Two-dimensional nature of fluctuations in $KMnF_3$ deduced from measurement data on the dielectric constant

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Anomalous behavior of the dielectric constant has been observed for the first time in the antiferrodistortion structural phase transition in $KMnF_3$. The logarithmic temperature dependence of the first fluctuation correlation to the dielectric constant indicates that the order-parameter fluctuations are of a quasi-two-dimensional nature.

A phase transition in $KMnF_3$ from the O_h^1 cubic phase to the D_{4h}^{18} tetragonal phase with a three-component order parameter and condensation of the soft mode Γ_{25} at the R point of the Brillouin zone, a model example of a structural phase transition which occurs at a temperature $T_c = 188.6$ K, has been studied for several years.[1] According to the data on diffuse scattering of x rays,[2] the fluctuations of the order parameter are quasi-two-dimensional in nature. This conclusion has so far not been confirmed, however, on the basis of an analysis of the systematic features of the thermodynamic quantities.[3] In the present letter we report an observation of an anomalous behavior in the dielectric constant during this structural phase transition and offer an interpretation of its temperature dependence in the cubic phase from the position that the fluctuations of the order parameter are quasi-two-dimensional in nature.

In the experiment we have carried out precision measurements of the temperature dependence of the dielectric constant of $KMnF_3$ single crystals. The measurement procedure is described in Ref. 4. The temperature dependence $C(T)$ of the capacitance of a $KMnF_3$ single-crystal parallel-plate capacitor for an electric field directed along the [100] cubic axis is represented by curve 1 in Fig. 1. The temperature dependence of the dielectric constant $\epsilon(T)$ was calculated taking into account the thermal expansion of the sample.[5,6] At room temperature $\epsilon = 8.65 \pm 0.15$. Since the dependence $\epsilon(T)$ in the cubic phase is linear far from T_c, we can single out the anomalous part, $\Delta\epsilon(T)$, of the dielectric constant above T_c, which is traced to a temperature $T_c + 60$ K (curve 2 in Fig. 1). The anomalous part of the dielectric constant, which does not exceed 1%, was not detected in the preceding experiments.[1,7] Figure 2 is a plot of the temperature dependence of $\Delta\epsilon$ in the coordinates $\Delta\epsilon = f(\log\tau)$, where $\tau = (T - T_c)/$

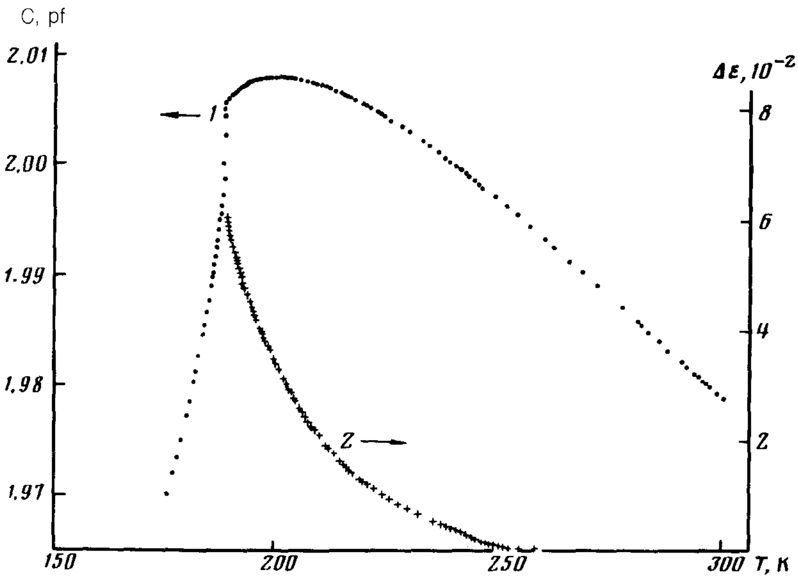

FIG. 1. Temperature dependence of the capacitance of a $KMnF_3$ single-crystal sample near the structural phase transition (curve 1). Temperature dependence of the anomalous part of the dielectric constant, $\Delta\epsilon$, in the cubic phase (curve 2).

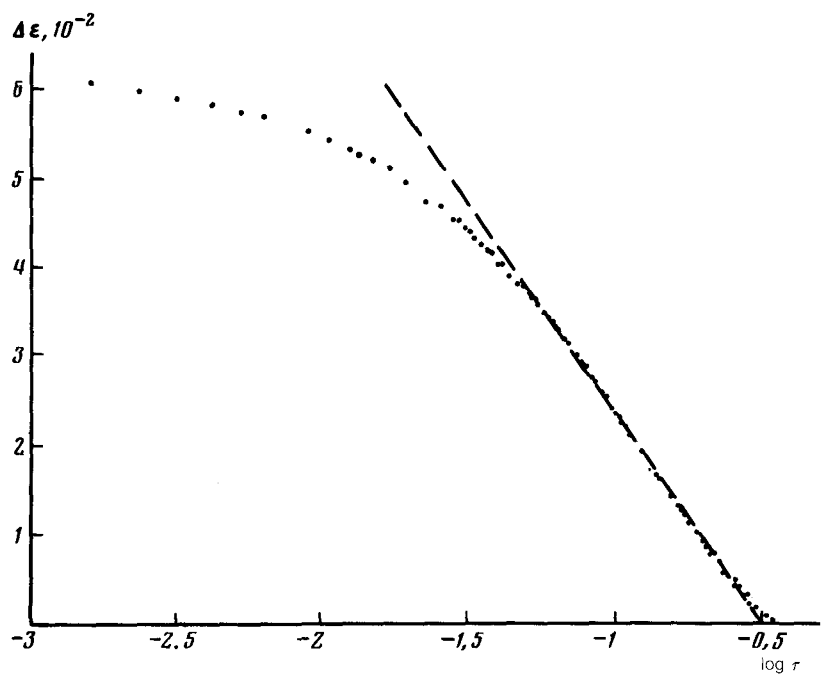

FIG. 2. Temperature dependence of $\Delta\epsilon$ in the $\Delta\epsilon = f(\log \tau)$ coordinates.

$T_c = \Delta T/T_c$ is the reduced temperature. In the temperature interval $10 < \Delta T < 60$ K, the temperature dependence of $\Delta\epsilon$ varies as log τ. This behavior corresponds to the first fluctuation correction to the dielectric constant for quasi-2D fluctuations of the order parameter indicated above. The order parameter in this case is linked with the electric field **E** through three invariants which are bilinear in the order-parameter components and in the electric-field components. The structure of these invariants is similar to the structure of the invariants which describe the relation between the order parameter and the deformation (see, e.g., Ref. 8). We easily see that above T_c only one of these invariants, in combination with the electric-field components, is responsible for the critical contribution to the dielectric constant. This combination does not break the symmetry of the high-temperature phase. Since the use of this invariant leads to only a renormalization of T_c proportional to E^2, we find

$$\frac{\partial \Delta\epsilon}{\partial T} = -\frac{1}{4\pi}\frac{\Delta c}{T_c}\frac{\partial^2 T_c}{\partial E^2},$$

where $\Delta\epsilon$ and Δc are the most singular contributions to the dielectric constant and the specific heat, respectively. The first fluctuation correction to the quasi-2D nature of the fluctuations gives the specific-heat index $\alpha = 1$, in agreement with the logarithmic dependence $\Delta\epsilon(\tau)$ obtained in our experiments. The data of Refs. 9 and 10 on the anomalous behavior of the elastic moduli of $KMnF_3$ support our finding that at $\Delta T > 10$ K we are dealing with the first fluctuation correction. Since the index of the first fluctuation correction for the specific heat is the same as that for elastic moduli, we expect that the anomaly of the elastic moduli is also proportional to $(T - T_c)^{-1}$. This behavior of the elastic moduli of $KMnF_3$ was in fact observed by Melcher and Plovnick[9] and Holt and Fossheim[10] but was reported by the latter authors as an inexplicable event.

The behavior of $\Delta\epsilon$ corresponding to the first fluctuation correction is seen in the temperature interval corresponding to approximately a ten-degree change in the reduced temperature τ a reasonable distance from the phase transition. In the case of low-dimensionality fluctuations of the order parameter, the fluctuation effects, as we know, are more pronounced than in the 3D case. A deviation from the behavior corresponding to the first fluctuation correction, which is observed upon approaching T_c, therefore seems to us to be a systematic behavior. The behavior of $\Delta\epsilon$ in the temperature interval $0.3 < \Delta T < 10$ K probably corresponds to the crossover region of the first fluctuation correction to the truly critical quasi-2D behavior. If we assume that at[11] $\Delta T \approx 0.2$ K there is a crossover to a 3D behavior, then the truly critical quasi-2D behavior may not materialize. In the temperature interval $0.3 < \Delta T < 6$ K the temperature dependence of $\Delta\epsilon$ corresponds to the specific-heat index $\alpha = 0.24 \pm 0.03$, in agreement with the value of α found from the data on the thermal expansion.[6] However, since there are two crossovers in this temperature interval, we cannot explain this value of α in terms of the asymptotic behavior of some degree of generality.

Finally, we note that in contrast with the generally accepted viewpoint,[12] the measurement of insulating materials can be used, according to the results of this study, as an effective method of studying critical static phenomena even in nonferroelectric structural phase transitions.

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