APPLIED SCIENCES AND ENGINEERING

Response to comment on "Giant electromechanical coupling of relaxor ferroelectrics controlled by polar nanoregion vibrations"

M. E. Manley[1]*, D. L. Abernathy[2], A. D. Christianson[2], J. W. Lynn[3]

Gehring *et al*. argue that a splitting observed by us in the transverse acoustic (TA) phonon in the relaxor ferroelectric $Pb[(Mg_{1/3}Nb_{2/3})_{1-x}Ti_x]O_3$ with x = 0.30 (PMN-30PT) is caused by a combination of inelastic-elastic multiple scattering processes called ghostons. Their argument is motivated by differences observed between their measurements made on a triple-axis spectrometer and our measurements on a time-of-flight spectrometer. We show that the differences can be explained by differences in the instrument resolution functions. We demonstrate that the multiple scattering conditions proposed by Gehring *et al*. do not work for our scattering geometry. We also show that, when a ghoston is present, it is too weak to detect and therefore cannot explain the splitting. Last, this phonon splitting is just one part of the argument, and the overall conclusion of the original paper is supported by other results.

INTRODUCTION

The difference between the triple-axis (BT7) measurements by Gehring *et al.* (*1*) on PMN-29PT and time-of-flight wide angular-range chopper Spectrometer (ARCS) measurements by us on PMN-30PT (*2*), which is a blurring of the transverse acoustic (TA) mode splitting, can best be explained by the angular resolution difference between instruments. For the ARCS spectrometer, the out-of-plane angular resolution is ±0.3° compared to ± 2° for BT7. This higher angular resolution on ARCS helps in resolving some features, including phonon splitting. Figure 1 shows what happens when data used to make Fig. 1F in (*2*) is integrated in the out-of-plane direction by an amount that is equivalent to the relaxed out-of-plane **Q** resolution of the BT7 spectrometer [L = [−0.14, 0.14] reduced lattice units (rlu)]. As shown in Fig. 1, this blurs the apparent splitting in the TA mode (main peak near 5 meV) relative to a tighter **Q** resolution integration (L = [−0.025, 0.025] rlu), and this occurs for both **Q** = [1.775, −2.225, 0] (Fig. 1A, away from poling direction) and **Q** = [2.225, −1.775, 0] (Fig. 1B, toward poling direction). As discussed in (*2*), there are some differences in the phonon energies parallel and perpendicular to the poling direction, but here, we see that the splitting is evident in both directions despite the small shifts in the phonon energies. The occurrence of splitting in both directions makes it unlikely to be a multiple scattering effect since these directions are not equivalent with respect to the scattering geometry.

Gehring *et al.* (*1*) propose an explanation for these differences based on inelastic-elastic multiple scattering processes called ghostons. The conditions for a ghoston are (*3*): (i) a Bragg condition $|\mathbf{k_i}| = |\mathbf{k_i} - \boldsymbol{\tau}|$ (Bragg first) or $|\mathbf{k_f}| = |\mathbf{k_f} + \boldsymbol{\tau}|$ (Bragg last) or for extinction $|\mathbf{k_f}| = |\mathbf{k_f} - \boldsymbol{\tau}|$ (Bragg last) and (ii) an excitation at $\mathbf{q} = \mathbf{Q} - \boldsymbol{\tau}$. Gehring *et al.* (*1*) propose possible extinction conditions for some $\mathbf{q} = \mathbf{Q} - \boldsymbol{\tau}$, but the Bragg constraints on $\mathbf{k_i}$ and/or $\mathbf{k_f}$ are not described. These Bragg constraints are central to determining multiple scattering and depend on instrument geometry (*4*).

Calculations that express the incident and final wave vectors for our geometry show that the conditions proposed by Gehring *et al.* (*1*) do not work. For example, the suggested Bragg last ghoston using a 5-meV phonon at $\mathbf{q} = \mathbf{Q} - \boldsymbol{\tau}$ = [0.25, −1.75, ±2] with **Q** = [2.25, −1.75, 0] and $\boldsymbol{\tau}$ = [2, 0, ±2] does not satisfy the extinction condition for our geometry. The parameters are

$$\begin{aligned} \mathbf{Q} &= [2.25, -1.75, 0]\text{rlu} \\ E &= 5\text{meV} \\ \mathbf{k_i} &= [2.2097, 0.2374, 0]\text{rlu} \\ \mathbf{k_f} &= [-0.0403, 1.9874, 0]\text{rlu} \\ \boldsymbol{\tau} &= [2, 0, \pm 2]\text{rlu} \end{aligned}$$

It follows that $|\mathbf{k_f}|$ = 1.9878 rlu and $|\mathbf{k_f} - \boldsymbol{\tau}|$ = 3.4803 rlu, which does not satisfy the Bragg last condition for extinction, $|\mathbf{k_f}| = |\mathbf{k_f} - \boldsymbol{\tau}|$ (*3*). Furthermore, this condition cannot be satisfied for any geometry for which $\mathbf{k_f}$ is in the [HK0] plane because the resulting $|\mathbf{k_f}|$ is not large enough to reach |L| = 2. The Bragg last condition is not satisfied for $\boldsymbol{\tau}$ = [4, 0, 0] either since it gives $|\mathbf{k_f} - \boldsymbol{\tau}|$ = 4.5026 rlu $\neq |\mathbf{k_f}|$.

We estimate the magnitude of potential ghostons by examining the downstream monitor on the ARCS spectrometer as a function of crystal rotation angle as Bragg scattering depletes the beam. For a single diffracted beam, this amounts to only a 1 to 2% depletion and implies that the diffracted beams responsible for ghostons are small compared to the incident neutron beam responsible for phonons. To test this, we identify a Bragg last ghoston for a 13.35-meV phonon at $\mathbf{q} = \mathbf{Q} - \boldsymbol{\tau}$ = [1.225, −2.775, ±1] with **Q** = [2.225, −1.775, 0] and $\boldsymbol{\tau}$ = [1, 1, ±1]. The parameters are

$$\begin{aligned} \mathbf{Q} &= [2.225, -1.775, 0]\text{rlu} \\ E &= 13.35\text{meV} \\ \mathbf{k_i} &= [2.2074, -0.2580, 0]\text{rlu} \\ \mathbf{k_f} &= [-0.0176, 1.5170, 0]\text{rlu} \\ \boldsymbol{\tau} &= [1, 1, \pm 1]\text{rlu} \end{aligned}$$

This results in $|\mathbf{k_f}|$ = 1.517 rlu and $|\mathbf{k_f} - \boldsymbol{\tau}|$ = 1.517 rlu, which satisfies the Bragg last condition for extinction, $|\mathbf{k_f}| = |\mathbf{k_f} - \boldsymbol{\tau}|$ (*3*). Hence, there is a ghoston extinction condition at 13.35 meV for the spectra shown in Fig. 1B. The fact that there is no identifiable dip in either spectrum at this energy confirms that the ghoston effect is relatively

[1]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA. [2]Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA. [3]NIST Center for Neutron Research, National Institute of Standards and Technology (NIST), Gaithersburg, MD 20899, USA.
*Corresponding author. Email: manleyme@ornl.gov

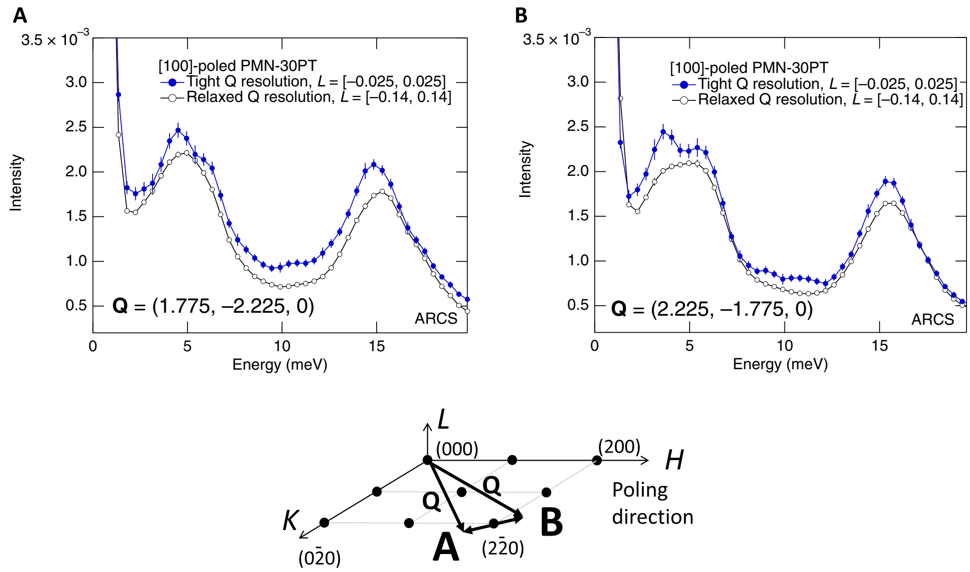

Fig. 1. Time-of-flight neutron scattering measurements of [100]-poled PMN-30%PT at 300 K. The solid blue data points show cuts made by integrating a narrow range in the out-of-plane direct [00L] in the range L = [−0.025, 0.025]. The open data points show cuts made using the same data sets but integrating over a wider range of L = [−0.14, 0.14]. (**A**) is in the direction away from the poling axis, and (**B**) is in the direction toward the poling axis (see orientation illustrated at bottom).

weak and therefore cannot explain the apparent splitting in the TA phonon. This is consistent with the original ghoston paper, which concludes that ghostons are too weak to have any effect on extracted dispersions (*3*).

Last, the TA phonon splitting interpretation of the neutron spectra is not a standalone result. It is one of several observations consistent with a coupling of the TA phonons to the polar nanoregions (*2*). For example, the observed softening of the lower branch is also consistent with bulk ultrasound measurements showing a large decrease in the transverse sound velocity along the same [110] direction with the same [100] poling (*5*). What the neutron scattering results show is that this effect extends down to the nanoscale and ultimately connects it to a coupling to the polar nanoregions. It is this shear softening that ultimately enhances the electromechanical coupling by easing the resistance to mechanical deformation.

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