

# On the origin of the electronic anisotropy in iron pnictide superconductors

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We use polarization-resolved Raman spectroscopy to study the anisotropy of the electronic characteristics of the iron-pnictide parent compounds  $A\text{Fe}_2\text{As}_2$  ( $A = \text{Eu}, \text{Sr}$ ). We demonstrate that above the structural phase transition at  $T_S$  the dynamical anisotropic properties of the 122 compounds are governed by the emergence of xy-symmetry critical collective mode foretelling a condensation into a state with spontaneously broken four-fold symmetry at a temperature  $T^*$ . However, the mode's critical slowing down is intervened by a structural transition at  $T_S$ , about 80 K above  $T^*$ , resulting in an anisotropic density wave state.

The properties of solids are defined by the symmetry of the underlying lattice. For the iron pnictide superconductors, the electronic nematicity, a tendency of the electronic ground state to deform spontaneously in proximity to the tetragonal to orthorhombic structural transition, remains the focus of intense research activity [1–3]. In the 122 family of iron pnictides, below the structural transition temperature  $T_S$  that breaks the four-fold rotational symmetry, the crystal lattice constants  $a$  and  $b$  differ by less than one percent while the anisotropy of the electronic characteristics, such as the DC resistivity and the optical conductivity, can be as large as 30 percent [4–6]. More importantly, even above  $T_S$ , where the underlying lattice remains tetragonal, large electronic nematic susceptibility has been suggested from resistivity anisotropy measurements in a “zero strain” limit [7] and from the observation of a splitting of the electronic bands derived from degenerate iron orbitals [8, 9]. However, the origin of nematicity remains controversial because of the strong interplay of multiple degrees of freedom.

Theoretical proposals suggest that either the magnetic moments of the iron spins, which order in close proximity to  $T_S$  into collinear stripes [3, 10], or the iron  $d$ -orbitals, which below  $T_S$  no longer have the four-fold symmetry, drive the nematicity [11–13]. Because the tetragonal to orthorhombic transition occurs in close proximity to the magnetic ordering transition (at  $T_{SDW}$ ) and leads to superconductivity with an unconventional order parameter, in-depth studies of symmetry and critical dynamics of nematic fluctuations are essential to gain an insight into both magnetism and superconductivity in pnictides [3, 14–17].

In this letter we use Raman spectroscopy to study the evolution of electronic anisotropy for representative parent compounds of the iron pnictide superconductors. We

demonstrate that critical long temporal collective orbital fluctuations of xy-symmetry develop in a broad range of temperatures, foretelling the new ground state of the same symmetry at temperature  $T^*$ . However, condensation into the emergent state is intervened by a first order transition at  $T_S$ , about 80 K above  $T^*$ , characterized by a broken four-fold symmetry, a doubling of the unit cell, an anisotropic density wave, and a collinear spin stripe order. We propose a microscopic interpretation of the observed anomalous evolution of the Raman susceptibility, elucidate reported anisotropy of electronic characteristics above and below  $T_S$ , and explain the implications on superconductivity for doped pnictide materials.

Polarization-resolved inelastic light scattering probes excitations of prescribed symmetries. The temperature evolution of the electronic Raman susceptibility reveals the dynamics of collective excitations and provides an unambiguous identification of their symmetry. Unlike most other symmetry sensitive probes requiring moderate to strong external perturbations, such as magnetic [18] or strain fields [7, 19], the photon field used in Raman scattering is weak. Thus, Raman spectroscopy presents an ideal tool to study the dynamics and symmetry of nematic fluctuations without introducing external symmetry breaking perturbations.

The Raman response function traces electronic density-density correlations driven by the incident and scattered photons of the chosen polarizations ( $e^I$  and  $e^S$  shown as blue and red arrows in Fig. 1)

$$\chi_{e^I e^S}(\omega) \propto -i \int_0^\infty dt e^{i\omega t} \langle [\tilde{\rho}(t), \tilde{\rho}(0)] \rangle. \quad (1)$$

In the effective mass approximation,  $\tilde{\rho} = (m/\hbar^2) \sum_{\alpha,\beta} e_\alpha^I e_\beta^S \sum_{b,\mathbf{k}} [\partial^2 \epsilon_{\mathbf{k}}^b / \partial k_\alpha \partial k_\beta] \hat{n}_{b\mathbf{k}}$ , with  $m$  denoting the electron mass and  $\hat{n}_{b\mathbf{k}}$  the occupation

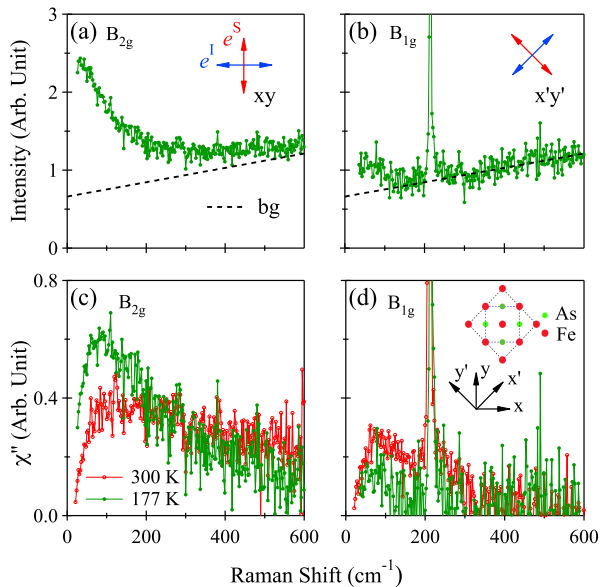


FIG. 1. (color online) Secondary emission (a and b) and Raman response (c and d) for  $\text{EuFe}_2\text{As}_2$  at 177 K (green) and 300 K (red) for  $xy$  (a and c) and  $x'y'$  (b and d) polarization geometries corresponding to  $B_{2g}$  and  $B_{1g}$  responses in the tetragonal phase, recorded with  $\lambda_L=476$  nm. Schematic diagram of the Fe-As layer is shown in (d), with the tetragonal 2-Fe unit cell in blue and the orthorhombic 4-Fe unit cell in black. The light polarization vectors are denoted with respect to the crystallographic orientations. The luminescence background indicated by black dotted lines in (a) and (b) is determined in Refs. [24, 25].

of the Bloch state at momentum  $\mathbf{k}$  in the band  $b$  with the dispersion  $\epsilon_{\mathbf{k}}^b$  [20–22]. All components of the symmetrized Raman tensor  $\chi_{e^i e^s}$  can be classified by irreducible representations of the crystallographic point group [23]. The two photon fields with cross polarization couple to quadrupole moments, and thus the scattering experiments probe the  $B_{2g}$  susceptibility if light polarization is aligned along  $a$  and  $b$  crystallographic directions, and the  $B_{1g}$  susceptibility if rotated by  $45^\circ$ .

For the Fe pnictides, a local charge transfer between degenerate  $d_{x'z}$  and  $d_{y'z}$  orbitals without a spin flip induces a quadrupole moment of  $B_{2g}$  symmetry on an iron site, Fig. 3(b), while no low energy excitation of  $B_{1g}$  symmetry is allowed without charge transfer between the irons. Therefore,  $\chi_{xy}$ , which bears four nodes along the crystallographic directions, delineates such singlet quadrupole excitations [21, 22]. An excitation with a spin flip excites a magnon to which, in the leading order, light cannot couple. In Figs. 3(a-d) we illustrate the relevant quadrupole excitation transition both in  $k$ -space and real space.

In Figs. 1(a) and 1(b) we compare the intensity measured in  $xy$  and  $x'y'$  geometries [24] from  $\text{EuFe}_2\text{As}_2$  pnictide just above  $T_S = 175$  K. The Raman response shown in Figs. 1(c), 1(d) and 2 is derived from the scattering

intensity by accounting for the Bose factor [25].

As shown in Fig. 1(d), the electronic response from intraband excitations using the  $x'y'$  polarization is indeed very weak. The broad feature centered at about  $75 \text{ cm}^{-1}$  is likely due to an interband transitions between the  $\alpha$  and  $\beta$  bands, as illustrated in Fig. 3 (a). The sharp mode at  $214 \text{ cm}^{-1}$  is an iron  $B_{1g}$  phonon [26] which only shows weak anomaly upon cooling across  $T_S$  [24].

In contrast, the response for the  $xy$  polarization is much stronger, the signal extends beyond  $1000 \text{ cm}^{-1}$ , it strengthens significantly upon cooling and develops a well defined maximum at low energy. In Fig. 2 we show that the temperature evolution of the  $B_{2g}$  Raman susceptibility for  $\text{EuFe}_2\text{As}_2$  and  $\text{SrFe}_2\text{As}_2$  is generic for the 122 compounds [27]. Upon cooling toward  $T_S$ , the data show gradual enhancement of the low energy response along with linear in temperature softening of the mode maximum frequency from about  $160 \text{ cm}^{-1}$  at room temperature to  $90 \text{ cm}^{-1}$  at  $T_S$ , denoted  $\omega_{max}(T)$  in Figs. 2 (d-e), an energy scale independent on the quasi-particle characteristics. No enhancement with cooling is observed for the susceptibility in any other symmetry channel. Such critical evolution of the anomalous  $B_{2g}$  susceptibility due to quadrupole deformation of the electron density at the Fermi surfaces is abruptly intervened at  $T_S$ , the entrance into the orthorhombic phase, when a density wave like gap and a coherence peak at about  $1070 \text{ cm}^{-1}$  develops. The power law in the density wave gap structure suggests that the gap is anisotropic, possibly with a  $d_{xy}$  symmetry.

From these data, we calculate and plot in Figs. 2 (d-e) the real part of the static Raman susceptibility using the Kramers-Kronig relations.  $\chi'_{xy}(0, T) \propto (T - T^*)^{-1}$  shows a mean-field-like enhancement with temperature  $T^*$  about 80 K below  $T_S$ . Remarkably, the mode's maximum frequency  $\omega_{max}(T)$  scales to the very same  $(T - T^*)$  behavior and does not soften near  $T_S$ , indicating that the critical slow down is intervened abruptly at  $T_S$ .

The  $B_{2g}$ -susceptibility data cannot be understood in a picture of non-interacting incoherent quasi-particle excitations because in a paramagnetic phase such excitations can only produce a featureless low frequency response. The spectrum of low-frequency anisotropic plasmas is typically featureless and has a cutoff at  $4\pi\hbar v_F/\lambda_L$  due to kinematic constraints of the scattering process, where  $\lambda_L$  is the excitation wavelength and  $v_F$  is Fermi velocity in the direction of photon propagation, relatively small for the layered iron pnictides [28, 29]. Hence, the presented  $\chi''_{xy}(\omega, T)$  data showing the emergence and softening of an anomalous quasi-elastic response above  $T_S$  is a manifestation of electronic correlations.

To describe the observed collective behavior we use an expression for interacting susceptibilities

$$\chi_{xy}(\omega, T) \propto \frac{\chi_{xy}^{(0)}(\omega, T)}{1 - g\chi_{xy}^{(0)}(\omega, T)} \quad (2)$$

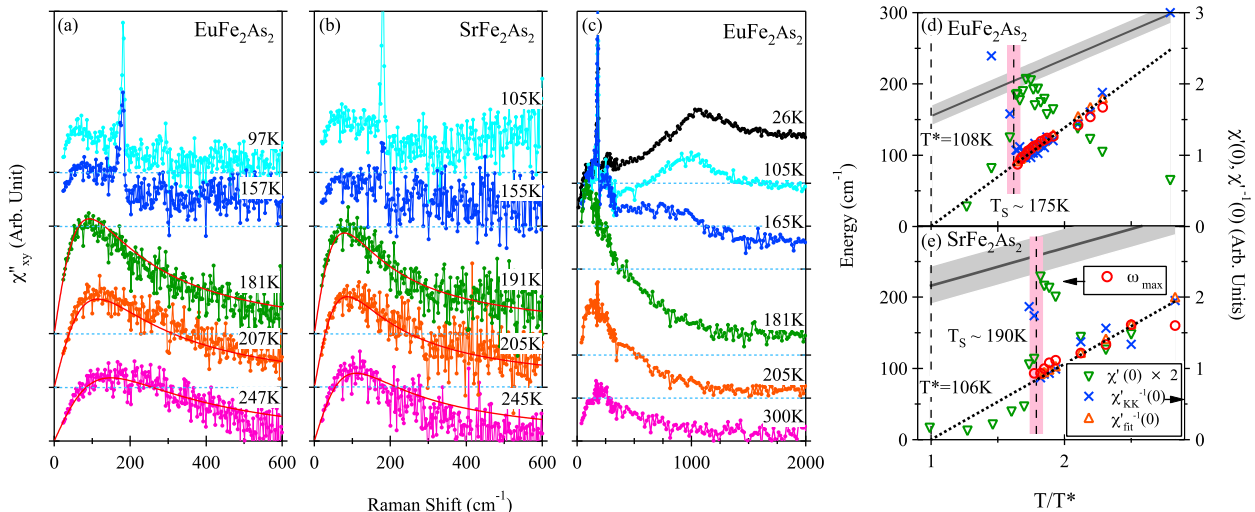


FIG. 2. (color online) Evolution of the Raman response for the  $xy$  polarization with cooling from the tetragonal into the orthorhombic phase (a and c) for  $\text{EuFe}_2\text{As}_2$  and (b) for  $\text{SrFe}_2\text{As}_2$ . The red curves are the fits to the phenomenological model Eq. (2) [24, 25]. The data show the development of an anomalous quasi-elastic peak above  $T_S$ , its abrupt disappearance at  $T_S$ , followed by the formation of a density wave gap with a coherence mode at about  $1070 \text{ cm}^{-1}$  and a new phonon at  $183 \text{ cm}^{-1}$ . Laser excitations  $\lambda_L$  of  $476 \text{ nm}$  (a and b) and  $647 \text{ nm}$  (c) were used. The temperature dependence of the  $\chi''_{xy}(\omega, T)$  response maximum  $\omega_{max}(T)$  (red circles), of the relaxation rate  $\Gamma_T$  (solid lines, shaded areas indicate the fitting error), the static Raman susceptibility  $\chi''_{xy}(0, T)$  (green triangles),  $\chi''_{KK}(0, T)$  from data (blue crosses), and  $\chi''_{fit}(0, T)$  from fit (orange triangles) for  $\text{EuFe}_2\text{As}_2$  (d) and  $\text{SrFe}_2\text{As}_2$  (e). The horizontal axis is scaled by  $T^*$ . The vertical black dashed lines at  $T/T^* = 1.6$  and  $1.8$  denote  $T_S$ , while the red shaded areas indicate a hysteresis.

where  $\chi_{xy}^{(0)}$  is the non-interacting susceptibility, and  $g$  is the coupling constant. We conjecture

$$\chi_{xy}^{(0)}(\omega, T) = \frac{C}{\pi} \log \frac{(\omega + i\Gamma_T)^2 - \Lambda^2}{(\omega + i\Gamma_T)^2}, \quad (3)$$

as this form yields expected featureless bare response  $\chi_{xy}^{(0)}(\omega, T) \propto \arctan(\omega/\Gamma_T)$ . Here  $\Gamma_T$  describes a temperature dependent electron-hole relaxation rate  $\gtrsim 2k_B T$  and  $\Lambda$  is the ultraviolet cutoff. At  $\omega \lesssim \Gamma_T$  Eqs. (3) and (2) yield the relaxational form

$$\chi_{xy}''(\omega, T) \propto \omega \Gamma_T / (\omega_{max}(T)^2 + \omega^2), \quad (4)$$

with  $\omega_{max}(T) = \Gamma_T [1/\tilde{g} - \log(\Lambda/\Gamma_T)]$  and  $\tilde{g} = Cg$ . Because  $\chi_{xy}^{(0)}(0, T)$  is logarithmically large, Eq. (2) guarantees the observed critical temperature dependence of the static susceptibility  $1/\chi_{xy}(0, T) \propto (1/\chi_{xy}^{(0)}(0, T) - g)$  with  $T^*$  defined by the coupling constant  $g$ ,  $\chi_{xy}^{(0)}(0, T^*) = 1/g$ .

The expression (2) enables us to fit the entire temperature evolution of the  $\chi_{xy}(\omega, T)$  susceptibility between the room temperature and  $T_S$  [24]. The data show critical enhancement of the susceptibility and slow down of the characteristic fluctuation frequency  $\omega_{max}(T)$ , which is abruptly intervened by the structural transition at  $T_S \approx 1.6 - 1.8 T^*$  (See Fig. 2). Above  $T_S$ , the observed anomalous temperature dependence of the  $B_{2g}$  susceptibility is arising from the critical collective fluctuations indicating the system's approach to a phase transition

breaking the four-fold symmetry. The susceptibility reveals how, counteracting the relaxation processes, long temporal correlations characterized by the same symmetry develop, foretelling inception of the new ground state at temperature  $T^*$ .

We now turn to the microscopic interpretation of the observed anomalous  $\chi_{xy}(\omega, T)$ . The body centered unit cell of 122 structure contains two Fe layers with two Fe sites in each one. In the long wavelength limit,  $B_{2g}$  and  $A_{2u}$  are two possible symmetries of the quadruple excitations, as illustrated in Fig. 3(c-d). The only Raman active quadrupole excitations are singlets of  $\Gamma_4^+$  ( $B_{2g}$ ) symmetry. Those quadrupole excitations precipitate the observed anomalous susceptibility. The critical behavior of  $\chi_{xy}(\omega, T)$  foretells that the system contemplates a phase transition at temperature  $T^*$  which would break the four-fold symmetry without a structural density wave instability and establish the ground state depicted in Fig. 3(c).

For the momentum at the Brillouin zone (BZ) corner, the  $T_S$  structural instability vector  $X = (\pi/a, \pi/a, 2\pi/c)$ , both the singlet quadrupole excitations and the spin triplet excitations obey a degenerate  $X_5^-$  symmetry, as illustrated in Figs. 3(e-h). The quadrupole-quadrupole and/or spin-spin interaction between nearest and next-nearest Fe sites favors such collinear stripe order as its ground state [30, 31]. Coupling of quadrupole and spin excitations to the lattice leading to a structural phase transition is naturally expected for non-symmetric fluc-

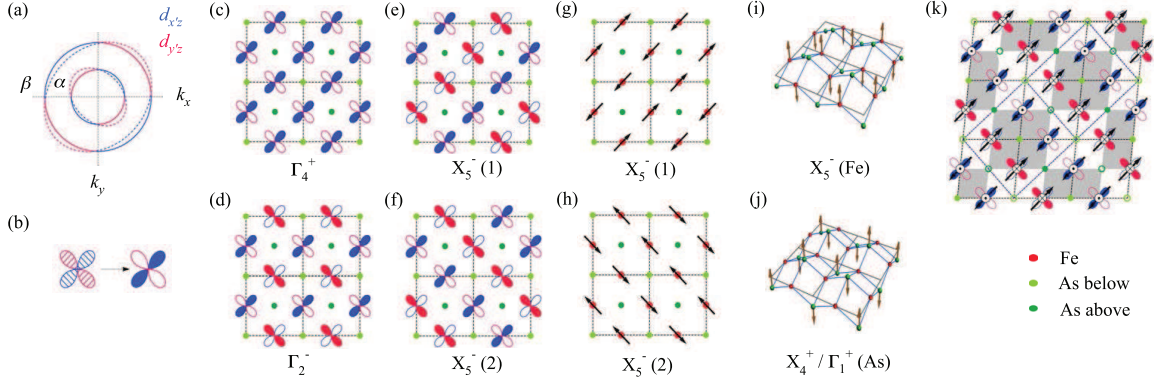


FIG. 3. (color online) (a) Schematic diagram of the  $\alpha$  (inner) and  $\beta$  (outer) Fermi surfaces at the BZ center  $\Gamma$ -point. The Fermi surfaces are distorted along the  $x'$  and  $y'$  directions due to the charge transfer fluctuations between the  $d_{x'z}$  and  $d_{y'z}$  orbitals. (b) A quadruple moment is caused by the charge transfer between the  $d_{x'z}$  and  $d_{y'z}$  orbitals. (c) and (d) Two possible long wavelength ( $\Gamma$ -point) configurations of quadrupoles with  $\Gamma_4^+$  ( $B_{2g}$ ) and  $\Gamma_2^-$  ( $A_{2u}$ ) symmetries. In the second Fe layer in the unit cell the  $d_{x'z}$  and  $d_{y'z}$  orbitals have the same/reversed occupation with the first layer for  $B_{2g}$  and  $A_{2u}$ , respectively. (e) and (f) The degenerated quadruple excitations that belongs to the  $X_5^-$ -symmetry at the corner. The second Fe layer has the reversed with the first layer orbital occupation. (g) and (h) The spin triplet excitation that belongs to the  $X_5^-$ -symmetry at the BZ corner. The second Fe layer has the reversed spin with the first layer. (i) and (j) The atomic displacement of the  $X_5^-$  type of Fe and  $X_4^+$  ( $D_{4h}$  phase)/ $\Gamma_1^+$  ( $D_{2h}$  phase) type of As shown in one Fe layer. The second Fe layer has the anti-phase/in-phase displacement with the first layer. (k) The possible quadruple, spin and lattice configuration in the orthorhombic ground state below  $T_S$ . The Fe  $X_5^-$ -symmetry displacement. Dots and crosses denote the Fe  $X_5^-$ -symmetry displacement up and down along the  $c$ -direction. The orthorhombic unit cell is shown by the blue dashed line. As ions above and below the Fe layer are distinguished in different colors. The shaded grey rhombuses show a checkerboard charge modulation of approximate  $xy$  symmetry.

tuations. If, as it is here, the electronic state is degenerate and can interact with a degenerate lattice displacement, the Jahn-Teller theorem predicts the lifting of the  $X_5^-$  quadrupole state degeneracy by interaction with  $X_5^-$  phonons [32]. The Fe  $X_5^-$  phonon mode, illustrated in Fig. 3(i), buckles the flat Fe layer, modulates the distance between quadrupoles and/or spins and therefore promotes the choice of quadrupole and spin collinear stripe order. The solution of vibronic cooperative  $X_5^- \otimes X_5^-$  Jahn-Teller problem results in a new ground state shown in Fig. 3(k). When the lattice symmetry breaks, the quadrupole and spin collinear stripe orders lock-in. Hence, for the parent compounds, the first order structural transition with doubling of the unit cell at  $T_S$ , 80 K above  $T^*$ , intervenes the formation of the contemplated long-range ordered electronic state. The new order induces long-range charge modulation observed at low temperatures as the  $d_{xy}$ -symmetry density wave gap. The distortion of the As layer is induced by coupling to the new electronic ground state of the same symmetry, a quadrupole- and/or magneto-striction, and is revealed by the appearance of the new Raman active As phonon mode below  $T_S$  in the spectra of  $xy$  polarization, Fig. 2(a-c) and 3(j). As the lattice modulations are buckle-like, the induced anisotropy of the lattice parameters remains weak.

The results obtained here for parent compounds might

bear implications on superconductivity in the Fe pnictides. As superconductivity evolves from the tetragonal phase where the  $B_{2g}$ -type fluctuations remain dynamic and strong, they may mediate the superconducting pairing of unconventional symmetry, or appear as long-lived in-gap collective modes.

In summary, we use polarization-resolved Raman spectroscopy to study the emergence and the competition of the collective orbital, spin and lattice fluctuations in  $\text{EuFe}_2\text{As}_2$  and  $\text{SrFe}_2\text{As}_2$  parent compounds of the iron pnictide superconductors. In the  $xy$  scattering geometry the Raman response exhibits a broad excitation spectrum with a peak at low energy showing critical behavior in a wide temperature range above the structural transition. This spectrum reveals dynamics of the relaxational mode due to orbital fluctuations of  $B_{2g}$ -symmetry and demonstrates the general interrelation between anticipated phase transitions breaking the discrete symmetry and critical fluctuations characterized by the same symmetry. For the studded parent compound the condensation of the emergent  $B_{2g}$ -mode is intervened by a structural density wave transition characterized by a doubling of the unit cell and a collinear spin stripe order.

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