

Fluctuation-enhanced electron-phonon coupling in FeSe

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The interactions among lattice, charge, and spin degrees of freedom fundamentally shape material properties. In FeSe, symmetry-breaking perturbations serve as highly sensitive probes of these couplings. Previous work has shown that defects and isoelectronic substitution can substantially alter these interactions, giving rise to additional phonon modes. In this study, uniaxial strain is employed as a tunable symmetry-breaking control parameter to probe the intrinsic lattice response in the absence of disorder. The temperature evolution of phonon excitations was examined with a temperature resolution of 3 K in the vicinity of the nemato-structural transition temperature T_s , under strain applied along the $\langle 110 \rangle$ and $\langle 100 \rangle$ crystallographic directions. A subtle asymmetry of the A_{1g}^{ph} mode appears in the unstrained crystal within a narrow temperature window around T_s , originating from the emergence of an additional mode in the fully symmetric channel. With applied strain, this feature becomes more distinctly resolved. The anomaly is attributed to modifications of the coupling between lattice and electronic degrees of freedom driven by the ordering fluctuations right above the nematic transition. These fluctuations may enhance the susceptibility of the electron-lattice system and enable two-phonon Raman scattering involving the acoustic mode near the X and R points. The presence of this two-phonon scattering depends on both the strength and the direction of the applied strain, indicating a high sensitivity of FeSe to local symmetry breaking.

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I. INTRODUCTION

FeSe is the simplest iron-based superconductor exhibiting $T_c = 9$ K at ambient pressure [1]. Superconductivity is substantially enhanced in bulk samples under pressure, with T_c reaching 36 K at 10 GPa [2,3], while thin films display even

higher T_c values of about 50 K [4,5]. Substituting 17% of Se with S raises T_c to 11 K [6], while intercalating alkali ions or molecular layers between the FeSe sheets can boost it above 40 K [7]. These results demonstrate that even small modifications of the structure can lead to substantial changes in the properties of FeSe. Consequently, since superconductivity is a highly sensitive indicator of interactions between electrons and the related excitations of the spin and lattice degrees of freedom, FeSe serves as an excellent platform for disentangling the various types of coupling mechanisms that remain under debate in iron-based systems. Among the different internal and external perturbations, the application of uniaxial stress specifically targets the interplay of the lattice with predominantly orbital degrees of freedom.

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Angle-resolved photoemission spectroscopy (ARPES) has revealed that the low-energy electronic structure of FeSe consists of three hole-like bands around the Γ point and two electron-like bands centered at M [8–13]. In the tetragonal phase, the two electron pockets at M are nearly circular and symmetry-equivalent, arising from hybridized d_{xz} and d_{yz} orbitals. Below the nemato-structural transition, this orbital degeneracy is lifted, producing pronounced anisotropic shifts and hybridization gaps near M . As a consequence, only one of the two electron bands continues to cross the Fermi level, while the other one is pushed above it, an effect commonly referred to as the “missing-electron-pocket problem” [8,10,14]. In twinned crystals, where orthorhombic domains of both orientations coexist, the ARPES intensity maps at M reveal two perpendicular, peanut- or propeller-shaped Fermi surface contours originating from the two domain variants. In detwinned crystals, where uniaxial strain aligns the domains to a single orthorhombic one, only one of these features remains visible [9], reflecting the intrinsic electronic reconstruction driven by nematic order. Simultaneously, the hole band at Γ sinks below the Fermi level and becomes anisotropic, consistent with orbital polarization between d_{xz} and d_{yz} states below T_s [8,9].

Building on these observations, an open question remains as to how the delicate balance between lattice and electronic degrees of freedom in FeSe responds to externally applied symmetry-breaking fields. The missing electron pocket at the M point and the pronounced orbital polarization indicate that the electron–phonon coupling (EPC) in this material is inherently anisotropic and strongly tied to local symmetry [15,16]. By selectively breaking the in-plane fourfold symmetry, a strain field directly interacts with the nematic order parameter and tunes the lattice potential without introducing embedded disorder. Ideally, uniaxial strain would allow us to disentangle the role of electron-electron and electron-phonon coupling in FeSe.

To address this, we employ uniaxial strain as a clean and reversible control parameter, focusing on the narrow temperature range around the nemato-structural transition, where EPC effects are expected to be strongest. By tracking the temperature evolution of zone-center phonons under strain applied along the $\langle 110 \rangle$ and $\langle 100 \rangle$ crystallographic directions, we demonstrate that the direction of the applied stress and related distortion symmetry dictate the lattice response and its coupling to electronic fluctuations.

Our measurements reveal an asymmetry between the low-energy and the high-energy wing of the A_{1g}^{ph} mode in a narrow temperature range around T_s . We analyze this effect in terms of an additional fully symmetric contribution associated with modifications of EPC near the nematic transition. The comparison with FeS and $\text{FeSe}_{1-x}\text{S}_x$ places this observation in the broader context of 11-type iron chalcogenides [17,18], where additional fully symmetric Raman features appear when phonons off the zone center are coupled by enhanced EPC. In pure FeSe, the additional contribution appears only close to T_s , suggesting that nematic fluctuations provide the relevant enhancement mechanism. Some of the vibrational properties of FeSe show the interdependence of lattice and electronic fluctuations near T_s . Altogether, FeSe emerges as a system where symmetry breaking, nematic fluctuations, and

EPC seem to be entangled, such that even subtle perturbations to the lattice or electronic structure lead to pronounced and anisotropic responses.

II. METHODS

The FeSe single crystals were grown using chemical vapor transport (CVT) [19]. The high-purity sample had a transition from a tetragonal to an orthorhombic structure at $T_s = 89.1$ K [20]. The measurements in the temperature range between 78 and 300 K were performed in a KONTI CryoVac He-flow cryostat capable of a base vacuum of 10^{-6} mbar. The sample was cleaved immediately before evacuating the cryostat.

We attached the sample to the actuators of a homemade strain cell (see Appendix E), using a two-component epoxy adhesive (Tokyo Measuring Instruments Laboratory EA-2A) developed for use in cryogenic environments (4–323 K). A schematic of the sample mounting is shown in Figs. 1(a) and 1(b). Throughout this work, all strain direction labels are defined with respect to the crystallographic tetragonal 2-Fe unit cell, with axes a and b ($b = a$). This convention is consistent with previous strain studies of Fe-based superconductors, where strain along the tetragonal $\langle 100 \rangle$ and $\langle 110 \rangle$ directions induces distortions of B_{1g} and B_{2g} symmetry, respectively [8,21]. The 1-Fe unit cell, commonly used to describe the electronic structure, is rotated by 45° with respect to the crystallographic 2-Fe cell [Fig. 1(e)]; therefore, the B_{1g} and B_{2g} labels are interchanged between the two notations [22]. In our experiment, tensile strain was applied in two configurations: along the tetragonal $\langle 110 \rangle$ direction [Fig. 1(a)], corresponding to $\varepsilon_{B_{2g}}$, and along the tetragonal $\langle 100 \rangle$ direction [Fig. 1(b)], corresponding to $\varepsilon_{B_{1g}}$.

The strain values were estimated from the response of a strain gauge glued to the body of the strain cell, while the sample itself was attached directly between the fixed and movable plates. Measurements were performed under three nominal tensile-strain conditions, denoted as zero strain ($\varepsilon_{\text{zero}}$), low strain (ε^{low}), and high strain ($\varepsilon^{\text{high}}$), corresponding to 0%, 0.13%, and 0.21% relative change, respectively. Since the strain gauge does not measure the microscopic strain directly transferred to the crystal, small deviations may arise from the epoxy layer, sample mounting, and strain-transfer efficiency. Therefore these values should be regarded as nominal strain levels. The analysis focuses on the qualitative dependence of the Raman response on strain direction and magnitude rather than on an exact determination of the microscopic strain in the sample.

Previous transport studies of FeSe showed that, for comparable $\varepsilon_{B_{2g}}$ strain, the shift of T_s is of order 1 K [23]. Since our temperature steps around T_s are 3 K, such a shift is below our experimental resolution. Thus we discuss our results relative to the unstrained T_s rather than assigning a separate T_s value for each strain configuration.

The Raman scattering experiment was performed using a Jobin Yvon T64000 triple spectrometer in back-scattering micro-Raman configuration. To achieve a resolution close to 1 cm^{-1} per pixel on the CCD detector, we used diffraction gratings with 1800 grooves/mm. In the back-scattering configuration, the incident and scattered photons propagate nearly parallel to the crystallographic c axis, and the polarizations are

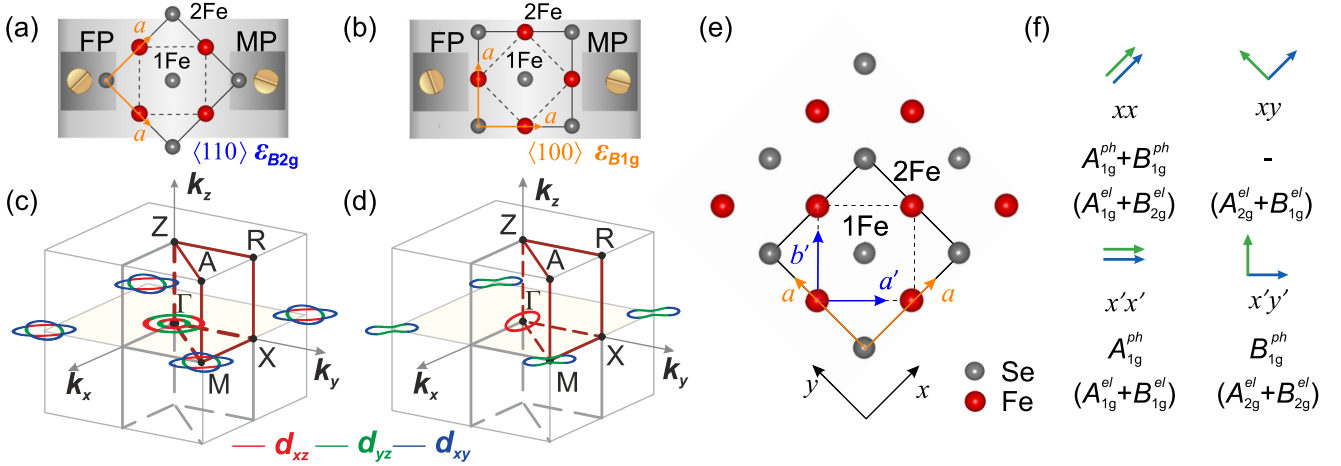


FIG. 1. (a) and (b) Diagram showing the applied strain direction, labeled with respect to the 2-Fe crystallographic unit cell. Fixed and movable sample plates integrated into a piezoelectric device are labeled as FP and MP, respectively. (c) Schematic representation of the FeSe 2-Fe Brillouin zone and Fermi surface in the tetragonal and (d) the nematic phase (adapted from Ref. [10]). (e) Crystal structure of FeSe in the ab plane ($b = a$). The solid line represents the 2-Fe crystallographic unit cell determining the symmetry of the lattice vibrations. The dashed line represents the 1-Fe unit cell with axes a' and b' relevant for electronic and spin excitations. (f) Selection rules for four main linear polarization configurations. The light polarizations x and y are aligned with the crystallographic axes (a, a). The Raman-active A_{1g}^{ph} and B_{1g}^{ph} phonons are projected in parallel $x'x'$ and perpendicular $x'y'$ scattering configurations, respectively. Electronic and spin excitations are preferably described in the 1-Fe cell, where B_{1g} and B_{2g} are swapped with respect to the xy system (symmetries in brackets).

in the ab plane as shown in Fig. 1(f). We use Porto notation for describing the light polarizations and assume that $b = a$ above and below T_s because of the very small orthorhombic distortion and $b \perp a$, $a' = (a + b)/\sqrt{2}$, and $b' = (a - b)/\sqrt{2}$ [Fig. 1(e)]. Then xx means that both incoming and outgoing polarizations $\mathbf{e}_i$ and, respectively, $\mathbf{e}_s$ are parallel to the a axis [Fig. 1(f)].

For excitation, we used a mixed-gas Ar^+/Kr^+ laser (Coherent Innova 70C) operating at the 514.5 nm emission line. The laser beam was focused using a microscope objective lens having an ultralong working distance and $\times 50$ magnification, allowing a spot size of approximately 6 μm . The power of the incoming polarized light was adjusted for each measurement to deliver 1 mW on the sample surface or 0.5 mW absorbed by the material, inducing heating of approximately 4 K. All temperatures shown here were corrected for this heating. The experimental Raman intensities were divided by the Bose-Einstein factor, yielding response functions $R\chi''(\omega)$. For describing the phonon lines, Voigt profiles (a convolution of a Lorentzian and a Gaussian profile) turned out to be superior to simple Lorentzians since the intrinsic linewidths are very small. The Gaussian and the Lorentzian profiles account for the resolution of the spectrometer (typically 1 cm^{-1}) and the linewidth of the phonon peaks, respectively.

III. RESULTS

A. Uniaxial strain, symmetry, and selection rules

The experimental geometry and the two strain configurations employed in this study are illustrated in Fig. 1 and described in the Methods section. In one configuration, the Fe-Fe bond direction is parallel to the strain direction [Fig. 1(a), B_{2g} , see Fig. 1(e) for definition]. In the other one, the strain is applied along the crystallographic a axis or at 45° with respect

to the Fe-Fe bond direction [Fig. 1(b), B_{1g} , see Fig. 1(e) for definition]. These two geometries were selected as they impose distortions of different symmetry: (i) strain along the Fe-Fe bond ($\varepsilon_{B_{2g}}$) couples directly to the nematic distortion that describes the transition of the quadratic 1-Fe unit cell into an orthorhombic one; (ii) strain along a transforms the 1-Fe tetragonal cell into a diamond or rhombohedron (B_{1g} symmetry; see Methods). This approach enables a direct comparison of different symmetry-breaking mechanisms and their impact on the lattice dynamics.

Since B_{2g} strain directly couples to the nematic order parameter, it is important to recall how the underlying electronic structure evolves across the nemato-structural transition. The corresponding Fermi surface topology above and below T_s is schematically illustrated in Figs. 1(c) and 1(d), respectively, using the 2-Fe Brillouin zone. In the tetragonal phase [Fig. 1(c)], FeSe hosts two hole-like bands centered at the Γ point and two symmetry-equivalent elliptical electron pockets at the M point (the Brillouin zone of the 1-Fe cell is twice as large and M becomes X). Below T_s [Fig. 1(d)], lifting of the d_{xz}/d_{yz} orbital degeneracy leads to an anisotropic reconstruction of the electronic states [8–10] and propellers oriented along $\mathbf{k}_x$ of the 1-Fe cell.

The selection rules governing phonon Raman scattering are dictated by the symmetry of the crystallographic 2-Fe unit cell. In our case, only in-plane polarizations (within the ab plane) are applied, as illustrated in Fig. 1(e), where solid and dashed lines represent the 2-Fe and 1-Fe unit cells, respectively. While the 1-Fe unit cell is often used to describe electronic and spin excitations, since the relevant bands and the magnetism derive from the Fe orbitals, the 2-Fe unit cell provides the appropriate framework for analyzing phonon modes.

Incident and scattered photons propagate approximately along the c axis. Under these conditions, the selection rules

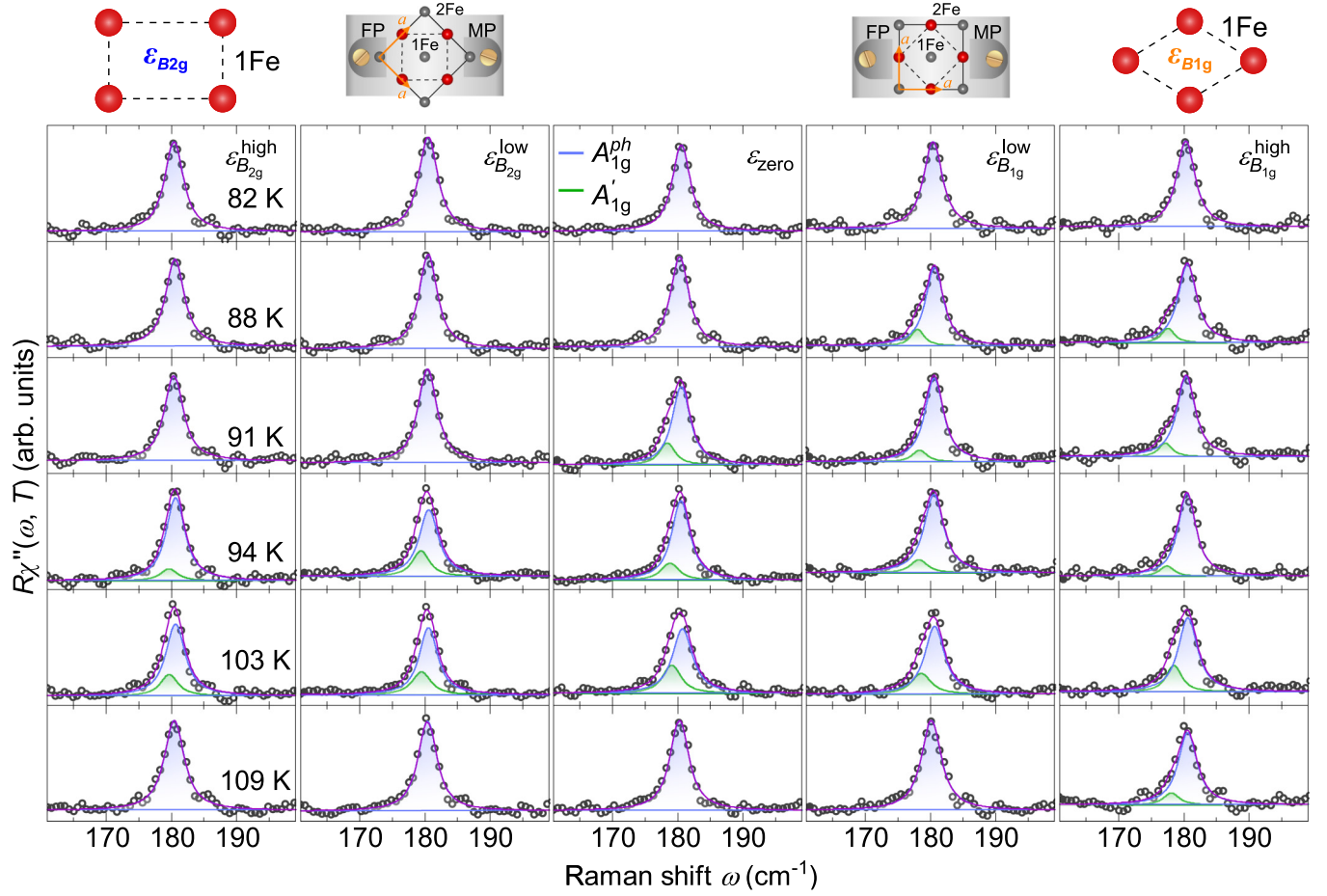


FIG. 2. Raman spectra in $x'x'$ scattering geometry. The spectra in the range of the A_{1g}^{ph} phonon line were measured as a function of strain and temperature, as indicated in the first row and column, respectively. In addition to the measurements, we described the experimental data by model fits (see text). The blue-shaded area marks the symmetry-allowed A_{1g}^{ph} mode, and the green-shaded area highlights an additional feature emerging near T_s , labeled A_{1g}' . The top panel above the spectra schematically illustrates the sample orientations for the $\epsilon_{B_{2g}}$ and $\epsilon_{B_{1g}}$ strain and the related distortions of the unit cell. The $\epsilon_{B_{2g}}^{high}$ and $\epsilon_{B_{2g}}^{low}$ denote spectra obtained under uniaxial tensile strain applied along the diagonal of the 2-Fe unit cell, while $\epsilon_{B_{1g}}^{high}$ and $\epsilon_{B_{1g}}^{low}$ correspond to spectra obtained under tensile strain along the a axis.

dictate that only A_{1g}^{ph} and B_{1g}^{ph} Raman-active modes are observable for the $P4/nmm$ (space group 129) symmetry. Fig. 1(f) shows which phonon modes are projected for a given scattering geometry. In the $x'x'$ (see Methods section) configuration, the scattering probes only the nondegenerate fully symmetric, in-phase vibration of Se atoms along the c axis (A_{1g}^{ph}) since there is no B_{2g} phonon. In the crossed $x'y'$ polarization, the observed B_{1g}^{ph} mode breaks the fourfold C_4 rotational symmetry and corresponds to the out-of-phase vibration of Fe atoms along the c axis.

B. Raman spectra of FeSe

The nemato-structural phase transition in FeSe occurs at $T_s = 89.1$ K (see Fig. 7 in Appendix A) [15,16,20], with the crystal symmetry decreasing from tetragonal $P4/nmm$ to orthorhombic $Cmma$ symmetry. The transition leaves $b = a$, but introduces a small deviation from 90° between a and b [15,18,24–26] of 0.23° , for $\delta = (a' - b')/(a' + b') \approx 0.002$ [27,28] (for the definition of a' and b' axes, see Fig. 1 and

Methods section). Although this deviation does not affect the selection rules, there are subtle spectral changes across this transition, and we performed detailed temperature-dependent Raman scattering measurements with temperature steps of 3 K in the vicinity of T_s .

We first examine the $x'x'$ spectra. Fig. 2 (spectra panels) shows the spectral region of the A_{1g}^{ph} phonon as a function of temperature and applied tensile strain. From top to bottom, we show six representative temperatures; from left to right, we show results for different strain directions and magnitudes. Specifically, the first two columns correspond to B_{2g} strain, inducing an orthorhombic distortion of the Fe plane, while the last two columns correspond to B_{1g} strain (see top panel). The spectra in the middle column represent the nominally unstrained case (ϵ_{zero}). This compilation enables a direct comparison of how distinct strain configurations affect the A_{1g}^{ph} phonon evolution with temperature. From left to right, the effects of strain become progressively more evident, illustrating its systematic impact on the phonon response (details in the Discussion section and Appendix B).

In the unstrained case (Fig. 2, middle column), we observe a change on the low-energy side of the A_{1g}^{ph} phonon, where a kink at approximately 3 cm^{-1} below the A_{1g}^{ph} line develops in the narrow range of temperatures. We label this new feature as A'_{1g} to denote its fully symmetric character in the FeSe compound (see Fig. 5). Upon cooling, the mode appears well above T_s and disappears quickly at a slightly strain-dependent temperature at or below T_s (see also Fig. 8) [20].

A similar behavior is observed for B_{2g} strain coupling directly to nematic order and enhancing the orthorhombic distortion of the Fe plane (Fig. 2, left columns). The additional A'_{1g} peak appears only at higher temperatures (94–106 K) compared to the unstrained case.

In contrast, B_{1g} strain produces a considerably different evolution (Fig. 2, right columns). For low-magnitude strain ($\epsilon_{B_{1g}}^{\text{low}}$), the A'_{1g} feature is observed over an extended temperature interval ranging from 88 to 106 K and appears as a well-defined shoulder on the low-energy side of the A_{1g}^{ph} . With increasing strain ($\epsilon_{B_{1g}}^{\text{high}}$), the feature emerges at the same temperature as for $\epsilon_{B_{1g}}^{\text{low}}$ but remains visible up to about 109 K, indicating a slight broadening of the anomaly window.

To be more quantitative, we described all spectra using Voigt profiles (see Methods section). None of the spectra in the range 88 to 106 K could be described reasonably well using a single line. The discrepancy was most evident for B_{1g} strain (Fig. 2, right columns), where the line shape directly indicates the presence of an additional peak on the low-energy side. When a second Voigt component was introduced, the fit quality improved significantly for both the unstrained and strained cases, within the corresponding temperature ranges discussed above. This observation is discussed in detail in the Discussion section, with the full analysis presented in Appendix B.

Figure 3 shows $x'y'$ Raman spectra in the energy range of the B_{1g}^{ph} mode at selected temperatures. The orthorhombic distortion ($\epsilon_{B_{2g}}$) causes the B_{1g}^{ph} phonon to shift to slightly higher energies at low temperatures, whereas rhombohedral distortion ($\epsilon_{B_{1g}}$) results in shifts of about 1 cm^{-1} toward lower energies. As the temperature increases, these strain-induced shifts progressively decrease, and the phonon energies converge toward those of the unstrained sample.

C. Temperature dependence of the phonon excitations

Using the results of the quantitative analysis, we now focus on the temperature dependences of the energies and linewidths of all detected excitations in the zero-strain regime, as well as under low and high strain conditions along the two directions considered. The extracted phonon energies and Lorentzian linewidths of the A_{1g}^{ph} and A'_{1g} modes are presented in Fig. 4, in the left and right panels, respectively. The temperature dependence of the linewidths for each strain configuration was further modeled using the standard expression for symmetric anharmonic decay [29]:

$$\Gamma_L(T) = \Gamma_L(0) \left(1 + \frac{2\lambda_{\text{ph-ph}}}{e^{\frac{\hbar\omega(0)}{2k_B T}} - 1} \right), \quad (1)$$

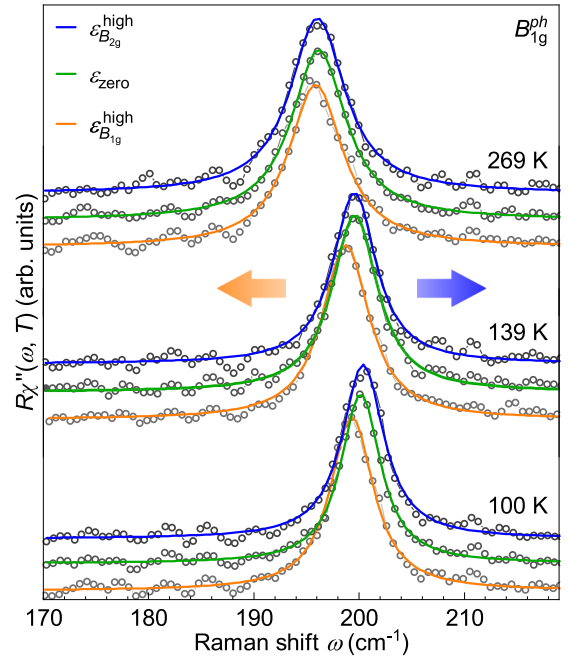


FIG. 3. Strain-dependent B_{1g}^{ph} Raman spectra at temperatures as indicated. At 269 K, strain has very little effect. Upon approaching the transition at T_s , B_{2g} strain leads to a blue shift (blue) of the phonon, while B_{1g} strain causes a red shift (orange) with respect to the unstrained case (green).

where $\Gamma_L(0)$ and $\omega(0)$ were obtained by extrapolating linewidths and energies to the zero-temperature limit, while the phonon-phonon coupling constant ($\lambda_{\text{ph-ph}}$) is a free parameter. The phonon energy $\omega(T)$ is influenced by both anharmonic decay and lattice contraction and is given by the following expression [30]:

$$\omega(T) = \omega(0) \left[1 - \gamma \frac{V(T) - V(0)}{V(0)} - \frac{\Gamma_L^2(0)}{2\omega^2(0)} \times \left(1 + \frac{4\lambda_{\text{ph-ph}}}{e^{\frac{\hbar\omega(0)}{2k_B T}} - 1} \right) \right]. \quad (2)$$

Here, the volume $V(T)$ was determined from experimental lattice parameters [3], and $V(0)$ was evaluated by extrapolating the data to the zero-temperature limit. Using $\lambda_{\text{ph-ph}}$ from Eq. (1), the Grüneisen parameter γ , which relates phonon energy and thermal expansion, was obtained from Eq. (2) as the only remaining free parameter.

Both the energies and linewidths of the A_{1g}^{ph} mode are well reproduced for all experimental strain conditions using the same set of parameters (dashed and solid black lines in Fig. 4), with $\lambda_{\text{ph-ph}} \approx 0.25$ and $\gamma \approx 0.4$. This consistency is expected since the A_{1g}^{ph} mode corresponds to vibrations of Se atoms along the c -axis, and the applied in-plane strain has only a minor effect on the out-of-plane bonding environment. Consequently, the oscillator potential of the phonons remains essentially unchanged.

The B_{1g}^{ph} mode, corresponding to out-of-phase vibrations of the Fe atoms along the c axis, exhibits a moderate and systematic response to strain. The phonon energy and linewidth

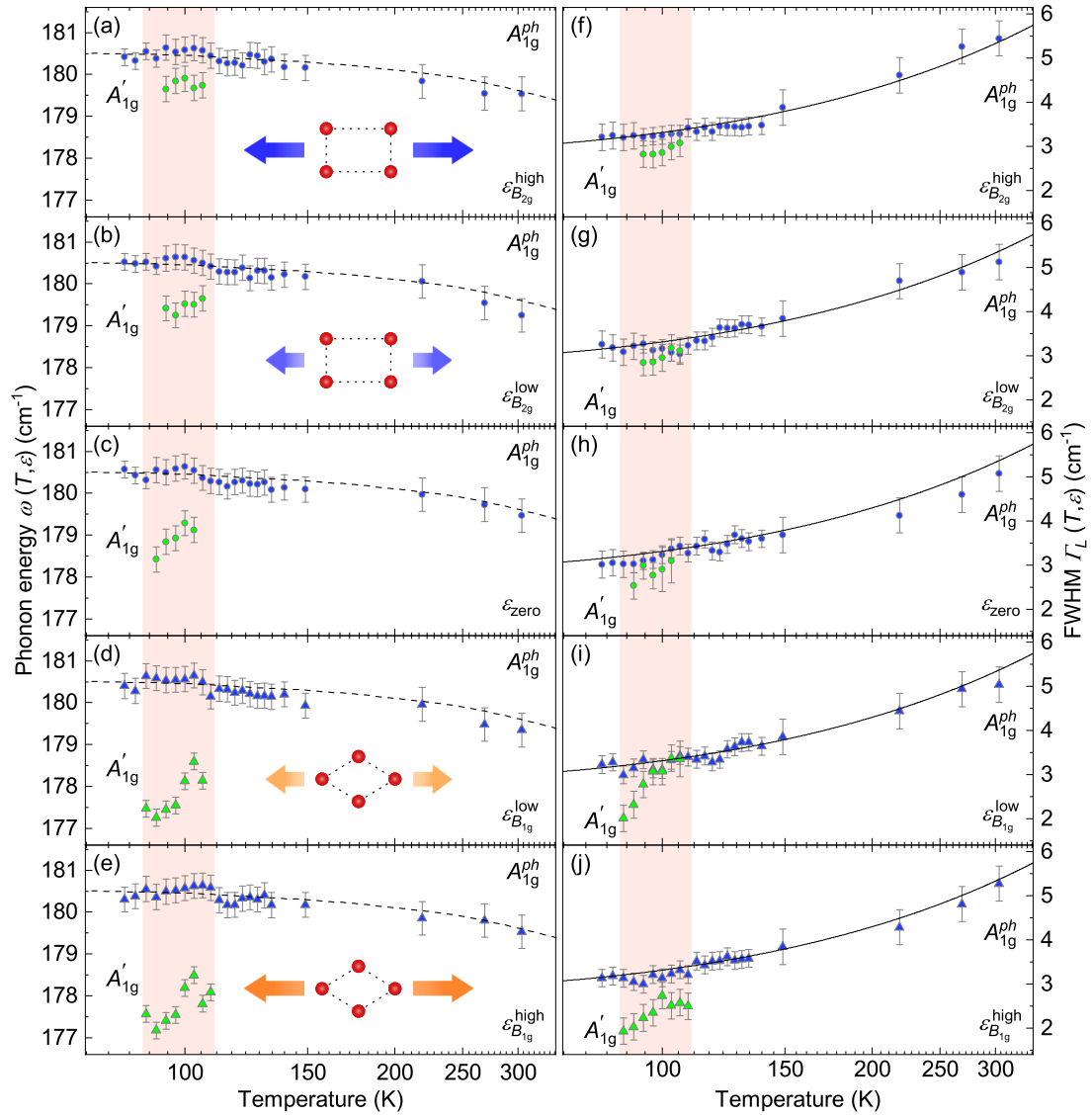


FIG. 4. Temperature dependence of the A_{1g}^{ph} and A'_{1g} mode energies (left) and Lorentzian linewidths (right) for different strain configurations. From top to bottom, the phonon energy and linewidth evolution for [(a) and (f)] $\varepsilon_{B_{2g}}^{high}$, [(b) and (g)] $\varepsilon_{B_{2g}}^{low}$, [(c) and (h)] ε_{zero} , [(d) and (i)] $\varepsilon_{B_{1g}}^{low}$, and [(e) and (j)] $\varepsilon_{B_{1g}}^{high}$. The black [(a)–(e)] dashed and [(f)–(j)] solid lines represent the temperature dependencies of the phonon energies [according to Eq. (2)] and linewidths [according to Eq. (1)], respectively.

evolve smoothly without any anomalous behavior, indicating that this mode is also only weakly affected by strain (see Fig. 9). The corresponding phonon-phonon coupling constants and Grüneisen parameters are summarized in Table I. $\gamma \approx 2$ is several times larger than that of the A_{1g}^{ph} mode and reflects the stronger coupling of B_{1g} vibrations to lattice contraction and in-plane bonding geometry, as out-of-phase Fe displacements modulate the overlap of the in-plane $3d$ orbitals of the Fe-Fe network.

We now turn to the most crucial part of the results, the analysis of the additional A'_{1g} mode and its evolution with strain and temperature. Figure 4 summarizes the behavior of the A_{1g}^{ph} and A'_{1g} peaks under different strain configurations.

We first examine the energy offset of the A'_{1g} mode with respect to the A_{1g}^{ph} mode. In the unstrained sample (ε_{zero}), the maximal energy difference of about 2 cm^{-1} occurs at the

lowest temperature where the A'_{1g} mode is detected. Applying a nematic distortion ($\varepsilon_{B_{2g}}$) reduces this offset, whereas $\varepsilon_{B_{1g}}$ strain enhances it, with its separation from the A_{1g}^{ph} mode exceeding 3 cm^{-1} at the lowest temperature. In all cases, the A'_{1g} mode shifts toward the A_{1g}^{ph} mode upon heating. This strain-dependent evolution indicates that the relative position of the A'_{1g} mode is controlled by the symmetry of the applied strain field and its coupling to the electronic degrees of freedom, an aspect analyzed in detail in the Discussion section.

The temperature dependence of the linewidth of the A'_{1g} mode is shown in Figs. 4(f)–4(j). For the ε_{zero} and $\varepsilon_{B_{2g}}$ strain configurations, the linewidth closely follows that of the A_{1g}^{ph} mode, with a similar broadening. In contrast, under $\varepsilon_{B_{1g}}$ strain, the peak becomes noticeably narrower at lower temperatures than in all other cases. This behavior likely reflects the strain-induced deviation from the nematic ordering.

TABLE I. Parameters of the B_{1g}^{ph} mode obtained from a least-squares fit of the temperature dependence of energies and linewidths [Fig. 9]. The phonon-phonon coupling constant (λ_{ph-ph}) and Grüneisen parameter (γ) were extracted for different strain directions and strengths.

Strain	λ_{ph-ph}	γ
$\varepsilon_{B_{2g}}^{high}$	0.18	2.0
$\varepsilon_{B_{2g}}^{low}$	0.16	2.0
ε_{zero}	0.2	2.0
$\varepsilon_{B_{1g}}^{low}$	0.13	1.8
$\varepsilon_{B_{1g}}^{high}$	0.13	1.8

IV. DISCUSSION

The most unexpected result of this study is the additional feature slightly below the A_{1g}^{ph} phonon energy, observed within a narrow temperature interval around the nematic transition at $T_s = 89.1$ K (Fig. 2). Although the additional A'_{1g} contribution is not directly resolved as a separate structure because of its small energy difference from the A_{1g}^{ph} phonon and its low intensity, the two-component analysis of the phonon line-shape performed for different strain configurations supports the presence of two overlapping contributions. This effect is most pronounced under high B_{1g} tensile strain, whereas for the other strain configurations, including nominally zero strain, the corresponding asymmetry is weaker and observed in a narrower temperature range.

To assess this behavior quantitatively, we fit one- and two-component models to the spectra, with each component described by a Voigt profile. Figure 5 shows representative spectra measured at $T = 106$ and 133 K under high B_{1g} strain, $\varepsilon_{B_{1g}}^{high}$. The A'_{1g} contribution is most clearly resolved at $T = 106$ K, where the two-component model provides an improved description of the low-energy asymmetry, as demonstrated by the residuals $\Delta R\chi''_{1,2}(\omega)$, their difference $\delta_{(1,2)}(\omega)$, and the corresponding $R^2_{1,2}$ values. By contrast, for the spectrum at $T = 133$ K, the two-component model no longer yields a better description of the measured data. This provides quantitative support for the presence of an additional contribution below the symmetry-allowed A_{1g}^{ph} phonon. The residual difference, $\delta_{(1,2)}(\omega, T)$, over the entire measured temperature range for three strain configurations, $\varepsilon_{B_{1g}}^{high}$, ε_{zero} , and $\varepsilon_{B_{2g}}^{high}$, is presented in Fig. 8 of Appendix B. This analysis shows that the A'_{1g} contribution is not an artifact, but is systematically observed in the vicinity of T_s and thus statistically significant.

FeS and $\text{FeSe}_{1-x}\text{S}_x$ provide useful reference systems in which additional Raman features associated with the fully symmetric response have been observed. In FeS, an overtone excitation inside the gap of the PDOS originates from a two-phonon scattering process involving acoustic branches close to the Brillouin zone boundary in the M and A regions [see Fig. 6(a)]. Its activation inside this gap has been attributed to enhanced EPC in FeS, where acoustic phonons acquire sufficient coupling strength to produce the overtone near twice

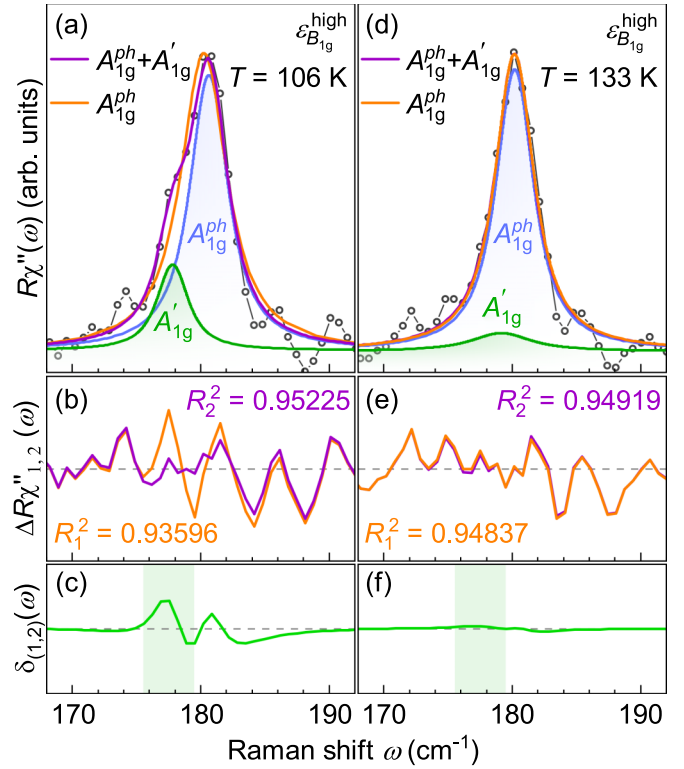


FIG. 5. Comparison of one- and two-component fits to the asymmetric A_{1g}^{ph} peak. Results are shown for the $\varepsilon_{B_{1g}}^{high}$ strain configuration at $T = 106$ K and $T = 133$ K. [(a) and (d)] Measured spectra (open symbols) together with the one-component (orange) and two-component (purple) models. The A_{1g}^{ph} phonon and A'_{1g} contributions comprising the two-component model are shown by the blue and green shading, respectively. [(b) and (e)] Residuals $\Delta R\chi''_1(\omega)$ and $\Delta R\chi''_2(\omega)$ for the one- and two-component fits, respectively. The corresponding $R^2_{1,2}$ values are indicated. [(c) and (f)] Difference between the residuals, $\delta_{(1,2)}(\omega) = \Delta R\chi''_1(\omega) - \Delta R\chi''_2(\omega)$. The dashed lines indicate zero residual, while the green shading marks the spectral region associated with the A'_{1g} contribution.

their zone boundary energy [17]. As shown in Fig. 6(a), the energies of the acoustic branches at M and A in FeS lie approximately at half of those of the $A_{1g}^{S,1}$ phonon line.

The situation in $\text{FeSe}_{1-x}\text{S}_x$ is notably different for small x : the PDOS does not have a gap, and an additional feature emerges in the A_{1g} channel even at the lowest substitution levels ($x \approx 0.04-0.05$) [18,31] (see Fig. 10, Appendix D). The appearance of this feature at such low x suggests that even weak sulfur substitution perturbs the lattice and electronic environment locally, thereby modifying the A_{1g} response. Therefore the persistence and evolution of these features favor a scenario where EPC of some acoustic branches may be enhanced at the Brillouin zone boundary.

Unlike in $\text{FeSe}_{1-x}\text{S}_x$, where the additional structure develops on the high-energy side of the A_{1g}^{ph} line (Fig. 10) and survives at all temperatures [31], in FeSe the additional mode appears on the low-energy side of the A_{1g}^{ph} mode in the small temperature interval around T_s (Fig. 4).

The acoustic branches near the X and R points of the phonon dispersion [2-Fe cell, see Fig. 6(c)] are plausible

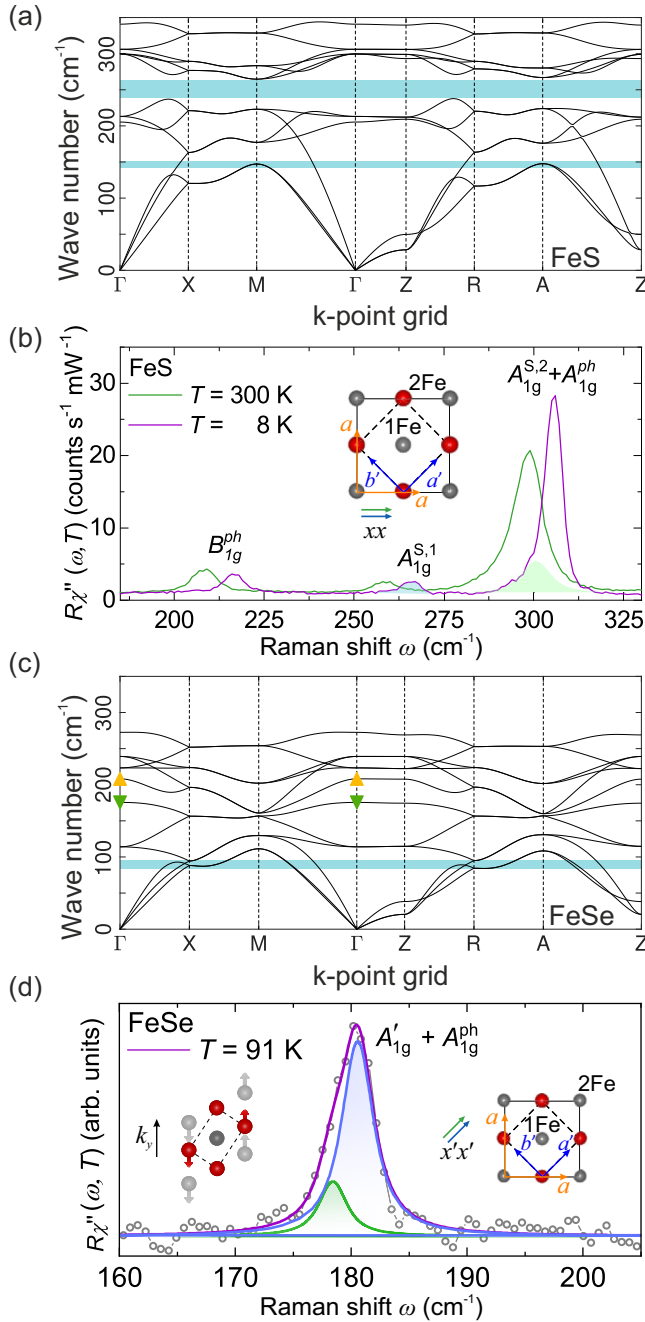


FIG. 6. Comparison of phonon dispersions and Raman spectra of FeS and FeSe. (a) Calculated phonon dispersion of FeS along high-symmetry directions of the Brillouin zone. The blue-shaded area marks the gap in the phonon density of states (PDOS). (b) Raman spectra of FeS in xx polarization, showing the main A_{1g}^{ph} mode and additional $A_{1g}^{S,1}$ and $A_{1g}^{S,2}$ features, the first of which lies within the PDOS gap. (c) Calculated phonon dispersion of FeSe along high-symmetry directions. A_{1g}^{ph} and B_{1g}^{ph} values at the Γ point are labeled with green and yellow triangles, respectively. (d) Raman spectra of FeSe in $x'x'$ polarization, where an additional A_{1g}' mode appears near the A_{1g}^{ph} peak. The left inset shows the atomic displacement pattern of the relevant acoustic mode at the X point in the 2-Fe Brillouin zone. Figures adapted from our previous studies (for details see Refs. [17,18]).

candidates for the origin of the two-phonon process, because twice their energy corresponds approximately to the observed A_{1g}' feature. Additionally, the longitudinal acoustic (LA) phonon at the X point has a purely in-plane displacement pattern of the atoms [inset of Fig. 6(d)] that produces the same modulation of the 1-Fe cell as the nematic distortion, which entails a reconstruction of the Fermi surface by lifting the d_{xz}/d_{yz} degeneracy of the bands at the M point. Thus, although the X -point LA phonon induces this reconstruction at all temperatures, the nematic susceptibility increases rapidly close to T_s and, for symmetry reasons, this phonon couples directly and more strongly to the fluctuations. Concomitantly, the interaction between two zone-boundary LA modes is enhanced around T_s , and the energy is renormalized down. Since two modes having the same symmetry always yield an A_{1g} excitation, the appearance of the A_{1g}' mode has a straightforward explanation.

In crystals detwinned along the Fe-Fe bond direction (B_{2g} strain), the applied strain aligns with the nematic distortion and promotes domain ordering. In this configuration, the additional mode is found to gradually merge with the A_{1g}^{ph} phonon [see Figs. 4(a) and 4(b)] and persists over a narrower temperature range, consistent with the suppression of critical fluctuations once the lattice becomes more ordered. Conversely, B_{1g} strain perturbs the orthogonal in-plane symmetry channel and modifies the local lattice environment and the anisotropy of EPC. As a result, the temperature range over which the A_{1g}' mode is observed broadens, and the peak becomes more sharply defined (Figs. 2 and 4). This contrast highlights the sensitivity of the FeSe lattice to the direction of applied strain and indicates that the emergence of the A_{1g}' feature and its energy separation from the A_{1g}^{ph} phonon mode are sensitive to the balance between nematic ordering and the modulation of EPC by lattice distortions.

Thus, while the phonon anomalies in FeS and FeSe may be associated with the acoustic branches near the zone boundary, the conditions enhancing the two-phonon Raman response are different. In FeS, the effect reflects an EPC-enhanced two-phonon response within a structurally stable phase, whereas in FeSe it appears only near T_s , where critical nematoelastic fluctuations and the temperature-dependent electronic susceptibility near the M point may enhance the coupling to acoustic lattice modes near the zone boundary.

These findings contribute to clarifying the complex behavior of interrelation between structural, magnetic, and electronic degrees of freedom in FeSe. Applying stress along the principal axis or the diagonal leads to distinctly different responses of the electronic system and the lattice. In the first case, the Fe plane is rhombohedrally distorted and the fluctuation range is extended due to the suppression of the nematic order. Consequently, the presence of the two-phonon response survives in a broader temperature range because of the fluctuation-induced EPC. In the second case, the nematic order is supported by the external strain, the fluctuation range narrows down, and the two-phonon response appears only in a smaller temperature range. Obviously, fluctuations and EPC are correlated here because of the eigenvector of the longitudinal acoustic mode near the X point of the Brillouin

zone. In the $\text{FeSe}_{1-x}\text{S}_x$ system, additional mechanisms may be at work such as local enhancement by disorder.

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J.B. and A.M. contributed equally to this work. J.B., A.M., R.H., and N.L. designed and set up the experiment. A.W. and C.P. prepared the samples. J.P., A.Š., and E.D.L. performed the DFT calculations. E.S.B., M.V.M., and Z.V.P. contributed to scientific discussions and interpretation of the results. J.B., A.M., T.B., B.V., M.O., N.P., and N.L. carried out the Raman scattering measurements. All authors contributed to data analysis, discussion of the results, and writing of the manuscript.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

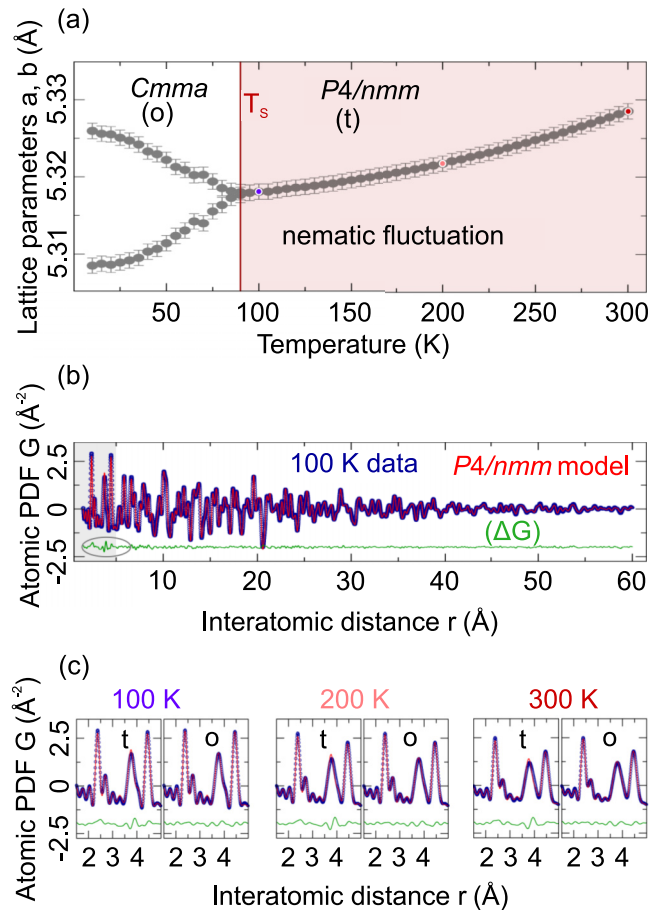


FIG. 7. Crystal and local structure of the FeSe sample used in this study. (a) Temperature dependence of the in-plane lattice parameters, with T_s as indicated. The global structure is orthorhombic (o) $Cmma$ below and tetragonal (t) $P4/nmm$ above 90 K. In the tetragonal phase, a known signature of nematic fluctuations is observed at the nanoscale, as exemplified in (b). (b) The difference between the synchrotron pair distribution function data and the tetragonal model fit at 100 K (green line, offset for clarity) displays subtle misfits on a unit-cell length scale (shaded region). This signature remains virtually unchanged at elevated temperature and can be rectified using the orthorhombic local symmetry model (c) [15,16]. (c) Comparison of tetragonal (t) and orthorhombic (o) local symmetry models for 100, 200, and 300 K.

APPENDIX A: CRYSTAL STRUCTURE OF FESE

To access the local structure on top of the average crystallographic structure, we studied a FeSe crystal by x-ray diffraction and extracted the pair-density distribution function (pdf) at three different temperatures. Figure 7(a) shows the in-plane lattice parameters as a function of temperature and the effect of the tetragonal-to-orthorhombic transition at $T_s \approx 90$ K. Above T_s , the average structure is tetragonal, described by the $P4/nmm$ space group, and orthorhombic below T_s , described by the $Cmma$. Figure 7(b) shows the synchrotron pair distribution function $G(r)$ at 100 K, where r is the interatomic distance. This experimental result is compared to the expectation from a model based on the tetragonal space group $P4/nmm$. The difference shows larger deviations $\Delta G(r)$ between experiment and model in the range $r \leq 5$ Å.

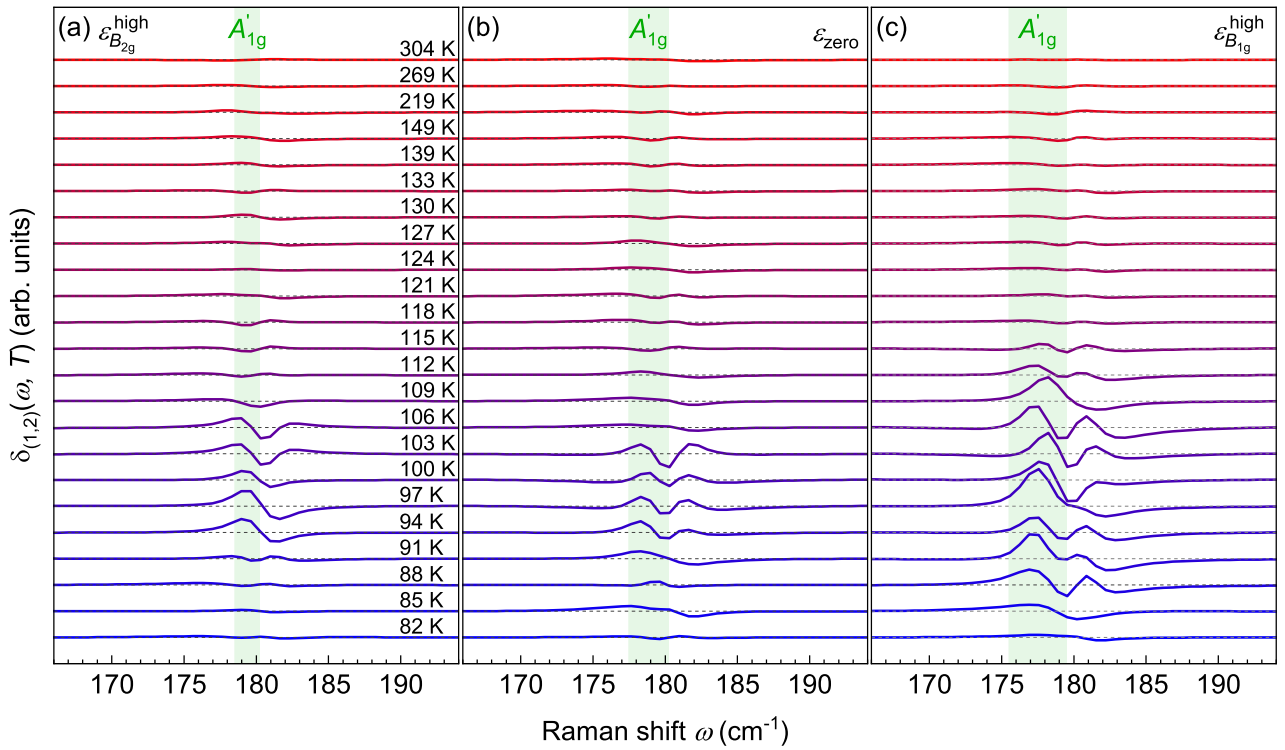


FIG. 8. Decomposition of the asymmetric A'_{1g} Raman peak. [(a)–(c)] Difference between the residuals obtained from one- and two-component descriptions of the A'_{1g} Raman mode using Voigt profiles over the measured temperature range (82–304 K). The corresponding residual difference, $\delta_{(1,2)}(\omega, T) = \Delta R\chi_1''(\omega, T) - \Delta R\chi_2''(\omega, T)$, is shown for three representative strain configurations: (a) $\varepsilon_{B_{2g}}^{\text{high}}$, (b) $\varepsilon_{\text{zero}}$, and (c) $\varepsilon_{B_{1g}}^{\text{high}}$. The curves are vertically offset for clarity. Dashed lines indicate zero residual difference, while the green shading marks the spectral region associated with the additional A'_{1g} contribution.

as highlighted by the shaded region, whereas for $r > 5$ Å the agreement is satisfactory. The range and the magnitude of $\Delta G(r)$ do not strongly depend on temperature, meaning that the (small) local deviations from the overall tetragonal structure survive up to room temperature as shown in Fig. 7(c). Obviously, the deviations are slow enough to be observable by a quasistatic method [15,16].

APPENDIX B: DECOMPOSITION OF UNRESOLVED A'_{1g} AND A'_{1g} MODES

The A'_{1g} peak in the temperature range around T_s exhibits a pronounced asymmetry on the low-energy side in the spectra of the twinned FeSe sample. Upon detwinning, it becomes evident that this asymmetry arises from the emergence of an additional peak.

In Fig. 8, the difference between the residuals obtained from one- and two-component descriptions of the A'_{1g} mode, $\delta_{(1,2)}(\omega, T) = \Delta R\chi_1''(\omega, T) - \Delta R\chi_2''(\omega, T)$, with each component represented by a Voigt profile, is shown. The comparison is presented over the full measured temperature range for (a) $\varepsilon_{B_{2g}}^{\text{high}}$ strain, (b) zero strain, and (c) $\varepsilon_{B_{1g}}^{\text{high}}$ strain. The green shading marks the spectral range associated with the A'_{1g} contribution. Positive values of $\delta_{(1,2)}(\omega, T)$ indicate that the two-component model yields smaller residuals than the one-component model, whereas values close to zero indicate comparable fit quality. The one-component model adequately

describes the spectra for all strain configurations except within a narrow temperature window around T_s . Within this window, systematic deviations emerge in the spectral range associated with the A'_{1g} contribution.

APPENDIX C: B'_{1g} MODE TEMPERATURE DEPENDENCE

The temperature dependences of extracted energies and linewidths of the B'_{1g} mode for different strain configurations are shown in Fig. 9. They are modeled analogously to the A'_{1g} mode, as described in the main text. Both the energy and linewidth change gradually without any anomalies. The extracted phonon–phonon coupling parameter ($\lambda_{\text{ph-ph}}$) and Grüneisen parameter (γ) are summarized in Tab. I of the main text for the free-standing sample ($\varepsilon_{\text{zero}}$) and for the B'_{1g} and B'_{2g} strain configurations.

APPENDIX D: TWO-PHONON EXCITATIONS IN FeS AND FeSe_{1-x}S_x

Figure 10 shows the evolution of the Raman spectra of FeSe_{1-x}S_x with increasing sulfur content x up to the QCP ($x \approx 0.2$) from our previously published study (see Ref. [18]). The spectra were recorded above the nemato-structural transition ($T > T_s$) in the $x'x'$ configuration, where only a fully symmetric mode is expected. In the FeSe compound, only a single phonon line A'_{1g} was observed. At small substitution

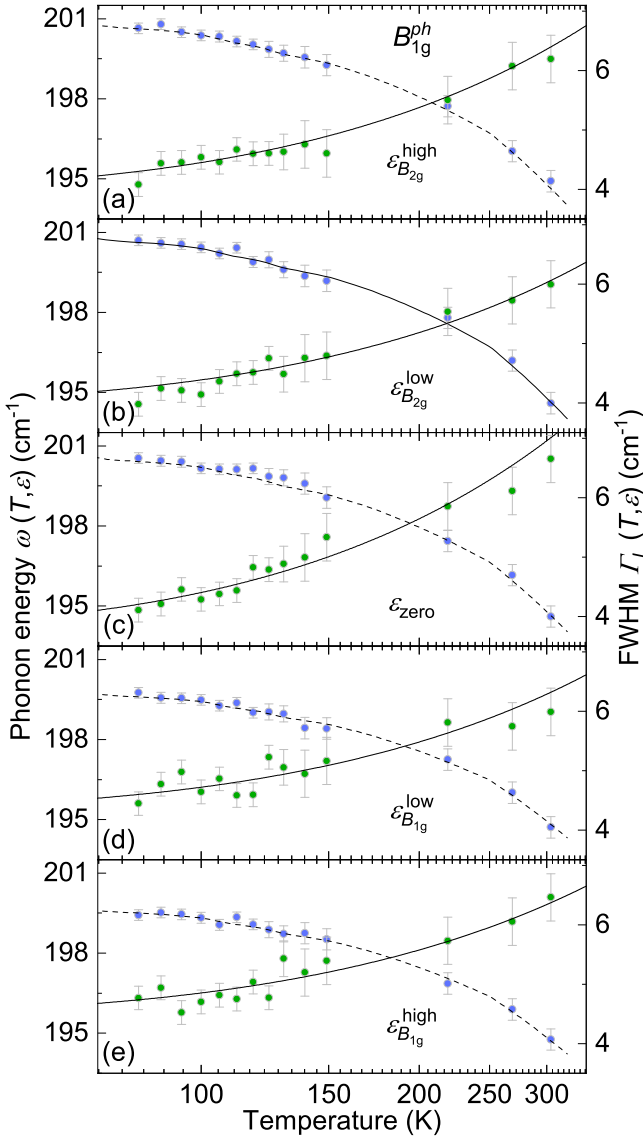


FIG. 9. Temperature dependence of the B_{1g}^{ph} mode energies and linewidths for different strain configurations. Panels from top to bottom show the phonon energy and linewidth evolution for (a) $\epsilon_{B_{2g}}^{high}$, (b) $\epsilon_{B_{2g}}^{low}$, (c) ϵ_{zero} , (d) $\epsilon_{B_{1g}}^{low}$, and (e) $\epsilon_{B_{1g}}^{high}$. The black dashed and solid lines represent the temperature dependencies of the energies and Lorentzian phonon linewidths according to Eqs. (2) and (1), respectively, in the main text.

levels ($x = 0.05$), new features emerge in the $x'x'$ configuration [32]. With increasing sulfur content, these features gain intensity, while the A_{1g}^{ph} mode softens, weakens in intensity, and becomes almost undetectable for $x = 0.23$ [18]. The spectrum of FeS [inset of Fig. 6(b)] contains additional features that were previously attributed to two-phonon scattering and to a projection of the phonon density of states [17].

APPENDIX E: STRAIN CELL

The strain cell was assembled in-house following the designs described in Refs. [33,34]. The cell consists of a precision frame formed by two titanium bridges connected by

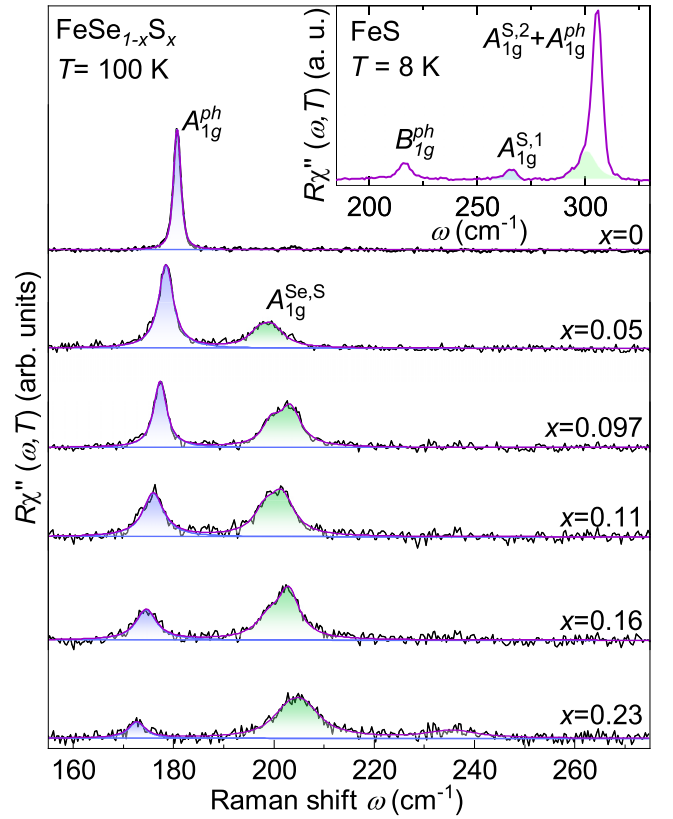


FIG. 10. Raman spectra of $\text{FeSe}_{1-x}\text{S}_x$ ($0 \leq x \leq 0.23$) and FeS. The spectra of $\text{FeSe}_{1-x}\text{S}_x$ are measured at $T > T_s$ in the $x'x'$ scattering configuration where only fully symmetric phonon excitations can be observed. Spectral lines other than the Raman-active phonons are labeled as $A_{1g}^{Se,S}$. (Inset) Raman spectra of FeS at 8 K, where two-phonon excitations are labeled as $A_{1g}^{S,1}$ and $A_{1g}^{S,2}$. These figures are adapted from our previous studies (for details see Refs. [17,18]).

three piezoelectric stacks, allowing controlled application of uniaxial strain. The two outer stacks connect the bridges and generate tensile strain, while the inner stack is attached to one bridge and carries the movable sample platform (MP) at its opposite end. The fixed platform (FP) is part of the second bridge. Extension of the outer stacks moves the MP away from the FP, whereas extension of the inner stack pushes the MP toward the FP, producing compression.

To monitor the applied strain, a strain gauge was attached between the fixed and movable platforms, beside the sample. The strain gauge was incorporated into an electronic feedback circuit controlling the voltage applied to the piezoelectric stacks, so that the strain remained constant during the measurements. The tensile-strain measurements discussed in the main text were performed at 0, 70, and 140 V, corresponding to nominal strain values of 0%, 0.13%, and 0.21%, respectively. These conditions are denoted as zero strain (ϵ_{zero}), low strain (ϵ^{low}), and high strain (ϵ^{high}). Since the focus of this work is the qualitative strain dependence of the Raman response, these values are treated as nominal strain levels. Although the piezoelectric driver allows operation down to -30 V, the resulting compression was negligible in the present configuration; therefore, only tensile-strain measurements were considered.

- [1] F.-C. Hsu, J.-Y. Luo, K.-W. Yeh, T.-K. Chen, T.-W. Huang, P. M. Wu, Y.-C. Lee, Y.-L. Huang, Y.-Y. Chu, D.-C. Yan, and M.-K. Wu, Superconductivity in the PbO-type structure α -FeSe, *Proc. Natl. Acad. Sci. USA* **105**, 14262 (2008).
- [2] P. Massat, Y. Quan, R. Grasset, M.-A. Méasson, M. Cazayous, A. Sacuto, S. Karlsson, P. Strobel, P. Toulemonde, Z. Yin, and Y. Gallais, Collapse of critical nematic fluctuations in FeSe under pressure, *Phys. Rev. Lett.* **121**, 077001 (2018).
- [3] S. Margadonna, Y. Takabayashi, Y. Ohishi, Y. Mizuguchi, Y. Takano, T. Kagayama, T. Nakagawa, M. Takata, and K. Prassides, Pressure evolution of the low-temperature crystal structure and bonding of the superconductor FeSe ($T_c = 37$ K), *Phys. Rev. B* **80**, 064506 (2009).
- [4] W. Qing-Yan, L. Zhi, Z. Wen-Hao, Z. Zuo-Cheng, Z. Jin-Song, L. Wei, D. Hao, O. Yun-Bo, D. Peng, C. Kai, W. Jing, S. Can-Li, H. Ke, J. Jin-Feng, J. Shuai-Hua, W. Ya-Yu, W. Li-Li, C. Xi, M. Xu-Cun, and X. Qi-Kun, Interface-induced high-temperature superconductivity in single unit-cell FeSe films on SrTiO₃, *Chin. Phys. Lett.* **29**, 037402 (2012).
- [5] S. Tan, Y. Zhang, M. Xia, Z. Ye, F. Chen, X. Xie, R. Peng, D. Xu, Q. Fan, H. Xu, J. Jiang, T. Zhang, X. Lai, T. Xiang, J. Hu, B. Xie, and D. Feng, Interface-induced superconductivity and strain-dependent spin density waves in FeSe/SrTiO₃ thin films, *Nat. Mater.* **12**, 634 (2013).
- [6] K. Matsuura, Y. Mizukami, Y. Arai, Y. Sugimura, N. Maejima, A. Machida, T. Watanuki, T. Fukuda, T. Yajima, Z. Hiroi, K. Y. Yip, Y. C. Chan, Q. Niu, S. Hosoi, K. Ishida, K. Mukasa, S. Kasahara, J.-G. Cheng, S. K. Goh, Y. Matsuda, *et al.*, Maximizing T_c by tuning nematicity and magnetism in FeSe_{1-x}S_x superconductors, *Nat. Commun.* **8**, 1143 (2017).
- [7] G. Hu, M. Shi, W. Wang, C. Zhu, Z. Sun, J. Cui, W. Zhuo, F. Yu, X. Luo, and X. Chen, Superconductivity at 40 K in lithiation-processed [(Fe,Al)(OH)₂][FeSe]_{1.2} with a layered structure, *Inorg. Chem.* **60**, 3902 (2021).
- [8] M. D. Watson, T. K. Kim, L. C. Rhodes, M. Eschrig, M. Hoesch, A. A. Haghighirad, and A. I. Coldea, Evidence for unidirectional nematic bond ordering in FeSe, *Phys. Rev. B* **94**, 201107(R) (2016).
- [9] M. D. Watson, A. A. Haghighirad, L. C. Rhodes, M. Hoesch, and T. K. Kim, Electronic anisotropies revealed by detwinned angle-resolved photo-emission spectroscopy measurements of FeSe, *New J. Phys.* **19**, 103021 (2017).
- [10] L. C. Rhodes, M. Eschrig, T. K. Kim, and M. D. Watson, FeSe and the missing electron pocket problem, *Front. Phys.* **10**, 859017 (2022).
- [11] Y. Suzuki, T. Shimojima, T. Sonobe, A. Nakamura, M. Sakano, H. Tsuji, J. Omachi, K. Yoshioka, M. Kuwata-Gonokami, T. Watashige, R. Kobayashi, S. Kasahara, T. Shibauchi, Y. Matsuda, Y. Yamakawa, H. Kontani, and K. Ishizaka, Momentum-dependent sign inversion of orbital order in superconducting FeSe, *Phys. Rev. B* **92**, 205117 (2015).
- [12] L. C. Rhodes, M. D. Watson, A. A. Haghighirad, D. V. Evtushinsky, and T. K. Kim, Revealing the single electron pocket of FeSe in a single orthorhombic domain, *Phys. Rev. B* **101**, 235128 (2020).
- [13] A. I. Coldea and M. D. Watson, The key ingredients of the electronic structure of FeSe, *Annu. Rev. Condens. Matter Phys.* **9**, 125 (2018).
- [14] M. Yi, H. Pfau, Y. Zhang, Y. He, H. Wu, T. Chen, Z. R. Ye, M. Hashimoto, R. Yu, Q. Si, D.-H. Lee, P. Dai, Z.-X. Shen, D. H. Lu, and R. J. Birgeneau, Nematic energy scale and the missing electron pocket in FeSe, *Phys. Rev. X* **9**, 041049 (2019).
- [15] R. J. Koch, T. Konstantinova, M. Abeykoon, A. Wang, C. Petrovic, Y. Zhu, E. S. Bozin, and S. J. L. Billinge, Room temperature local nematicity in FeSe superconductor, *Phys. Rev. B* **100**, 020501(R) (2019).
- [16] B. A. Frandsen, Q. Wang, S. Wu, J. Zhao, and R. J. Birgeneau, Quantitative characterization of short-range orthorhombic fluctuations in FeSe through pair distribution function analysis, *Phys. Rev. B* **100**, 020504(R) (2019).
- [17] A. Baum, A. Milosavljević, N. Lazarević, M. M. Radonjić, B. Nikolić, M. Mitschek, Z. I. Maranloo, M. Šćepanović, M. Grujić-Brojin, N. Stojilović, M. Opel, A. Wang, C. Petrovic, Z. V. Popović, and R. Hackl, Phonon anomalies in FeS, *Phys. Rev. B* **97**, 054306 (2018).
- [18] N. Lazarević, A. Baum, A. Milosavljević, L. Peis, R. Stumberger, J. Bekaert, A. Šolajić, J. Pešić, A. Wang, M. Šćepanović, A. M. M. Abeykoon, M. V. Milošević, C. Petrovic, Z. V. Popović, and R. Hackl, Evolution of lattice, spin, and charge properties across the phase diagram of FeSe_{1-x}S_x, *Phys. Rev. B* **106**, 094510 (2022).
- [19] A. Wang, A. Milosavljevic, A. M. M. Abeykoon, V. Ivanovski, Q. Du, A. Baum, E. Stavitski, Y. Liu, N. Lazarevic, K. Attenkofer, R. Hackl, Z. Popovic, and C. Petrovic, Suppression of superconductivity and nematic order in Fe_{1-y}Se_{1-x}S_x ($0 \leq x \leq 1$; $y \leq 0.1$) crystals by anion height disorder, *Inorg. Chem.* **61**, 11036 (2022).
- [20] A. Baum, H. N. Ruiz, N. Lazarević, Y. Wang, T. Böhm, R. Hosseinian Ahangharnejhad, P. Adelman, T. Wolf, Z. V. Popović, B. Moritz, T. P. Devereaux, and R. Hackl, Frustrated spin order and stripe fluctuations in FeSe, *Commun. Phys.* **2**, 14 (2019).
- [21] M. S. Ikeda, T. Worasaran, J. C. Palmstrom, J. A. W. Straquadine, P. Walmsley, and I. R. Fisher, Symmetric and antisymmetric strain as continuous tuning parameters for electronic nematic order, *Phys. Rev. B* **98**, 245133 (2018).
- [22] N. Lazarević and R. Hackl, Fluctuations and pairing in Fe-based superconductors: Light scattering experiments, *J. Phys.: Condens. Matter* **32**, 413001 (2020).
- [23] M. Ghini, M. Bristow, J. C. A. Prentice, S. Sutherland, S. Sanna, A. A. Haghighirad, and A. I. Coldea, Strain tuning of nematicity and superconductivity in single crystals of FeSe, *Phys. Rev. B* **103**, 205139 (2021).
- [24] T. Hanaguri, K. Iwaya, Y. Kohsaka, T. Machida, T. Watashige, S. Kasahara, T. Shibauchi, and Y. Matsuda, Two distinct superconducting pairing states divided by the nematic end point in FeSe_{1-x}S_x, *Sci. Adv.* **4**, eaar6419 (2018).
- [25] A. V. Chubukov, R. M. Fernandes, and J. Schmalian, Origin of nematic order in FeSe, *Phys. Rev. B* **91**, 201105(R) (2015).
- [26] S.-H. Baek, D. V. Efremov, J. M. Ok, J. S. Kim, J. van den Brink, and B. Büchner, Orbital-driven nematicity in FeSe, *Nat. Mater.* **14**, 210 (2015).
- [27] A. E. Böhrer and A. Kreisel, Nematicity, magnetism and superconductivity in FeSe, *J. Phys.: Condens. Matter* **30**, 023001 (2018).
- [28] S. Rößler, M. Coduri, A. A. Tsirlin, C. Ritter, G. Cuello, C. Koz, L. Muzica, U. Schwarz, U. K. Rößler, S. Wirth, and

- M. Scavini, Nematic state of the FeSe superconductor, [Phys. Rev. B **105**, 064505 \(2022\)](#).
- [29] P. G. Klemens, Anharmonic decay of optical phonons, [Phys. Rev. **148**, 845 \(1966\)](#).
- [30] H.-M. Eiter, P. Jaschke, R. Hackl, A. Bauer, M. Gangl, and C. Pfleiderer, Raman study of the temperature and magnetic-field dependence of the electronic and lattice properties of MnSi, [Phys. Rev. B **90**, 024411 \(2014\)](#).
- [31] W. Zhang, S. Wu, S. Kasahara, T. Shibauchi, Y. Matsuda, and G. Blumberg, Quadrupolar charge dynamics in the nonmagnetic FeSe_{1-x}S_x superconductors, [Proc. Natl. Acad. Sci. USA **118**, e2020585118 \(2021\)](#).
- [32] V. Gnezdilov, Y. G. Pashkevich, P. Lemmens, D. Wulferding, T. Shevtsova, A. Gusev, D. Chareev, and A. Vasiliev, Interplay between lattice and spin states degree of freedom in the FeSe superconductor: Dynamic spin state instabilities, [Phys. Rev. B **87**, 144508 \(2013\)](#).
- [33] C. W. Hicks, M. E. Barber, S. D. Eddins, D. O. Brodsky, and A. P. Mackenzie, Piezoelectric-based apparatus for strain tuning, [Rev. Sci. Instrum. **85**, 065003 \(2014\)](#).
- [34] J. Schmidt, V. Bekeris, G. S. Lozano, M. V. Bortulé, M. M. Bermúdez, C. W. Hicks, P. C. Canfield, E. Fradkin, and G. Pasquini, Nematicity in the superconducting mixed state of strain detwinned underdoped Ba(Fe_{1-x}Co_x)₂As₂, [Phys. Rev. B **99**, 064515 \(2019\)](#).