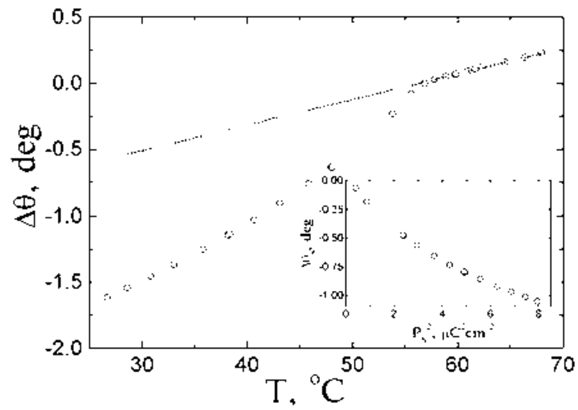

Fig.2. Temperature dependence of the indicatrix rotation in DTGS is relative to its valueat T_C. The inset represents relationship between the spontaneous part of the indicatrix rotation and the spontaneous polarization in the ferroelectric phase.

Discussion

Basing on the above results, one can well fairly evaluate the experimental accuracy of determining the indicatrix rotation in DTGS as being of the order of 10^{-3}deg. As far as we know, it is the highest value ever achieved in this kind of measurements. Our present results also strongly support the point of view of the authors [3] that accounting consistently for the imperfection parameters of optical apparatus is a necessary condition for obtaining reliable indicatrix rotation data, even if one utilizes calcite polarizers of the highest commercial grade.

The $\Delta\theta$ vs T dependence appears to be linear in the paraphase (the slope being 1.66×10^{-2}deg°C^{-1}), while a spontaneous increment $\Delta\theta_S$ appearing at lower temperatures can quite naturally be prescribed to the onset of spontaneous polarization, due to a quadratic electrooptical effect ($\Delta\theta_S = M P_S^2$, where $M = -30.3$ m^{4}C^{-2} - see fig. 2, inset). Such the M coefficient correlates with the value $M = -21.3$ m^{4}C^{-2} reported in [5] for the pure TGS, though is rather different from $M = -14.0$ m^{4}C^{-2} derived with less accurate method [3]. The deviations from linearity in the $\Delta\theta_S$ vs P_S^2 dependence observed at temperatures far from T_C may be presumably referred to higher-order electrooptical effects.

The authors are grateful to Prof. L.A.Shuvalov and Dr. A.I.Otko for samples. Two of us (O.S.K. and O.A.B.) wish to thank Prof. M.O.Romanyuk for helpful discussion.

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