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# Dielectric dispersion of relaxor ferroelectrics

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The dielectric dispersion of lead magnesium niobate and Ta-bearing strontium barium niobate was measured and analyzed using 'universal' law of dielectric response. The experimental results were well explained by the 'universal' law. The temperature dependence of relaxation frequency indicated that thermally fluctuating dipoles exist in polar microregions. The dielectric response of relaxor ferroelectrics could be explained by an enlargement of polar microregions, freezing process of fluctuating dipoles and dielectric relaxation which follows the 'universal' law.

## 1. INTRODUCTION

Relaxor ferroelectrics (RF) are characterized by a broad maximum in the dielectric constant as a function of temperature, together with a dielectric relaxation in the radio frequency range. The model of RF has been proposed by many research groups[1-5]. Most of models proposed the formation of randomly oriented polar microregions (PMR) in paraelectric crystals[1]. Cross[2] suggested that PMR are thermally fluctuating at high temperatures, and Viehland et al.[3] consequently suggested that the fluctuations have a freezing temperature similar to spin glasses.

The fluctuation of PMR should be detectable as a frequency dependence of dielectric response (dielectric dispersion). Some work[3,6,7] has been done on the dielectric dispersion of RF but a clear evidence of the fluctuation has not been obtained yet. Moreover, most of analyses of dielectric dispersion postulated the existence of relaxation time distribution. This postulate seems to be acceptable from the view point of composition fluctuation proposed by Smolensky[5]. However, the departure from the Debye behavior observed in a vast range of materials could be explained by advanced theories[8,9] which do not postulate the relaxation time distribution.

In this paper, the dielectric dispersion of lead magnesium niobate ($Pb(Mg_{1/3}Nb_{2/3})O_3$,PMN) and Ta-bearing strontium barium niobate with tungsten bronze structure ($Sr_{0.5}Ba_{0.5}Nb_{1.6}Ta_{0.4}O_6$, SBNT) are measured, and analyzed using 'universal' dielectric response theory[8] in order to clarify the thermal fluctuation and freezing behavior of PMR in RF.

## 2.. 'UNIVERSAL' LAW OF DIELECTRIC RESPONSE

The dielectric response of condensed matter has been known to depart from Debye behavior.

$$\chi(\omega) \propto (1+i\omega\tau)^{-1} \qquad (1)$$

where $\omega$ is a angular frequency and $\tau$ is a relaxation time. Jonscher[10] indicated that there exists for a wide range of materials a remarkable universality of dielectric response.

$$\chi''(\omega) \propto \omega^{n-1} \qquad \text{with } 0 < n < 1$$

Ngai et al.[8] consequently explained the universality using a concept of low-energy excitation of 'correlated states'. Their conclusion was consistent with the empirical expression of dielectric response function suggested by Williams and Watts[11].

$$\Psi(t) = d\left[-\exp(t/\tau)^{\beta}\right]/dt \qquad (2)$$

where $\beta$=1-$n$. Dielectric response in frequency domain is a Fourier transform of the above function. In Ngai's theory, the temperature dependence of relaxation peak frequency $\omega_p$ is represented by

$$\omega_p \propto \exp\left[-E_A^* / kT\right] \qquad (3)$$

where $E_A^*$ is an apparent activation energy of $E_A^* = E_A/(1-n)$, and $k$ is the Boltzmann's constant.

## 3. EXPERIMENTAL

Powder of PMN was prepared by the columbite precursor method[12] in order to produce phase-pure compositions. It was sintered at 1473K for 4 hours. Ceramics of SBNT were prepared as described by Tsurumi et al.[13]

The dielectric dispersion of the specimen was measured using vector impedance analyzers (HP4192A,HP4191A). The dimension of specimens was changed with frequency to avoid an electric resonance of a measuring circuit and specimens.

We use the 'universal' theory mentioned in the previous section to analyze the dielectric dispersion of PMN and SBNT.

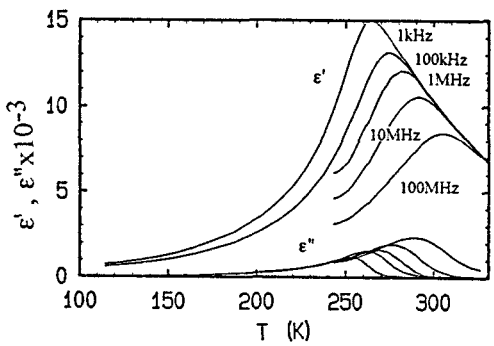


Fig.1 Temperature dependence of dielectric constant of PMN

## 4. RESULTS AND DISCUSSION

### 4.1 PMN

The temperature dependence of dielectric constant measured at different frequencies was consistent with well-known behavior of RF (Fig.1). Figure 2 shows the frequency dependence of dielectric constant. The solid lines in the figures were those calculated from the Williams and Watts function (equation 2). The experimental results were well explained by the 'universal' law This means that the dielectric dispersion of RF is not special, and the distribution of relaxation time is not necessary to

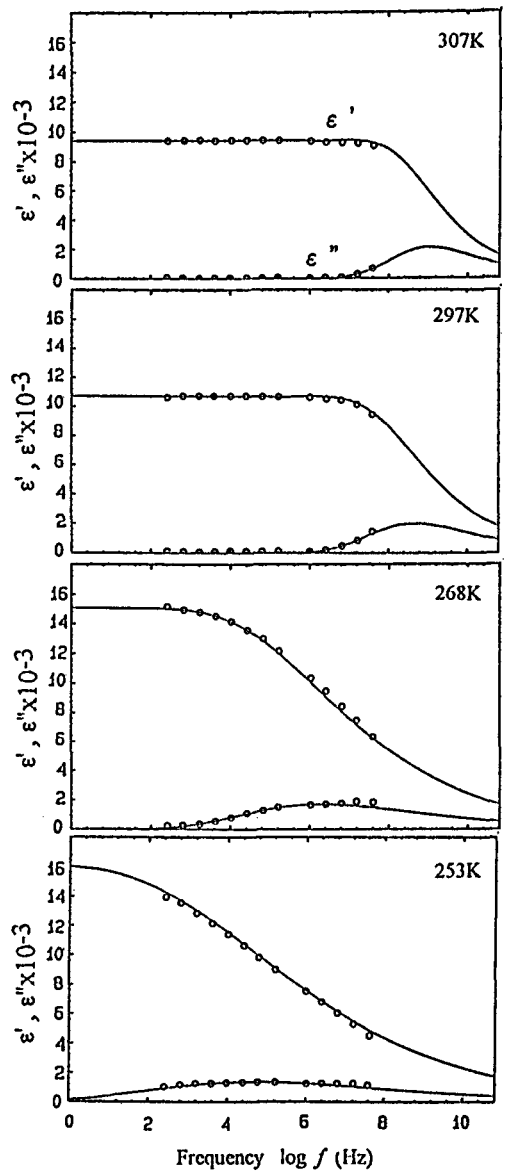


Fig.2 Dielectric dispersion of PMN

explain the experimental results.

Figure 3 is a change of β(=1-$n$) as a function of temperature. The low β values (0.15-0.3) indicate a strong correlation of fluctuating dipoles exists in the material. The temperature dependence of β implies that the correlation increases with decreasing temperature, which may be due to the increase in the volume of PMR[2,3]. Figure 4 shows a relation between the relaxation peak frequency and 1/(1-$n$)$T$. It was found that the temperature dependence of relaxation frequency obeys the equation (3) with an activation energy
$E_A$=0.048eV . This result evidences the thermally fluctuating dipoles in PMN suggested by Cross[2] and their freezing behavior. However, we could not find the Vogel-Fulcher relationship[3] which evidences the existence of the freezing temperature.

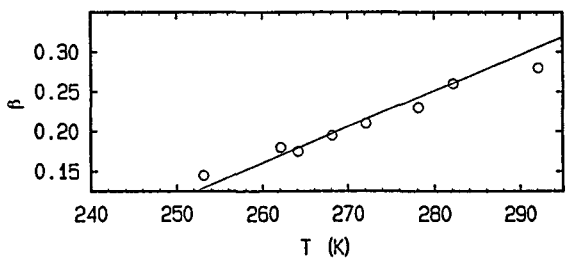


Fig. 3 The β value of PMN as a function of temperature

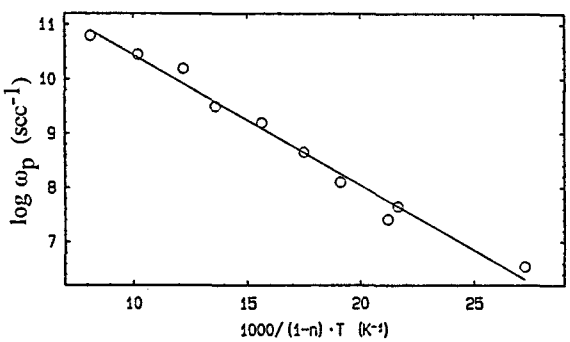


Fig. 4 Relation between $\omega_p$ and 1/(1-n)$T$ of PMN

## 4.2 SBNT

The SBNT ($Sr_{0.5}Ba_{0.5}Nb_{1.6}Ta_{0.4}O_6$) has a broad maximum of dielectric constant at about 210K[13]. Figure 5 shows dielectric dispersion of SBNT measured around the peak temperature of dielectric constant ($T_m$). The observed results were well

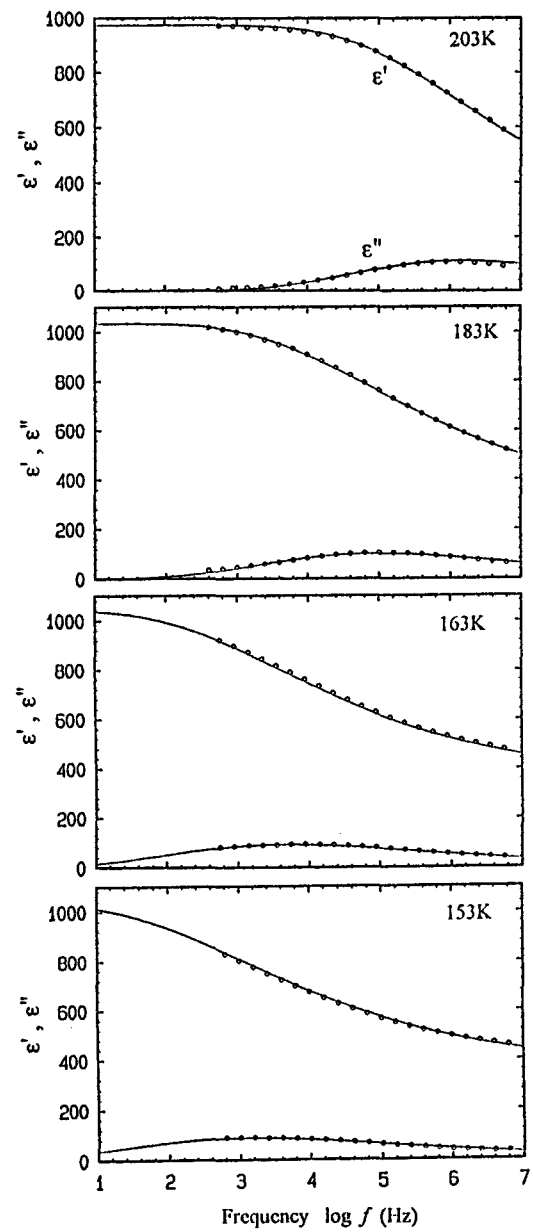


Fig. 5 Dielectric dispersion of SBNT

explained by the 'universal' theory as in the case of PMN. The β value was shown in Fig.6 as a function of temperature. The change of β value was less than that of PMN, indicating that in SBNT the temperature dependence of the dipole correlation is small.

Figure 7 is a relation between $\omega_p$ and $1/(1-n)T$ of SBNT. The straight line with an activation energy of $E_A$=0.050eV indicates the thermally fluctuating dipoles also exists in SBNT.

### 4.3 DIELECTRIC RESPONSE OF RF

From the analysis of dielectric dispersion of RF, it is found that the dielectric dispersion of RF is determined by the thermally fluctuating dipoles in PMR and their freezing process. A large dielectric relaxation is observed when the measuring frequency is close to the fluctuating frequency. Therefore, $T_m$ increases with measuring frequency up to microwave region[14], and no clear peak is observed in optical frequency region[4].

In the freezing process, the correlation of fluctuating dipoles may be enhanced due to the increase in the volume of PMR in paraelectric matrix. This enhancement changes the β value in the Williams and Watts function which determines the shape of dielectric loss peak. On the other hand, the electrically induced dipoles should increase with the enlargement of PMR in which dipoles are fluctuating. Therefore, the induced polarization and the real part of dielectric constant increases with decreasing temperature above $T_m$. However, if the frequency of the fluctuation becomes lower than the measuring frequency in the freezing process (below $T_m$), the real part of dielectric constant decreases according to the theory of dielectric relaxation.

The dielectric properties of RF could be explained by a superimposed phenomenon of the enlargement of PMR, freezing of the fluctuating dipoles in PMR and dielectric relaxation in the measuring frequency range.

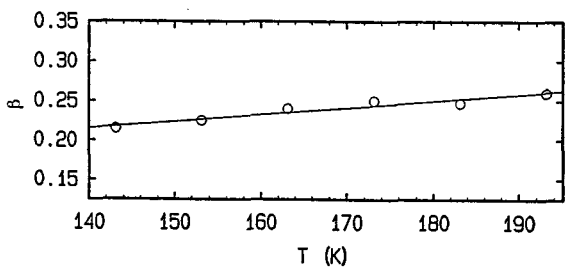


Fig. 6 The β value of BSNT as a function of temperature

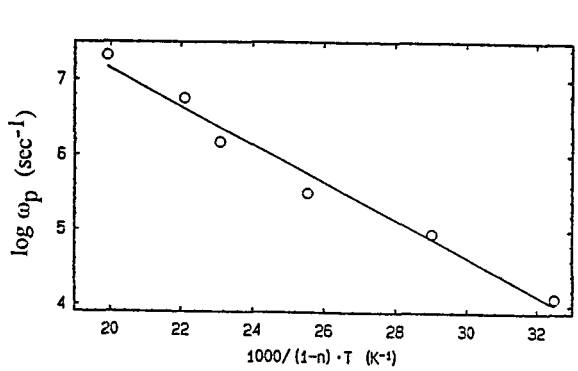


Fig. 7 The relation between $\omega_p$ and $1/(1-n)T$

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