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Frequency dependence of dielectric nonlinearity in PMN relaxor system

Cene Filipič [a,*], Joachim Hemberger [b], Zdravko Kutnjak [a], Adrijan Levstik [a], Alois Loidl [b]

[a] Jožef Stefan Institute, PO Box 3000, 1001 Ljubljana, Slovenia
[b] Experimentalphysik V, Universität Augsburg, D-86135, Augsburg, Germany

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Abstract

The linear ε_1^* and the third harmonic complex dielectric constant ε_3^* in $PbMg_{1/3}Nb_{2/3O_3}$ (PMN) single crystal were measured as a function of frequency and temperature in a small electric field. The dielectric dispersion of ε_3^* was studied and the characteristic relaxation time together with the static dielectric nonlinearity were determined as a function of the temperature. The static dielectric nonlinearity β_s is increasing with decreasing temperature in the temperature range 266–242 K, while the relaxation time follows the Vogel–Fulcher law.

Keywords: Dielectric properties; Ferroelectric properties; PMN

1. Introduction

Recently the investigation of relaxors is getting on intensity. Some aspects of the nature of the freezing transition are revealed. In zero bias electric field relaxors show the transition into the nonergodic phases,[1–4] but if relaxors are cooled in the electric field higher than critical, a long range ferroelectric phase is formed.[5,6]

With recently introduced spherical random-bond random-field (SRBRF) model[7,8] it is possible to describe temperature dependencies of the Edwards-Anderson order parameter q_{EA} and the quasistatic nonlinear dielectric ratio $a_3 = \varepsilon_3/\varepsilon_1^4$ in PMN relaxor. The SRBRF model explains also the crossover in the temperature dependence of a_3 from decreasing into the increasing behavior when approaching the freezing transition T_f from above[9] in zero bias electric field and the crossover from the glass-like behavior of a_3 into the ferroelectric-like monotonous decreasing behavior under the bias electric field higher than critical $E > E_C$.[10] SRBRF model predicts also the peak of the temperature dependence in the static dielectric nonlinearity a_3 at the freezing transition. Because of dispersions in ε_1 and ε_3, which characteristic relaxation times rapidly increases with decreasing temperature at temperatures much above T_f, it is experimentally not feasible to determine the static nonlinear susceptibility in the vicinity of the freezing temperature.

In the present work we will present the measurements of $\varepsilon_1^*(\omega, T)$ and $\varepsilon_3^*(\omega, T)$ in PMN single crystal and show that the dielectric nonlinearity β is the function of the frequency and temperature in the temperature interval between 266 and 242 K. The temperature dependence of the static dielectric nonlinearity β_s and the characteristic relaxation time will also be determined.

2. Experimental procedure

Measurements were done on (111) plate of PMN monocrystal with sputtered Ag electrodes. The method of measurements of ε_1 and ε_3 was described before.[11] The dielectric response was measured in the frequency interval of 10^{-2}–3.10^2 Hz. The amplitude of measuring ac electric field applied on the sample was 170 V/cm. The measurements were done in cooling run with the cooling rate of 0.5 K/min.

* Corresponding author. Tel.: +386-1-4773-420; fax: +386-1-2519-385.
E-mail address: cene.filipic@ijs.si (C. Filipič).

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3. Results and discussion

Fig. 1 shows $|\beta| = |\varepsilon_3(\omega)|/|\varepsilon_1(\omega)|^3|\varepsilon_1(3\omega)|\varepsilon_0^3$ as a function of the temperature at five frequencies. The dielectric nonlinearity $|\beta|$ shows the dispersion below ≈260 K.

Fig. 2 shows ε_3' and ε_3'' as a function of the frequency at 252.5 K. The curve in Fig. 2 represents the fit to the expression.[12–14]

$$\varepsilon_3^*(\omega) = \varepsilon_{3\infty} + \frac{\Delta\varepsilon_3}{(1 + (i\omega\tau)^\alpha)^3}. \quad (1)$$

The parameters of the characteristic relaxation time τ, the distribution parameter α, and the dielectric strength $\Delta\varepsilon_3$ of the imaginary part of the nonlinear dielectric constant were determined from the fits of ε_3'' to Eq. (1). From the experimental values of the ε_3' the high frequency dielectric constant $\varepsilon_{3\infty}$ was determined. Fig. 3 shows several Cole–Cole plots in PMN at different temperatures. It should be mentioned that the Eq. (1) does not describe the experimental data of $\varepsilon_3^*(\omega)$ in PMN at very high frequencies ($\omega\tau \gg 1$). The extrapolation to $\omega \to 0$ was also made with a linear plot through the measured points at low frequencies (Fig. 3), thus giving the static nonlinear dielectric constant ε_{3s}.

The temperature dependence of the static dielectric nonlinearity $\beta_s = \varepsilon_{3s}/\varepsilon_{1s}^4\varepsilon_0^3$ is shown in Fig. 4. The values of ε_{1s} were taken from the measurements of the field-cooled dielectric constant.[3] β_s is increasing with the decreasing temperature in the temperature interval between 266 and 242 K. This is in accordance with the prediction of the SRBRF model above T_f. To determine β_s at temperatures lower than 240 K the measurements of $\varepsilon_3^*(\omega)$ at frequencies much lower than 0.01 Hz would be necessary. The logarithm of the reciprocal characteristic relaxation time $1/2\pi\tau$ determined from the ε_3'' vs frequency (Fig. 2) by the fits to the Eq. (1) is presented as a function of the temperature in Fig. 5. The solid curve in Fig. 5 is a fit to the Vögel–Fulcher

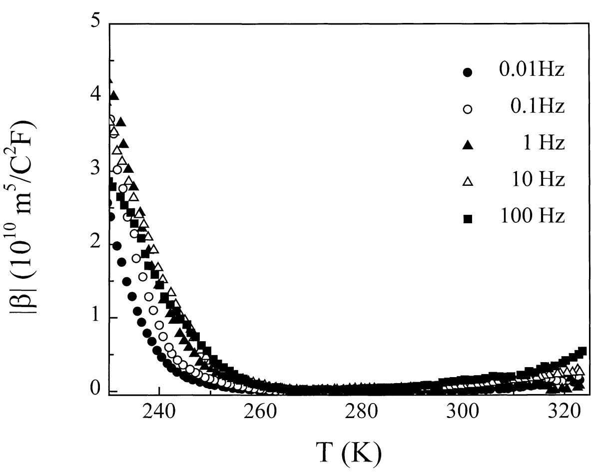

Fig. 1. The temperature dependence of $|\beta|$ at five frequencies.

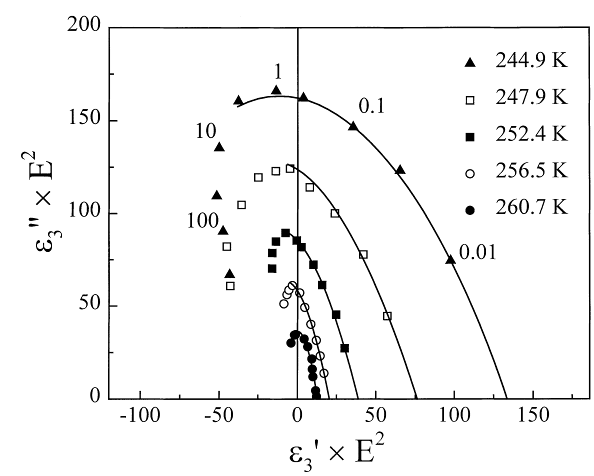

Fig. 3. Measured values of ε_3'' plotted vs ε_3' at five temperatures.

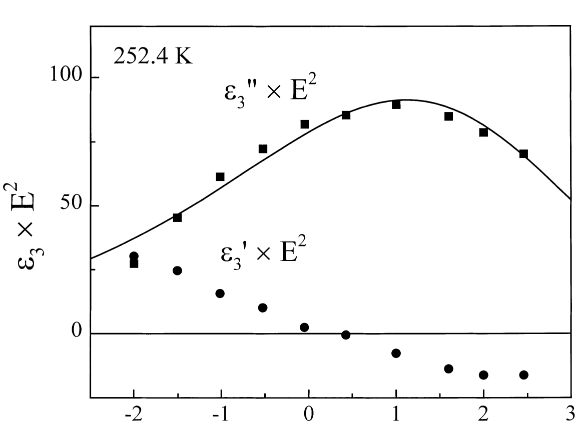

Fig. 2. ε_3' and ε_3'' vs frequency at 252.4 K.

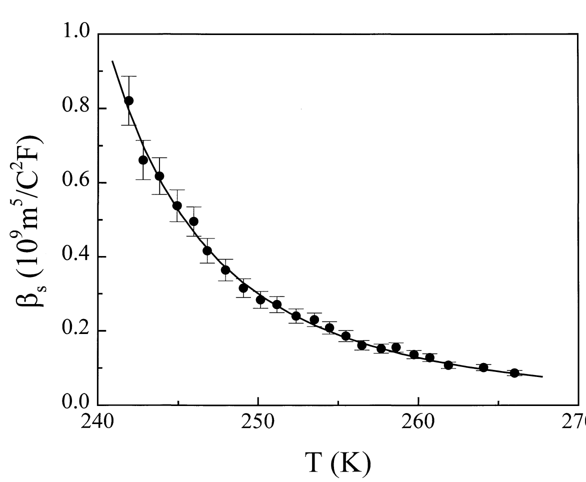

Fig. 4. The temperature dependence of β_s.

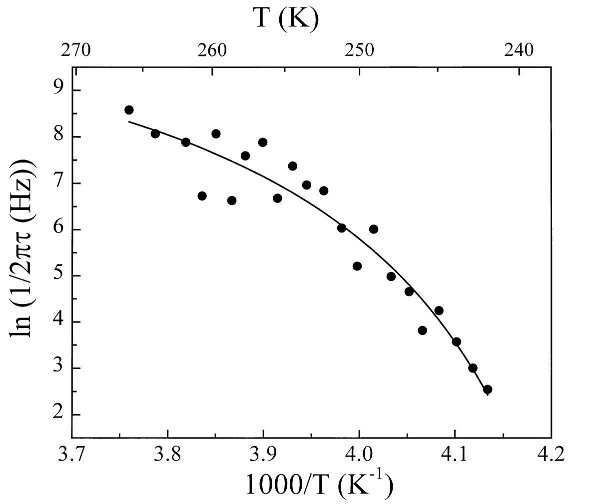

Fig. 5. Temperature dependence of the logarithm of the reciprocal characteristic relaxation time $1/2\pi\tau$ in PMN.

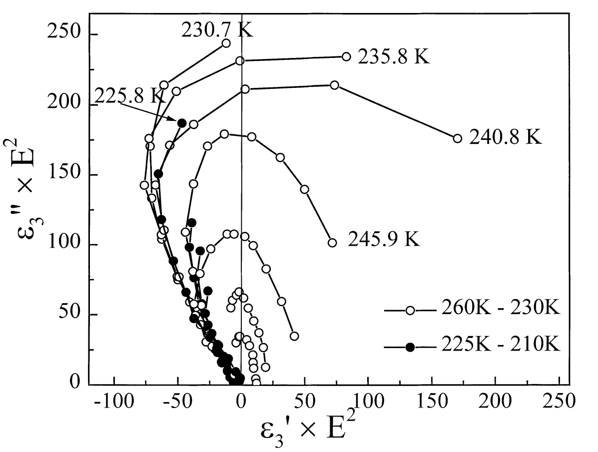

Fig. 6. Measured values of ε_3'' plotted vs ε_3' in the temperature range of 260–210 K.

expression with $T_0 = 227 \pm 5$ K, $U = 139 \pm 64$ kK, and $\tau_0 = 1.1 \times 10^{-6}$ s. The value of T_0 is close to the value determined previously.[3]

Fig. 6 shows measured values of ε_3'' plotted vs ε_1'' in PMN at eleven temperatures in the temperature interval 260–210 K with the temperature step of 5 K. The Cole–Cole plots below T_0 are indicated by (●) and the plots above T_0 by (○). The solid lines connect the data measured at the same temperature. Fig. 6 demonstrates a rapid increase of the characteristic relaxation time with decreasing temperature, which effectively preclude the observation of ε_{3s} in PMN through the freezing temperature. These makes confirmation of the existence of the peak in β predicted by the SRBRF model impossible by the present experimental technique. However, it should be noted that the observed temperature dependence of the β_s is qualitatively in agreement with the results of the SRBRF model in the measured temperature range.

4. Conclusions

The linear and nonlinear dielectric constant of the PMN single crystal were studied as a function of the frequency and temperature. The static nonlinear dielectric constant ε_{3s} and the characteristic relaxation time τ were determined as a function of the temperature. ε_{3s} and thus β_s are increasing with decreasing temperature in the temperature range of 266–242 K according to the SRBRF model, while τ shows Vögel–Fulcher type behavior.

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