PHYSICAL REVIEW B VOLUME 51, NUMBER 9 1 MARCH 1995-I

Critical acoustic behavior of the relaxor ferroelectric $Na_{1/2}Bi_{1/2}TiO_3$ in the intertransition region

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(Received 25 May 1994)

A cubic-tetragonal-trigonal sequence of phase transitions in the relaxor ferroelectric $Na_{1/2}Bi_{1/2}TiO_3$ (NBT) has been studied by Brillouin scattering. Pronounced anomalies for both the hypersonic velocity and damping have been found that peak just between two phase transitions in contrast to the ordinary well-known behavior with anomalies in the vicinity of every transition point. The fact that these anomalies are centered in the intertransition region and extend beyond both transitions is attributed to the fluctuations of two coupled order parameters associated with disorder in the Na- and Bi-cation distribution in a manner characteristic of relaxor ferroelectrics. Such a phenomenon seems to be typical of relaxor materials and to be the basis of the diffuse phase-transition dynamics.

I. INTRODUCTION

Diffuse phase transitions in relaxor ferroelectrics are still far from being understood in spite of their long history of investigation.[1] Recent papers devoted to the best-known ferroelectric of this type, $PbMg_{1/3}Nb_{2/3}O_3$ (PMN), describe the low-temperature phase $T \leq T_c$ (=212 K) at the microscopic level, including a structural model[2] and an analysis of the specific behavior initiated by the random-field-induced domain states.[3] Dynamic aspects of the diffuse phase transition, judging by a central peak in light scattering,[4] are most pronounced at higher temperature in the region of a unique broad dielectric anomaly with maximum centered at $T \sim 260$ K at low frequencies.[5,6] Correlation between superparaelectric moments[7] occurs at even higher temperatures. It originates in clusters with $\langle 111 \rangle$ distortions which appear upon cooling from about 600 K.[2]

PMN has a ferroelectric phase transition at 212 K, but it is evident only in an external field (Ref. 3 and references therein). PMN also exhibits unusual pretransitional behavior with pronounced anomalies in dielectric, acoustic, and other phenomena. An extensive pretransition region is typical for relaxor ferroelectrics and is a keystone in understanding diffuse phase transitions. In this connection we have searched for analogous behavior in related crystals to find an example of more pronounced development of the precursor structures.

Similar evolution from paraelectric to ferroelectric phases in the complex perovskite-type $AB'_{1-x}B''_xO_3$ or $A'_{1-x}A''_xBO_3$ compounds is evidenced by a broad central peak in light scattering from PMN (Ref. 4) in the first group and $Na_{1/2}Bi_{1/2}TiO_3$ (NBT) (Refs. 8 and 9) in the second. However, central-peak analysis is difficult because the peak is superimposed on low-frequency Raman modes with unknown and probably complex intensity temperature dependence. Therefore we studied acoustic behavior of some relaxor ferroelectrics in a wide temperature interval. Preliminary PMN results show correlation between central-peak behavior and sound velocity and damping anomalies.[4,10,11] We prefer Brillouin scattering to ultrasonic methods, for which heavy damping hinders continuous measurements at low frequencies, as seen for example in comparing Brillouin-scattering data[10,11] with ultrasonic measurements for PMN.[12]

In this paper we consider Brillouin-scattering measurements in NBT. Most relaxor ferroelectrics have pairs of different B cations, but NBT has unlike valency Na^+ and Bi^{3+} cations in A positions. At room temperature NBT is ferroelectric with a trigonal structure which is the result of a phase transformation sequence which proceeds,[13-16] unlike PMN, from cubic to tetragonal at T_{c1}, then to trigonal at T_{c2}, and finally spontaneous polarization appears at T_{c3} at the transition to the ferroelectric phase. This cubic-tetragonal-trigonal sequence is confirmed by neutron-diffraction[16] and x-ray measurements.[15] This interesting and different transition sequence is the basis of our choice of NBT for these Brillouin-scattering measurements.

II. EXPERIMENTAL

A cubic sample of NBT with edges about 5 mm was illuminated along [001] by an argon-ion laser with $\lambda = 514.5$ nm. We used the same sample employed before in Raman scattering.[8,9] Backscattering spectra were analyzed by a five-pass Fabry-Perot interferometer. We present both the Brillouin shift (or hypersonic velocity) and damping for longitudinal-acoustic phonons propagating along [001]. We report the shift and the linewidth at half-maximum. We also follow the temperature dependence of the Brillouin spectrum background from the NBT crystal. This background intensity peaks strongly near the extrema of the elastic anomalies.

Figure 1 shows that at higher temperature (655 K) this background greatly exceeds the Brillouin doublet peak intensity. Even at 78 K the signal is less than background. This background reduces the quality of our five-pass interferometer results to that of the worst single-pass ones for ordinary samples, especially at temperatures of interest near the sound velocity and damping extrema. Use of a narrow-band (1 Å) interference filter

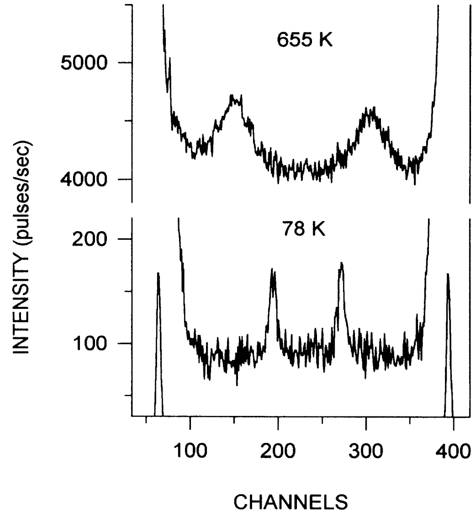

FIG. 1. Two Brillouin spectra at different temperatures are shown to illustrate the experimental conditions to measure three values: the frequency shift, the linewidth at half maximum, and the intensity of the background. The free spectral ranges of the interferometer are 30.20 GHz for $T=655$ K and 34.62 GHz for $T=78$ K.

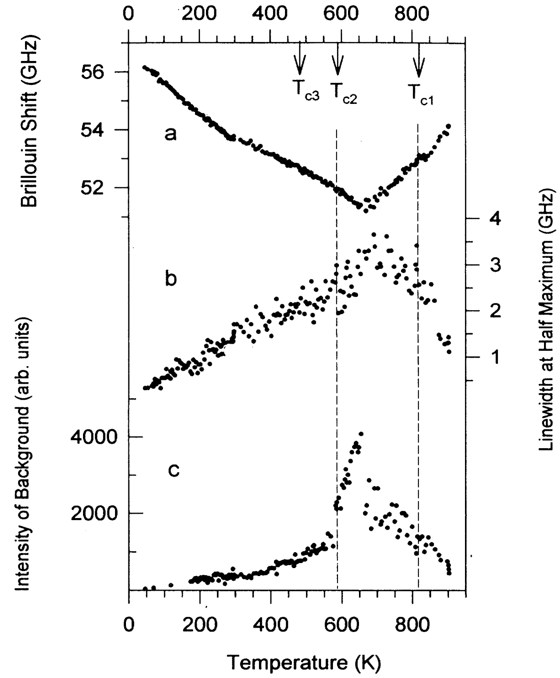

FIG. 2. Temperature dependence of different characteristics in NBT: (a) Brillouin shift of the longitudinal acoustic phonons vs T; (b) Linewidth of Brillouin components at half maximum (or damping) vs T; the instrumental broadening was subtracted; (c) Intensity of the background in the Brillouin spectra vs T.

did not improve the situation radically. Probably a very strong central peak is the main origin of this background. Although this background causes significant spreads in our shift and linewidth values, we obtained good reliability of the main anomalies (a dip in sound velocity and a peak in sound damping, Fig. 2).

We used the green line of our argon-ion laser and our NBT sample is yellow-green at room temperature, so we were concerned about green light absorption at higher sample temperatures. However, the laser beam intensity exciting the crystal was not significantly reduced even at the highest temperatures. In any case, a shift of the fundamental absorption edge upon heating mostly affects intensity measurements. Our shift and linewidth measurements covered too small a frequency range to be affected, except in very special cases. We carried out only qualitative analysis of the background intensity. Qualitatively similar behavior was found for both the background intensity (a green laser) and the central-peak intensity in Raman scattering (a red laser[8,9]). Accordingly we are sure that our Brillouin shift and linewidth data are correct.

The ferroelastic phase transition at T_{c1} reduces the cubic symmetry to tetragonal, but the a/c cell parameter ratio changes only slightly, going abruptly to $a/c=0.999$ at T_{c1} and decreasing to 0.998 on cooling down to T_{c2}.[15] Because of this small change, we observed no splitting or even broadening of the Brillouin components from the ferroelastic domain structure, even though such domains were clearly seen under a microscope.[14] Accordingly we expect that application of uniaxial pressure to create a single ferroelastic domain would not have affected our spectra significantly.

The tetragonal-trigonal symmetry change and the transitions to the antiferro- and ferroelectric states also do not affect the Brillouin spectrum. This implies either that the antiferro- and ferroelectric microdomains are small compared to the optical wavelength, or that these distortions also are too small to affect the Brillouin splitting and linewidth. In the latter case, application of an electric field to create a single ferroelectric domain would not have affected the Brillouin spectrum. In the former case, a shift might have been observable, but such a large field could have drastically altered the phase diagram. For instance, in PMN an anomaly in ac dielectric permittivity corresponding to a ferroelectric transition appears only upon simultaneous application of a large dc electric field (Ref. 3 and references therein).

The acoustic behavior of NBT thus remains pseudocubic even at low temperatures, where the velocity and damping recover the values they had in the paraphase. Such pseudocubic behavior in lower-symmetry phases is typical for relaxor materials. For instance, the ferroelectric phase of PMN has regions with trigonal distortions[2] but the acoustic properties can be described by a cubic set of elastic constants (Ref. 10 and references therein).

III. COMPARISON OF BRILLOUIN AND OTHER EARLIER NBT RESULTS

Figures 2(a) and 2(b) show, respectively, the temperature dependence of shift and linewidth for the Brillouin components in NBT and reveal a huge dip in sound velocity and an unprecedented damping peak. Figure 2(c) presents a very large background intensity peaking in the same temperature range. To analyze these unusual elastic and background anomalies we superimpose these results on a transition sequence scheme indicated by results from the literature.

Neutron-diffraction data[16] indicate that NBT crystals possess two instabilities at different points in the Brillouin zone. A first-order phase transition occurs at $T_{c1}=820$ K from a cubic paraphase to a tetragonal ferroelastic phase. The tetragonal distortion corresponds to an irreducible representation at the M point of the cubic phase Brillouin zone. The appearance, development and disappearance of the M-point superlattice reflections observed in neutron diffraction[16] is illustrated in Fig. 3.

The next transition which is nominally at $T_{c2}=610$ K (Ref. 12) is to a trigonal phase. The trigonal structure is not a subgroup of the tetragonal structure, but its distortion corresponds to an irreducible representation at the R point of the cubic phase Brillouin zone. The temperature given as T_{c2} in older publications is 593 K, which is shown as the limit of the tetragonal phase in Fig. 3 and which is the point of onset of phase coexistence in heating runs. There is a sharp cusp at 640 K in the M-point neutron reflection intensity in heating runs, which signals the completion of the phase transition. All our Brillouin-scattering measurements were carried out during heating runs.

The neutron-scattering data indicate that the phase transitions at T_{c1} and T_{c2} are initiated by some irreducible representations at the M and R points of the cubic phase Brillouin zone, respectively.[16] It is well-known that in both these cases the anomalies for longitudinal-acoustic phonons are described by $Q\eta^2u$-type coupling, squared in order parameter and linear in strain, where Q is an electrostriction coefficient.[17] However, neither the steplike anomaly for sound velocity nor the Landau-Khalatnikov maximum for damping could be observed with certainty in the vicinities of T_{c1} and T_{c2} which are located on opposite sides of an extensive dip in sound velocity and a broad damping peak. The velocity dip and damping peak look like reflections of each other in a horizontal mirror plane [Figs. 2(a) and 2(b)]. The usual velocity step and Landau-Khalatnikov maximum at phase transitions do not appear as reflections of each other.

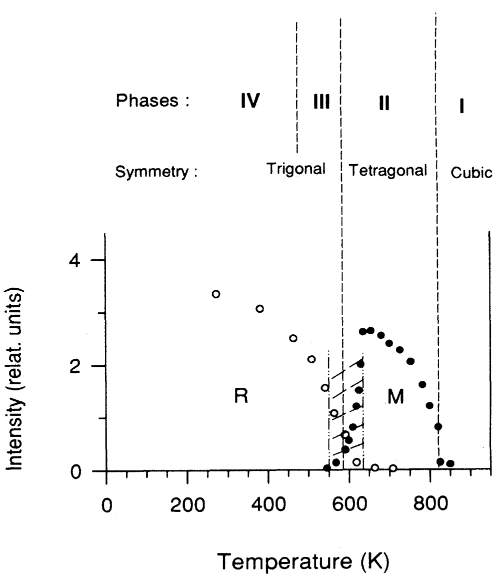

FIG. 3. Intensity of the superlattice reflections from M and R points of the cubic Brillouin zone obtained in neutron-scattering measurements (Ref. 16) vs T to show the range of phase coexistence (shaded region). The phase sequence in NBT is shown at the top; I: paraelectric and paraelastic, II: ferroelastic, III: antiferroelectric (?), IV: ferroelectric. The corresponding crystal symmetries are shown in the middle. All data were obtained on heating runs.

Dielectric measurements by ourselves and others give another sequence of anomalies which correspond only partially to the structural instabilities. The dielectric behavior of NBT is summarized in our recent paper.[18] The lowest-temperature permittivity anomaly is small and corresponds to a third transition, in a region centered at $T_{c3}=473$ K where the remanent polarization disappears. This ferroelectric transition is far below the nearest structural instability point, T_{c2}.

Another dielectric anomaly was found in our NBT sample near 640 K (Fig. 4) and had been seen by others, with some spread of reported permittivity peak temperatures (Ref. 18 and references therein). This anomaly has a characteristic cusp shape similar to that of the M-point reflection intensity which also cusps at 640 K, so this anomaly may result from the tetragonal-trigonal transition centered at T_{c2}. From T_{c2} down to T_{c3} there is an enigmatic phase in which some workers report double hysteresis loops typical of antiferroelectric behavior, but others deny that an antiferroelectric phase exists (see Ref. 18 for discussion and other references).

Our largest dielectric hump was found near the cubic-tetragonal transition temperature T_{c1} and was attributed

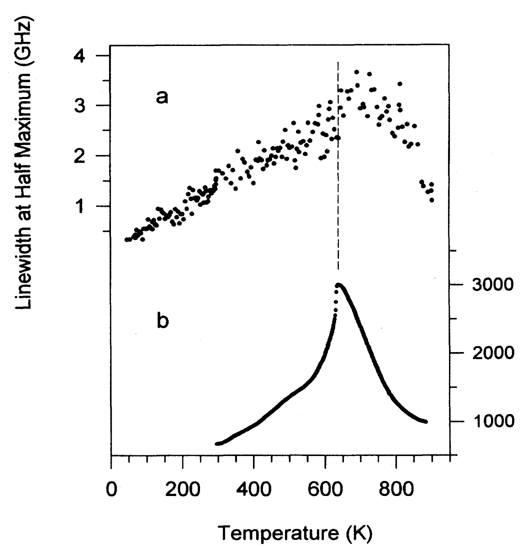

FIG. 4. A comparison of the dielectric maximum in NBT measured at 100 kHz (b) with the behavior of acoustic-phonon damping (a). The dashed line indicates the temperature difference between dielectric and acoustic damping maxima.

to superparaelectric clusters in the tetragonal phase.[18] This hump was observed from 20 Hz to its disappearance above 10 kHz with corresponding peak positions ranging from 720 to 870 K. No particular anomaly was observed right at the first-order transition temperature $T_{c1}=820$ K reported by Vakhrushev *et al.*[16] Similar dielectric permittivity humps in other perovskite crystals and ceramics have been attributed to partial blocking of current flow by surface layers or electrodes.[19,20] Accordingly, the degree and nature of connection of this special high-temperature relaxation behavior with the ferroelastic phase transition is uncertain.

To summarize the comparison of our Brillouin results with neutron-diffraction and dielectric measurements, the broad anomalies we observed in sound velocity, damping, and background do not correspond to any of the transitions indicated by neutron and dielectric results. The closest correspondence is a very rough resemblance of the background intensity in Fig. 2(c) to the neutron *M*-point reflection intensity in Fig. 3. The comparison of linewidth with permittivity in Fig. 4 shows that the peak values are not far apart, but the linewidth peak is an order of magnitude broader and so it cannot reasonably be attributed to the transition at T_{c2}. To explain these broad anomalies, we employ comparisons with other systems as described in Sec. IV.

IV. DISCUSSION OF NBT IN RELATION TO OTHER SYSTEMS

Recent papers show that relaxor ferroelectrics provide a unique opportunity for observing fluctuation contributions,[2,3,6] which we believe are responsible for the broad anomalies we observed in Brillouin scattering from NBT. For instance, the diffuseness of the ferroelectric phase transition in PMN was attributed to random-field-induced clusters originating from charged compositional fluctuations.[3] In NBT such fluctuations could originate in randomly placed Na^{+} and Bi^{3+} cations and we expect that they contribute to the acoustic anomalies also. Figure 2(b) shows considerable anomalous damping gradually developing far above T_{c1} which can be reasonably attributed to fluctuations.

The usual Landau theory is not valid if large fluctuations exist, so we would not expect to find separately the relaxation portion of the acoustic anomaly which should be a sharp anomaly near the phase transition. For example, it was shown for critical ultrasonic attenuation at the liquid-helium λ point that inescapable mixing of the Landau-Khalatnikov (or order parameter) and fluctuation contributions occurs in this system which has large fluctuations.[21] A similar "hybridization" may prevent us from observing relaxation contributions of the usual form as seen in many ferroelectrics and ferroelastics even in the presence of the considerable dispersion characteristic of Brillouin scattering in these materials.[17,22]

The "hybridization" describes for the helium system[21] near the λ point should occur in NBT near *two* transitions, from the tetragonal phase to the cubic phase at T_{c1} and to the trigonal phase at T_{c2}. However, a linear superposition of two anomalies at the T_{c1} and T_{c2} phase transitions cannot fit our data. The velocity minimum and damping maximum extremes both occur near $T_{\mathrm{extr}}\approx 670$ K which is between T_{c1} and T_{c2} and closer to T_{c2}. This leads us to suggest an interaction of fluctuations. Each type of fluctuation contributes to acoustic anomalies with the same $\eta^2 u$-type coupling between the fluctuating order parameter and strain.

In what ways is the hybridization in NBT similar to that in liquid helium, and in what ways does it differ? In liquid helium, and other pure systems with second-order transitions, as one goes away from the critical point there is a dominant phase (possibly broken up into domains large enough so they do not affect Brillouin scattering significantly) which near the critical point has small temporal fluctuations of the other phase(s) not restricted to any particular spatial regions. These fluctuations are only significant in the near vicinity of the transition.

In NBT we do not have a pure system because disordered placement of unlike-valence cations leads to electric and elastic fields. This disorder is quenched, at least for times shorter than the conductivity relaxation time. This disorder and the resulting fields prevent buildup of large domains. Instead, there are microdomains or superparaelectric clusters whose boundaries fluctuate in time, and which may rearrange completely in a time longer than the conductivity relaxation time (if the conductivity is due to these cations). If such complete rearrangement occurs, it is likely that it occurs throughout the tetragonal phase temperature range, with partial rearrangement extending into the cubic and trigonal phases where tetragonal clusters are expected to persist. Then the "pure fluctuation" state which in a pure system exists only exactly at T_c, exists in this impure system over a wide temperature range, with greater correlation length and longer time constant than for typical fluctuations.

To our knowledge, no theory deals explicitly with such impure-system fluctuations. Perhaps the closest analogy is the behavior right at T_c for a pure system with a second-order phase transition. We have provided here a qualitative discussion of the NBT behavior based on this analogy. This behavior can be expected to exhibit dispersion effects. Figure 4 compares temperature dependences for the largest frequency-independent dielectric anomaly (lower curve) with that for hypersonic damping (upper curve). As discussed above, the position of this dielectric maximum near 640 K is probably determined by competition of coupled order parameters initiating different structural instabilities. The hypersonic damping maximum peak near 670 K is probably connected with fluctuation of these coupled order parameters. The shift of damping maximum (measurements at about 5×10^{10} Hz) relative to the dielectric anomaly (measurements at 10^5 Hz) is clearly seen. The shift may be due to the frequency dependence of fluctuations coupling electrostrictively to strain. This behavior is very similar to that in PMN which also shows a shift between the dielectric maximum[2,6,7] and the hypersonic damping peak.[10,11]

Fluctuations of the coupled order parameters or hybridized fluctuations manifest themselves in the spectra of low-frequency light scattering also. A preliminary indication of increased central-peak intensity between T_{c1}

and T_{c2} was obtained in the Raman-scattering study.[8,9] In the present work we followed the behavior of the background in the Brillouin spectra which consist of the scattered light transmitted through an interference filter of 1-Å half-width together with the Brillouin components. This background corresponds to the peak intensity of the central component if there is only one broad central peak as suggested by the Raman spectra.[8,9] Figure 2(c) shows three special features of this background. The major feature is the broad background peak, which is qualitatively similar to the broad linewidth peak in Fig. 2(b) and the broad Brillouin shift dip in Fig. 2(a). The second feature is the background increase when approaching T_{c2} from below and T_{c1} from above, which is comparable to the evolution of both the central peak in Raman scattering[8,9] and overdamped soft modes detected in neutron scattering.[16] However, these effects are small in comparison with the dominant anomaly in low-frequency light scattering between T_{c1} and T_{c2}. The third feature is the "sloping mesa" between 593 and 640 K which corresponds nearly to the tetragonal and trigonal phase coexistence region. The discontinuities characterizing this feature are not seen in the acoustic anomalies in Figs. 2(a) and 2(b).

The shaded region in Fig. 3 shows the coexistence range of the R- and M-point superlattice reflections, which corresponds to the trigonal and tetragonal phases. The acoustic anomalies have no obvious relation to this coexistence region centered near 600 K because their extrema [Figs. 2(a) and 2(b)] are significantly shifted to higher temperatures. Although the neutron measurements can separate information from different points in the Brillouin zone so superlattice structures of different types (M and R) were studied independently[16] (Fig. 3), the light-scattering spectra show an integral response.

We have found an obvious correlation of the neutron-scattering intensity[16] with our Brillouin background intensity and the central peak in Raman scattering[8,9] as T_{c1} is approached from above. However, this effect in light scattering is weak enough in comparison with the increase of the background between T_{c1} and T_{c2} so that we can clearly define it as a critical process. We have no data now on the location in the Brillouin zone to which this critical process corresponds. It may be at some other point than the M or R points, in partial analogy with the situation in proton glass discussed below.

Brillouin scattering revealed fluctuations in $Rb_{1-x}(NH_4)_xH_2PO_4$ (RADP) mixed crystals[23,24] which in the $0.22<x<0.74$ "proton glass" range exhibit features qualitatively different from both the ferroelectric ($x=0$) or antiferroelectric ($x=1$) pure substances. Also, neutron studies revealed[25] a growth of short-range correlations upon cooling in the deuterated isomorph of RADP. These fluctuations were neither ferroelectric (Γ point at Brillouin zone center) nor antiferroelectric (Z point at the zone boundary). Instead, a characteristic response in the neutron scattering appeared at wave vectors $q\Sigma$ on the Σ lines connecting the Γ point to the Z point. Perhaps, in partial analogy to these results, neutron scattering in the correct geometry in NBT will someday show that the fluctuations do not occur at a Brillouin-zone point corresponding to its ordered phase.

Both proton glass and NBT have competing interactions caused by random cation placement. In proton glass the competing ferroelectric and antiferroelectric interactions provide local order which is neither ferroelectric and antiferroelectric. This order can be characterized by an Edwards-Anderson order parameter (at least on a short-time scale). This order parameter provides a Landau-Khalatnikov term as the main contribution for the acoustic anomalies of the longitudinal phonons in RADP,[23,24] compared to which the contribution of dynamic fluctuations was negligible.[23]

In NBT the tetragonal and trigonal ordering tendencies are not completely frustrated by the random cation placement. However, the strong short-range interactions from the random ion placement compete with the interactions causing the transitions, giving the relaxor state, in partial analogy to the proton glass state in the RADP family. In contrast to RADP, there is strong evidence for critical behavior in NBT resulting from the dominance of the fluctuation contribution to the acoustic anomalies.

Returning now to the unusual, possible antiferroelectric, region between T_{c2} and T_{c3}, we emphasized recently[18] the similarity in the behavior of NBT and other complex perovskites like $PbCo_{1/2}W_{1/2}O_3$ and $PbYb_{1/2}Ta_{1/2}O_3$. A theoretical interpretation recently appeared[26] for the polarization response of crystals with structural and ferroelectric instabilities. A simple model with two coupled structural and ferroelectric order parameters "shifts" the main dielectric anomaly from the ferroelectric transition to the structural one. $PbCo_{0.5}W_{0.5}O_3$ is considered as the clearest example. We cannot apply this model directly because NBT has two structural instabilities rather than one. However, the above "shift" makes it seem likely that the dielectric response of NBT is modified by the coupling between different order parameters. This may be the reason why the dielectric anomaly near 640 K has no obvious connection with any particular instability.

Comparing results for NBT and other relaxor materials, we would like to emphasize the importance of the short-range random-ion-caused interactions in promoting similar relaxor behavior in both crystals. For instance, the hypersonic response of PMN is very similar to that of NBT. Both exhibit a broad sound velocity dip and a corresponding broad damping maximum.[4,10,11] Thus we expect the relaxor state in PMN to be the result of competing interactions also. However, in PMN the ordering tendencies are completely frustrated under normal conditions. Only in an external electric field did a ferroelectric anomaly of the dielectric constant appear at $T_c \sim 212$ K which corresponds to the lower transition with respect to the relaxor state.[3,27] In $PbIn_{1/2}Nb_{1/2}O_3$ (PIN), the upper antiferroelectric phase transition occurs only in samples with an ordered distribution of the In and Nb cations.[28] It may be that the dynamics of partly disordered PIN will be similar to that in NBT. Hence PIN is a very desirable object for future Brillouin or ultrasonic measurements.

Numerous perovskite structure compounds have a succession of phase transitions.[29] Most of them have large fluctuation contributions evidenced by broad central peaks in light scattering, (i.e., in $BaTiO_3$ and $KNbO_3$) (Ref. 30) or by display of precursor order in the behavior of different properties (i.e., in Raman spectra).[31] However, as far as we know, no one has shown acoustic anomalies to be shifted to an intertransition region. NBT does show formally a sequence of perovskite-type phase transitions with symmetry reduction but phenomenologically it is affiliated with the relaxor ferroelectrics. The competing but interacting fluctuations in NBT manifest themselves clearly in the appearance of acoustic anomalies between phase transitions. Such behavior seems to be characteristic of relaxor materials even if the "surrounding" phase transitions are frustrated. The charged compositional heterogeneity provides a fruitful ground for development and coupling of the competing order parameters.

V. CONCLUSIONS

We have shown that the relaxor ferroelectric NBT exhibits correlated anomalies in the sound velocity and damping as well as in the background intensity. The latter anomaly is connected qualitatively with the central peak in light scattering. Figures 2(a)–2(c) show that these anomalous behaviors extend far above T_{c1} due to fluctuations, as for many other perovskite phase transitions. However, a simple superposition of critical anomalies at T_{c1} and T_{c2} does not explain the experimental dependences. There may be special mixed fluctuations or fluctuations of the coupled order parameters (which we also call "hybridized" fluctuations) with a characteristic extremal response at a temperature that has no direct relation to the Landau-like behavior of the order parameters obtained from neutron scattering. We illustrate in Fig. 3 the absence of a simple relation between the anomalies discussed above and "the region of coexistence of two independent phases."[16]

The main conclusion of this paper concerns the existence of acoustic anomalies above the temperatures of the two frequency-independent dielectric anomalies in NBT. This work continues related investigations of PMN,[10,11] in which acoustic anomalies were found near 290 K, above the characteristic relaxor dielectric peak near 260 K and far above the frustrated ferroelectric transition[3] at 212 K. In both crystals very similar anomalies were found in the sound velocity and damping far above the temperatures at which the remanent polarization or the ferroelectric state disappear. The relaxor ferroelectric states in both NBT and PMN seen to be final results of slightly different temperature evolution in which unusual but similar acoustic anomalies were found as a common feature in these two different types of relaxor ferroelectrics.

ACKNOWLEDGMENTS

The authors are grateful to T. V. Kruzina for single-crystal growth. One of us (I.G.S.) wishes to thank the Physics Department for financial support and for kind hospitality during a long stay at the Montana State University. I.G.S. greatly appreciates the financial support through an NRC CAST Grant. This work was supported by NSF Grant No. DMR-9017429.

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