Dielectric relaxation in ferroelectric TAAP near the Curie temperature

LETTER TO THE EDITOR

Dielectric relaxation in ferroelectric TAAP near the Curie temperature

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Abstract. We have measured the relaxation time in ferroelectric TAAP near the Curie temperature. The dielectric relaxation in this crystal could be described to a good approximation by the Debye equations. The relaxation time exhibits critical slowing down characteristic of an order–disorder transition.

Telluric acid ammonium phosphate (TAAP) having the chemical formula $Te(OH)_8 \cdot 2NH_4H_2PO_4 \cdot (NH_4)_2HPO_4$ is a monoclinic crystal (Pn) which is ferroelectric at room temperature and undergoes a second-order phase transition to a non-polar phase at the Curie temperature of 45 °C (Guilliot Gauther *et al* 1984). Recent Raman spectroscopic studies of this crystal suggest that the phase transition takes place predominantly through the order–disorder nature of the hydrogen bonds of the type N–H—O (Shashikala *et al* 1990). In this letter we report studies of the dielectric relaxation near the Curie temperature.

Optical quality single crystals of TAAP were grown at room temperature (25 °C) by water evaporation from the aqueous solution of telluric acid, ammonium hydrogen phosphate and diammonium hydrogen phosphate mixed in required proportions. All the measurements were carried out on {101} crystal plates. The dielectric constant and loss were measured as a function of temperature in the range 10 kHz to 0.4 MHz using a HP4275 model multifrequency *LCR* meter. The dielectric data were collected while the temperature was varied at the rate of four degrees per hour.

For a normal dielectric obeying Debye type relaxation we have (Daniel 1967):

$$\varepsilon'(\omega) = \varepsilon(\infty) + (\varepsilon(0) - \varepsilon(\infty))/(1 + \omega^2\tau^2) \tag{1}$$

$$\varepsilon''(\omega) = (\varepsilon(0) - \varepsilon(\infty))\omega\tau/(1 + \omega^2\tau^2). \tag{2}$$

Thus we have,

$$\varepsilon''(\omega)/\varepsilon'(\omega) = \tan\delta = (\varepsilon(0) - \varepsilon(\infty))\omega\tau/(\varepsilon(0) + \varepsilon(\infty)\omega^2\tau^2) \tag{3}$$

where ε' and ε'' are the real and imaginary parts of the dielectric constant; $\varepsilon(0)$ and $\varepsilon(\infty)$ are the static and high frequency dielectric constants; ω is the angular frequency of the electromagnetic field; τ is the relaxation time and $\tan\delta$ is the loss factor. Experimentally
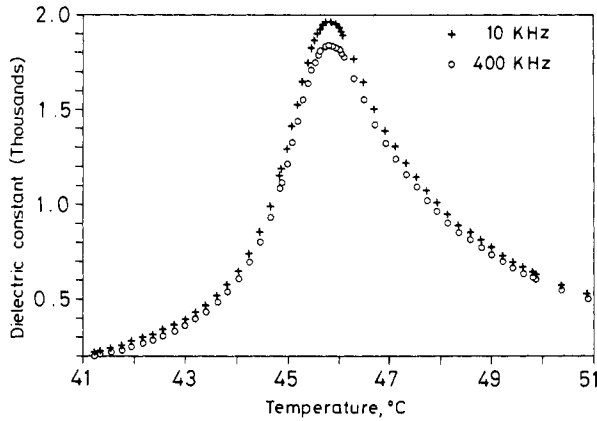
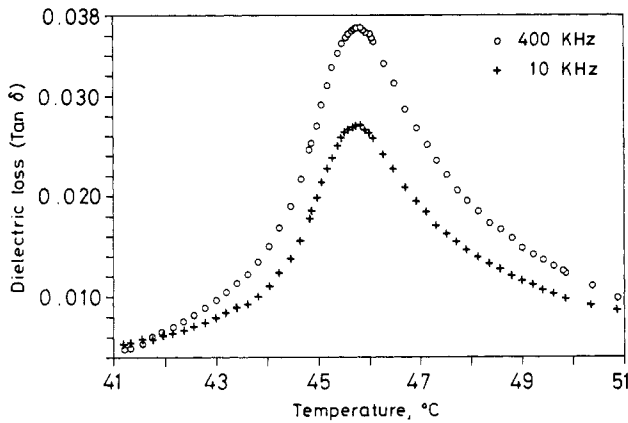**Figure 1.** Temperature dependence of the dielectric constant of TAAP.

Figure 2. Temperature dependence of the dielectric loss of TAAP.

one measures the quantities $\varepsilon'(\omega)$ and the loss tangent $\tan\delta$. Eliminating $\varepsilon(0)$ from equations (1) and (3) we have:

$$\tau = \varepsilon'(\omega)\tan\delta / \omega\big(\varepsilon'(\omega) - \varepsilon(\infty)\big).$$

The relaxation time can thus be calculated from the measured values of $\varepsilon'(\omega)$ and $\tan\delta$.

Figures 1 and 2 show the temperature dependence of $\varepsilon'(\omega)$ and $\tan\delta$ at 10 kHz and 0.4 MHz as a function of temperature. As can be seen from figure 1 there is not much dielectric dispersion in this frequency range. The temperature dependence of τ at 0.4 MHz is shown in figure 3. τ is found to obey relations of the form:

$$\tau = A'/(T - T_c) \qquad \text{(in the PE phase)}$$
$$\tau = A''/(T - T_c) \qquad \text{(in the FE phase)}.$$

The values of A' and A'' for the various frequencies in the region 10 kHz to 0.4 MHz is shown in table 1.
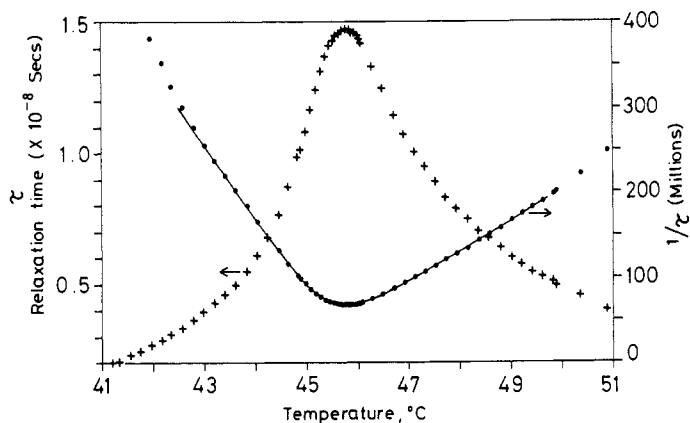
Figure 3. Temperature dependence of the relaxation time and its inverse at 0.4 MHz of TAAP.

Table 1. Values of A' and A'' as a function of frequency.

Frequency (kHz)	A'	A''
10	0.10×10^{-5}	0.52×10^{-6}
40	0.23×10^{-6}	0.10×10^{-6}
100	0.92×10^{-7}	0.33×10^{-7}
400	0.23×10^{-7}	0.29×10^{-8}

It is well known that the dielectric relaxation in many of the order–disorder ferroelectrics can be described to a good approximation by the mono-dispersive Debye type relaxation equations (Blinc and Zeks 1974). The phenomenon of critical slowing down is also observed in these ferroelectrics. The temperature variation of relaxation time in TAAP clearly shows that the phenomenon of critical slowing down occurs in TAAP as well and thus confirms the order–disorder nature of the phase transition in this crystal.

References

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Guilliot Gauther S, Peuzin J C, Oliver M and Rolland G 1984 *Ferroelectrics* **52** 293
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