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FERROELECTRICITY IN $NaNO_2$

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It is well known that sodium nitrite ($NaNO_2$) has the orthorhombic structure (C_{2v}^{20}) at room temperature[1] and this structure changes to the more highly symmetric one (D_{2h}^{25}) at about 160°C.[2,3] The piezoelectric effect observed in the low-temperature form disappears at this temperature.[2] Although the structural investigations of $NaNO_2$ crystal have been carried out well by several researchers,[4,5] other properties, in particular the dielectric properties, have scarcely been noticed.

We have studied the dielectric properties of this crystal and found that it is ferroelectric along the b axis below the 160°C transition point. Crystals were prepared by the fusion method, where $NaNO_2$ powder had been dried in advance in 10^{-3} mm Hg vacuum at about 120°C during 48 hr, since this substance is considerably deliquescent. Crystals were transparent, slightly yellowish, and easily cleaved along the (101) plane. Figure 1 shows the dielectric constant vs temperature relation measured at the frequency of 100 kc/sec, using air-drying silver paste electrodes. The dielectric constant in the direction of b axis, ϵ_{010} (dimension of specimen: thickness 0.0835 cm, area 0.537 cm^2), is 7.4 at room temperature and reaches 780 at 164°C on heating and 1100 at 162°C on cooling, respectively, where the rate of temperature change was about 0.5°C/min. Above this transition temperature, the Curie-Weiss law holds with the Curie-Weiss temperature of 433°K and the Curie constant of 5.0×10^3°K. The dielectric constant perpendicular to the b axis, ϵ_{101} (dimension of specimen: thickness 0.114 cm, area 1.032 cm^2), is 6.4 at

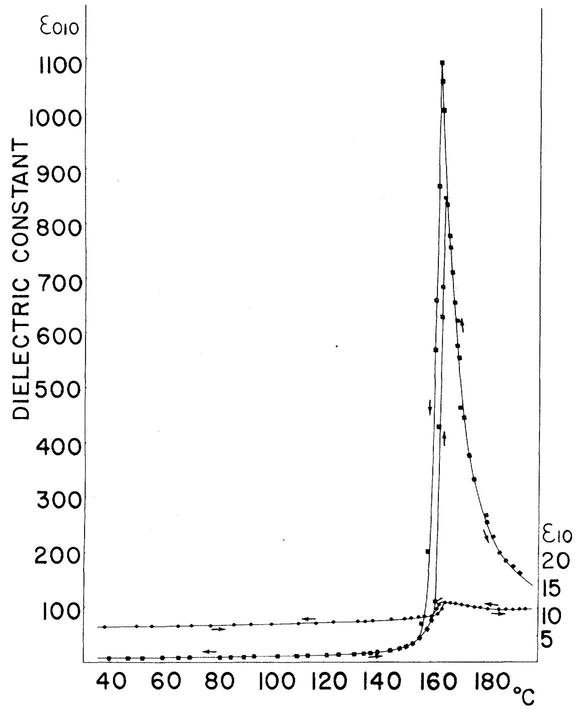


FIG. 1. Dielectric constant ϵ_{010} and ϵ_{101} (100 kc/sec) vs temperature. Squares: ϵ_{010}; circles: ϵ_{101}.

room temperature and remains low at high temperatures, though a small anomaly was observed at the transition.

The pyroelectricity was clearly observed along the b axis below the transition temperature and furthermore its polarity could be easily reversed by applying a dc field to the crystal. We obtained about 7 μcoul/cm^2 as the value of spontaneous polarization at room temperature from the pyroelectric measurement, using a sample which had been cooled to room temperature from 180°C applying the dc field of 1.4 kv/cm. The typical D-E hysteresis loop was observed below the transition temperature in the direction of b axis at 50 cps. An example is shown in Fig. 2, where the coercive field and the spontaneous polarization are seen to be 2.3 kv/cm and 6.4 μcoul/cm^2 at 143°C, respectively. This value of spontaneous polarization nearly coincides with that obtained from the measurement of pyroelectricity. The coercive field derived from the hysteresis loop varied with temperature and strongly depended on the applied field strength. At room temperature, the loop did not open out even for the field of 25 kv/cm, because of its large coercive field. A double hysteresis loop could be observed just above the transition tem-

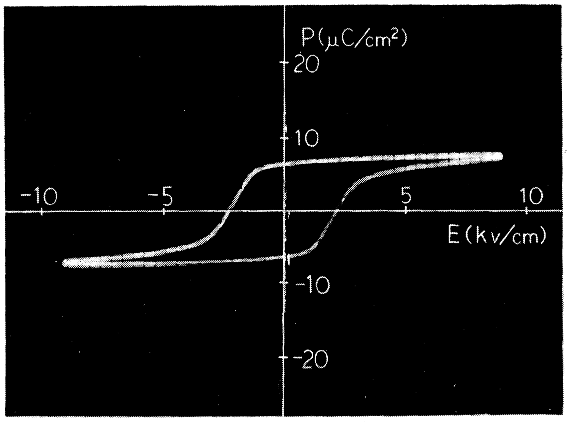
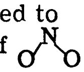


FIG. 2. Hysteresis loop (50 cps) in the direction of b axis at 143°C.

perature.

Judging from these experimental results, it is clear that $NaNO_2$ is ferroelectric along the b axis below the 160°C transition point, and the transition seems to be of the first kind, but even then the jump at the transition would be very small. The ferroelectricity is considered to come from the favorable arrangement of  groups in the crystal. The ordered arrangement of these groups causing the crystal to be polar in the low-temperature form seems to become disordered at the transition temperature.

Details of our studies will be reported shortly.

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SUSCEPTIBILITY OF THE ISING MODEL OF AN ANTIFERROMAGNET

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The susceptibility versus temperature curve for an antiferromagnet is usually characterized by the appearance of a maximum and the precise relationship of this maximum to the associated anomaly in the specific heat is of great interest. The elucidation of this problem using even a comparatively simple model of an antiferromagnet, such as the Ising model, is difficult and the behavior in the critical region has long been a subject of speculation. The work summarized below, however, leads to the conclusion that the susceptibility, χ, versus temperature, T, plot for an antiferromagnet should have a vertical tangent (χ increasing with T) at the same temperature at which the specific heat anomaly occurs. Since χ must vanish as T becomes infinite, this form of the singularity implies that the observed maximum in χ occurs above the true critical temperature.

These conclusions are based on a study of the susceptibility of Ising models. No exact closed expressions for the susceptibility of two- or three-dimensional Ising models have yet been given but series have been published by various authors.[1-3] The best method of obtaining these seems to be that employed by Oguchi[2] and developed by Domb and Sykes.[3] Extrapolation of these series has been attempted, most recently by Park,[4] but it has proved difficult to draw firm conclusions in the critical region. We have been able to rewrite the general expansion in such a way that the major part of the susceptibility is known if the configurational energy of the lattice is known. The remaining part has a much simpler configurational interpretation than the original expansion. In terms of the antiferromagnetic "high-temperature" variable $v = \tanh |J|/kt$, where J is the interaction energy between parallel spins, we find that the susceptibility per spin may be written

$$\chi(v) = \frac{m^2}{kT}[1+(q-1)v]^{-2}\{1+(q-2)v+v^2 - 2v\left|\frac{U(v)}{J}\right| + 8\sum_r(-1)^r d_r v^r\}, \qquad \text{(A)}$$

where m is the magnetic moment per spin, q is the coordination number, and $U(v)$ is the configurational energy per spin. The positive coefficients d_r enumerate a well defined class of closed graphs on the lattice with at most two odd vertices; d_0 to d_4 vanish identically on all lattices and d_5 and d_6 vanish on most loose-packed lattices.

By using this result, the series expansion for the susceptibility of the plane triangular lattice has been extended up to the term in v^{12}, that of the plane quadratic lattice up to the term in v^{14}, and that of the honeycomb lattice up to the term in v^{24}. The general result holds equally for three-dimensional lattices and the corresponding series can also be considerably extended.

Numerical evaluation of the function defined by $\Sigma(-1)^r d_r v^r$ shows that it contributes only a few

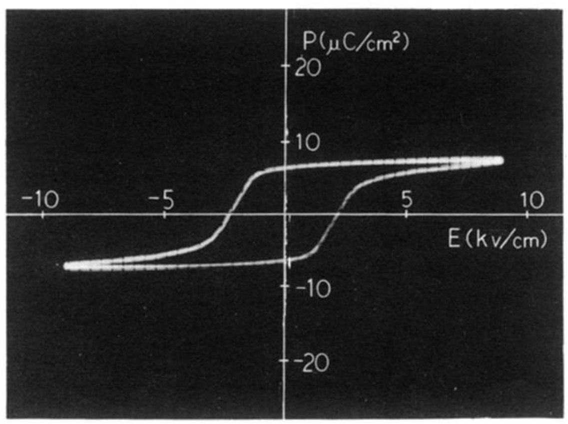

FIG. 2. Hysteresis loop (50 cps) in the direction of b axis at 143°C.