Phase transition of chemically doped uniaxial relaxor ferroelectric

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Abstract
We report a neutron scattering study of the ferroelectric phase transition in Sr0.585Ce0.025Ba0.39Nb2O6 (SBN-61:Ce). We find no evidence for a soft transverse optic phonon. We do, however, observe anisotropic diffuse scattering. This scattering has inelastic and elastic contributions. In the paraelectric phase the susceptibility associated with the elastic diffuse scattering from SBN-61:Ce increases on approaching the transition temperature. In the ferroelectric phase the lineshape of the elastic scattering is consistent with the form expected for the ferroelectric domain walls. In contrast to the macroscopic observations, the scattering properties of Ce-doped crystal do not exhibit important changes with respect to those of pure Sr0.61Ba0.39Nb2O6.

Keywords: phase transitions, neutron scattering, disordered ferroelectric

(Some figures may appear in colour only in the online journal)

I. Introduction

Relaxor ferroelectrics, relaxors for short, are disordered crystals with outstanding dielectric, piezoelectric and electro-optic properties [1]. It is generally believed that disorder is a key ingredient in the physics of relaxors, but a consistent model of their behaviour has still not been developed [2–9]. Even the basic criteria for a disordered ferroelectric to become a relaxor have not yet been established.

Sr1−xBaxNb2O6 (SBN), a group of disordered uniaxial ferroelectrics with an unfilled tetragonal tungsten bronze structure [10]. The phase transition of SBN is strongly affected by the relative amount of Sr/Ba ions or via doping with rare-earth ions. The effects of the latter chemical modification are most often studied for Sr0.61Ba0.39Nb2O6 (SBN-61) as large and high-quality single crystals of this composition are available. The observation for SBN-61 is that even a minuscule concentration of Ce3+ ions strongly affects the ferroelectric phase transition [11]. More specifically, the anomaly in dielectric permittivity ε broadens and its frequency dispersion becomes much more pronounced in SBN-61:Ce as compared to the pure crystal. The peak in ε shifts towards lower temperatures upon increasing the doping level. The ferroelectric polarisation is history-dependent near the transition temperature [12, 13]. There is a strong change in polarisation aging under light illumination in SBN-61:Ce [14]. Thus, the macroscopic properties of doped SBN-61 crystals have much in common with relaxors.

The phase transition of pure SBN-61 has been recently studied by neutron scattering [15–17]. On approaching from above the transition temperature, TC, a slow dynamic diffuse scattering (DS) emerges in the neutron spectra [16, 17]. This appears to account for divergent dielectric permittivity [16] and has a characteristic energy scale in the μeV range [17]. Well below TC the DS appears in neutron spectra as an
elastie component and can be fully ascribed to ferroelectric domain walls (DW) [16]. Thus, despite inherent chemical disorder the scattering from SBN-61 is similar to the one from classic order-disorder ferroelectric. It is therefore tempting to establish if doping, which considerably modifies the macroscopic properties of SBN-61, leads to substantial changes in the microscopic properties as seen by neutrons. We report here a neutron scattering study of the ferroelectric phase transition in Ce-doped Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ single crystal. In stark contrast with modifications to the macroscopic properties induced by doping, we do not find important changes in the scattering properties with respect to the parent pure material.

II. Sample and the experimental details

Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ has a tetragonal structure. Above $T_c$ the space group of SBN is 4/mmm, while below the ferroelectric phase transition temperature the space group is 4mm. The unit cell contains 5 formula units, so that five Sr/Ba ions are distributed over six available positions. These two ions have considerably different ionic radii ($r_{\text{Sr}^{2+}} = 1.12$ Å and $r_{\text{Ba}^{2+}} = 1.34$ Å) [18]. The structure of SBN is a three-dimensional network of distorted Nb/O octahedra connected together so that there are pentagonal, square and triangular tunnels. The smallest triangular tunnels are not occupied. Only strontium ions reside at the square tunnels. Finally, strontium and barium ions are distributed over the largest pentagonal channels. As the Sr$^{2+}$ and Ba$^{2+}$ ions have formally the same charge there are no strong electrostatic forces related to the distribution of the Sr and Ba ions. The possible charge irregularity is then due to the presence of unfilled positions in the triangular channels.

An obvious charge disorder is introduced in SBN upon doping it with trivalent cerium. Ce$^{3+}$ ions occupy Sr$^{2+}$ sites in the lattice [19], although there is evidence for their slight shifts with respect to the nominal positions of strontium [20]. The Ce$^{3+}$ ions are likely to cause substantial electrostatic forces related to simultaneous charge and chemical randomness. Such a modification brings SBN closer to the Pb-based cubic relaxors, such as PbMg$_{1/3}$Nb$_{2/3}$O$_3$ (PMN).

We performed a neutron scattering study of Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ doped by 2.5 atomic % Ce, Sr$_{0.585}$Ce$_{0.025}$Ba$_{0.39}$Nb$_2$O$_6$ (SBN-61:Ce). The crystal was grown by using a modified Stepanov process [21]. A congruently melting composition of Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ was doped by 0.1 wt.% CeO$_2$. High-quality single crystals grow even at this rather large concentration of Ce ions [22]. The ferroelectric phase transition temperature $T_c$ for this crystal was estimated from dielectric measurements to be $T_c \sim 348$ K. The precise determination of $T_c$ for doped crystals is problematic as the increase in polarisation or in linear optical birefringence turns out to be less sharp in temperature [13, 23]. Nevertheless, the $T_c \sim 348$ K is also inferred from birefringence results for a crystal with the number of Ce ions nearly identical to our sample [23].

We used a single crystal of SBN-61:Ce with dimensions of $3 \times 3 \times 1.3$ cm$^3$. The mosaic spread was within the resolution of the spectrometer. The sample was aligned in the $<0, 0, 1>\sim<1, 1, 0>$ scattering plane. This experimental geometry is sketched in figure 1. The lattice parameters of SBN-61:Ce at $T = 300$ K are $a = b = 12.43$ Å, and $c = 3.93$ Å. The crystal was mounted in a displex refrigerator that enabled the temperature to be controlled between 20 K and 500 K.

The experiments were conducted with the cold neutron 3 axis spectrometer TASP [24], situated at the end of a curved guide at the SINQ facility (PSI, Switzerland). The energy of the scattered neutrons was kept fixed to 8.04 meV and a PG filter was installed in front of the analyser. Most of the data were collected with the collimation in the horizontal plane from reactor to detector as open-80′-sample-80′, improving the energy resolution of 0.40 meV. Some data were taken with a tighter collimation: open-20′-sample-20′, improving the resolution to 0.2 meV.

III. Results

III.A. Low-energy excitations across $T_c$

Figure 2 shows the neutron spectra for a wave-vector transfer $Q = (0.35, 0.35, 2)$ at temperatures above, near and below the phase transition $T_c \sim 348$ K. These data show two peaks. One of them is centered at zero energy transfer, while the other one is an inelastic peak. The inelastic peak shifts towards higher energies and broadens at higher wavevector transfers $Q = (q, q, 2)$. Qualitatively, the most pronounced changes in these spectra are the significant variations in elastic intensity and a reduction in the width of the inelastic peak on decreasing the temperature. No intensity that could be associated with a
The stiffness $\omega$ of the IDS do not the peak intensity. The data were taken with collimation as open. The stiff-ness of the TA phonon across the $Q=(0.35, 0.35, 2)$ temperature range. One should therefore conclude that the critical

dynamics in SBN-61:Ce is concentrated at low energies.

We now describe the temperature evolution of the compo-nents in the low-energy neutron spectra from SBN-61:Ce. The parameters of the DHO function used to approximate the TA phonon are shown in figure 3 as a function of wavevector for several temperatures. As is clearly seen in figure 3(A) the stiffness of the TA phonon across the $T_c$ does not change within the errors. No scattering that could be associated with an optic phonon was detected. This suggests that there is no softening of a low-energy optic branch. In other words, we do not find any evidence for a noticeable decrease in the frequencies of the phonons on approaching the transition temperature. It might happen, however, that such a softening of an optic phonon is not directly observed in neutron spectra due to the unfavourable structure factor around a particular reciprocal lattice point. This hidden softening could still significantly distort the lineshape of the TA phonon and thus change its structure factor and/or damping [29, 30]. As follows from the tempera-ture evolution of these parameters shown in figures 3(B) and (C) this is not the case in SBN-61:Ce. The (squared) structure factor of the TA phonon is fairly temperature-independent. The damping of the TA phonon simply decreases towards lower temperatures; this behaviour is just as expected for a weakly anharmonic system. The energy resolved diffuse scattering is weak, as can be seen from figure 2. As a function of wavevector the intensity and the width $\Gamma_{IDS}$ of the IDS do not show any regular dependence. The intensity of the IDS just gradually diminishes at lower temperatures.

Altogether, our results show that energy-resolved compo-nents in the neutron spectra of SBN-61:Ce do not play an impor-tant rôle in the ferroelectric phase transition of this crystal. As is obvious from figure 2 the most noticeable changes in the spectra are in the elastic, limited in energy by the spectrometer resolution, component. At $Q=(0.35, 0.35, 2)$ the peak intensity of the EDS varies by a factor 3 in the considered tempera-ture range. One should therefore conclude that the critical dynamics in SBN-61:Ce is concentrated at low energies.
III.B. Shape of neutron diffuse scattering from SBN-61:Ce

SBN-61:Ce is a considerably disordered crystal. In such a case there might be several equally important sources of EDS. To get a hint on the origin of the EDS that showed a strong temperature dependence we analyse the distributions of the EDS from SBN-61:Ce near several Bragg peaks above and below the ferroelectric phase transition. Corresponding false colour plots for the data taken at $T = 375$ K and at $T = 300$ K are shown in figure 4. Around the (0,0,2) and the (1, 1, 2) Bragg positions at $T = 375$ K the EDS in the form of lines extended along the $<1, 1, 0>$ direction is observed. However, no similar EDS is detected around the (1, 1, 1) and (3, 3, 0) Bragg peaks. This shape of the EDS is generally associated with pronounced quasi-1D ionic correlations along the ferroelectric $c$ axis. Indeed, in the latter case a diffuse scattering would form the planes (sheets) in the reciprocal space that are orthogonal to the direction with pronounced correlations. In the geometry of our experiments these sheets appear as sharp lines perpendicular to the $c$ axis. A similar distribution of the EDS is generally associated with the complex pattern of the ionic displacements along the $c$ axis. Let us compare the EDS intensities taken at $T = 375$ K and at $T = 300$ K along the $<1, 1, 0>$ direction around these three Bragg positions (See figure 4). The reduction in the EDS intensity on changing from the (0,0,2) to the (1,1,2) point is a factor of 10 and the intensity around the (1, 1, 1) peak in turn is much weaker. As the polarisation factor $\gamma_q^2$ stays nearly the same for the (0, 0, 2) to the (1, 1, 2) positions (see table 1), the difference in the EDS intensity is likely due to the complex pattern of the ionic displacements.

At $T = 300$ K, in the ferroelectric phase, the EDS becomes much more condensed underneath the Bragg peaks. This is easily observed in the data collected near the (0, 0, 2) Bragg peak (see figures 4(a) and (b)). The nearly temperature-independent halo around the (0, 0, 2) position is very weak as compared to the EDS. This scattering is likely due to the

Figure 3. (A)–(C) Wavevector dependence of the parameters of DHO inferred from the data taken in the vicinity of the (0,0,2) Bragg peak at several temperatures. (D) A dispersion curve of the TA phonon obtained from simple fits by a Lorentzian. The apparent difference in the quality of the results shown in figures 3(A) and (D) is due to a broader range of parameters spanned by the dispersion curve plotted in figure 3(D). The error bars for the parameters shown in figures 3(A) and (D) are comparable and amount to $\sim 2.5\%$ on average. The lines in the panel (C) are the fits to the phenomenological expression $\gamma_q(T) = \gamma_q(0) - q^2 + \gamma_q(0) - q^4$ and serve as a guide to the eye. rlu stands for reciprocal lattice unit.
contribution from acoustic phonons to the elastic intensity. Importantly, no additional asymmetric in wavevectors component in diffuse scattering has been detected in the vicinity of the reciprocal lattice positions. This means that possible Huang scattering in SBN-61:Ce is negligible. As the maps of the elastic neutron scattering were collected around the Bragg peaks with different parities in (H, K, L), we conclude that the rather high doping level by Ce did not cause considerable excessive strain in the crystal. Thus, the temperature-dependent EDS originates from chain-like ferroelectric correlations. The behaviour of this EDS in paraelectric (section III.C) and ferroelectric (section III.D) phases is considered below.

III.C. Critical scattering in the paraelectric phase

The elastic neutron scattering was studied as a function of temperature by performing scans along the <q, q, 0> direction across the (0, 0, 1) and (0, 0, 2) Bragg peaks. This choice was dictated by the following reasons. Spontaneous polarisation in SBN:Ce appears along the c axis, thus the geometric condition for a transverse geometry is best fulfilled for the <q, q, 0> direction with respect to the (0, 0, L)-type Bragg peaks. Also, the neutron structure factors for the (0, 0, 1) and (0, 0, 2) Bragg peaks differ strongly. Probing simultaneously the scattering around these two reflections ensures reliable data collection and parametrisation.

Figure 5 shows elastic neutron scans taken at several temperatures above the ferroelectric phase transition. These scans consist of two obvious components. The intense Bragg peak, whose width is determined by experimental resolution, and less intense but extended in wavevectors diffuse scattering centered at q = 0. At higher temperatures this scattering is very broad and identifying its lineshape is difficult. On cooling towards \( T_c \) the diffuse intensity rapidly increases. This tendency is just opposite to the behaviour of the IDS discussed in section III.A. Thus, the broader component of transverse scans shown in figure 5 is elastic diffuse scattering, the EDS.

The energy width of the EDS in SBN-61:Ce could not be resolved in our experiments. This requires an assumption about its intrinsic nature; whether this diffuse scattering is caused by static or dynamic fluctuations. Following the approach used for pure SBN-61 [16], we assume the EDS is a critical scattering caused by dynamic, although very slow fluctuations. In this case, the neutron intensity can be approximated as

### Table 1. Auxiliary information on the geometry of our experiments on SBN-61:Ce.

| \( \tau \) | \( |\tau| \), Å\(^{-1}\) | \( \angle(\tau, <0,0,1>)^\circ \) | \( |Q \cdot \textbf{u}|^2 \) |
|---|---|---|---|
| 001 | 1.46 | 0 | 2.1 |
| 002 | 2.92 | 0 | 8.5 |
| 112 | 3.00 | 14 | 8.4 |
| 111 | 1.63 | 26 | 2.1 |
| 330 | 2.15 | 90 | 0 |

Note: \( \tau \) stands for a particular reciprocal point vector. \( |\tau| \) is a distance from the origin of reciprocal space to the appropriate Bragg peak. \( Q \cdot \textbf{u} \) is a square of the absolute value of the polarisation factor. For simplicity, vector \( \textbf{u} \) is normalised to unity. Mutual arrangements of the vectors are sketched in figure 1.
An inspection of Figure 5 shows the elastic scattering in SBN-61:Ce taken along the \( <1, 1, 0> \) direction above \( T_c \). The lines are the fits as described in the text. The shaded areas emphasise the Bragg peaks. Their widths are defined by spectrometer resolution and were calculated with ResLab 4.2 [28]. The data presented in figure 5 are obtained with open-20'–20'–20' collimation. The inset shows the overall intensity in the scan.

\[
I_{\text{EDS}} \sim \chi(q = 0, T) \cdot T \cdot \frac{\kappa}{\kappa^2 + q^2},
\]

where \( \chi(q = 0, T) \) is the susceptibility associated with the EDS taken at the \( q = 0 \), \( \kappa = 1/\xi \) is the inverse of the correlation length, and \( T \) is the temperature. In the range from 405 K down to ~350 K the elastic scans collected around the \((0, 0, 1)\) and the \((0, 0, 2)\) positions were fitted to a Gaussian Bragg peak together with a Lorentzian curve convoluted with the Gaussian resolution width. Examples of these fits are presented in figure 5 for \( T = 405 \) K and \( T = 365 \) K. Within this approach the susceptibility and the width of the EDS were obtained as a function of temperature.

Figure 6(a) shows the temperature dependence of the integrated intensity of the EDS scattering \( I_{\text{EDS}}/T \sim \chi(q = 0, T) \) from SBN-61:Ce measured around the \((0, 0, 1)\) and the \((0, 0, 2)\) positions. \( \chi(q = 0, T) \) probed around both Bragg reflections increases on approaching the phase transition from above. Furthermore, as discussed in sections III.A and III.B, we did not observe any other strongly temperature-dependent scattering in the vicinity of \( T_c \). Taken together these observations imply that fluctuations producing the EDS account for the dielectric anomaly associated with the ferroelectric transition of SBN-61:Ce.

On approaching the phase transition one expects not only the diverging susceptibility of critical fluctuations, but also condensing of the associated intensity at the propagation vector. The width of the EDS from SBN-61:Ce, however, does not decrease to zero near \( T_c \), as shown in figure 6(b). In contrast to the susceptibility, the width of the EDS probed in the vicinity of the \((0, 0, 1)\) and the \((0, 0, 2)\) Bragg peaks exhibits a dissimilar temperature evolution. At higher temperature their ratio is about 1.3, but progressively becomes unity on approaching \( T_c \). This unusual behaviour is possibly related to a complicated pattern of ionic displacements producing neutron EDS from SBN-61:Ce.

III.D. Diffuse scattering in the ferroelectric phase: domain Walls

The distribution of neutron elastic scattering in the ferroelectric phase of SBN-61:Ce is very similar to that in the paraelectric phase (see section III.B). However, below \( T_c \) the EDS becomes considerably sharper in wavevectors and much more intense near \( q = 0 \). Thus, separating the EDS from the Bragg peaks in the ferroelectric phase is more complicated. Furthermore, just below \( T_c \) there are two contributions to the EDS. First, below \( T_c \) the sample breaks up into ferroelectric domains. This causes intense diffuse scattering from the domain walls. In addition, just below \( T_c \) there should be a contribution to the EDS caused by the critical scattering. The integrated susceptibility of the DS follows the real part of dielectric permittivity \( \varepsilon' \) closely for \( T > T_c \). Similar behaviour is expected for \( T < T_c \). An inspection of figure 6(a) suggests that the contribution from the critical scattering should become negligible for \( T \lesssim 300 \) K. We can expect the EDS from SBN-61:Ce to be dominated by scattering from the ferroelectric domain walls below this temperature. With these assumptions we concentrate on the treatment of the EDS in the temperature range 150–300 K.

To analyse the lineshape of the EDS we need to recall the results on the diffuse scattering from the domain walls [16, 29, ...]
For uniaxial ferroelectrics an analytical expression for the scattering intensity was obtained under the following assumptions. If no electric field is applied to a specimen, below $T_c$, the crystal breaks into the ferroelectric domains. They are expected to be macroscopically large along the direction of spontaneous polarisation. These domains are likely to form the so-called 180° structure with their normals orthogonal to the unique ferroelectric axis $c$. It is assumed that the spontaneous polarisation is proportional to the ionic displacement field along $c$ as $P_c \sim u(x)$, where $u(x) = u_0 \tanh(x/\lambda)$. This leads to the one-dimensional scattering function $S(q_x) = \frac{D \lambda^2}{q \sinh^2(q \lambda/2)}$. Extending this result to three dimensions in wavevector space requires further approximations. Namely, the domain walls are assumed to be randomly arranged in the $(a)-(b)$ plane of the crystal and so interference effects are negligible in the scattering. Taken together, these arguments lead to the following scattering function for the diffuse scattering induced by the domain walls:

$$S(Q, \omega) = D \frac{1}{|q|} \frac{\lambda^2}{\sinh^2(q \lambda/2)} \frac{\kappa_z}{q^2 + \kappa_z^2} \cdot \delta(\omega)$$

(3)

here $\lambda$ is a half of the average thickness of the domain walls, $D$ is a constant proportional to their density, the reduced wavevector $q$ runs from the respective Bragg peak, $q = Q \pm \tau$. $\delta(\omega)$ accounts for the elastic nature of the scattering. A Lorentzian with small $\kappa_z$ accounts for the large size of the domains along the unique ferroelectric direction. In our fits $\kappa_z$ was fixed to the value 0.0005 rlu to balance the calculation time and numeric precision of the modeling.

The scattering function given by equation (3) was convoluted with the spectrometer resolution function by using ResLib4.2 library [28] and a constant background was added. The best fit results for SBN-61:Ce are shown in figure 7. The model equation (3) reproduces the data taken around the Bragg peaks (0, 0, 1) and (0, 0, 2) in the temperature range 220–300 K. The fitted curves describe the data spanning more than 3 orders of magnitude in intensity. Such an agreement further validates our approach. The domain wall density (which is just the scale factor in the model) was found to be temperature-independent. The temperature dependence of the width of the ferroelectric domain walls is shown in figure 8.

In the addressed temperature range the width probed by neutron scattering near the (0, 0, 1) and (0, 0, 2) Bragg peaks is the same. This suggests that the pattern of displacements remains unchanged through the domain walls. The width of the domain walls in SBN-61:Ce is close to the lattice constant and is similar to the values found in other uniaxial ferroelectrics [29, 35–37].

IV. Discussion and conclusions

As a rule, phase transitions are related to a condensation of low-energy modes. In particular, on approaching the ferroelectric phase transition the transverse polarisation fluctuations should slow down and the associated susceptibility diverges.

The assumptions on the shape of the ferroelectric domains in SBN are in agreement with observations made by piezoresponse force microscopy [37].

Figure 7. The EDS below the phase transition in SBN-61:Ce shown together with fits to the domain wall model. The data are shown by blue circles and the fits to equation (3) are given by red lines. The insets emphasise the low-intensity parts of the scans. The shaded areas denote the portion of the scans where the intensity would be influenced by the Bragg peak. These data points were not considered in the fits. The data were taken with collimation as open-20°-20°.

Figure 8. The width of the ferroelectric domain walls in SBN-61:Ce in the temperature range 150–300 K. The vertical lines denote the phase transition $T_c$ and the temperature where we assume the contribution from critical scattering to the EDS becomes negligible. The horizontal line denotes the value of the lattice constant.
The spontaneous polarisation appears in SBN crystals along the c axis. Thus, in pure and Ce-doped SBN-61 [16] we studied in details the low-energy transverse modes propagating along the <q, q, 0> direction. Getting a few steps forward, the scattering properties of the two crystals are very similar.

We were able to model the intensity of the TA phonon in Ce-doped SBN-61 assuming a DHO lineshape and sinusoidal dispersion relation (see section III.A). This approach allows for more reliable estimates of the damping, but is different from that previously employed in other studies of SBN [16, 35]. To compare more closely the TA dispersion in SBN-61:Ce with that in SBN crystals of other compositions the phonon frequencies were also obtained via simple Lorentzian fits. These results are shown in figure 3(D) for several temperatures.

The critical scattering observed in the paraelectric phase of Ce-doped SBN-61 is consistent with the chain-like correlations of ionic displacements. However, the displacements pattern has additional modulation leading to an unexpected difference in width of the EDS probed around different Bragg positions, but along the same direction in reciprocal space. On approaching the transition temperature this width of the EDS decreases and the associated susceptibility increases.

In the ferroelectric phase the EDS from Ce-doped SBN-61 is produced by the domain walls. The width of this DW inferred from the data taken around the (0, 0, 1) and (0, 0, 2) Bragg reflections is essentially the same and well below $T_c$ does not depend noticeably on the temperature.

In summary we compare the properties of unmodified and Ce-doped SBN-61, as seen by neutron scattering. The dispersion curve of the TA phonon propagating along the <q, q, 0> direction does not change on doping by Ce [16, 35]. The critical scattering observed in the paraelectric phase of Ce-doped SBN-61 is similar to that one from undoped SBN crystals and in both cases its distribution is consistent with the chain-like correlations of ionic displacements. An additional charge disorder introduced by Ce$^{3+}$ ions does not lead to a noticeable change in ferroelectric DW thickness, which was found to be ~10 Å for both crystals. Despite the significant differences in macroscopic properties, the lattice dynamics, critical dynamics and domain structure of Ce-doped SBN-61 surprisingly is almost identical to that of SBN-61.

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References

[1] Blinc R 2011 Advanced Ferroelectricity (Oxford: Oxford University Press)
[2] Levanyuk A P and Blinc R 2013 Phys. Rev. Lett. 111 097601
[3] Takenaka H, Grinberg I and Rappe A M 2013 Phys. Rev. Lett. 110 147602
[4] Sherrington D 2013 Phys. Rev. Lett. 111 227601
[5] Guzmán-Verri G G, Littlewood P B and Varma C M 2013 Phys. Rev. B 88 154106
[6] Cowley R A, Gvasaliya S N, Lushnikov S G, Roessli B and Rotaru G M 2011 Adv. Phys. 60 229
[7] Phelan D et al 2014 Proc. Natl Acad. Sci. 111 1754
[8] Manley M E, Lynn J W, Abernathy D L, Specht E D, Delaire O, Bishop A R, Sahul H and Budai J D 2014 Nat. Commun. 5 3683
[9] Guzmán-Verri G G and Varma C M 2015 Phys. Rev. B 91 144105
[10] Jamieson P B, Abrahams S C and Bernstein J L 1968 J. Chem. Phys. 48 5048
[11] Dec J, Kleemann W, Woike T and Pankrath R 2000 Eur. Phys. J. B 14 627
[12] Granzow T, Woike T, Wöhrlecke M, Imlau M and Kleemann W 2004 Phys. Rev. Lett. 92 065701
[13] Kleemann W, Dec J, Shvartsman V V, Kutzjak Z and Braun T 2006 Phys. Rev. Lett. 97 065702
[14] Granzow T, Dörfler U, Woike T, Wöhrlecke M, Pankrath R, Imlau M and Kleemann W 2002 Europhys. Lett. 57 597
[15] Borisov S, Okuneva N, Vakhrushev S, Naberezhnov A, Volk T and Filimonov A 2013 Phys. Solid State 55 334
[16] Gvasaliya S N, Cowley R A, Ilevela L I, Lushnikov S G, Roessli B and Zheludev A 2014 J. Phys.: Condens. Matter 26 185901
[17] Ondrejkovic P, Kempa M, Kulda J, Frick B, Appel M, Combet J, Dec J, Lukasiewicz T and Hlinka J 2014 Phys. Rev. Lett. 113 167601
[18] Shannon R D 1976 Acta Crystallogr. A 32 751
[19] Chemaya T, Maksimov B, Verin I, Ilevela L and Simonov V 1998 Crystallogr. Rep. 43 986
[20] Wingermühle J, Meyer M, Schirmer O F, Pankrath R and Kremer R K 2000 J. Phys.: Condens. Matter 12 4277
[21] Ilevela L 2009 Bull. Russ. Acad. Sci. Phys. 73 1338
[22] Ilevela L, Volk T, Isaakov D, Gladkii V, Polozkov N and Lykov P 2002 J. Cryst. Growth 237–9 700
[23] Kleemann W, Dec J, Lehnen P, Woike T and Pankrath R 2000 AIP Conf. Proc. 535 26
[24] Semadeni F, Roessli B and Boni P 2001 Physica B 297 152
[25] Buixaderas E, Savinov M, Kempa M, Veljko S, Kamba S, Petzelt J, Pankrath R and Kapphan S 2005 J. Phys.: Condens. Matter 17 653
[26] Buixaderas E, Gregora I, Hlinka J, Dec J and Lukasiewicz T 2013 Phase Trans. 86 217
[27] Giles N C, Wolford J L, Edwards G J and Uhrin R 1995 J. Appl. Phys. 77 976
[28] Zheludev A 2001 resLib4.2
[29] Cowley R A, Axe J D and Iizumi M 1976 Phys. Rev. Lett. 36 806
[30] Cowley R 1980 Adv. Phys. 29 1
[31] Krivoglaz M 1996 Diffuse Scattering of X-Rays and Neutrons by Fluctuations (Berlin: Springer)
[32] Bruce D A 1981 J. Phys. C: Solid State Phys. 14 5195
[33] Andrews S R and Cowley R A 1986 J. Phys. C: Solid State Phys. 19 615
[34] Collins M A, Blumen A, Currie J F and Ross J 1979 Phys. Rev. B 19 3630
[35] Prokert F and Schalge R 1978 Phys. Status Solidi b 87 179
[36] Currie J F, Blumen A, Collins M A and Ross J 1979 Phys. Rev. B 19 3645
[37] Peng J and Bursill L A 1982 Phil. Mag. A 45 911