Neutron Scattering Signature of Phonon Renormalization in Nickel (II) Oxide

Qiang Sun¹, Bin Wei¹,³, Yaokun Su², Douglas L. Abernathy⁴, and Chen Li¹,²,*

¹Department of Mechanical Engineering, University of California, Riverside, Riverside, CA 92521, USA
²Materials Science and Engineering, University of California, Riverside, Riverside, CA 92521, USA
³Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo 454000, China
⁴Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA

Abstract

The physics of mutual interaction of phonon quasiparticles with electronic spin degrees of freedom, leading to unusual transport phenomena of spin and heat, has been a subject of continuing interests for decades. Understanding phonon properties in the context of spin-phonon coupling is essential for engineering functional phononic and spintronic devices. By means of inelastic neutron scattering and first-principles calculations, anomalous scattering spectral intensity from acoustic phonon was identified in the exemplary collinear antiferromagnetic nickel (II) oxide, unveiling strong correlations between spin and lattice degrees of freedom that renormalize the polarization of acoustic phonon. Anomalously large neutron scattering spectral intensity from acoustic phonons observed at small momentum transfer decays with increasing temperature and is successfully modeled with a modified magneto-vibrational scattering cross section, suggesting the presence of phonon driven of spin precession. On the other hand, TA phonon intensity that are “forbidden” by the scattering geometry is observed at wide span of momentum transfer, indicating a renormalization of phonon eigenvector.

Introduction

Phonon, as the quanta of lattice vibrations, is known to strongly couple with spin degrees of freedom in a variety of magnetic materials, leading to many intriguing novel phenomena. In magnetic insulators, the large electron band gap accompanied with high energy of crystal electric field excitations prevents the direct coupling of phonon/spin system to the electronic system, whereas the transport processes are affected predominately by spin-phonon coupling. Generally, there are two most well-known spin-phonon coupling processes [1-5], that is, the modulation process and Raman spin relaxation process. The former refers to the dynamic modulation of exchange coupling between magnetic ions induced by phonons, and the latter refers to the relaxation of spin through emitting two incoherent (thermal) phonons. While the spin-phonon coupling is known to result in modifications on magnon dispersions [6,7] by the modulation process, the effect on phonon systems is less well understood, e.g., the spin exchange-driven renormalization of phonon energy is only phenomenologically characterized by a spin-phonon coupling coefficients in many magnetic systems [8-10]. On the other perspective, the interaction of magnons and acoustic phonons has been extensively studied in various magnetic systems [11-16], unveiling the formation of hybrid magneto-acoustic modes, which manipulate the acoustic phonon dispersion and deploy angular momentum to acoustic phonons [17]. However, these are constrained to the vicinity of crossing points of magnon-phonon dispersions, thus do not provide details on acoustic phonon properties for the whole branch, which can be critical in transport processes.

Antiferromagnetic (AFM) insulators have been of particular interest for applications in next-generation signal processing devices due to their ultra-low dissipation in spin transportation. The collinear AFM nickel oxide (NiO (II)), with a Néel temperature (T_N) of 523 K, is a promising candidate for spintronic applications at ambient temperature. It presents a simple face center cubic (FCC) crystal structure in the paramagnetic (PM) phase and a slight rhombohedral distortion of 0.09° deviating from 60° in the AFM phase at 5K [18]. The rhombohedral distortion angle is much smaller than that of MnO (0.72° at 5K) [18], and both are well
predicted by first principles calculations based on Hubbard-U approximations [19]. While such subtle distortion together with the asymmetry of Born effective charge on Ni^{2+} ion give rise to the splitting of two TO phonons [20,21] that were supposed to be degenerate in FCC crystal symmetry, its role on affecting acoustic phonon eigenstates remains unrevealed. In contrast to optical phonons, acoustic phonon with large group velocity play critical role in transport processes. Moreover, a sudden increase of thermal conductivity beyond T_N has been reported for NiO [22], hinting the existence of strong spin-phonon interactions and motivating our investigations on the acoustic phonon properties.

Here, we report inelastic neutron scattering (INS) experiments and atomistic simulations that demonstrate the appearance of anomalous intensity from phonon both at low Q and high Q region. INS measurements on single crystals have been proved to be a powerful probing technique in resolving phonon and magnon dispersions because they have different momentum transfer (Q) dependence, where INS intensities from coherently excited phonons (magnons) are higher (lower) at higher Q. However, with spin-phonon coupling, both phonon dispersion and intensity can be modified and “magneto-vibrational” modes, which do not follow the Q and temperature dependence of phonons, are formed [23-27]. Therefore, a theoretical simulation of phonon dynamic structure factor will shed light on identifying such modes in a system with strong spin-phonon coupling. Strong INS intensity that follows acoustic phonon dispersions is observed at low Q region, suggesting its magnetic origin. Unusual temperature dependence of such intensity is identified and associated to the temperature dependence of spin order in the crystals, indicative of strong coupling between phonon and spin. Furthermore, strong INS intensity from transverse acoustic (TA) phonons is observed in longitudinal scans, where the TA intensity were supposed to be “forbidden” by the scattering geometry. The “forbidden” intensity from TA modes increase at higher Q, showing a signature of its lattice scattering origin and suggesting renormalizations of acoustic phonon polarizations.

Experiments

Time-of-flight INS measurements are performed on single crystal NiO with the Wide Angular Range Chopper Spectrometer (ARCS) at the Spallation Neutron Source (SNS). Sample is placed in an Al foil sachet and mounted in low-background electrical resistance vacuum furnace. Four-dimensional dynamical structure factors S(Q,E) are obtained at T = 10, 300, 540, and 640 K using incident energy of 150 meV, which covers multiple Brillouin zones (BZ) and measures magnon and phonon simultaneously. Data reduction is done with MANTID [28]. The data is normalized by the proton current on target and corrected for detector efficiency by the vanadium scan. The data was sliced along high symmetry Q-directions in reciprocal space to produce two-dimensional energy-momentum views of dispersions. Since no detectable difference can be found in binning experimental data (10, 300K) with distorted rhombohedral or FCC lattice coordinates, the slight distortion in the AFM phase was neglected and the FCC crystal structure was used. Slices along high symmetry directions of [0,0,L] are shown in Fig. 1 and the results along [-H,H,0] and [-H,H,H] are shown in Fig. S1 in Supplementary Information.

First-principles phonon calculations (details in SM) based on local spin density and Hubbard-U approximation (LSDA+U) exhibits good agreements with measured phonon dispersions. The relaxed rhombohedral structure with a distortion angle of 0.15° , which is larger than the experiment value in Ref [18], was used in phonon dispersion calculations. The interatomic force constants, phonon polarization vectors, band structure and dynamical structure factors S(Q,E)_{phonon} are obtained using Phonopy [29] and our own code. Magnon dispersions and dynamical structure factor S(Q,E)_{magnon} are calculated using the Heisenberg model that includes up to second nearest neighbor exchange interactions as implemented in SpinW [30]. The exchange interactions of J_{NN} = -1.4 meV and J_{NNN} = 19.5 meV are obtained by fitting of the INS results. The total S(Q,E) was evaluated by a weighted summation of phonon and magnon contributions and convoluted with ARCS instrumental resolution. The calculated total scattering function shows excellent agreement with the experimental one, as shown in Fig. 1a,b and Supplementary Information Fig. S1.
The measured and calculated dynamic structure factor, $S(Q, E)$, of NiO at 10 K. (a, d) The dynamic structure factor of NiO measured by INS on ARCS, SNS along the [0, 0, 1] direction in the reciprocal space. The intensity is integrated over ±0.1 (r.l.u) along perpendicular axes and scaled by multiplying $E$. (b) Simulation of phonons and magnons with the same $Q$ integration ranges and instrument resolution function. Both experimental data and theoretical calculations are plotted on logarithmic scale. (c) Calculated phonon and magnon dispersion along $\Gamma - X$ with BSW notation [31] for phonon and magnon [32]. (e) Constant $Q$ cuts at various equivalent $Q$ points, labelled by red dashed lines in (d).

Results and Discussion

In Fig.1a, pronounced spectral intensity below 45 meV is observed in the first BZ along [0, 0, 1] in AFM phase. Such intensity follows the dispersion of acoustic phonons, but it does not follow the $Q^2$ dependence of coherent one-phonon INS process, as shown by the simulation in Fig. 1b. Instead, strong intensity is shown at $0 < L < 1$; the mode weakens at $1 < L < 3$ and strengthens again at $L > 3$. Such trend is visualized by constant $Q$ cuts at various equivalent reciprocal points, as presented in Fig.1d,e. This is highly unusual because phonon INS cross section is expected to be smaller at low $Q$. The anomalously strong intensity and its $Q$ dependence suggest that it cannot arise solely from lattice scattering. On the other hand, the magnetic INS cross section, which is subject to the Ni$^{2+}$ magnetic form factor, is expected to decrease with $Q$. Therefore, the intensity of this mode at $L < 4$ can be understood as a combined contribution from magnetic and lattice scattering processes, where magnetic scattering gives diminishing intensity at larger $Q$ while lattice scattering increases with $Q$. This also suggests that the acoustic-phonon-like intensity in the first BZ ($L < 2$) may predominantly originate from magnetic scattering. For simplicity, such modes will still be referred as phonon modes.

Interestingly, strong spectral intensities from acoustic phonon modes are also observed in the first BZ along [-1, 1, 1] and [-1, 1, 0], as shown in Supplemental Materials (Fig. S1), indicating such behavior was not limited to [0, 0, 1] direction. To elucidate the appearance of anomalous phonon intensity in small $Q$ regime, volumetric views of simulated (one-phonon) and measured dynamical structure factors at 10K are shown in Fig. 2 a,c. The LA, TO, and LO phonon branches are well captured by the lattice dynamical structure...
factor calculation. Comparing the calculated intensity with experimental data at 10 K in the vicinity of (0, 0, 2) and (-2, 2, 0), one can observe that the calculated intensity of LA modes is 2 orders of magnitude stronger than that in the measurement, indicating the calculation cannot predict spectral intensities here while retaining reasonable agreement of LA phonons at the cropped slice (Fig. 2 a,c). This further indicates that such intensity in low $Q$ regime cannot be solely from one-phonon coherent scattering, hinting the magnetic origin of these modes.

Temperature dependence of such spectral intensity is also consistent with the proposed magnetic origin. At elevated temperature, both static and dynamic correlations of magnetism are weakened by increased thermal fluctuations. Indeed, the magnetic INS cross section is directly related to the thermal average of spin correlations (Supplemental Materials Eq. 3). Hence, it is natural to expect such anomalous intensities to weaken with increasing temperature if they have magnetic origin. On the other hand, INS spectral intensities from phonon lattice scattering are supposed to increase at high temperatures because, at low $Q$ regime, the Debye-Waller factor contribution is trivial and the temperature-dependent lattice scattering cross section is only subject to the Bose-Einstein statistics (Supplemental Materials Eq. 1). As shown in Fig. 2 a,b by the cropped cross section, the measured INS spectrum in lower order BZs reveals dramatic weakening at elevated temperature. Such trend can also be observed along [0, 0, 1] and [-1, 1, 0] directions in lower order BZs (Fig. 2 a,b) and powder INS measurement (Fig. S3).

**FIG 2** Anomalous temperature dependence of phonon intensity reveals strong coupling between acoustic phonon modes and spin. Volumetric view of the measured (a) (10 K), (b) (640 K), and calculated (c) (10 K) lattice scattering spectral intensity of coherent one phonon scattering in the (110) plane near $Q=(0,0,0)$. Black lines indicate the limits of the cropped cross section. The intensity is in arbitrary unit. The spectral intensity of experiment data and calculation has been rescaled by multiplying $E$. (d1-d4) 1D spectral cuts at $L = 1, 3, 5, 7$ (r. l. u.) with $Q$ integration ranges of $\pm 0.2$ on perpendicular directions. Symbols and colored curves represent experimental data and Lorentzian fits at 10, 300, 540, and 640 K. (e1-e4) Temperature dependence of mode intensity from Lorentzian fits of phonon modes at equivalent BZ boundaries along [0,0,1]. There is no magnetic Bragg peak along this direction, so that the spectral weights from magnons are negligible comparing to that of phonons in the experiment data below 40 meV and the peak areas represent the intensities of TA and LA. Error bars indicate fitting errors.
To analyze the temperature dependence of spectral intensities in detail, integrated mode intensities are extracted from Lorentzian fitting of S(E) cuts at equivalent BZ boundaries along [0, 0, 1] direction. As shown in d1,e1, both TA and LA intensities decrease with increasing temperature at \( Q = (0,0,1) \). Again, this is unexpected because such descending trend cannot be explained by phonon scattering. More importantly, the spectral intensities of acoustic modes at (0,0,1) is stronger in AF phase and the intensities drastically decrease from 300 to 540 K through the phase transition temperature. This further suggests the anomaly is related to magnetic order and the temperature trend can be explained by the weakening spin correlation due to thermal fluctuations.

Equally striking is the pronounced TA mode intensity observed below 30 meV at 10 K (Fig. 1a, Fig. 2a, Fig. S1) in a broad range of \( Q \). This is completely unexpected because the TA modes are “forbidden” according to the scattering geometry, in which the momentum transfer is perpendicular to the TA phonon eigenvectors, making the INS lattice scattering cross section zero for these modes (Supplemental Materials Eq.1). It is expected that the simulation present non-zero intensity of TA modes near Bragg points (Fig. 1b) due to finite integration width along other perpendicular \( Q \) directions. Furthermore, the intensity is not likely to result from the AFM-striction induced lattice distortion, which has been included in our phonon calculations. One plausible explanation is the formation of magnon polarons [11,12,15], which emerge from magnon-phonon hybridization and possess characteristics of both magnon and phonon. Because the magnon group velocity is much larger than that of phonon, the intersections of magnon and acoustic phonon dispersion in this system only exist in a small range of \( Q \) in the vicinity of the lattice BZ boundaries, which corresponds to magnetic BZ centers. Because such “forbidden” intensity is found not only around BZ boundaries but also elsewhere in reciprocal space without magnon-phonon crossing, such intensity cannot be solely from the magnon-phonon hybridization. Moreover, ignoring the small rhombohedral lattice distortion in AFM phase, the symmetry of all phonons at long wavelength limit is of irreducible representation \( \Gamma_{15} \) in Bouckaert-Smoluchowski-Wigner (BSW) notation [31]. Following the compatibility relations, the representation \( \Gamma_{15} \) splits into \( \Delta_1 \bigotimes \Delta_5 \) along [0, 0, 1] direction. In comparison, the magnon symmetry is of \( \Delta'_1 \bigotimes \Delta'_2 \) [32], none of which are compatible with that of the phonon modes, as shown in Fig. 1c. As a result, magnons are not expected to hybridize with acoustic modes in NiO, and the anomalous spectral intensity from acoustic branches cannot be attributed to magnon-phonon hybridization.

Also surprising is that at higher order BZs, the temperature dependence of TA and LA intensity shows different behaviors. As shown in Fig. 2 d2-d4,e2-e4, at equivalent BZ boundaries of higher \( Q \) at \( L > 2 \), TA modes behave similarly to that in the first BZ and its intensity decreases with temperature, whereas LA intensity increases monotonically. This suggests that, at higher \( Q \), the weakening of “forbidden” TA modes with temperature still shows their relation to the magnetic order. On the other hand, the temperature dependence of LA intensity at \( L = 3, 5, 7 \) agrees well with the phonon INS simulation, showing normal phonon behavior (Fig. 2 e2-e4). The different temperature dependences of TA and LA modes imply that they likely couple differently to the spin degree of freedom.

The appearance of phonon INS intensity at small \( Q \) is reminiscent of magneto-vibrational scattering (MVS), which is elastic in spin system, inelastic in phonon system, and proposed based on the assumption of no correlation between lattice and spin. While the MVS cross section has the same \( Q^2 \) dependence as coherent one phonon scattering, it also contains a term related to the magnetic form factor \( |F(Q)|^2 \), giving slight intensity at small \( Q \) (see Supplementary Material). A detailed comparison of phonon spectral intensities between the experiment and MVS models is shown in Fig. 3a,b. Clearly, the calculated lattice + MVS (see Supplementary Material) still fails for lower order BZs, suggesting the MVS model cannot satisfactorily explain the observations. On the other hand, the appearance of low-\( Q \) anomalous intensity cannot originate from neutron scattered by phonon orbital magnetic moments [33–35], because they are only on the order of nuclear magneton [34], and the corresponding magnetic cross section will be at least 6 orders of magnitude smaller than that of typical magnetic neutron scattering by electronic dipoles. Therefore, anomalous intensity at low-\( Q \) region may still originate from magnetic INS by electronic dipoles through a modified MVS process, in which lattice and spin are strongly correlated.
A magnetoelastic-correlated picture, in which the atomic displacements induced by phonon modulates the magnitude of magnetic moment (spin precession driven by phonon), may explain the phonon INS intensity anomalies at small \(Q\). Following the methodology discussed in Ref. [24], a modified MVS (mMVS) model, which contains an extra term related to the driving coefficient, was derived (see Supplementary Information). As shown in Fig. 3a, the calculated lattice + mMVS intensity can reproduce the LA intensity at lower order BZs, indicating such anomaly can be attributed to the effect of spin precession driven by phonon, and revealing strong dynamic correlations between magnetic moment and phonon induced lattice displacements. Meanwhile, one may expect such effect applies to TA phonons even though they are “forbidden” by the scattering geometry. This is because the mMVS model is finite under non-trivial driving coefficient even when \(Q \cdot e = 0\) [24]. Moreover, the driving coefficient of TA and LA modes can be similar in magnitude. This can be deduced from the non-collinear frozen phonon calculations (presented in Supplementary Information), which reveal the similar driving effect of TA and LA phonon induced atomic displacements to the magnitude of magnetic moment (Fig. S3 a). Such effect is analogous to the typical magnetoelastic coupling through dynamic modulation of exchange coupling strengths, which are of similar scales among various phonon branches by DFT calculation in NiO [36]. Henceforth, the anomalously strong TA intensity at small \(Q\) may originate from the effect of spin precession driven by phonon as well.

However, at large \(Q\), the appearance of the “forbidden” TA intensity cannot be modeled by mMVS. This is because the mMVS cross section is subject to \(|F(Q)|^2\), and will approach zero at large \(Q\). In fact, similar “forbidden” phonon modes have been observed in Fe\(_{35}\)Ni\(_{65}\) [23,24] by INS. Despite the success of mMVS model in explaining the “forbidden” TA modes in Fe\(_{35}\)Ni\(_{65}\), the observed \(Q\)-dependence is completely different in NiO. While in Fe\(_{35}\)Ni\(_{65}\) such mode shows a decrease in intensity at higher \(Q\), our measurement presents an ascending trend, as can be seen in Fig. 3 b. Clearly, the lattice + mMVS model still fails for TA modes at large \(Q\) (\(L > 5\)). In contrast, the experimental intensity from LA modes at \(L > 5\) was successfully reproduced the lattice scattering simulation. This indicates the LA intensity at larger \(Q\) is predominantly from lattice INS by phonons and is consistent with temperature dependent analysis above. The gigantic discrepancy between experiment intensity from TA modes and lattice scattering simulation indicates that such “forbidden” TA modes are related to the spin-phonon coupling beyond the scope of mMVS model.

In the case of NiO, the “forbidden” TA intensity at \(L > 5\) must result from lattice scattering instead of magnetic scattering, because magnetic INS always weakens towards the increase of \(Q\), following the magnetic form factor. From the experiment data shown in Fig. 1a, magnon spectral intensity decrease with the increase of \(Q\) and vanish at \(L > 5\), which is consistent with the calculated magnetic form factor (Fig. S6). It should be noted that the lattice counterpart of MVS (neutrons create or annihilate magnetic excitations via lattice scattering) can be safely ignored because the hyperfine coupling between the nuclear and electronic moments are weak [37]. Therefore, the “forbidden” TA phonon intensity must originate from lattice INS. The scattering intensity of this mode may have similar scattering origin as the “forbidden” intensity observed in iron chalcogenides [26]. For iron chalcogenides, the “forbidden” intensity vanishes under the spin-flip channel by spin polarized INS measurements, indicating it primarily originates from lattice INS by phonons. It’s worthwhile mentioning that, such intensity cannot result from the instrument resolution or data binning (Fig. S4), as discussed in SM. If such INS intensity has pure lattice origin, a renormalization of phonon eigenvector is necessary to explain the observed “forbidden” modes.

Renormalization of phonon eigenvector is usually associated with phonon eigen-energy renormalization, which majorly comes from phonon-phonon coupling and spin-phonon coupling in magnetic insulators. To estimate the phonon energy renormalization contributed by spin-phonon coupling, calculations of quasi-harmonic approximation (based on the AFM spin configuration) are carried out to account for energy renormalization contributed by phonon-phonon coupling (see SM). Comparing experimental acoustic phonon energy at BZ boundary with calculations of quasi-harmonic approximation, one can observe the phonon energy of TA (LA) shows a 3%-4% (1%-2%) softening from 10 to 640 K (Fig. 3 c,d). Moreover, the TA phonon energy dramatically decreases above \(T_N\), showing a coincidence with the intensity change
of the “forbidden” TA modes cross $T_N$. This suggests both phonon energy and polarization are renormalized by the spin-phonon coupling.

![Graph](image)

**FIG.3** $Q$-dependence of TA and LA phonon intensity at 10 K. (a,b) The $Q$-dependent spectral intensity comparisons between measurement and simulated $S(Q,E)$ of LA and TA modes along [00L] are presented. The mode spectral intensities are obtained by subtracting a background and integrating a width of 13 meV following the calculated phonon dispersion. The red (Blue) circles represent experimental spectral intensities for LA (TA) modes. Data near Bragg points is masked. Atomic motion corresponding to TA and LA modes at BZ boundary are sketched in the insets. (c,d) The temperature-dependent acoustic phonon energy ratio at BZ boundary plotted with quasi-harmonic approximation calculations (QHA). Phonon energies are obtained from Gaussian fitting of the experiment data. Red and blue dots denote LA and TA phonon energy ratios $E(T)/E(10K)$ at temperature $T$. The error bars denote fitting errors.

Such renormalization process is at least related to two factors, i.e., the spin order and the symmetry breaking. The former is suggested by the temperature-dependent intensity variation of the “forbidden” mode, as discussed above. On the other hand, the renormalization of eigenvector needs to be related to symmetry breaking, because phonon eigenvectors are obtained by solving the dynamical matrix, which substantially follows the symmetry of the lattice. An early work suggested that the “forbidden” intensity in Fe$_{65}$Ni$_{35}$[23,24] may result from slow local orthorhombic distortions [38], which modify the dynamical matrix, thereby cause the renormalization of eigenvectors. It’s worthwhile emphasizing that, the static distortion induced by the AFM-striction cannot explain the observed “forbidden” TA modes, henceforth the symmetry breaking needs to be dynamic, as was pointed out in [26]. Therefore, the renormalization of phonon eigenvector is most likely from dynamical symmetry breaking, which is subject to the spin order.

In such context, some possible origins, including spin order induced anharmonicity of the crystal potential energy surface and electronic excitation-phonon coupling, are discussed in the following. Firstly, while the renormalization of eigenvectors can be closely related to the anharmonicity of the crystal potential energy surface [39], frozen phonon calculations show that both TA and LA modes are quite harmonic regardless of its spin configuration (Fig. S3 b). Therefore, rather than the anharmonic crystal potential induced by the static spin order, the renormalization of TA phonon eigenvectors can be related to the direct coupling
between TA phonons and spin. Secondly, renormalization of phonon eigenvectors induced by magnetoelastic coupling have been found in bulk YIG [40] and YbB$_2$ [25,41]. In both cases such effect can be related to the interaction between phonons and magnetic excitations. In YbB$_2$, the temperature dependent phonon intensity was successfully explained by the symmetry compatibility between phonons and magnetic excitations, which were assumed to be of the same symmetry as crystal field excitations. However, the symmetry of phonon and magnon are not compatible along [0, 0, 1] in the case of NiO, as mentioned previously. This triggers us to examine the symmetry compatibility between phonons and crystal field excitations. Surprisingly, in NiO, the irreducible representation of the first state of crystal field excitation is $\Gamma'_5$, and along [0, 0, 1] direction, it splits into $\Delta'_2 \otimes \Delta_5$, which is compatible with the symmetry of TA modes, $\Delta_5$. However, the first crystal field excitation state (~1 eV) cannot be excited thermally, henceforth phonons are not expected to couple with the crystal field excitations without external stimuli. To summarize, the magnon-phonon hybridization, spin-induced anharmonic crystal potential, or electronic excitation-phonon interactions are not likely to be at the origin of such effect.

Indeed, such renormalization effect may originate from direct coupling between phonons and spin order. Interestingly, this can be reflected by the in-zone intensity of TA modes (Fig. 3 b), which is maximal at BZ boundaries ($L = 1, 3, 5, 7$). The BZ boundary TA modes are non-propagating modes, whose displacement pattern shares the same spatial periodicity as the static spin order. Therefore, such renormalization effect is expected to be the most prominent at BZ boundaries because the coupling between phonons and sublattice magnetization is in phase. Above $T_N$, with the loss of long-range spin order, such renormalization effect moderates and the corresponding “forbidden” intensity decreases.

There can be two possible mechanisms that enable the direct coupling between phonons and spin order in NiO [42]. Firstly, the direct coupling between phonon orbital magnetic moment [33-35] and magnetization may renormalize phonon eigenvectors by minimizing the corresponding Zeeman energy (discussed in SM). However, such energy can be too small (<10$^{-4}$ meV) to perturb the phonon system or result in any eigenvector change. Secondly, the coupling between phonon and orbital states on the magnetic ions may be many orders of magnitude stronger than the previous mechanism [42]. Moreover, it is suggested that such renormalization effect can result from dynamic symmetry breaking induced by the interplay between phonon and electron orbit degree of freedom [26], implying perturbations to orbital states can be critical in affecting phonon characteristics. Specifically in the present case, the renormalization of phonon eigenvector can result from dynamical unit cell symmetry breaking, which may come from the coupling between phonon induced atomic displacements and the orbital state on the magnetic ion Ni$^{2+}$.

**Conclusion**

In conclusion, anomalously large spectral intensity that follows the dispersions of acoustic phonon modes is observed at low Q. The magnetic origin of such intensity is suggested by its Q and temperature dependences, which fail to be modeled with the coherent one-phonon lattice scattering. Such intensity can be successfully calculated with the modified MVS cross section, unveiling strong correlation between magnetic moment and lattice displacements induced by phonons. On the other perspective, TA phonon that is “forbidden” by the scattering geometry, is observed at entire span of Q that are accessible in the measurement. The “forbidden” intensity increase with Q, suggesting that the intensity is from lattice excitation by lattice INS. This suggests acoustic phonon eigenvectors renormalization, which result from dynamic symmetry breaking that is subjected to the spin order.

**Reference:**

[1] J. H. Van Vleck, Physical Review **57**, 426 (1940).
[2] R. D. Mattuck and M. W. P. Strandberg, Physical Review **119**, 1204 (1960).
[3] R. Orbach, Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences **264**, 458 (1961).
[4] T. Ray and D. K. Ray, Physical Review **164**, 420 (1967).
[5] A. S. Ioselevich and H. Capellmann, Phys Rev B Condens Matter **51**, 11446 (1995).
[6] J. Oh et al., Nat Commun 7, 13146 (2016).
[7] K. Park, J. Oh, J. C. Leiner, J. Jeong, K. C. Rule, M. D. Le, and J.-G. Park, Physical Review B 94 (2016).
[8] M. G. Cottam and D. J. Lockwood, Low Temperature Physics 45, 78 (2019).
[9] E. Aytan, B. Debnath, F. Kargar, Y. Barlas, M. M. Lacerda, J. X. Li, R. K. Lake, J. Shi, and A. A. Balandin, Applied Physics Letters 111 (2017).
[10] D. J. Lockwood and M. G. Cottam, Journal of Applied Physics 64, 5876 (1988).
[11] C. Kittel, Physical Review 110, 836 (1958).
[12] T. Kikkawa, K. Shen, B. Flebus, R. A. Duine, K. I. Uchida, Z. Qiu, G. E. Bauer, and E. Saitoh, Phys Rev Lett 117, 207203 (2016).
[13] H. Man et al., Physical Review B 96 (2017).
[14] F. Godejohann et al., Physical Review B 102 (2020).
[15] J. Li, H. T. Simensen, D. Reitz, Q. Sun, W. Yuan, C. Li, Y. Tserkovnyak, A. Brataas, and J. Shi, Phys Rev Lett 125, 217201 (2020).
[16] Y. Li, C. Zhao, W. Zhang, A. Hoffmann, and V. Novosad, APL Materials 9 (2021).
[17] J. Holanda, D. S. Maior, A. Azevedo, and S. M. Rezende, Nature Physics 14, 500 (2018).
[18] A. M. Balaguurov, I. A. Bobrikov, S. V. Sumnikov, V. Y. Yushankhai, and N. Mironova-Ulmane, JETP Letters 104, 88 (2016).
[19] A. Schrönn, C. Rödl, and F. Bechstedt, Physical Review B 86 (2012).
[20] H. Uchiyama, S. Tsutsui, and A. Q. R. Baron, Physical Review B 81 (2010).
[21] Y. Wang, J. E. Saal, J.-J. Wang, A. Saengdeeijing, S.-L. Shang, L.-Q. Chen, and Z.-K. Liu, Physical Review B 82 (2010).
[22] F. R. B. Lewis and N. H. Saunders, Journal of Physics C: Solid State Physics 6, 2525 (1973).
[23] P. J. Brown, I. K. Jassim, K. U. Neumann, and K. R. A. Ziebeck, Physica B: Condensed Matter 161, 9 (1990).
[24] P. J. Brown, B. Roessli, J. G. Smith, K. U. Neumann, and K. R. A. Ziebeck, Journal of Physics: Condensed Matter 8, 1527 (1996).
[25] P. A. Alekseev, J. M. Mignot, K. S. Nemkovski, A. V. Rybina, V. N. Lazukov, A. S. Ivanov, F. Iga, and T. Takabatake, J Phys Condens Matter 24, 205601 (2012).
[26] D. M. Fobes et al., Physical Review B 94 (2016).
[27] Z. Xu et al., arXiv preprint arXiv:1803.01041 (2018).
[28] O. Arnold et al., Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 764, 156 (2014).
[29] A. Togo and I. Tanaka, Scripta Materialia 108, 1 (2015).
[30] S. Toth and B. Lake, J Phys Condens Matter 27, 166002 (2015).
[31] L. P. Bouckaert, R. Smoluchowski, and E. Wigner, Physical Review 50, 58 (1936).
[32] A. P. Cracknell and S. J. Joshua, Mathematical Proceedings of the Cambridge Philosophical Society 66, 493 (2008).
[33] S. Park and B. J. Yang, Nano Lett 20, 7694 (2020).
[34] D. M. Jurasek and N. A. Spaldin, Physical Review Materials 3 (2019).
[35] M. Hamada, E. Minamitani, M. Hirayama, and S. Murakami, Phys Rev Lett 121, 175301 (2018).
[36] P. Maldonado and Y. O. Kvashnin, Physical Review B 100 (2019).
[37] A. T. Boothroyd, Principles of Neutron Scattering from Condensed Matter (Oxford University Press, 2020).
[38] S. Lipinski, K. U. Neumann, and K. R. A. Ziebeck, Journal of Physics: Condensed Matter 6, 9773 (1994).
[39] X. He, D. Bansal, B. Winn, S. Chi, L. Boatner, and O. Delaire, Phys Rev Lett 124, 145901 (2020).
[40] H. Matthews and R. C. LeCraw, Physical Review Letters 8, 397 (1962).
[41] A. V. Rybina et al., Journal of Physics: Conference Series 92 (2007).
[42] D. M. Jurasek, P. Narang, and N. A. Spaldin, Physical Review Research 2 (2020).