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SPECIFIC FEATURES OF HYPERSONIC DAMPING IN RELAXOR FERROELECTRICS

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A comparative analysis of the hypersonic damping behavior in a wide temperature range has been carried out in a relaxor ferroelectric $\text{PbMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$ (PMN) and in a related crystal $\text{Na}_{1/2}\text{Bi}_{1/2}\text{TiO}_3$ (NBT). The damping of longitudinal acoustic phonons was obtained from Brillouin scattering data. The main very broad maxima in hypersonic damping are found to be shifted to higher temperatures with respect to the corresponding main dielectric anomalies. Additional sharp peaks in the hypersonic damping are connected with an inner structural rearrangement in both crystals. This inner evolutionary process contributes to the hypersonic damping regardless of whether there is a real phase transition like in NBT or such a phase transition is frustrated like in PMN. Comparing NBT with PMN we suggest that the dynamics of these materials is determined by two coupled order parameters.

Keywords: Relaxor ferroelectrics, diffuse phase transition, Brillouin scattering.

INTRODUCTION

Now there is a great need for re-investigation of the dynamic behavior of relaxor ferroelectrics with so-called diffuse phase transition. The interest in such complex systems like relaxors has increased due to the recent achievement in understanding of other disordered systems with competing interactions. Let us clarify this situation on an example of a well-known relaxor like PMN. According to the former approach,^{1,2} a main dielectric anomaly with the mean Curie temperature $T_c \sim 260$ K was identified with a ferroelectric phase transition which occurred to be diffused for some reason. For example, the composition fluctuations considered as the most probable origin capable to extend a phase transition region. It seemed that the acoustic anomalies in PMN in the region of T_c could be explained in the same way.³ However, contradictions in this approach have stimulated a search for the other models.

Two models seem to be most important: a model of the glass-like behavior^{4,5} and another quite recent model of the random-field-induced domain states.⁶ In these cases a ferroelectric phase transition according to the ordinary conception does not occur in the former Curie region. Instead the PMN system evolves from a more or less normal disordered state at high temperatures to a new state which is a glass-like phase below $T \sim 250$ K^{4,5} or random-field-induced clusters with a frustrated ferroelectric phase transition at 212 K.⁶ Dynamic aspects of such an

evolution are expected to be rather unusual even in comparison with a heavy overdamped soft modes which are supposed in the former model. Really, a broad central peak was found in light scattering in PMN just in the temperature region of our interest.⁷

One can note that the study of acoustic anomalies appears to be very fruitful if dealing with a central peak and no ordinary soft modes. A good example would be a search for a missing “true” soft mode in ferroelectric-ferroelastic $\text{Gd}_2(\text{MoO}_4)_3$ where such a mode was reconstructed from both the acoustic anomalies in Brillouin scattering and the analysis of low-frequency Raman spectra.^{8–10} Therefore one can hope that the study of acoustic modes in relaxors will contribute to understanding the transformation dynamics in such compounds.

In the present paper, we give a comparative analysis of the Brillouin scattering data in two complex perovskite-like compounds, namely, in well-known PMN and in NBT with a different transformation scheme, although the final ferroelectric state seems to be very similar in both compounds. We suppose that the study of different disordered compounds within the perovskite-like family gives us a chance to find common features in the evolutionary dynamics in the systems of these types.

EXPERIMENTAL

We used a backscattering geometry to obtain the Brillouin spectra from acoustic phonons with the largest wave vectors. An argon ion laser with $\lambda = 514.5$ nm was used to excite Brillouin scattering. Scattered light was analyzed by a Fabry-Perot interferometer operating in a five-pass regime (spectra of NBT and several spectra of PMN) or in a three-pass regime (spectra of PMN in an external electric field and other spectra of PMN). The same results were obtained for PMN in both cases. We followed the temperature behavior of the shift and linewidth at half-maximum of the Brillouin components.

Nearly cubic samples of NBT and PMN with edges about 5 mm were used for studying the behavior of longitudinal acoustic phonons with the wave vector along [001] in both cases without any electric field. To reveal the effect of an external electric field, a sample of PMN with a thickness of about 1 mm was cut. The sample as illuminated along [110] which was an edge about 5 mm long also and an electric field was applied along [110].

ANALYSIS AND DISCUSSION

According to X-ray¹¹ and neutron scattering¹² data, NBT crystals possess a sequence of structural transitions from the high temperature cubic phase to a tetragonal at T_{c1} and then to a trigonal structure at T_{c2} . It seems that a sequence of dielectric anomalies in NBT does not obviously correlate with the structural transitions (see Reference 13 and references therein). A spontaneous polarization appears at some point T_{c3} in the trigonal phase which is more than 50 K below T_{c2} . This temperature, T_{c3} , should be considered as a transition point to the ferroelectric phase. A main dielectric anomaly attributed formerly to the Curie temperature occurs at some temperature in the tetragonal phase just about 50 K above T_{c2} .

FIGURE 1 The temperature dependence of hypersonic damping and the real part of the dielectric permittivity in NBT.

A very broad dip in the behavior of the hypersonic sound velocity seems to have no simple connection with the structural transitions, either.¹⁴ Minimum points of the dip are located in the intertransition region between T_{c2} and T_{c1} . This velocity anomaly in NBT is very similar to that in PMN.^{15,16} And both of them, NBT and PMN, show the correlation between velocity anomalies and corresponding main dielectric maxima. One should note that this conclusion is correct for both hypersonic sound velocity and damping. However, the analysis of damping seems to be preferable due to an additional thin structure of the damping maxima while additional special features of the velocity dips remain under limit of errors.

Therefore, we show only damping anomalies in NBT (Figure 1) and PMN (Figure 2) in the present short paper. Also Figures 1 and 2 represent the corresponding dielectric anomalies measured in the same samples as 100 kHz. There is an obvious shift of the main damping maximum to the higher temperature with respect to the main maximum of dielectric anomaly. The difference is about 30 K in PMN and 50 K in NBT. It seems that the maximum values of different properties show some characteristic points in the evolution of the systems when the response of systems is probed by different methods at different frequencies. The only question is whether the evolution to a ferroelectric-like state in both crystals is similar or not.

However, the damping behavior in our relaxor materials appears to be even more complicated. Besides the main damping maxima, there are some additional peaks on the slopes. NBT exhibits an obvious sharp peak of additional damping in the region of the upper phase transition T_{c1} (Figure 1). This result was confirmed by our measurements several times. Probably, there is an additional structure of damping at lower temperature, too, but now it is difficult to treat these results without a doubt. In contradistinction to NBT, an additional peak of the hypersonic

FIGURE 2 The temperature dependence of hypersonic damping and real part of the dielectric permittivity in PMN.

damping in PMN occurs on the low-temperature slope of the main damping maximum (Figure 2). At first we found this new anomaly using a three-pass Fabry-Perot interferometer.¹⁶ Figure 2 represents new data obtained with a five-pass interferometer. So, the existence of this additional anomaly is clearly proved. The anomaly is found in the temperature region of a ferroelectric phase transition induced by the external electric field.⁵ Dielectric response of PMN in the electric field shows an additional peak at the same temperature, $T \sim 212$ K. One should emphasize that the additional anomaly in the sound damping appears without any electric field.

We have no possibility to discuss in this short paper a phase transition sequence in NBT in detail. The recent idea of two coupled order parameters¹⁷ seems to be very fruitful in this case. Even a puzzling state in NBT between T_{c2} and T_{c3} with double hysteresis loops finds its explanation in a simple model with two coupled structural and ferroelectric order parameters as a result of the special relation between coefficients. In this case both the dielectric response and the acoustic anomalies appear to be modified by the coupling between different order parameter, so the main dielectric and sound damping maxima in NBT have no obvious relation to any particular structural transition.

Note that there is a sharp peak in the hypersonic damping as a similar response in both crystals: in NBT with respect to a structural cubic-tetragonal phase transition and in PMN with respect to a frustrated ferroelectric phase transition. Simultaneously there is no appreciable dielectric response in the corresponding temperature regions. The latter comparison shows that hypersonic damping is very sensitive to a change of the inner structure of relaxor materials including pretransitional phe-

FIGURE 3 The influence of an external electric field on the hypersonic damping in PMN in the region of an induced ferroelectric phase transition.

nomena. Therefore we suppose that the complex behavior of hypersonic damping in PMN evidences an evolution which is determined by some coupled order parameters. However, the situation in PMN is more complicated in comparison with NBT because the order parameter activity is completely frustrated due to disordering and random fields.

The pertinent question whether an electric field ought to affect the damping of Brillouin component arises after what the dielectric response of PMN has shown in an external electric field (See Reference 6 and references therein). Figure 3 compares the hypersonic damping in PMN without and in the electric field. As one can see, the external field $E = 3 \text{ kVcm}^{-1}$ does not affect the sharp additional peak of damping appreciably while the hypersonic damping increases in an induced ferroelectric phase as temperature decreases. Further study by Brillouin and ultrasonic method is needed to better understanding the sound propagation in this unusual phase. Also it is desirable to know more about the local atomic arrangements in this phase and their changes in an electric field.

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