In-plane electronic anisotropy resulted from ordered magnetic moment in iron-based superconductors

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We study the in-plane electronic anisotropy in the parent compounds of several families of Fe-based superconductors (BaFe₂As₂, EuFe₂As₂, NaFeAs, LiFeAs, FeSe, and LaFeAsO) by polarization-resolved Raman scattering. We measure intensity of the fully symmetric c-axis vibration of As atom mode in the XY scattering geometry and notice that the mode’s intensity is significantly enhanced below the magnetostructural transition only for compounds showing magnetic ordering. In particular, we find that the intensity ratio of this As phonon in the XY vs. XX scattering geometries is proportional to the square of the ordered magnetic moment. We relate this As phonon intensity enhancement below the Néel temperature in iron pnictides to in-plane electronic anisotropy induced by the collinear spin-density wave order.

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The lattice, orbital, and magnetic degrees of freedom are strongly coupled in the Fe-based superconductors. This is best evidenced by the observation, in most parent compounds, of a magnetic transition from paramagnetic to collinear antiferromagnetic (AFM), occurring at a temperature \( T_N \) slightly below the temperature \( T_S \) at which a structural transition from tetragonal to orthorhombic phase occurs. The interplay between these degrees of freedom is complex and led to a chicken-egg problem for which there is still no consensual view [1,2].

The electronic structure is directly affected by an electronic band folding accompanied by the formation of a collinear spin-density wave (SDW) gap [3–6]. As a result, a significant electronic anisotropy was found for properties measured along the two planar orthogonal Fe-Fe directions (Fig. 1) below the magnetostructural transition, notably in electrical transport [7], optical conductivity [8,9], thermopower [10], local density-of-states (DOS) imaging [11], and quasiparticle band dispersions [12].

Raman scattering offers a unique way to study the electronic anisotropy below the magnetostructural transition of the iron-based superconductors [6,13]. For example, one can study the interband transitions along the two planar orthogonal Fe-Fe directions (Fig. 1) in a detwinned sample [6] or investigate the As fully symmetric phonon in a twinned sample [14].

In relation to this study, it has been noticed that the Raman coupling vertex to the As fully symmetric phonon: the c-axis vibration of As atom, which modulates the Fe-As bond angle of the Fe-As tetrahedra (Fig. 1), is forbidden for \( XY \) scattering geometry in the tetragonal phase, whereas the coupling becomes finite in the orthorhombic phase. However, because orthorhombicity of the lattice constants \( \delta \) for all studied materials is weak, the emerging As mode’s intensity due to geometrical lattice anisotropy is expected to be small. Surprisingly, a significant intensity of the As phonon in the nearly forbidden \( XY \) scattering geometry has been observed for Ba(Fe₁−χAu)_₂As₂ below \( T_N \), in contrast to the weak signals at temperatures between \( T_S \) and \( T_N \) [14]. Similar results were reported for Ba(Fe₁−χCo)_₂As₂ [15]. In addition, the temperature dependence of integrated As phonon intensity was reported to be proportional to square of the magnetic moment \( M(T)^2 \) below \( T_N \) [14]. The origin of this anomalous intensity enhancement was related to the in-plane electronic anisotropy induced by the collinear SDW order.
the 122 system, a detailed study of this relation among temperature and doping dependence of the As phonon in anisotropy and the magnetic order parameter by studying the we observe a strong intensity for the fully symmetric As mode conductors. For all compounds showing magnetic ordering, that temperature dependence of the Raman vertex amplitude is proportional to magnitude of the magnetic order parameter

\[ M \propto |M| \]

Furthermore, the Fano model analysis of data revealed the geometrical lattice anisotropy, and ordered magnetic moment

\[ \frac{1}{T} \propto \frac{1}{T^2} \]

Single crystals of materials listed in Table I were grown in-plane electronic anisotropy. As shown in Fig. 2(a), sharp Raman phonon peaks at 186 cm\(^{-1}\) and 237 cm\(^{-1}\), corresponding to a \( A_{1g} \) and \( B_{2g} \) modes, respectively, are detected in the XX scattering geometry. However, as expected for the tetragonal structure of LiFeAs, these modes have no intensity in the XY scattering geometry. Similar Raman results are reported for the tetragonal \( FeSe_{1+y}Te_{0.2}Se_{0.4} \) single crystal [30].

If anisotropy develops in the orthorhombic phase, the \( A_{1g} \) anion mode may acquire a finite intensity [\( (\bar{a}' - \bar{b}')/2 \)] in the XY scattering geometry related to the anisotropy of the in-plane polarizability associated to this \( A_{1g} \) anion mode, because \( \bar{a}' \) and \( \bar{b}' \) are the polarizability derivatives along the two Fe-Fe orthogonal directions (\( X' \) and \( Y' \)) in the orthorhombic phase. Since the lattice anisotropy \( \delta \) is small (Table I), the intensity due to geometrical anisotropy is expected to be weak. For example, for the FeSe material, which exhibits a structural phase transition at 90 K [31,32] but no long-range magnetic ordering, we observe a \( A_{1g}(Se) \) phonon at 180 cm\(^{-1}\) and a \( B_{1g}(Fe) \) phonon at 208 cm\(^{-1}\) for the XX polarization [Fig. 2(b)]. Although the intensity of the \( A_{1g}(Se) \) phonon with the \( XY \) polarization is finite at 20 K [inset of Fig. 2(b)], it is only 2% of the corresponding intensity recorded for the XX polarization (Table I) [13,33].

**Table I.** Summary of \( T_N \), \( T_N \) (in Kelvin), lattice anisotropy [\( \delta = (a-b)/(a+b) \)], intensity ratio of \( A_{1g} \) phonon in \( XY \) vs. \( XX \) geometries, and ordered magnetic moment/Fe M (in \( \mu_B \)) for compounds studied in this paper.

| Sample      | \( T_N/T_S \) | \( \delta (\%) \) | \( I_{XY}/I_{XX} \) | M  |
|-------------|---------------|------------------|--------------------|----|
| \( FeSe \)  | 175/175       | 0.5 [22]         | 3.3                | 0.98 [23] |
| \( BaFe_2As_2 \) | 135/135     | 0.4 [24]         | 3.1                | 0.87 [25] |
| \( LaFeAsO \) | 155/157      | 0.24 [20]        | 0.54               | 0.36-0.6 [25] |
| \( NaFeAs \) | 55/40        | 0.18 [26]        | 0.16               | 0.09 [25] |
| \( FeSe \)  | 90/1        | 0.25 [27]        | 0.017              | –   |
| \( LiFeAs \) | –/–          | 0.01             | 0                  | –   |
In contrast, BaFe$_2$As$_2$ with strong magnetic ordering clearly shows the 181 cm$^{-1}$ $A_g$(As) mode [13,15,34,35] in the $XY$ scattering geometry below $T_N$ [Fig. 2(c)]. Similar observation is made for NaFeAs [Figs. 2(e) and 2(f)], which also encounters both a structural and a magnetic phase transition: (i) We observe only a weak intensity between $T_S$ and $T_N$, and (ii) the 162 cm$^{-1}$ $A_g$(As) phonon mode appears in the $XY$ spectra only below $T_N$. LaFeAsO [36–38] is another system with split structural and magnetic phase transitions. In this case as well, we detect sizable intensity for the $A_{g}^1$ (in-phase La and As) mode at 166 cm$^{-1}$ and the $A_{g}^2$ (out-of-phase La and As) mode at 209 cm$^{-1}$ in the $XY$ scattering geometry below $T_N$ [Fig. 2(d)].

To quantify the intensity of the $A_g$(As) phonon in the $XY$ scattering geometry below $T_N$ in different families of Fe-based superconductors, we study the ratio between the $A_g$(As) mode intensity in the $XY$ and $XX$ scattering geometries $I_{XY}/I_{XX}$. This ratio is proportional to $|\langle \hat{a}' - \hat{b}' \rangle/\langle \hat{a}' + \hat{b}' \rangle|^2$, which is a direct measure of the in-plane polarizability anisotropy of the $A_g$(As) mode. Based on Table I, the ratio $I_{XY}/I_{XX}$ is significant only for compounds with long-range magnetic ordering. For example, the ratio $I_{XY}/I_{XX}$ is 300% for BaFe$_2$As$_2$, 16% for NaFeAs and 50% for LaFeAsO, as compared to 2% for FeSe, i.e., 1–2 orders of magnitude smaller. Such behavior cannot be solely explained by weak geometrical lattice orthorhombicity $\delta$, instead, the observation relates the mode’s intensity to magnetic order parameter. This is best evident from Fig. 2(g), where we show that the $I_{XY}/I_{XX}$ ratio of the $A_g$(As) phonon intensity for different Fe-based families is proportional to the square of the magnetic moment/Fermi level.

In conclusion, we revealed a significant intensity enhancement of the emergent $A_g$(As) phonon mode in the $XY$ scattering geometry below $T_N$ only for parent compounds of Fe-based superconductors showing magnetic order. We demonstrate that the ratio of the As phonon intensity in the $XY$ and $XX$ scattering geometries $I_{XY}/I_{XX}$ is proportional to the square of the magnetic ordered moment $M^2$. We conclude that the generic $A_g$(As) phonon intensity enhancement below $T_N$ in iron pnictides is due to the in-plane electronic anisotropy induced by the collinear SDW order: a larger ordered moment in the magnetic phase results in larger in-plane electronic anisotropy, which in turn causes larger As phonon intensity ratio $I_{XY}/I_{XX}$ below $T_N$.

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