Fractal nature of large-scale nonuniform states in ferroelectrics with a diffuse phase transition

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(Submitted 28 December 1994)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 3, 214–216 (10 February 1995)

The angular distribution of the anomalous component of the scattered-light intensity, $I(\theta)$, in disordered $PbSc_{1/2}Ta_{1/2}O_3$ crystals has been measured. A power-law distribution $I(\theta) \propto \theta^{-\alpha}$, where $\alpha=3.2$, was observed as the intensity was varied over four orders of magnitude. The physical nature of the optical irregularities, whose length scales are broadly distributed, is discussed in terms of a fractal model. © *1995 American Institute of Physics.*

Models with fluctuations of the local phase-transition temperature in disordered ferroelectrics with a diffuse phase transition have led to the prediction[1,2] that anomalous peaks in the intensity of light scattered at small angles may arise in these cases. Narrow peaks of this sort have indeed recently been observed in several disordered ferroelectrics [$PbSc_{1/2}Ta_{1/2}O_3$ (PST), $PbSc_{1/2}Nb_{1/2}O_3$, and $PbZn_{1/3}Nb_{2/3}O_3$)], on the temperature dependence[3,4] and the field dependence.[5] The sharp increase in the small-angle intensity of scattered light, $I(\theta)$, down to the smallest scattering angles which can be reached experimentally, $\theta_{min} \sim 20'$, implies internal processes of a percolation type. However, not enough experimental information has been available for a quantitative description of statistical characteristics of the spatially nonuniform stochastic structures that arise.

In this paper we are reporting careful measurements of the intensity of small-angle scattering of light by several PST crystals with various degrees of ordering (s) of the Sc^{3+} and Ta^{5+} ions (PSTI, $s=0.95$, maximum of the dielectric constant at $T_c=43\,°C$; PSTII, $s=0.7$, $T_c=23\,°C$). These results make it possible, for the first time, to draw the conclusion that the experimentally observed structures are of a fractal nature. The degree of ordering s of the ions was monitored by x-ray diffraction, on the basis of the intensities of superstructural lines. The measurements were carried out on single-crystal samples which were parallelepipeds cut along the [100] crystallographic direction. The length of the samples along the light propagation direction was 0.2–1 mm. The layout of the apparatus used to measure the temperature dependence of the intensity of small-angle light scattering is described in Ref. 1.

Figure 1 shows, in full logarithmic scale, experimental distributions $I(\theta)$ for crystals of PSTI (a) and PSTII (b) at various scattering angles. The points conform well to a straight line with a slope $\alpha=3.2$ over the interval of scattering angles accessible in the intensity measurements, $20' < \theta < 10°$. This result means that the spatially nonuniform

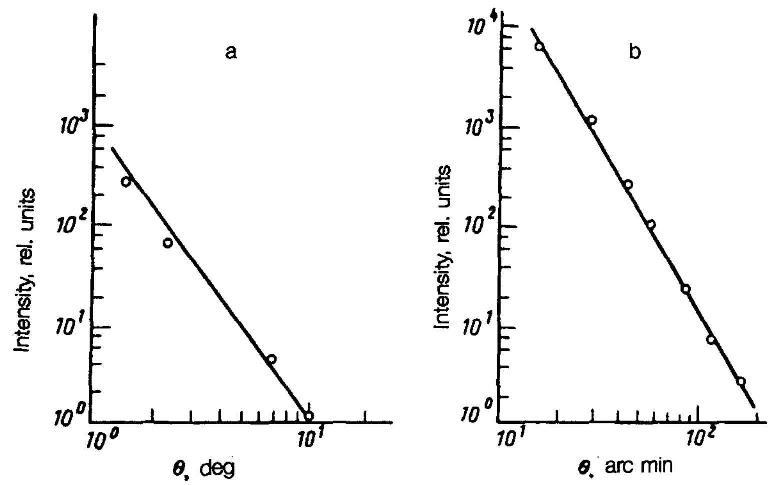

FIG. 1. Angular distribution of the scattered-light intensity at the temperature of the anomalous peak. a—PSTI crystal; b—PSTII crystal.

structure is a fractal entity, at least over the size interval 20–0.6 μm, and that it does not consist of randomly arranged 3D (Euclidean) nonuniformities of finite scale, since in the latter case the small-angle scattering would be described by a power $\alpha=4$ (Porod's law).[6]

Knowledge of the value of the Porod index α does not, of course, definitively determine the fractal geometry, but the fact that we have a value $3<\alpha<4$ means that we can classify this observable scattering structure as a "surface" fractal, since for "bulk" fractals the value of α is equal to their Hausdorff dimension d_f and should be less than 3 (Ref. 7).

For surface fractals, the power α is related to the Hausdorff dimension d_s $(3>d_s>2)$ of a "roughened" surface of a 3D irregularity by the relation[7] $\alpha=6-d_s$. In our case we find $d_s=2.8$, so the surface is very choppy.

To what sort of physical object would the surface fractal observed in a light-scattering experiment correspond? Clearly, the physical properties at the boundary of the object would have to vary greatly over distances on the order of the wavelength of the light. Otherwise, the Porod index α would be greater than 4 (Ref. 6). As possible candidates for the role of such a fractal it is natural to consider domain walls and/or phase boundaries, since we know that in completely ordered PST crystals with $s=1$ a first-order ferroelectric phase transition occurs. This property is probably retained for local phase transitions in disordered crystals. In turn, the formation of roughened domain walls and phase boundaries due to pinning by defects is characteristic of appreciably disordered solids (Ref. 8, for example).

Another fact which deserves consideration is that a near-surface layer which is more ordered than the interior exists in disordered PST crystals.[4] Its boundary may also have a fractal structure, which would form during nonequilibrium growth of the crystal.

Just which of these possibilities is actually realized in practice requires further research.

This study had financial support from the Russian Fund for Fundamental Research (Project 93-02-14156).

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Translated by D. Parsons