# Anomalously narrow peak in the dielectric constant of $Cd_2Nb_2O_7$ near the diffuse phase transition

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An unusually narrow peak is observed in the dielectric constant of the ferroelectric $Cd_2Nb_2O_7$ at 201 K, above the broad peak of the diffuse ferroelectric phase transition. Possible causes of this anomaly, observed in this study for the first time, are discussed.

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Potassium pyroniobate ($Cd_2Nb_2O_7$), which at room temperature has symmetry space group $O_h^7$, undergoes a variety of phase transitions of a nature which has not been conclusively established.[1–4]

We have made more careful measurements of the temperature dependence of $\epsilon$ and tan $\delta$ in the range 193–213 K at heating and cooling rates of 0.05 K/min. Capacitance measurements were made on an E4-7 $Q$ meter with a measuring field of 200 V/m at frequencies of 50 and 400 kHz.

Figure 1 shows the temperature dependence of $\epsilon$ for a crystal oriented along the [111] direction. At a temperature of 201 K one sees an unusual, anomalously narrow peak in $\epsilon$, which was detected for the first time in the present study; the value of $\epsilon$ at the peak is higher than in the region of the diffuse ferroelectric transition. The width of this new peak is 1–1.5 K. Such narrow peaks in $\epsilon$ have not previously been observed in materials having diffuse phase transitions. The temperature dependence of tan $\delta$ displays a kink in the region of the anomalous peak in $\epsilon$ and a small maximum at 205 K (Fig. 1). The value of $\epsilon$ in the vicinity of the anomalous peak turns out to depend on the previous history of the sample.

In the range of frequencies studied, 50–400 kHz, we did not detect an appreciable change in the height of the anomalous peak in $\epsilon$ or a shift in its temperature position with frequency, whereas the temperature of the $\epsilon$ peak in the region of the diffuse phase transition is strongly shifted with frequency.[3,5]

The temperature and height of the anomalous peak in $\epsilon$ under heating and cooling do not coincide; the hysteresis in the temperature of the peak is ~0.6 K. This sort of behavior is typical of a first-order phase transition which is close to second-order transition. The hysteresis in the value of $\epsilon$ is much smaller above the anomalous peak than immediately below it (Fig. 2a). Above the anomalous peak, $\epsilon$ obeys the Curie-Weiss law during both cooling and heating, with a Curie-Weiss constant $C = (3.3 \pm 0.3) \times 10^4$ K. At temperature below the peak, the Curie-Weiss law is obeyed with lower accuracy and in a narrower interval (~1 K), with $C = (5 \pm 1) \times 10^4$ K.

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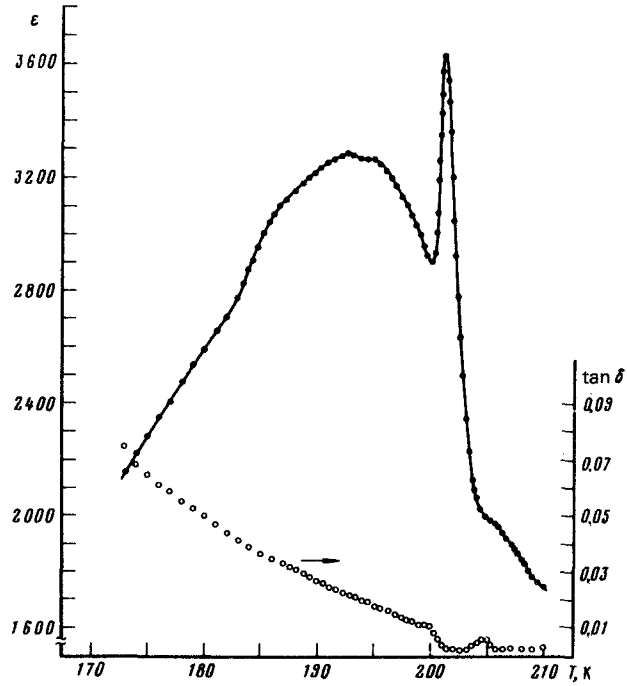


FIG. 1. Temperature dependence of the dielectric constant $\epsilon$ and of the loss tangent tan $\delta$ for a $Cd_2Nb_2O_7$ crystal oriented along the [111] direction.

After a heating of the samples to a temperature of 700–800 K, the position of the anomalous peak is shifted upward by $\sim 1.5$ K, but 5–10 days later the peak returns to its original position, indicating that the state of the defects exerts a substantial influence and that is changes with temperature. The application of a small static electric field to the sample shifts the temperature of the anomalous peak upward at a rate of $10^{-4}$ deg m/V, with a deviation from linearity appearing in several measurements in fields as low as $2\times10^4$ V/m (Fig. 2b).

We should point out that a change in the domain structure occurs in the investigated temperature range. At temperatures between 201 and 205 K the observed domain structure has diffuse boundaries apparently consisting of subdomains, i.e., a strong inhomogeneity arises in the system. The formation of a superstructure with a large period in this temperature interval is not ruled out. At 201 K a distinct domain structure appears in the crystal.[5]

The narrow peak in $\epsilon$ at 201 K can be interpreted as resulting from a transition from a phase with a fine, diffuse domain structure stabilized by defects to a phase with a distinct ferroelectric domain structure. We note that an improper ferroelectric phase occurs at temperatures above the familiar proper ferroelectric phase transition in cadmium pyroniobate. The data of different authors[1,3,6] disagree as to the temperature interval in which this phase exists. The existence region of this phase in crystals having

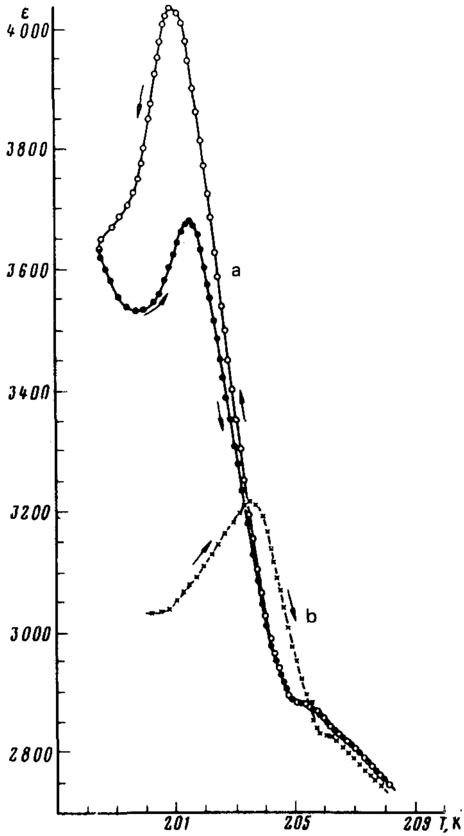


FIG. 2. Thermal hysteresis of the anomalous peak in the dielectric constant of a $Cd_2Nb_2O_7$ crystal oriented along the [001] direction (a), and the temperature dependence of the peak in an external electric field $E = 2\times 10^4$ V/m (b). The samples were annealed before each cycle of measurements.

an anomalous peak in $\epsilon$ cannot be determined solely by low-field dielectric measurements such as we have made here. In addition to this improper phase, there is another noncubic phase that exists below 205 K, according to the data of Refs. 2 and 5. In improper ferroelectrics, transitions between phases having a nonzero proper order parameter are known to be accompanied by anomalies in $\epsilon$. However, there are no known phase transitions of this class that are accompanied by dielectric-constant anomalies like the peak in $\epsilon$ observed in the present study.

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