“Forbidden” phonon: dynamical signature of bond symmetry breaking in the iron chalcogenides

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Investigation of the inelastic neutron scattering spectra in $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$ near a signature wave vector $\mathbf{Q} = (1, 0, 0)$ for the bond-order wave (BOW) formation of parent compound $\text{Fe}_{1+y}\text{Te}$ [1] reveals an acoustic-phonon-like dispersion present in all structural phases. While a structural Bragg peak appears in the mode in the low-temperature phase of $\text{Fe}_{1+y}\text{Te}$, it is absent in the high-temperature tetragonal phase, where Bragg scattering at this $\mathbf{Q}$ is forbidden by symmetry. Notably, this mode is also observed in superconducting $\text{FeTe}_{0.55}\text{Se}_{0.45}$, where structural and magnetic transitions are suppressed, and no BOW has been observed. The presence of this “forbidden” phonon indicates that the lattice symmetry is dynamically or locally broken by magneto-orbital BOW fluctuations, which are strongly coupled to lattice in these materials.

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Since the discovery of high-temperature superconductivity (HTSC) in cuprates, elucidation of the connection between the electronic and lattice degrees of freedom has been of considerable interest in regard to the driving mechanisms behind HTSC. The iron-based superconductors (FeSCs) share many similarities with the cuprates; both have parent phases featuring antiferromagnetic (AFM) ordering, structural distortions, strong magnetic fluctuations and broken electronic symmetry [1–5]. Understanding the complex lattice dynamics in FeSCs is of critical importance for understanding the connection between these different orders, their relation to the superconductivity, and the connection between the two types of HTSCs. The compounds in the iron-chalcogenide series, featuring the simplest structure and the strongest electronic correlation among the Fe-HTSC, provide a good opportunity to study these dynamics.

The iron-chalcogenides $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$, with a maximum $T_c$ of $\sim 14.5$ K at optimal doping, consist of a continuous stacking of Fe square-lattice layers, separated by two half-filled chalcogen (Te,Se) layers [6–8]. Initially predicted by band structure calculations to be a metal [9], the non-superconducting parent material $\text{Fe}_{1+y}\text{Te}$ instead exhibits non-magnetic character in resistivity, indicative of charge carrier incoherence near the Fermi level at high temperatures [10–12]. Large local magnetic moments of about $4 \mu_B$, which indicate full involvement of three electronic bands, are revealed by Curie-Weiss behavior in magnetic susceptibility [11–13]; nevertheless, angle-resolved photoemission (ARPES) studies show significant spectral weight near the Fermi energy [14–16].

These electronic and magnetic properties of Fe(Te,Se) are very sensitive to non-stoichiometric Fe at interstitial sites, particularly evident in the parent compound [17–24]; at low concentrations, a first-order magnetostructural transition is observed from paramagnetic tetragonal ($P4/nmm$) to monoclinic ($P2_1/m$) with bicollinear AFM order (propagation vector $\mathbf{q} = (0.5, 0, 0.5)$) and metallic resistivity [18–21]. For intermediate 0.06 $\lesssim y \lesssim 0.12$ the magneto-structural transition splits into a sequence of transitions. Our recent neutron studies of $\text{Fe}_{1+y}\text{Te}$ in the intermediate range uncovered evidence that the lowest-temperature transition coincides with the formation of a bond-order wave (BOW), indicative of ferro-orbital order in the ground state [1], which stabilizes the bicollinear AFM order in the low-$T$ phase, common to $y \lesssim 0.12$.

In this letter, we report inelastic neutron scattering measurements on $\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x$ samples, aimed at elucidating the dynamics associated with the newly discovered BOW state. We have studied two compositions of the $\text{Fe}_{1+y}\text{Te}$ parent compound, as well as the optimally-doped superconductor $\text{FeTe}_{1-x}\text{Se}_x$ ($x = 0.45$). In all three samples, we observe an acoustic-phonon-like mode that appears to disperse out of the $\mathbf{Q} = (1, 0, 0)$ reciprocal lattice point, even in the tetragonal structure where such a Bragg peak is forbidden by crystal symmetry; only a weak reflection, manifest of the BOW state, develops at this wave vector in the parent compounds at low $T$.

At all temperatures, the mode appears to be ungapped and sharp within instrumental resolution, demonstrating it is not a conventional soft mode; furthermore, we have confirmed that it is not a result of magnetic spin-flip scattering. The presence of this phonon suggests a dynamical breaking of crystal symmetry, potentially related to the magneto-orbital BOW fluctuations in these materials.

Neutron scattering measurements were carried out using the Hybrid Spectrometer (HYSPEC) [25, 26] at the Spallation Neutron Source, Oak Ridge National Labo-
In Fig. 1, we present inelastic neutron scattering intensity maps at $\hbar\omega = 4.0(5)$ meV in Fe$_1$Te (a) and $\hbar\omega = 5(1)$ meV in FeTe$_{0.55}$Se$_{0.45}$ (b) in the $(h,k,0)$ zone, measured on HYSPEC using $E_r = 24$ meV and $E_i = 50$ meV, respectively. Panels (c) and (d) show the longitudinal and (e) and (f) the transverse line cuts of the data in (a) and (b), respectively, across the symmetry equivalent forbidden Bragg positions. The lines are Gaussian fits, revealing the longitudinal phonon velocities near $(2,0,0)$, $v_L = 36(2)$ meV/r.l.u. in Fe$_1$Te and $v_L = 44(8)$ meV/r.l.u. in FeTe$_{0.55}$Se$_{0.45}$, compared to $v = 21(1)$ meV/r.l.u. and $v = 22(2)$ meV/r.l.u., respectively, for the forbidden mode [r.l.u. is in units of $\alpha = 1.645(5)$Å$^{-1}$].

In all panels, the intensity is plotted using a linear-log color scale. The color bar at the top right indicates the intensity range from 0 to 1 in arbitrary units. The inelastic scattering intensity maps reveal unusual features near the Bragg positions, suggesting the presence of multiple scattering, not uncommon in samples of this size.

FIG. 1. (Color Online) Neutron scattering intensity along $(1,0,l)$ as a function of energy transfer $\hbar\omega$, showing the phonon dispersions from $(100)$, $(101)$ and $(102)$ at 5 K (left column) and 300 K (right column) for (a–b) Fe$_{1.06}$Te, (c–d) Fe$_{1.1}$Te, and (e–f) FeTe$_{0.55}$Se$_{0.45}$.

In Fig. 2 we show constant-energy inelastic data covering a large region of the $(h,k,0)$ plane for the Fe$_1$Te and FeTe$_{0.55}$Se$_{0.45}$ samples. The data reveal the presence of ring-like contours of inelastic scattering intensity consistent with the dispersion of excitations around the forbidden Bragg peaks at the symmetry equivalent positions in different Brillouin zones, $(\pm 2n \pm 1, 0, 0)$, $(0, -2n - 1, 0)$, and $(n, n, n)$. These contours are consistent with the observed acoustic-phonon-like dispersions along $(1,0,l)$, as shown in Fig. 1. However, at 300 K the structure is tetragonal (P4/mmm) [18] and the $(100)$ Bragg reflection is symmetry-forbidden; nevertheless, a gapless, acoustic-phonon-like mode is still observed dispersing out of the forbidden Bragg position (Fig. 1b, 1d). A similar mode is also observed in superconducting FeTe$_{0.55}$Se$_{0.45}$ throughout the whole temperature range (Figs. 1e–1f). In FeTe$_{0.55}$Se$_{0.45}$ the structural and magnetic transitions observed in the parent compound are suppressed, and it is therefore tetragonal at all temperatures, so that Bragg scattering at $(100)$ is never allowed. In Figs. 1e–1f, residual elastic scattering appears to be present at $(100)$, but elastic slices (data not shown) reveal an unusual structure to the $(100)$ peak, which changes depending on the incident energy, suggesting that these elastic features are a result of multiple scattering, not uncommon in samples of this size. The observed forbidden mode appears to be ungapped within the experimental limit, $\lesssim 1$ meV, mainly imposed by the presence of this spurious double scattering.

In the three fol-

The observed forbidden mode appears to be ungapped
yielding the acoustic velocity $\pi v = 32(2)$ meV/r.l.u.

The neutron scattering cross-section of a phonon with index $\nu$ and wave vector $q$ measured at a wave vector $Q = q + \tau$ near the reciprocal lattice vector $\tau$ and at a temperature $T$,

$$\frac{d^2\sigma}{dE d\Omega} = N \frac{k_f}{k_i} |g_{Qj}|^2 \frac{h}{2\omega_j(q)} \frac{\delta(h\omega - h\omega_j(q))}{1 - e^{-h\omega_j(q)/T}}$$

is proportional to the square of the structure factor,

$$g_Q^j = \sum_j \frac{b_j}{\sqrt{M_j}} e^{-W_j(q)\epsilon_j/\nu_j} e^{iQ \cdot \mathbf{r}_j} \mathbf{e}_j(q),$$

where $e^{-2W_j(q)}$ and $b_j$ are the Debye-Waller factor and the scattering length of an atom of mass $M_j$ at a position $r_j$ in the unit cell ($N$ is the number of unit cells); $\omega_j(q) = \omega_j(q + \tau)$ and $\mathbf{e}_j(q) = \mathbf{e}_j(|q + \tau|)$ are the mode frequency and polarization vectors, which are given by the eigenvalues and the eigenvectors of the dynamical matrix [29, 30]. Due to lattice periodicity, $q$ can be constrained to the first Brillouin zone.

In the long-wavelength limit of acoustic phonons, all atoms in the unit cell move uniformly together, and the magnitude of the phonon structure factor approaches that of the static structure factor at the Bragg position, $\tau$, from which the dispersion originates, $|g_{Qj}|^2 \sim \delta q^2$. The magnitude of the phonon structure factor approaches $\omega_j(q)$ when $q$ is small, $q < 2\pi |Q|/c$, for $\leq q < 2\pi |Q|/c$, for $\leq q < 2\pi |Q|/c$, and to within zero, at most linearly in $q$, as $q \to 0$ and $h\omega_j \to 0$. This is in contradiction to what is seen in Fig. 3b, where the integrated intensity of $\chi''(Q, \omega)$ increases as $h\omega_j$ decreases. Analysis of Fe$_{1.1}$Te and Fe$_{0.06}$Te data (Figs. 1a–1d) leads to similar conclusions [28].

In order to understand the possible origin of the observed “forbidden phonon” mode, we performed LDA frozen phonon calculations, which reveal a coupling between magnetic fluctuations and a spin imbalance of neighboring Fe atoms, suggesting the possibility that a forbidden phonon mode could result from magnetic scattering induced by thermal atomic vibrations [28]. To test this, polarized neutron measurements were performed on the Fe$_{1.06}$Te sample, as shown in Fig. 4, where spin-flip (SF) and non-spin flip (NSF) scattering was measured at 300 K and 2.5 K using $^3$He polarizers and $Q \parallel B$ (guide field). With a median flipping ratio of $\sim 30$ during these experiments, our results indicate a lack of SF magnetic scattering from either the expected (200) phonon or the

\[ n = 0, 1, 2 \]

The line cuts presented in Fig. 2c, 2d, reveal both the longitudinal acoustic phonon branch near $\pm 2\pi n, 0, 0$ and the small “forbidden mode” peaks near $\pm 2\pi (1, 0, 0)$, whose intensity distribution with respect to the wave vector direction is consistent with that of a longitudinal acoustic phonon, as discussed in more detail below. However, the forbidden peak position reveals a dispersion with velocity, $v = 21(1)$ meV/r.l.u. in Fe$_{1.1}$Te and $v = 22(2)$ meV/r.l.u. in Fe$_{1.06}$Te, which is marked by (nearly) twice slower than the respective longitudinal phonon velocities, $v_L = 36(2)$ meV/r.l.u. and $v_L = 44(8)$ meV/r.l.u., and is close to that of the transverse acoustic modes. Hence, the combination of polarization and dispersion of the forbidden mode is inconsistent with that expected for a phonon in an ideal lattice. In addition, the forbidden phonon intensities at symmetry equivalent positions do not display the expected scaling with wave vector, $\sim Q^2$; one should expect that the position-normalized phonon intensity at symmetry equivalent positions, $I/Q^2$, should be constant as a function of $Q$, whereas we observe a significant decrease [28].

In Fig. 3, we present a set of line-cuts along $(1, k, 0)$ of $\chi''(Q, \hbar \omega)$ in Fe$_{0.055}$Se$_{0.45}$ at multiple energy transfers, $2$ meV $\leq \hbar \omega \leq 6$ meV (Fig. 3a), which we use for an analysis of the transverse acoustic-phonon-like dispersion near $(100)$ (Fig. 1f). Line cuts were fit to a two-gaussian function, where the gaussians were constrained to be symmetric around $k = 0$. Figures 3b–3c show the results of fitting, the total integrated intensity and dispersion, respectively. The dispersion is fit to $\hbar \omega = v |\sin(\pi k)|$, with a median flipping ratio of $\sim 30$ during these experiments, our results indicate a lack of SF magnetic scattering from either the expected (200) phonon or the
“forbidden” (100) mode. A similar polarized neutron experiment was performed on the FeTe$_{0.55}$Se$_{0.45}$ sample [28] at HB-1, yielding consistent results. While the absence of spin-flip scattering indicates the mode primarily originates from atomic displacements, this does not exclude a scenario in which the atomic displacements originate from magnetic/orbital fluctuations, as we discuss below.

A previous example of a “forbidden” phonon was found in Fe$_{65}$Ni$_{35}$ invar, in which a transverse acoustic (TA) mode is observed in a position where it is forbidden by the scattering geometry, i.e. $Q \cdot \xi(q) = 0$, cf. Eqs. (1–2) [31]. An early explanation suggested this mode could result from the breaking of the cubic crystal symmetry of the dynamical matrix by slow local orthorhombic distortions [32]. However, this mode has several properties divergent from those expected for a conventional phonon. The mode does not exhibit the expected $Q^2$-dependence, but instead shows a decrease in intensity at higher $Q$. The mode also shows a significant decrease in intensity at temperatures well above the magnetic ordering temperature ($T_c \approx 550$ K). Finally, there is a polarization ratio associated with this mode, where some contribution to the mode is structural and some is magnetic [33]. These results suggested that the scattering intensity is in part a result of magnetic scattering, but the strong coupling of the magnetic and lattice degrees of freedom also results in magnetically-driven phonon scattering.

The magnetic fluctuations in the iron chalcogenides are well known to be strong, even in the absence of long-range magnetic order in these materials [34, 35], and previous studies have emphasized the strong coupling between the electronic spin, orbital, and lattice degrees of freedom [1]. The location of the “forbidden” phonon, which we observe near the wave vector of ferro-orbital ordering in the parent FeTe material [1] clearly suggests involvement of the orbital degrees of freedom. This is consistent with the magneto-vibrational scenario set forth by our LDA frozen phonon calculation, similar to the scenario offered to explain the “forbidden” phonon in Fe-Ni invar, but with an additional factor of the orbital hybridization, a crucial ingredient. Specifically, fluctuations of the orbital/magnetic nature associated with the spin imbalance between Fe atoms might lead to vibrations of forbidden character [28].

Recently, another example of a “forbidden” phonon has been observed in La$_{2-x}$Ba$_x$CuO$_4$ ($x = 0.125$), where the mode has been attributed to CuO$_6$ octahedral tilt fluctuations [30]. A possible electronic coupling for the observed mode is not ruled out. It could be that an acoustic phonon-like mode near a structurally forbidden Bragg reflection is a universal feature indicative of coupling between electronic and lattice degrees of freedom in both cuprates and iron chalcogenides.

In summary, we have observed an acoustic phonon-like mode dispersing from a position where Bragg scattering is forbidden by crystal symmetry in both the non-superconducting end-member and optimally-doped superconducting member of the iron-chalcogenide family. The mode intensity does not follow the expected behavior for phonon scattering near a forbidden Bragg reflection. Frozen phonon LDA calculations suggest that this mode might originate from slow electronic magnetic/orbital fluctuations associated with Fe spin moments leading to a dynamical breaking of the crystal unit cell symmetry.

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Supplementary Material

“Forbidden” phonon: dynamical signature of bond symmetry breaking in the iron-chalcogenides

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1. Frozen phonon calculation for FeTe

Here we present the results of the frozen phonon calculations, which demonstrate strong coupling between fluctuations of the total magnetic moment localized on Fe atoms and their atomic displacements. LDA+U calculations were performed based on the magnetic super-cell, which includes 4 Fe and 4 Te atoms, corresponding to the bicollinear antiferromagnetic structure of parent compound FeTe (Fig. S1A). An effective on-site Coulomb interaction of \( U=2 \) eV and effective on-site exchange interaction of \( J=0.8 \) eV were used. In order to elucidate the relationship between iron local-spin moments and acoustic phonons, we shift Fe atoms along the – x direction, as illustrated in Fig. S1B and S3a, and compute the resulting magnetic moments on Fe and Te atoms. The results are listed in Table 1 and summarized in Fig. S2. We observe that the local magnetic moments on the Fe atoms change nearly linearly with the shift in their atomic positions. We see that a significant, up to 8% (in units of lattice spacing) shift of Fe atomic positions is associated with only a small, \( \sim \pm 0.06 \mu_B \) imbalance in the magnetic moments. From the calculations, it also appears that small anisotropic effects exist due to the out-of-plane Te atoms, and they also bear small, position-dependent magnetic moments. From here we conclude that small fluctuations of magnetic moments can result in significant nuclear displacements detectable by neutron scattering.
**Table 1.** Summary of the results of the frozen phonon calculations in FeTe.

| Atoms in SC | M (0% Shift) | M (-x) (1% Shift) | M (-x) (2% Shift) | M (-x) (4% Shift) | M (-x) (8% Shift) |
|-------------|--------------|--------------------|--------------------|--------------------|--------------------|
| Fe1         | 2.62025      | 2.61165            | 2.60654            | 2.59396            | 2.56181            |
| Fe2         | 2.62026      | 2.61205            | 2.60723            | 2.59537            | 2.55145            |
| Fe3         | -2.62025     | -2.61165           | -2.60654           | -2.59396           | -2.56181           |
| Fe4         | -2.62026     | -2.61205           | -2.60723           | -2.59537           | -2.55145           |
| Te1         | 0.00161      | 0.00221            | 0.00281            | 0.00408            | 0.00626            |
| Te2         | -0.00161     | -0.00153           | -0.00135           | -0.00119           | -0.00038           |
| Te3         | -0.00161     | -0.00221           | -0.00281           | -0.00408           | -0.00626           |
| Te4         | 0.00161      | 0.00153            | 0.00135            | 0.00119            | 0.00038            |

**Figure S1.** Schematics of the crystal structure of FeTe. The rectangular supercell corresponding to the bicollinear magnetic structure contains four different Fe and Te sites. Zig-zag lines connect the equivalent Fe sites in the static low-temperature structure.
2. Characterization of magnetic scattering in $\text{Fe}_{1.06}\text{Te}$, $\text{Fe}_{1.1}\text{Te}$ and $\text{FeTe}_{0.55}\text{Se}_{0.55}$ samples

Here we present inelastic data that provide characterization of magnetism in the different samples studied in this work, and in particular, demonstrate that the parent samples have unique compositions of excess Fe. In Fig. S3 we compare inelastic intensity at $\hbar\omega = 7.5$ meV, an energy at which a maximum in magnetic scattering is observed in the parent compounds and the magnetic resonance occurs in the superconducting material. We present slices in both the $(HK0)$ and $(HOL)$ zones, except for $\text{Fe}_{1.06}\text{Te}$ which was only measured in the $(HOL)$ zone. In both $\text{Fe}_{1.06}\text{Te}$ and $\text{Fe}_{1.1}\text{Te}$ we observe “rods” of magnetic scattering along $l$ at $\hbar \approx \pm 0.5$, with broad maxima around half-integer $l$. Weak diffuse scattering centered at $\mathbf{Q} = (0.5,0,0)$ in the $(HK0)$ zone observed in $\text{Fe}_{1.1}\text{Te}$ is a signature of ferromagnetic-square-plaquette-type correlations [see Ref. 34 in main text]. In superconducting $\text{FeTe}_{0.55}\text{Se}_{0.45}$, magnetic scattering is strongest near $\mathbf{Q} = (0.5,0.5,0)$, Fig. S3e, indicating a different type of plaquette correlations [35], and therefore only a weak diffuse “tail” signal is observed in the $(HOL)$ zone (Fig. S3f). In Figs. S3g-h, we present structural and magnetic order parameters as a function of temperature, on warming and cooling for $\text{Fe}_{1.1}\text{Te}$, and on warming for $\text{Fe}_{1.06}\text{Te}$. Samples were oriented in the $(HOL)$ zone, where $(100)$ and $(\approx 1.5,0,0.5)$ were measured on HYSPEC with a small range of sample rotation. We present the integrated intensity around these two peaks, which illustrates the ordering temperatures of the bond-order wave and antiferromagnetism.
Figure S3. (a) Schematics from LDA frozen phonon calculations illustrating correlation between Fe atoms displacement along the x direction in the $a - b$ plane and the size of the local magnetic moment. Plus (minus) symbols indicate Te atoms above (below) the Fe plane. Circles represent Fe atoms, where diameter indicates magnetic moment. (b – f) Inelastic neutron scattering maps at $\hbar \omega = 7.5$ meV, 5 K: (b), (d), and (f) show Fe$_{1.06}$Te, Fe$_{1.1}$Te, and FeTe$_{0.55}$Se$_{0.45}$ in the $(H,0,L)$ zone, respectively, and (c) and (e) show FeTe$_{0.55}$Se$_{0.45}$ and Fe$_{1.1}$Te in the $(H,K,0)$ zone, respectively. (g – h) Order parameters for Fe$_{1.06}$Te and Fe$_{1.1}$Te: integrated intensity around (g) $(1,0,0)$ and (h) $(\approx 1.5,0,0.5)$ for Fe$_{1.06}$Te (squares) on warming and Fe$_{1.1}$Te (circles) on both cooling and warming. Error bars in all figures represent one standard deviation.

3. Additional analyses of the line cuts of the data in the $(hk0)$ plane shown in Figure 2 of the main text.

Presented in Figure S4 are the line cuts of the data shown in Figure 2 of the main text, which compare the transverse velocity of the allowed phonons near $(2n,0,0)$ lattice Bragg peaks and that of the forbidden mode near $(2n + 1,0,0)$ positions in the $(hk0)$ scattering plane in Fe$_{1.1}$Te and FeTe$_{0.55}$Se$_{0.45}$. The data were fit to the two Gaussian peaks, equally offset from the center (the Bragg position). The fitted peak positions correspond to nearly identical transverse acoustic velocities in both cases. In Fe$_{1.1}$Te (a) the transverse phonon velocity is 25.5(8) meV/rlu and that of the forbidden mode is 25.5(29) meV/rlu, while these are 34(2) meV/rlu and 30(3), respectively, in FeTe$_{0.55}$.
It should be noted that velocities refined from the line cuts measured in FeTe$_{0.55}$Se$_{0.45}$ (b) with $E_l = 50$ meV are less reliable because of the significant systematic error associated with rather coarse energy resolution of this configuration. Nevertheless, the refined transverse velocity of the forbidden mode compares favorably with that obtained from the transverse scans shown in Figure 3 of the main text, $v = 31.7(15)$ meV/rlu, where data collected using $E_l = 24$ meV is shown.

Figure S4. Transverse line cuts quantifying the sound velocity near the forbidden, $(2n + 1,0,0)$, and the allowed, $(2n,0,0)$, lattice Bragg positions in Fe$_{1.1}$Te (a) and FeTe$_{0.55}$Se$_{0.45}$ (b). In all figures error bars represent one standard deviation.

Figure S5. The wave-vector-normalized intensity of phonon at symmetry-equivalent positions near allowed and forbidden Bragg peaks in Fe$_{1.1}$Te and Fe$_{1.1}$Te$_{0.55}$Se$_{0.45}$ in (hk0) zone, obtained from the line fits of longitudinal scans shown in Figure 2, (c) and (d) of the main text. Note the logarithmic intensity scale.

Figure S5 presents the wave-vector-normalized phonon intensity, $I(Q)/Q^2$ at symmetry-equivalent positions near the forbidden Bragg peaks in Fe$_{1.1}$Te, and allowed
and forbidden peaks in Fe$_{1.1}$Te$_{0.55}$ Se$_{0.45}$ in (hk0) zone, obtained by fitting the longitudinal line scans shown in Figure 2, (c) and (d) of the main text. As expected, this quantity is roughly constant for an allowed phonon near (2n, 0,0). However, it decreases markedly for the forbidden mode, even though the absolute intensity increases with $Q$. This increase, however, is noticeably slower than $\sim Q^2$ expected for an acoustic phonon at symmetry-equivalent positions in reciprocal lattice.

4. Analysis of the acoustic mode intensity in Fe$_{1.06}$Te and Fe$_{1.1}$Te and in (hk0) zone

Here we present analyses of the (100) forbidden-phonon dispersion in Fe$_{1.06}$Te, Fe$_{1.1}$Te, and FeTe$_{0.55}$ Se$_{0.45}$ in the (hk0) scattering plane similar to that in Fig. 3 of the main text. In all cases there is no decrease of the integrated $\chi''$ intensity with the decreasing energy [i.e. $Q$ moving towards (100)].

**Figure S6.** (100) mode in Fe$_{1.1}$Te in the (h0l) scattering plane. In all figures error bars represent one standard deviation.
Figure S7. (100) mode in Fe$_{1.1}$Te in the (hk0) scattering plane.

Figure S8. (100) mode in Fe$_{1.06}$Te in the (h0l) scattering plane.
**Figure S9.** (100) mode in FeTe$_{0.55}$ Se$_{0.45}$ in the (hk0) scattering plane.