Nematic Fluctuations and Phase Transitions in LaFeAsO: a Raman Scattering Study

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Raman scattering experiments on LaFeAsO with splitted antiferromagnetic (TAFM = 140 K) and tetragonal-orthorhombic (TS = 155 K) transitions show a quasi-elastic peak (QEP) in B2g symmetry (2 Fe tetragonal cell) that fades away below ∼ TAFM and is ascribed to electronic nematic fluctuations. A scaling of the reported shear modulus with the T−dependence of the QEP height rather than magnetism in the latter [16, 17]. However, even for FeSe the magnetic scenario may still apply [18]. In general, the primary order parameter that drives the structural/nematic transition at TS and the dominating mechanism of TAFM/TS separation in parent FeSCs are not fully settled yet.

Raman scattering was recently employed as a probe of nematic fluctuations in FeSCs and their parent materials. In A(Fe1−xCo2−x)2As2 (A = Ca, Sr, Ba, Eu) [1, 4, 19, 21, 22, 24, 25], Ba1−pKpFe2As2 [25], FeSe [26, 27] and NaFe1−xCo2As [28], a quasi-elastic peak (QEP) with B2g symmetry (considering the 2 Fe tetragonal cell, see Fig. 1(a)) has been observed and interpreted in terms of either charge/orbital [21, 22, 24, 26, 29] or spin [23, 24, 30] nematic fluctuations. An unambiguous experimental identification of the nature of the fluctuations generating the B2g Raman QEP (charge/orbital or magnetic) is challenging due to the inherent coupling between the corresponding degrees of freedom. Despite such extensive investigations in several materials, no Raman study of the nematic fluctuations in the key parent compound LFAO has been carried out yet. In this work, we fill this gap and investigate in detail the temperature dependence of both electronic and phonon Raman scattering in LFAO.

Details of the synthesis procedure and basic characterization of the crystal employed in this work, showing TS = 155 K and TAFM = 140 K, are described elsewhere [12, 32]. A fresh ab surface with a large area was obtained by cleaving the crystal and immediately mounting it at the cold finger of a closed-cycle He cryostat. The polarized Raman spectra were taken in quasi-backscattering geometry using the 488.0 nm line as exciting source focused into the ab surface with a spot of ~ 50 μm diameter. A triple 1800 mm−1 grating spectrometer

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equipped with a $LN_2$-cooled multichannel CCD detector was employed. The instrumental linewidth was $\sim 4$ cm$^{-1}$. Figure 1(a) illustrates a square lattice of the Fe atoms and sets the conventions for polarizations. The 2 Fe tetragonal (space group P4/mmm) and 4 Fe orthorhombic (space group Cmcm) unit cells and axes in the $ab$ plane are also represented.

Symmetry analysis indicates that four Raman-active phonons are accessible by our experimental geometry in both tetragonal ($A_{1g}$ and $B_{2g}$) and orthorhombic ($A_{g}$ and $2B_{1g}$) phases. Illustrations of such modes are given in Fig. 1(b) (see also Ref. [33]). The raw Raman spectra in the phonon region at distinct linear polarizations are given in Figs. 1(c) ($T = 20$ K) and 1(d) ($T = 290$ K). The $B_{1g}$ modes observed at 203 and 317 cm$^{-1}$ at $T = 290$ K are ascribed to Fe and O vibrations along $c$, respectively [33], while the $A_{1g}$ modes at 164 and 208 cm$^{-1}$ are ascribed to As and La vibrations along $c$. The position of the 164 cm$^{-1}$ mode is comparable to that reported for the As mode in NaFeAs (163 cm$^{-1}$ [34]) and in AFe$_2$As$_2$ ($A = $ Ca, Sr, Ba) (180-190 cm$^{-1}$ [35, 36]). The $T$-dependence of this phonon was investigated in detail (see SM). Its linewidth at low-$T$ is resolution-limited, suggesting a high crystalline quality, and shows a maximum at $T \sim T_{AFM}$ with no anomaly at $T_S$. Frequency anomalies are observed for this mode at both $T_S$ and $T_{AFM}$. Finally, an enhancement in $XY$ polarization is observed below $T_{AFM}$, which is similar to related systems [1-4] and is due to the coupling of this phonon with anisotropic electronic states in the magnetic phase [3].

The Raman response $\chi''(\omega, T)$ is related to the raw intensity $I$ through the relation $I = (1 + n)\chi''(\omega, T) + D$, where $n \equiv 1/(e^{\omega/k_B T} - 1)$ is the Bose-Einstein statistical factor and $D$ is an intensity offset (for details, see SM). Figures 2(a-e) show $\chi''(\omega, T)$ in $XY$ polarization corresponding to $B_{2g}$ symmetry in the 2 Fe tetragonal cell. These measurements were made with much less laser power ($\sim 3$ mW) than for the data shown in Fig. 1 (~10 mW), in order to minimize laser heating effects [4], and were also taken with $4\times$ less exposure times due to the large number of investigated temperatures. These limitations resulted in poorer signal-to-noise in the data shown in Figs. 2(a-e). An 8-point-average smoothing is applied in these data for better visualization of the broad electronic Raman signal. A linear component for $\chi''_{B_{2g}}(\omega, T)$ is observed in the frequency region below 600 cm$^{-1}$, which is enhanced below $T_{AFM}$. Measurements performed on an extended frequency region show this component is part of broad peaks at $\sim 2400-3000$ cm$^{-1}$ (see SM). A similar structure was found in BaFe$_2$As$_2$ and attributed to two-magnon scattering [7]. An additional scattering channel, which is most evident at low frequencies ($\omega \lesssim 150$ cm$^{-1}$), is observed in this symmetry and enhances on cooling down to $\sim 140$ K, fading away on further cooling. This contribution is satisfactorily fitted by a quasi-elastic peak (QEP) 

$$(\chi'')_{QEP}^{B_{2g}}(\omega, T) = A(T)\omega\Gamma(T)/(\omega^2 + \Gamma(T)^2)$$

dashed lines in Figs. 2(a-d)), corresponding to a Lorentzian lineshape for $(\chi'')_{QEP}^{B_{2g}}(\omega, T)/\omega$. It can be seen from Fig. 2(f) that the relatively large noise in the Raman response above $\sim 150$ cm$^{-1}$ have little influence on the determination of the QEP fitting parameters $A(T)$ and $\Gamma(T)$. The Raman response for other symmetries accessible by our experimental setup are given in SM at selected temperatures, also showing contributions from two-magnon scattering.

Figures 3(a) and 3(b) show the $T$-dependence of the Lorentzian $B_{2g}$ QEP area $A$ and width $\Gamma$, respectively. Only data between $\sim 120$ and 200 K are shown, co-
responding to the $T$–interval where this signal is sufficiently strong to warrant reliable Lorentzian fits within our statistics. Figure 2(c) shows $A/\Gamma$, corresponding to the QEP height, while Fig. 2(d) is a zoom in of Fig. 2(b) near $T_S$. Between 280 and 120 K, the $B_{2g}$ QEP area and height show a maximum at $T_{\text{max}} = 143$ K, slightly above $T_{\text{AFM}}$, nearly vanishing below 120 K. Concerning the widths, the $B_{2g}$ QEP gradually sharpens on cooling down to $T_S$. Below $T_S$, $\Gamma_{\text{QEP}}$ further sharpens from $\sim 40$ to $\sim 30$ cm$^{-1}$.

As for the other FeSCs [4, 21, 22, 24–26, 29–31], we ascribe the $B_{2g}$ QEP in LFAO to electronic nematic fluctuations. In principle, the Raman intensity may be dominated either by charge/orbital or spin nematic fluctuations. The significant residual nematic fluctuations observed between $\sim 120$ K and $T_{\text{AFM}}$ (see Figs. 3(a,c)) are consistent with $^{75}$ As NMR measurements that show coexisting AFM and paramagnetic regions in this $T$-interval and attempted scalings of this curve to the $B_{2g}$ QEP height to a Curie-Weiss-like behavior between $T_S$ and $\sim 200$ K, yielding $\theta_{\text{CW}} = 137(3)$ K (see text). The solid line in (d) is a guide to the eyes.

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unit volume is expected to be reduced on cooling, the inverse tendency is found for the remaining tetragonal domains, leading to $T_{\text{max}} < T_S$. Still, the nematic fluctuations in LFAO are clearly sensitive to $T_S$, as demonstrated by the sharpening of the $B_{2g}$ QEP below $T_S$ (see Fig. 3(d)). In fact, this is a manifestation of slower nematic fluctuations in the orthorhombic phase. This is again qualitatively consistent with $^{75}\text{As}$ NMR results that show a slowing down of the magnetic dynamics below $T_S$ [40] and may be also related with the enhancement of the magnetic correlation length below $T_S$ observed in inelastic neutron scattering measurements [32].

We discuss our results considering separately the independent scenarios where charge/orbital or spin nematic fluctuations dominate the intensity of the $B_{2g}$ Raman QEP. Starting with the charge/orbital scenario (scenario A), the bare static nematic susceptibility $\chi_n^{(0)}(T)$ and $(\chi''_{\text{QEP}}(\omega,T))$ are directly connected by a Kramers-Kronig transformation $\chi_{\text{nem}}(T) = (2/\pi) \int_0^\infty (\chi''_{\text{QEP}}(\omega,T))/\omega d\omega$ [21,22], corresponding to the QEP area $A(T)$ in our analysis. An attempted scaling of $\chi_{\text{nem}}(T)$ obtained in this way and the polycrystalline shear modulus $C_S$ extracted from Ref. [11], i.e., $C_S(T)/C_S(300\text{K})=1-bA(T)$ [22], is given in Fig. 4, where $b$ is a free parameter (see footnote [42]). In our analysis, we tentatively varied $b$ to scale $A(T)$ to $C_S(T)$ either at $T \gtrsim T_S$ (empty triangles in Fig. 4) or at $T \sim 200\text{K} \gg T_S$ (filled triangles). However, no value for $b$ yielded a satisfactory scaling for the entire investigated interval $T_S < T \lesssim 200\text{K}$. The lack of scaling between the shear modulus and the QEP area, interpreted under scenario A, indicate that the charge/orbital fluctuations do not drive the structural transition at $T_S$, and an additional electronic nematic degree of freedom, presumably the magnetic one, is driving the phase transitions in LFAO [5]. This reasoning closely follows that presented in Ref. [21] for BaFe$_2$As$_2$.

We now explore the alternative scenario where spin nematic fluctuations dominate the intensity of the $B_{2g}$ Raman QEP (scenario B). In this case, the dynamical electronic nematic susceptibility is not given directly by $(\chi''_{\text{QEP}}(\omega,T))$, and therefore a Kramers-Kronig transformation does not apply to extract $\chi_{\text{nem}}(T)$. Instead, $\chi_{\text{nem}}(T)$ is proportional to the slope of $(\chi''_{\text{QEP}}(\omega,T))$ in the limit $\omega \to 0$ [11,133], namely the QEP height $A(T)/\Gamma(T)$. In this scenario, $\Theta_{\text{m}}=137(3)$ K, obtained from the fit of $A(T)/\Gamma(T)$ to a Curie-Weiss-like behavior $A/\Gamma = C/(T-\Theta_{\text{m}})$ over the interval $T_S < T \lesssim 200\text{K}$ (solid line in Fig. 3(c)), is the bare nematic transition temperature in the absence of the magento-elastic coupling that induces the transition at higher temperatures. Figure 4 displays a scaling of the polycrystalline shear modulus to the peak height, $C_S(T)/C_S(300\text{K})=1-b\ 'A(T)/\Gamma(T)$, showing an excellent agreement for the entire investigated interval. Therefore, independently of the assumption on the detailed nature of the Raman $B_{2g}$ QEP, our analysis supports the scenario where the nematic transition is magnetically driven.

The thermal evolution of the relaxation rate $\Gamma^{B_{2g}}$ provides further insight into the nematic transition. At $T \sim 200\text{K}$ one has $\Gamma^{B_{2g}} \sim 10\text{ meV} (\sim 80\text{ cm}^{-1})$, see Fig. 3(b), which is on the same energy scale of the optical phonons (see Fig. 1). However, the nematic fluctuations slow down continuously on cooling (see Fig. 3(b)). Presumably, as the nematic fluctuation rate become significantly smaller than the typical optical phonon frequencies, local and instantaneous orthonhombic distortions are expected to rise and accompany the electronic nematic correlations. We suggest that at $T_S$ the growing lattice strain caused by the local orthorhombic distortions finally drive the formation of a long-range orthorhombic phase, i.e., the so-called nematic phase. Immediately below $T_S$ the nematic fluctuations are slowed down further (see Fig. 3(d)). This is likely associated with changes in the $J_a$ and $J_b$ nearest-neighbor exchange integrals, partially releasing the magnetic frustration and allowing for increased magnetic correlation lengths [32].

Further inspection of our results gives insight into the large separation between $T_S$ and $T_{\text{AFM}}$ (15 K) compared to their near coincidence in BaFe$_2$As$_2$. We note that at $T = 163\text{K}$, for instance, the maximum of $(\chi''_{\text{QEP}}(\omega,T))$, corresponding to the QEP linewidth $\Gamma$, is 43(2) cm$^{-1}$ for LFAO (see Fig. 2(c) and 3(d)), much smaller than $\sim 100\text{ cm}^{-1}$ for BFA at this temperature [21]. Such slower nematic fluctuations in LFAO preempt the stabilization of orthorhombic domains significantly above $T_{\text{AFM}}$. This scenario may also give insight into the nematic transition of other FeSCs. For instance, a $B_{2g}$ QEP has also been reported [20] for FeSe, which also gradually sharpens on cooling, reaching $\Gamma \sim 30\text{ cm}^{-1}$ at $T_S$, which is comparable to the observed $\Gamma$ for LFAO in the nematic phase (see Fig. 3(d)).

In summary, polarized Raman scattering in LaFeAsO reveals a quasi-elastic $B_{2g}$ scattering channel from nematic fluctuations above $\sim T_{\text{AFM}}$. An analysis of the $T$-dependence of this signal supports the conclusion that magnetism is the primary order parameter driving the phase transitions in this material. Relatively slow electronic nematic fluctuations preempt $T_S$ and arguably signal the separation between $T_S$ and $T_{\text{AFM}}$.

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[35] There is no report on single crystal shear moduli in LFAO, to the best of our knowledge. The polycrystalline shear modulus is expected to become critical and display strong temperature dependence, and in our analysis we assume that it dominates the temperature-dependence of the polycrystalline shear modulus.
Temperature-dependence of the Arsenic \( A_g \) phonon

Figure S1(a) shows the temperature-dependence of the relative intensity of the As mode in \( XY \) with respect to \( YY \) polarizations (see also Fig. 1 of the main text). An enhancement of this mode in \( XY \) polarization is observed below \( T_{AFM} \). This is similar to observed in related systems [1–4] and can be ascribed to the coupling of this phonon with anisotropic electronic states in the magnetic phase. [5] Figures S1(b) and S1(c) show the frequency \( \omega_{0}^{As} \) and linewidth \( \Gamma_{As} \) of this mode. Frequency anomalies are observed at both \( T_S \) and \( T_{AFM} \), the latter being likely due to spin-phonon coupling, [6] while the phonon linewidth shows a maximum at \( T \sim T_{AFM} \) with no anomaly at \( T_S \). The linewidth of this mode at low-\( T \) is resolution-limited, suggesting a high crystalline quality and homogeneity, at least within the relatively small sample region probed by the laser spot.

Extraction of the Raman response from the raw intensities

As discussed in the main text, the raw intensity \( I \) (after subtraction of the dark noise) and the Raman response \( \chi''(\omega,T) \) are related by the expression \( I = (1+n)\chi''(\omega,T) + D \), where \( n \) is the Bose-Einstein statistical factor and \( D \) represents an intensity offset. The offset, which is frequency-independent over the limited spectral interval of interest to this work (\( \omega < 600 \text{ cm}^{-1} \)), may be due to a combination of residual stray light in the spectrograph stage of our instrument and luminescence. \( D \) was subtracted from the raw intensity prior to the Bose-Einstein correction in order to avoid distorting the Raman susceptibility spectra. Non-systematical variations of \( D \) the order of 20 % were observed as the laser spot moved slightly along the sample surface at different temperatures. Figure S2 shows the raw intensity spectra in \( XY \) polarization for \( T = 280 \text{ K} \) and 150 K. At \( T = 280 \text{ K} \), the intensity follows a linear \( \omega \)-dependence down to the lowest frequencies. This indicates that no significant intrinsic Raman intensity is present for \( \omega \rightarrow 0 \) at \( T = 280 \text{ K} \), which otherwise would be highly amplified by the Bose-Einstein statistical factor in the low-\( \omega \) region leading to a non-linear contribution to the raw intensity. Thus, the raw intensity for \( \omega \rightarrow 0 \) at \( T = 280 \text{ K} \) was taken as the offset \( D(280 \text{ K}) \). At \( T = 150 \text{ K} \), a significant quasi-elastic intensity is also observed, associated with the nematic fluctuations (see main text). The offset \( D(150 \text{ K}) \) was then taken as the extrapolation to \( \omega = 0 \) of a linear fit taken in the region \( 300 < \omega < 600 \text{ cm}^{-1} \) (see Fig. S2). This procedure was repeated for all
FIG. S1: Temperature dependence of the As A$_g$ mode: (a) intensity in $XY$ polarization relative to $YY$ polarization ($I_{XY}/I_{YY}$), (b) frequency, and (c) linewidth. The shaded area marks the $T_{AFM} < T < T_S$ interval.

FIG. S2: Raw intensity spectra in $XY$ polarization at $T = 280$ and 150 K. The solid lines show linear fits to the data above 300 cm$^{-1}$ and the arrows mark the extrapolation of these lines at $\omega \to 0$, taken as the intensity offset $D(T)$.

FIG. S3: Raman response $\chi''(\omega,T)$ at selected temperatures for $XY$ (a), $YY$ (b) and $X'Y'$ (c) polarizations. The corresponding symmetries defined according to the tetragonal unit cell are indicated.

investigated temperatures and polarizations.

**Raman response at different polarizations; two-magnon scattering**

Although the focus of this work is on the $B_{2g}$ quasi-elastic peak (QEP) obtained in $XY$ polarization, the electronic Raman signal of LaFeAsO was also investigated for the other accessible linear polarizations. Figures S3(a-c) show the Raman response for $B_{2g}$, $A_{1g} + B_{1g}$ and $B_{1g}$ symmetries, respectively, at selected temperatures and $\omega < 600$ cm$^{-1}$. Besides the phonon modes and the $B_{2g}$ QEP discussed in the main text, an additional signal that increases almost linearly with frequency in this range is observed for all polarizations. Figures S4(a-d) show the spectra at $T = 40$ K over an extended frequency region. Broad peaks were observed at $\sim 2400$ and $\sim 3100$ cm$^{-1}$ for $XY$ and $X'Y'$ polarizations, respectively, while the spectra at $YY$ and $Y'Y'$ polarizations...
seem to contain a combination of these contributions. It is important to notice that the linear component observed for $\omega < 600 \text{ cm}^{-1}$ in $XY$ polarization is actually the lower-frequency limit of this broad scattering. Further insight into the nature of this contribution is gained by the $T$-dependence of the slope of the linear component in the frequency range $300 < \omega < 600 \text{ cm}^{-1}$ in $XY$ polarization, using the data of Fig. 2 of the main text. Notably, this slope increases significantly below $T_N$. Thus, the linear contribution is not associated with nematic fluctuations, and can be safely excluded from the computation of the instantaneous nematic susceptibility. In fact, for $\text{BaFe}_2\text{As}_2$, broad signals centered at high frequencies were also observed and ascribed to two-magnon scattering. This identification is consistent with the temperature-dependence shown in Fig. S5.

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