Magneto-elastic coupling in Fe-based superconductors

S.-F. Wu,1,2,3, † W.-L. Zhang,1 V. K. Thorunvalde,1 G. F. Chen,2,4 G. T. Tan,5 P. C. Dai,5,6 Y. G. Shi,2 C. Q. Jin,2,4 T. Shibauchi,7 S. Kasahara,8 Y. Matsuda,8 A. S. Sefat,9 H. Ding,2,3,4 P. Richard,2,3,4,‡ and G. Blumberg1,10,†

1Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08854, USA
2Beijing National Laboratory for Condensed Matter Physics, and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
3School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China
4Collaborative Innovation Center of Quantum Matter, Beijing, China
5Center for Advanced Quantum Studies and Department of Physics, Beijing Normal University, Beijing 100875, China
6Department of Physics and Astronomy, Rice University, Houston, Texas 77005, USA
7Department of Advanced Materials Science, University of Tokyo, Kashiwa, Chiba 277-8561, Japan
8Department of Physics, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan
9Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
10National Institute of Chemical Physics and Biophysics, 12618 Tallinn, Estonia

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We used polarization-resolved Raman scattering to study the magneto-elastic coupling in the parent compounds of several families of Fe-based superconductors (BaFe2As2, EuFe2As2, NaFeAs, LiFeAs, FeSe and LaFeAsO). We observe an emergent Ag-symmetry As phonon mode in the scattering geometry whose intensity is significantly enhanced below the magneto-structural transition only for compounds showing magnetic ordering. We conclude that the small lattice anisotropy is insufficient to induce the in-plane electronic polarizability anisotropy necessary for the observed phonon intensity enhancement, and interpret this enhancement below the Néel temperature in terms of the anisotropy of the magnetic moment and magneto-elastic coupling. We evidence a Fano lineshape in the XY scattering geometry resulting from a strong coupling between the Ag(As) phonon mode and the B2g symmetry-like electronic continuum. Strong electron-phonon coupling may be relevant to superconductivity.

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 TN slightly lower than the temperature TS at which a structural transition from tetragonal to orthorhombic phase occurs upon cooling. 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 the structural and magnetic transitions, notably through nematic transport properties [3–5], as well as by an electronic band folding accompanied by the formation of a spin-density-wave gap [6–9].

The As height and the related Fe-As-Fe angle are widely believed to play crucial roles in shaping the magnetic and electronic properties of the Fe-based superconductors [10–23]. Both parameters are modulated by the c-axis motion of the As atom corresponding to a fully symmetric phonon mode (A1g) [24–30]. First-principles calculations show that the inclusion of the Fe spin ordering in the calculation of the As phonon mode frequency allows a good agreement with the energy of the As phonon density-of-states measured by neutron scattering [31–37], suggesting significant magneto-elastic coupling.

Raman scattering can directly probe the As phonon behavior upon cooling across the magneto-structural transitions. As a signature of the magneto-elastic coupling, a finite intensity of the As phonon in nearly forbidden scattering geometries below the magneto-structural transition has been reported in CaFe2As2 [38], EuFe2As2 [39], Ba(Fe1−xCox)2As2 [40–42], Ba(Fe1−xAu2x)2As2 [43], LaFeAsO [44]. In particular, the phonon shows an asymmetric line-shape below TN in Ba(Fe1−xCo2x)2As2, suggesting strong magneto-elastic coupling [40, 41]. However, the details behind this behavior, and its possible link to superconductivity, have not been stated satisfactorily. It has long been suggested that the electron-electron correlations enhance the electron-phonon coupling in the Fe-based superconductors and that the fully symmetric As vibration is related to the superconducting properties [22, 30, 45, 46].

In this Letter, we use polarized Raman scattering to study the temperature dependence of the magneto-elastic coupling for the fully symmetric phonon associated with the c-axis motion of the As atom for typical “122”, “111”, “1111” and “11” systems of Fe-based superconductors. For all compounds showing magnetic ordering, we observe strong intensity for the nearly forbidden XY scattering channel as a result of significantly enhanced anisotropy of the in-plane electronic polarizabil-
ity, while no such enhancement is found for compounds without magnetically-ordered state. Because the lattice anisotropy $\delta = (a - b)/(a + b)$ below $T_S$ is relatively small, we conclude that magneto-elastic coupling below $T_N$ is essential. We interpret the $A_g$ phonon intensity enhancement below $T_N$ in terms of strong coupling to the anisotropic in-plane magnetic moment. The study of the polarization dependence of the As phonon suggests that the mode is coupled to the non-symmetric $XY$-like electronic continuum. The asymmetric line-shape of $A_g$ phonon is described by a Fano model with a magneto-elastic coupling constant proportional to the magnetic order parameter. As the coupling between the $XY$-like electronic continuum and magnetism may survive in the superconducting compounds, our results emphasize the role played by the electron-phonon coupling in enhancing $T_c$.

Single crystals of materials listed in Table I were grown as described in Refs. [47–52]. The corresponding structural phase transition temperature ($T_S$) and magnetic phase transition temperature ($T_N$) are summarized in Table I. Raman measurements on BaFe$_2$As$_2$, NaFeAs, EuFe$_2$As$_2$, LiFeAs, FeSe were performed using the spectrometer described in Refs. [39, 53]. The measurements on LaFeAsO were performed in a back scattering geometry using a T64000 triple-stage spectrometer.

The phononic Raman scattering intensity is proportional to $I \propto |\tilde{\epsilon}_{g} \cdot \mathbf{R} \cdot \tilde{\epsilon}_{g}|^2$, where $\tilde{\epsilon}_{g}$ and $\tilde{\epsilon}_{g}$ are the polarization unit vectors of the incoming and scattering light, respectively, and $\mathbf{R}$ is the Raman tensor [62]. For the $D_{4h}$ point group the $XX$, $XY$, $X'Y'$ and $X'Y'$ polarization geometries probe $A_{1g} + B_{1g}$, $A_{2g} + B_{2g}$, $A_{1g} + B_{2g}$ and $A_{2g} + B_{1g}$ symmetry excitations, respectively. In the orthorhombic phase with $D_{2h}$ point group symmetry, the unit cell rotates by $45^\circ$; the $A_{1g}$ and $B_{2g}$ representations of the $D_{4h}$ point group merge into the $A_g$ representation of the $D_{2h}$ point group, and $A_{2g}$ and $B_{1g}$ ($D_{4h}$) merge into $B_{1g}$ ($D_{2h}$). In the orthorhombic phase, the $XX$ and $XY$ polarization geometries probe $A_g + B_{1g}$ and $A_g$ symmetry excitations, respectively [9].

Before investigating the behavior of the $A_{1g}/A_g$ symmetry As phonon across the magneto-structural transi-

| Sample          | $T_S/T_N$ (Kelvin) | $\delta$ (%) | $I_{XX}/I_{XY}$ | $M$ |
|-----------------|--------------------|--------------|-----------------|-----|
| EuFe$_2$As$_2$  | 175/175            | 0.5          | 0.98            | 55  |
| BaFe$_2$As$_2$  | 135/135            | 0.4          | 0.87            | 57  |
| NaFeAs          | 55/40              | 0.18         | 0.09            | 57  |
| LaFeAsO         | 155/137            | 0.24         | 0.36            | 57  |

*TABLE I.* Summary of $T_S$, $T_N$ (in Kelvin), lattice orthorhombicity ($\delta = (a - b)/(a + b)$), intensity ratio of $A_g$ phonon in $XY$ and $XX$ geometries, and ordered magnetic moment/Fe M (in $\mu_B$) for compounds studied in this manuscript.

![FIG. 1.](image)

(Color online) (a) Definition of the crystallographic directions in the tetragonal 2-Fe unit cell above $T_S$ (light red shaded area) and 4-Fe orthorhombic magnetic unit cell below $T_N$ (black solid lines). (b) Schematic diagram of the magnetic structure. Red arrows: Fe local moments forming collinear AFM order. Blue arrows: $c$-axes vibrations of the fully symmetric As phonon mode. The red and black solid lines illustrate the super-exchange paths of the nearest Fe neighbors, $J_{1a}$ and $J_{1b}$. The green solid lines illustrate the super-exchange path of the next-nearest Fe neighbors, $J_2$.

Accordingly, the $A_{1g}$-symmetry mode is forbidden in the $XY$ scattering geometry in the tetragonal phase. This is the case for LiFeAs, which shows no structural nor magnetic transition. As shown in Fig. 2(a), sharp Raman phonon peaks at 186 cm$^{-1}$ and 237 cm$^{-1}$, corresponding to a $A_{1g}$(As) and a $B_{1g}$(Fe) 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. If anisotropy develops in the orthorhombic phase, the $A_g$ anion mode may acquire a finite intensity $|\langle \tilde{\alpha}' - \tilde{\beta}' \rangle |^2$ in the $XY$-scattering geometry related to the anisotropy of the in-plane polarizability associated to this $A_g$ anion mode because $\tilde{\alpha}'$ and $\tilde{\beta}'$ are the polarizability derivatives along the two Fe-Fe orthogonal directions ($X'$ and $Y'$) in the orthorhombic phase. Since the lattice orthorhombicity $\delta$ is small (Table I), the intensity is expected to be weak. For example, for the FeSe material, which exhibits a structural transition at 90 K [63, 64] but no long-range magnetic ordering, we observe a $A_g$(Se) phonon at 180 cm$^{-1}$ and a $B_{1g}$(Fe) phonon at 208 cm$^{-1}$ for the $XX$ polarization. Although the intensity of the $A_g$(Se) phonon with the XY polarization is finite at 20 K, it is only 2% of the corresponding intensity recorded for the $XX$ polarization [Table I].

In contrast, BaFe$_2$As$_2$ with magnetic ordering clearly shows the 181 cm$^{-1}$ $A_g$ (As) mode [42, 65–67] 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 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 [44, 68, 69] (Fig. 2(d)) is another system with split $T_S$ and $T_N$ transitions. In this case as well, we detect sizable intensity for the $A_g$(in-phase La and As) mode at 166 cm$^{-1}$ and the $A_g$(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) peak intensity in the XY and XX scattering geometries $I_{XY}/I_{XX}$. This ratio is proportional to $(\langle a′ - b′\rangle/\langle a′ + b′\rangle)^2$, which is a direct measure of the in-plane polarizability anisotropy of the $A_g$(As) mode. Based on Table 1, 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 to 2 orders of magnitude smaller. Such behavior cannot be solely explained by weak lattice orthorhombicity $\delta$, and indicates that the intensity of the $A_g$(As) phonon in the XY scattering geometry is mainly controlled by the magneto-elastic coupling, for which we argue that the strength originates from the anisotropy of the magnetic interactions in the Fe-As plane that are modulated by the $c$-axis motion of the As atoms. We can estimate the strength of the magneto-elastic coupling by comparing the $I_{XY}/I_{XX}$ intensity ratios in the magnetically-ordered compounds to that in FeSe. As compared to FeSe, the coupling strength values are 200, 25 and 8 in BaFe$_2$As$_2$, LaFeAsO and NaFeAs, respectively.

In Fig. 2(g), we show that the $I_{XY}/I_{XX}$ ratio of the $A_g$(As) phonon intensity for different Fe-based materials scales linearly with the square of the magnetic moment $M$, indicating that the magneto-elastic coupling constant is proportional to the ordered magnetic moment $M$. In Ref. [23], the magneto-elastic coupling was explicitly calculated within a tight-binding Slater-Koster formalism. The study predicts a large enhancement of the As mode in the XY scattering geometry, consistent with experimental observation [Fig. 2], due to the anisotropy of the Slater-Koster energy integrals in the magnetically ordered state.

Above we have established that the intensity enhancement of the $A_g$(As) phonon mode in the XY scattering geometry depends on the presence of ordered magnetic moment. We now address the coupling between the $A_g$(As) phonon and the $B_{2g}$-like electronic continuum below $T_N$. In Figs. 3(a-b) we present the polarization dependence of the spectra for BaFe$_2$As$_2$ and EuFe$_2$As$_2$ at 15 K. The line-shape of the $A_g$(As) phonon in the XX and ZZ scattering geometries is symmetric, in contrast to asymmetric interference Fano shape observed in the XY and X’X’ geometries [62, 70]. The polarization analysis suggests that interfering with the phonon electronic continuum must have $B_{2g}$ symmetry. A $B_{2g}$-like continuum is allowed to couple to $A_g$(As) phonon in the orthorhombic phase because below the $D_{4h} \to D_{2h}$ transition the $A_{1g}$ and $B_{2g}$ representations merge into the
FIG. 3. (Color online) Raman spectra in the \(XX\), \(XY\), \(X'X'\) and \(XY'\) geometries for (a) \(\text{BaFe}_2\text{As}_2\) and (b) \(\text{EuFe}_2\text{As}_2\) at 15 K. The inset in (b) shows spectra for ZZ polarization from a polished \(ac\) surface of \(\text{EuFe}_2\text{As}_2\) for the in the magnetic state at 100 K excited with 752 nm laser line [72]. The red line is a fit of the \(A_g\) phonon with a Lorentzian function. (c) \(T\)-dependence of Raman spectra for \(\text{BaFe}_2\text{As}_2\) in the \(XY\) scattering geometry (shifted for clarity). The solid red lines are Fano-shape fits [43]. The spectral resolution is about 0.85 cm\(^{-1}\).

\(A_g\) irreducible representation. For \(XY\) and \(X'X'\) geometry, where the \(B_{2g}\)-like continuum is represent, the bare \(A_g(\text{As})\) phonon is coupled to the \(B_{2g}\)-like electronic continuum, giving rise to an asymmetric Fano line-shape.

In addition, we argue that the density-of-states of the \(B_{2g}\)-like continuum is temperature-dependent. In Fig. 3(c), we show temperature evolution of the Raman spectra in \(XY\) scattering geometry for \(\text{BaFe}_2\text{As}_2\). Just below \(T_N\) the \(A_g(\text{As})\) phonon instantly appears with a visibly asymmetric line-shape. The mode sharpens and becomes more symmetric upon cooling, which we attribute to a decrease in the electronic density-of-states at the Fermi level, contributing to the \(B_{2g}\)-like continuum, due to the spin-density-wave gap formation [6–9].

To quantify the electron-phonon interaction, we constructed a Fano model (Eq. 3 in Ref. [43]) with a magneto-elastic coupling constant proportional to the magnetic ordered moment. The Fano interference is between the \(A_g(\text{As})\) phonon mode and the \(B_{2g}\)-like electronic continuum below \(T_N\). Such model well describes the data for \(\text{BaFe}_2\text{As}_2\), Fig. 3(c). Same approach also describes all spectral features for \(\text{Ba(Fe}_{1−x}\text{Au}_x)\text{As}_2\) [43]. Hence, the polarization-resolved Raman spectroscopy implemented here represents an all-inclusive tool to study the magnetism and the magneto-elastic interaction in the Fe-based superconductors.

Finally, we discuss the implications of the electron-phonon coupling to superconductivity. Early calculations show that when magnetic moments are included [73], the electron-phonon coupling is enhanced by 50% as compared with non-magnetic calculations [74]. According to more recent calculations, the electron-phonon matrix element is rather enhanced four times in the ordered AFM state due to the presence of a \(d_{xz}/d_{yz}\) Fermi surface near the zone corner [75, 76]. In particular, the Eliashberg spectral function \(\alpha^2F\) is enhanced 4 times around 22 meV, which corresponds to the \(A_{1g}\) mode energy [75, 76]. Thus, one cannot rule out the possibility that the enhanced intraband electron-phonon coupling in the AFM phase, if sufficiently large, could enhance the paring temperature.

In conclusion, we revealed a significant intensity enhancement of the emergent \(A_g(\text{As})\) phonon mode in the \(XY\) scattering geometry below \(T_N\) only for parent compounds of Fe-based superconductors showing magnetic order. We argue that the in-plane electronic polarizability anisotropy necessary for the \(A_g(\text{As})\) phonon intensity enhancement originates from the anisotropy of the magnetic interactions in the Fe-As plane that are modulated by the \(c\)-axis motion of the As atoms. In particular, we demonstrate a magneto-elastic coupling between the \(A_g(\text{As})\) phonon and the \(B_{2g}\)-like electronic continuum that is essential to \(A_g(\text{As})\) phonon intensity enhancement. The asymmetric line-shape of \(A_g(\text{As})\) phonon is well explained by a Fano model with a magneto-elastic coupling constant proportional to the ordered magnetic moment. Our results identify strong electron-phonon coupling in the magnetic phase of Fe-based superconductors, which could enhance the paring temperature.

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