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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2000

Dielectric Properties and Low Field Switching of Partially Deuterated TGS

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The dependence of ferroelectric parameters (saturation polarization, transition temperature, Curie constant,...) with deuteration in mixed crystals of $(DTGS)_x(TGS)_{1-x}$ has been characterized by means of hysteresis loops and dielectric constant measurements. Although the transition character is known to be second order for all compositions, a slight evolution towards a tricritical point was ascertained.

INTRODUCTION

It is well known[1-3] that deuteration of mixed ferroelectric crystals $(DTGS)_x(TGS)_{1-x}$ changes their structural characteristics and consequently some other physical properties.

Hysteresis loops and dielectric constant measurements were made in order to determine the transition temperature T_C (x) as a function of the deuteration percentage and to investigate the relationship between the degree of deuteration and the type of ferroelectric transition undergone. This relationship was earlier investigated[4] for the isomorphous system $(DTGSe)_x(TGSe)_{1-x}$, showing an evolution from a tricritical point ($x= 0$) to a first order transition with an appreciable thermal hysteresis ($x=0.90$).

Switching[6-8] as a temperature dependent process must show as well changes with sample composition for measurements performed at room temperature (RT).

EXPERIMENTAL

Samples of $(DTGS)_x(TGS)_{1-x}$ crystals, with different nominal deuteration percentage ($x = 0.5, 0.7, 0.9$) have been studied. Another pure TGS sample was used as reference in order to compare the observed behavior. The deuterated samples had thickness 1.3 to 1.6 mm and areas 3 to 4 mm². The pure TGS sample was a regular disk of 1.2 mm and 20 mm² respectively.

The sample holder was a copper cylinder with three concentric compartments. The inner compartment contained the sample in contact with the electrodes and the thermocouple whose signal was measured by a Keitley voltmeter model 196. In the intermediate cylinder were located the electric connections to a temperature controller, Unipan type 680, with an accuracy of ± 0.1 K. The outside cylinder was used for refrigeration.

Hysteresis loops were obtained by using a DDP bridge and observed with an Nicolet 310 oscilloscope. Recording data process was fully automated.

Dielectric constant measures were performed with a Hewlett-Packard LCR meter 4284A applying a voltage of 1V and a frequency of 1 KHz. The data were as well automatically recorded.

Switching measurements were performed applying rectangular bipolar voltage pulses from a Hewlett-Packard 33120A generator that, after amplification by a Kepco bipolar amplifier, resulted in an almost linear signal[7]. The pulse time is longer than switching time and the rise time of the pulse is about 500 µs. Both, the applied voltage and the transient current were observed and measured by means of a H.P. 54603B oscilloscope.

RESULTS AND CONCLUSIONS

Figure 1 shows the behavior of the spontaneous polarization P_S as a function of temperature T for $(DTGS)_x(TGS)_{1-x}$ with $x=0, 0.5, 0.7$ and 0.9. All the curves display the second order transitions and they tend to converge at lower temperature.

We can characterize the second order transition of deuterated $DTGS$ making use of the standard effective field approach[5]:

$$e = \frac{T}{T_C} \tanh^{-1} p - p(1 + gp^2 + hp^4)$$ (1)

where $e \equiv E/\beta N_\mu$, and $p \equiv P/N_\mu$ (being P dipolar polarization and $N_\mu \equiv P_{S0}$ the saturation polarization), and g and h dimensionless coefficients that characterize the transition. For $e=0 \Rightarrow p = p_S$ and we can write

$$\frac{T}{T_C} = \frac{p_S}{\tanh^{-1} p_S}(1 + gp_S^2 + hp_S^4) = \frac{p_S}{\tanh^{-1} p_S}(1 + g'p_S^2),$$

defining, a linear relation between g' and p_S^2 $\quad g' = g + hp_S^2$ (2)
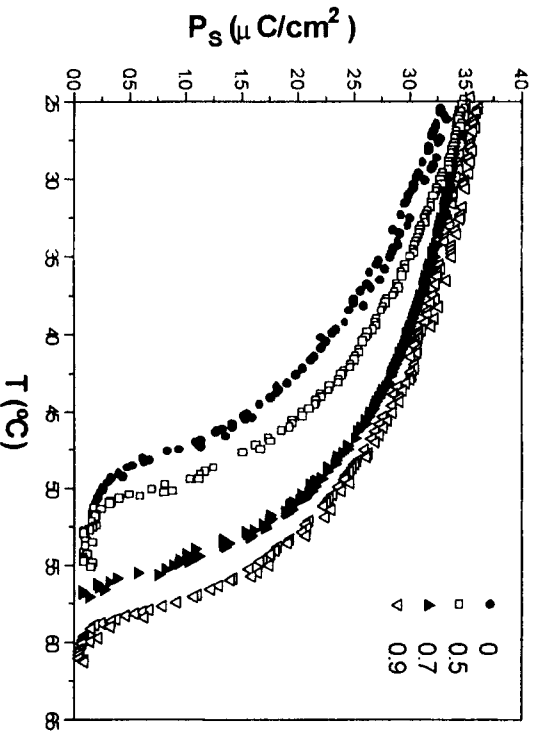
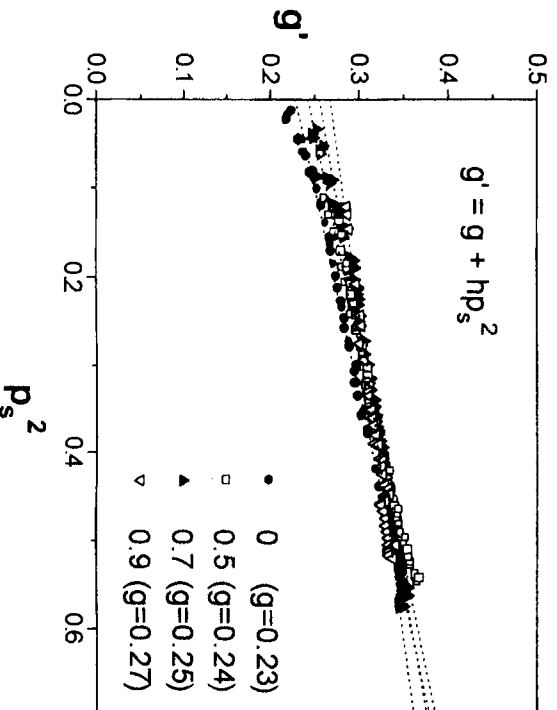FIGURE 1. P_S vs. T for $(DTGS)_x (TGS)_{1-x}$ at different compositions.

FIGURE 2. Plots of g' vs. p_s^2. The plots have been fitted linearly in order to determine the coefficient g for each composition.

Figure 2 shows that the linear relationship (2) between g' and p_s^2 is fulfilled experimentally for each one of the four compositions. In spite of the great similarity in their behavior we can note a slight evolution in the character of the transition that can be also observed in the polarization curves (Figure 1). The g values, all them lower than 1/3 as correspond to the second order transition, experiment a monotonic increase with x suggesting that the ordinary critical point evolves towards a tricritical point.
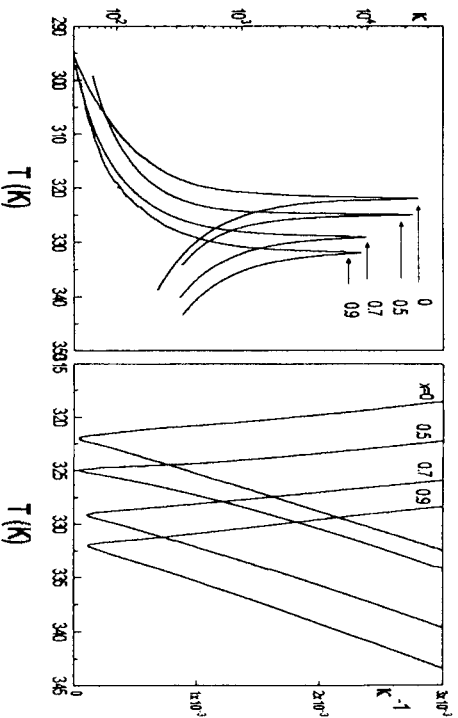

FIGURE 3.a). Dielectric constant and b) Inverse of dielectric constant vs. temperature for each composition.

Figure 3 a) shows the plots of dielectric constant κ vs. T for samples with different compositions. All of them show, as expected, pronounced peaks in the vicinity of their T_C, so the transition temperature can be properly determined for each molar fraction x. It can be checked[1] that the transition temperature rises with the increase of x, in a small range of around 10ºC between 0 and 100% of deuteration. and it may be noted as well that deuteration seems to produce a decrease of the κ peak values[4].

Figure 3 b) plots the inverse of dielectric constant, κ^{-1}, vs. T. The slopes of these plots ($T>T_C$) are very similar[2], so they suggest that there is a little change of $C(x)$.
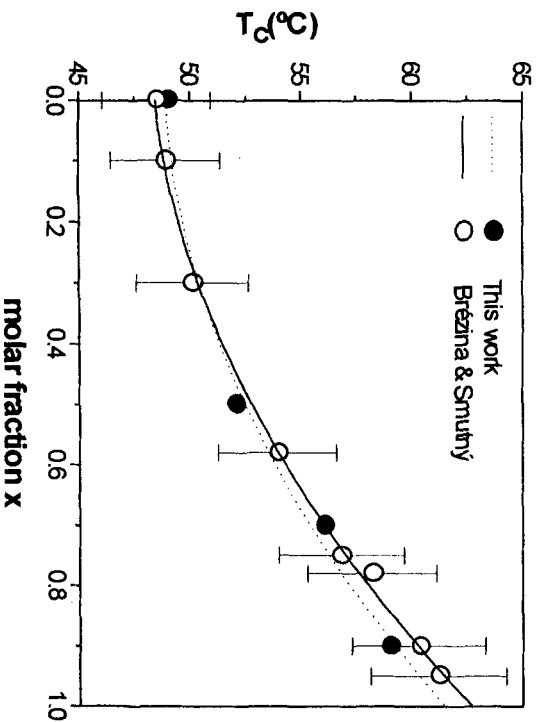
FIGURE 4. Plot of Curie temperature vs. composition x. Error bars have been taken from Brezina's data (Ref.1)

Figure 4 plots $T_C(x)$ obtained from the dielectric constant maximum peaks vs. x (Fig.3a). The second order fit might be explained in terms of an effective elementary dipolar moment $\mu_{eff}(x)$ due to the interactions between dipoles containing H atoms and those containing D atoms for intermediate compositions of mixed crystals.

From the expression of spontaneous polarization (1), $e = 0$

$$P_S^2(x) \approx 3N^2 \mu_{eff}^2(x) \left[\frac{T_C - T}{T} \right] \qquad (3)$$

We can evaluate $\mu_{eff}(x)$ from the slope of the linear fits to $P_S^2(x)$ vs. $(T_C - T)/T$ for every composition and, as it can be observed in Fig.5, $\mu_{eff}(x)$ so obtained is a linear function of x. Consequently this gives a non linear dependence of the Curie temperature

$$T_C(x) = \frac{\beta(x) N \mu_{eff}^2(x)}{k_B} . \qquad (4)$$

where $\beta(x)$ is the mean field coefficient that undergoes negligible changes with composition.

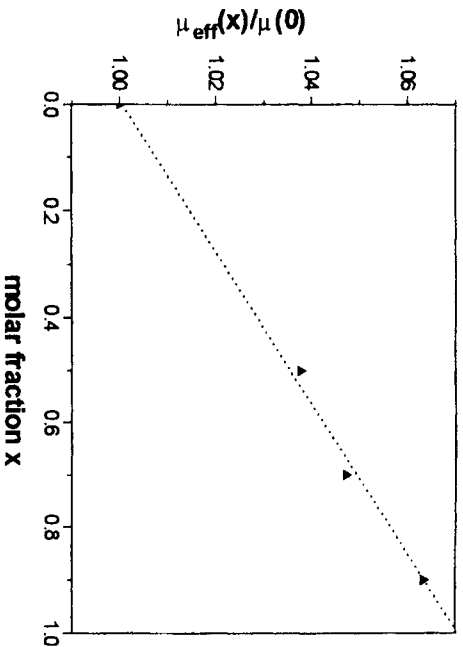C. ARAGÓ *et al.*

$\mu_{eff}(x)/\mu(0)$

molar fraction x

FIGURE 5. Plot of the normalized effective dipolar moment $\mu_{eff}(x)/\mu(0)$ vs. composition. (Note that glycine CH_2 groups are not deuterated)

Figure 6 shows the switching behavior of the deuterated samples. All they present the two well known[6-8] regimes (a 3/2 power regime for $E_m \ll E_m^*$, and a linear regime for $E_m \ll E_m^*$) in the plot of j_m vs. E_m, E_m being j_m the maximum switching current density and E_m the applied field value at which this maximum occurs. E_m^* denotes the applied field value corresponding to the change in regime. Note that previous results by Fatuzzo & Merz[6] and others fitted using an activation field relationship are different because they are taken with very short pulses of much larger peak voltages instead of approximately linear ramps as in our work.

We can see that deuteration acts in the opposite sense that increasing temperature does[8], retarding the whole switching process and giving rise to a larger coercive field as it can be seen at Figure 7. This might be understood in terms of a growing effective elementary dipolar moment making more difficult to switch dipoles at the domain walls.
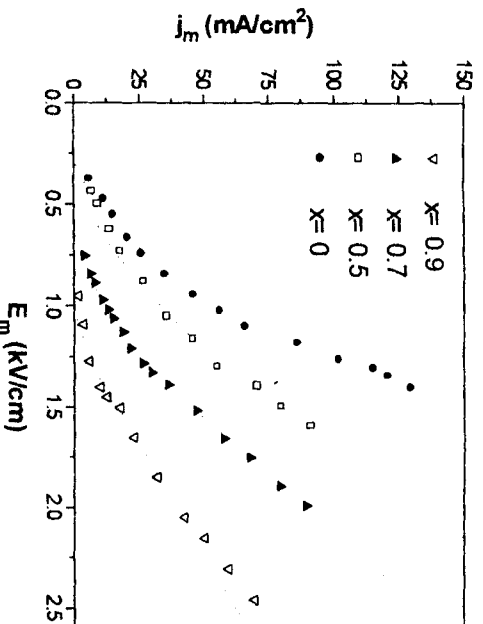
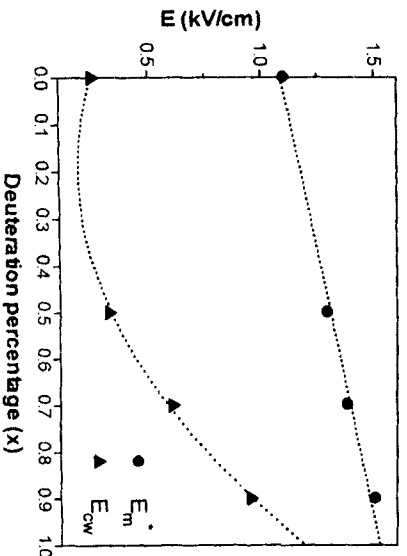FIGURE 6. Plots of the maximum switching current density j_m vs. the maximum of applied field values E_m.

FIGURE 7. Coercive field E_{cw} corresponding to domain wall motion and E_m, the field value at which change between regimes occurs, vs. composition.

In summary, we have characterized the transition of mixed crystals $DTGS_xTGS_{1-x}$ through hysteresis loops and dielectric constant measurements, and we have checked that the results of the measurements are in agreement. While remaining second order, the behavior of the parameter $g(x)$ indicates a monotonous evolution towards a tricritical point from $g(0)=0.23$ to $g(0.9)=0.27$.

Besides the switching behavior of deuterated compositions keep concordance with the transition temperatures earlier obtained and it indicates that deuteration enlarges the coercive field making more difficult the whole switching process.

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