

Dielectric Properties of Dirty Ferroelectric SBN

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Diffuse phase transition of a rare earth ion-doped SBN40 ($\text{Ba}_{0.4}\text{Sr}_{0.6}\text{Nb}_2\text{O}_6$; Pr^{3+} or Nd^{3+}) has been carefully examined by the measurements of the spontaneous polarization P_s , the dielectric constant ϵ and X-ray diffraction. P_s decreases gradually with increasing temperature and ϵ has a broad maximum near the transition temperature T_c . With increasing rare earth ion concentration M , 1) the broadening of ϵ increases, 2) the temperature dependence of P_s becomes more diffuse, 3) T_c and T_p (the inflexion point of P_s) decrease in proportion to M^{-1} . In order to discuss the temperature and M dependence of P_s and ϵ , we adopt the order parameter expansion of the free energy including odd powers as well as even powers of the polarization and the expansion of the reciprocal susceptibility in terms of $T - T_c$ which was introduced by L. Benguigui:¹⁾

$$\chi^{-1} = \chi_0^{-1} + B_2(T - T_c)^2 + B_3(T - T_c)^3. \quad (1)$$

B_2 and B_3 are the expansion coefficients, and were determined by the method of least square. It turned out that B_2/B_3 is proportional to M^{-1} . The calculated values of χ^{-1} are in good agreement with the experimental results in the temperature range of $T_c \pm 5^\circ\text{C}$ for pure SBN40, and $T_c \pm 15^\circ\text{C}$ for the samples with the rare earth ion. However, except the above temperature regions, χ^{-1} is a linear function of temperature. The ratio of the slopes of the χ^{-1} vs T curve at low and high temperature regions is about 2:1 in our experiments. Figure 1 shows the temperature dependence of P_s . Marks show the calculated values and the lines are the experimental results. The calculated values of P_s agree with the experimental results in the temperature range of $T_c \pm 20^\circ\text{C}$ for the samples containing the rare earth ion. Furthermore, by using the expressions for B_2 and B_3 ,¹⁾ we can write the

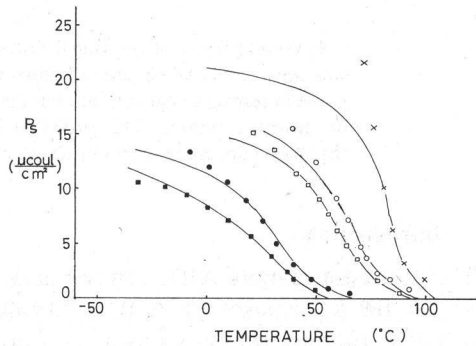


Fig. 1. Temperature dependence of the spontaneous polarization P_s and the calculated values of P_s for various samples. The lines show the experimental results. \times : pure SBN40, \circ : SBN40 + Pr^{3+} (0.3 wt%), \bullet : SBN40 + Pr^{3+} (1.0 wt%), \square : SBN40 + Nd^{3+} (0.3 wt%), \blacksquare : SBN40 + Nd^{3+} (1.0 wt%)

inflexion point as

$$T_p = T_c + B_2/6B_3. \quad (2)$$

It is clear that the calculated values of T_p given by eq. (2) agree with the experimental values within 5% error in all samples. As mentioned above, in the case of the rare earth ion-doped SBN40, temperature dependence of the dielectric properties is well explained by the order parameter expansion of free energy and the expansion of χ^{-1} in terms of $T - T_c$. Furthermore, X-ray experiments were carried out to clarify the microscopic mechanism of the diffuse phase transition. The equi-intensity contour of X-ray critical diffuse scattering around (0 0 4) reflection in SBN40 + Pr^{3+} (0.3 wt%) shows a strongly anisotropic distribution. Considering the piezoelectric coupling in χ^{-1} , we can explain the strong anisotropy near T_c in SBN40 + Pr^{3+} (0.3 wt%).

Reference

- 1) L. Benguigui: Solid State Commun. 14 (1974) 669.