Coupling of the A1g As-phonon to magnetism in iron pnictides

Belen Valenzuela
Instituto de Ciencias Materiales de Madrid
(ICMM-CSIC)
In collaboration with:

Noel A. García-Martínez (ICMM-CSIC)

Sergio Ciuchi (Università dell’Aquila)

Maria J. Calderón (ICMM-CSIC)

Emmanuele Cappelluti (CNR Roma)

Elena Bascones (ICMM-CSIC)

BNL, New York 2013
Index

• Introduction: Interplay of charge, spin and lattice dynamics in iron superconductors
  – A1g As-phonon Raman response

• Our work:
  – Electron-phonon Hamiltonian plus interactions.
  – Phonon Raman response using charge-phonon theory.
  – Results: Raman intensities of the A1g As-phonon in the A1g, B1g and B2g symmetries. Hardening/softening.
The orthorhombic transition follows the magnetic transition with \((\pi,0)\) ordering.

Both, chemical doping and pressure suppress the SDW and induce SC.

**BNL, New York 2013**
• Experimentally several phononic signatures show unconventional behavior in the magnetic state: Choi et al. PRB’08, Le Tacon et al. PRB’08, PRB’09, Chauviere et al. PRB’09, PRB’11, Akrap et al. PRB’09, Zhang et al. JACS’10, Schafgans et al. PRB’11, Nakajima et al. PNAS’11, Kim et al Nat. Mat.’12, Liu et al. PRL’13…

• Theoretically (Ab-initio calculations):

  • iron-magnetism is present also above $T_N$? what about the estimations of the electron-phonon coupling?

  • The electron-phonon coupling has been shown to be enhanced by magnetism. (Yndurain et al PRB’09, Boeri et al. PRB’10)

  • Role for the mechanism of superconductivity?

Zbiri et al., Phys. Cond. Matt’10

BNL, New York 2013
It has been claimed that the height of the As atom affects:

- the band structure at the Fermi level
- the magnetic moment, magnetic ordering
- the superconducting critical temperature, sc gap
Raman A1g As-phonon

A1g = LL
B1g = x’y’
B2g = xy
Below the Magneto-structural transition, the A1g As-phonon intensity increases a lot for B1g (x’y’)

A1g=LL
B1g=x’y’
B2g=xy
Below the Magneto-structural transition the A1g As-phonon intensity increases a lot for B1g (x’y’)

A1g As-phonon intensity in B1g is 1.5 bigger than in A1g in the magnetic state of BaFe$_2$As$_2$

The structural transition cannot explain these features, Why then?
Raman A1g As-phonon

Rahlenbeck et al PRB’09
(BaFe$_2$As$_2$): Below the Magneto-structural transition **softening** and narrower scattering rate

Choi et al PRB’08
(CaFe$_2$As$_2$): Below the Magneto-structural transition **hardening** and narrower scattering rate

BNL, New York 2013
Our work:
Coupling of the A1g arsenide phonon to magnetism in iron pnictides

N. García-Martínez, B.V, M.J. Calderón, S. Chiuci, E. Cappelluti, E. Bascones,
arXiv:1307.7065

BNL, New York 2013
Our model: Microscopic Hamiltonian

6 electrons in 5 d orbitals in a tetrahedral environment with crystal field 100-200meV:

\[
\begin{align*}
H &= H_{TB} + H_{ph} + H_{int} \\
\end{align*}
\]
Tight-binding

\[ H_0 = \sum_{\mathbf{k}, \mu, \nu, \sigma, r} F_{\mu \nu}^{r}(\mathbf{k}) t_{\mu \nu}^{r} C_{\mu \sigma}^{\dagger} C_{\nu \sigma} + \sum_{i, \mu, \sigma} \epsilon_{\mu} C_{i \mu \sigma}^{\dagger} C_{i \mu \sigma} \]

✓ Focus on Fe-pnictogen planes, square lattice, Fe unit cell
✓ Five Fe d-orbitals; pnictogen included through hoppings;
direct (Fe-Fe) + indirect (Fe-pnictogen-Fe) hoppings
✓ Symmetry of the orbitals considered through Slater-Koster
parameters to describe the hoppings (pd\(\sigma\), pd\(\pi\), dd\(\sigma_1\), dd\(\pi_1\), dd\(\delta_1\))
✓ Straightforward change of pnictogen position (angle \(\alpha\))

MJ Calderon, B.V, E Bascones PRB’09

BNL, New York 2013
Phonon Hamiltonian

\[ H_{\text{ph}} = \sum_{\mathbf{q}} \omega_{\mathbf{q}} a_{\mathbf{q}}^\dagger a_{\mathbf{q}} + \sum_{\mathbf{k}, \mathbf{q}, \mu, \nu, \sigma, M} g_{\mu\nu}^M(\mathbf{k}, \mathbf{q}) c_{\mathbf{k}+\mathbf{q}\mu\sigma}^\dagger c_{\mathbf{k}\nu\sigma} \left( a_{\mathbf{q}} + a_{\mathbf{-q}}^\dagger \right) \]

We are just interested in Raman processes i.e. \( \mathbf{q} = 0 \)

The electron-phonon coupling is split in two contributions \( g^g \) (geometrical) and \( g^{pd} \) (through energy integrals \( pd\sigma \) and \( pd\pi \)). Both have local and non-local contributions:

Geometrical phonon coupling:
\[ g_{\mu\nu}^{\alpha,\text{non-loc}}(\mathbf{k}) = \sum_r F_{\mu\nu}^r(\mathbf{k}) \frac{\partial t_{\mu\nu}^r}{\partial \alpha} \delta \alpha \]
\[ g_{\mu\nu}^{\alpha,\text{loc}} = \frac{\partial \epsilon_{\mu}^{\text{ind}}}{\partial \alpha} \delta \alpha \]

Energy integral phonon coupling:
\[ g_{\mu\nu}^{pd,\text{non-loc}}(\mathbf{k}) = \sum_r F_{\mu\nu}^r(\mathbf{k}) \delta pd\sigma \left( \frac{\partial t_{\mu\nu}^r}{\partial pd\sigma} + \frac{\partial t_{\mu\nu}^r}{\partial pd\pi} \frac{pd\pi_0}{pd\sigma_0} \right) \]
\[ g_{\mu\mu}^{pd,\text{loc}} = \delta pd\sigma \left( \frac{\partial \epsilon_{\mu}^{\text{ind}}}{\partial pd\sigma} + \frac{\partial \epsilon_{\mu}^{\text{ind}}}{\partial pd\pi} \frac{pd\pi_0}{pd\sigma_0} \right) \]

With:
\[ \delta pd\sigma = pd\sigma_0 \frac{1}{f(R_0)} \frac{\partial f(R)}{\partial R} \frac{\partial R}{\partial h} \delta h \]

We assume: \( f(R) = 1/R^4 \) valid for p & d orbitals localized

BNL, New York 2013
(\pi,0) \textit{Magnetic phase diagram}

\[ H = H_{TB} + H_{\text{int}}(U,J) \]

We calculate the \textbf{magnetic U-J_{H}/U phase diagram} applying mean field theory to the \textit{electronic} Hamiltonian.

We study this line $J=0.25U$.

xy and yz become \textbf{half-filled gap states}
Band structure for the non-magnetic and magnetic regime in the itinerant region

Spectral weight reorganization at low energies due to magnetism in a multiorbital system.

We have added a renormalization factor of 3 (in accordance with ARPES).

BNL, New York 2013
Raman response for the A1g As-phonon
In the PM state, B1g and B2g Raman response are zero because they are orthogonal to the symmetry of the A1g As-phonon.

**Our proposal:** Due to the electron-phonon coupling the B1g Raman response of the A1g As-phonon is large in the magnetic state since the tetragonal symmetry is broken by the (\(\pi,0\)) magnetic ordering:

---

**BNL, New York 2013**
Charge-phonon theory I:

The Raman response is calculated using the Charge-phonon theory

\[ \chi^\lambda (\Omega) = \chi^\lambda_{el-el} (\Omega) + \Delta \chi^\lambda_{ph} (\Omega) \]

This contribution has been calculated in B.V, Calderon, Leon, Bascones PRB'13

\[ \chi^\lambda_{el-el} (\Omega) \]

\[ \Delta \chi^\lambda_{ph, M, M'} (\Omega) = \chi^\lambda_M (\Omega) D_0 (\Omega) \chi^\lambda_M (\Omega) \]

Our main calculation

\[ D_0 (\Omega) = \frac{1}{(\Omega-\Omega_0)+i\Gamma_0} \]

\[ \Omega_0 \text{ and } \Gamma_0 \text{ are the phonon frequency and the phonon scattering rate} \]

BNL, New York 2013
Charge-phonon theory II:

\[ \Delta \chi_{ph,M,M'}^{\lambda}(\Omega) = \chi_{M}^{\lambda}(\Omega) D_0(\Omega) \chi_{M'}^{\lambda*}(\Omega) \]

\[ \chi_{M}^{\lambda}(\Omega) = -\frac{\pi}{V} \sum_{k\sigma\sigma'} \gamma_{nn'}^{\lambda}(k) g_{nn'}^{M*}(k) (f(E_n(k)) - f(E_{n'}(k))) \]

\[ \times (\delta(\Omega + E_n(k) - E_{n'}(k)) - \delta(-\Omega + E_n(k) - E_{n'}(k))) \]
Charge-phonon theory II:

\[ \Delta \chi_{ph,M,M'}^\lambda (\Omega) = \chi_M^\lambda (\Omega) D_0 (\Omega) \chi_{M'}^\lambda (\Omega) \]

\[ \chi_M^\lambda (\Omega) = -\frac{\pi}{V} \sum_{k\sigma nn'} \gamma^{\lambda}_{nn'}(k) g_{nn'}^M(k) (f(E_n(k)) - f(E_{n'}(k))) \times (\delta(\Omega + E_n(k) - E_{n'}(k)) - \delta(-\Omega + E_n(k) - E_{n'}(k))) \]

\[ \text{Im} \Delta \chi_{ph,M}^\lambda (\Omega) = -I_M^\lambda \frac{(q_M^\lambda )^2 - 1 + 2\left( \frac{\Omega - \Omega_0}{\Gamma_0} \right) q_M^\lambda}{(q_M^\lambda )^2 \left( 1 + \left( \frac{\Omega - \Omega_0}{\Gamma_0} \right)^2 \right)} \]

\[ I_M^\lambda = \left( \chi_M' (\Omega_0) \right)^2 \]

\[ q_M^\lambda = -\frac{\chi_M''(\Omega_0)}{\chi_M'(\Omega_0)} \]

Charge phonon theory related to Fano theory. All the physics is encoded in the mixed bubble:

E. Cappelluti, et al, PRB’12

BNL, New York 2013
Results: $A_{1g}$ As-phonon Raman intensity

There is a critical value of $U$ where $I_{B_{1g}} > I_{A_{1g}}$ as in BaFe$_2$As$_2$.

$I_{A_{1g}}$ increases a lot in the magnetic state as in 122 compounds.

But the resulting intensity cannot be calculated due to uncertainties in the values of the phonon couplings $g_\alpha$ and $g_{pd}$.
Phonon hardening/softening and life-time

\[ \Delta \Omega_M = \Pi'_M(\Omega_0, U) - \Pi'_M(\Omega_0, U = 0) \quad \Delta \Gamma_M = -(\Pi''_M(\Omega_0, U) - \Pi''_M(\Omega_0, U = 0)) \]

(a) \( M = \alpha \)  \( M = pd \)

Softening of the phonon frequency in the magnetic state.

Hardening just for large values of \( U \) when e-ph coupling goes via \( g_{pd} \)

(b)

Narrowing or broadening appear depending on parameters
Phonon hardening/softening and life-time

\[ \Delta \Omega_M = \Pi'_M(\Omega_0, U) - \Pi'_M(\Omega_0, U = 0) \]
\[ \Delta \Gamma_M = -(\Pi''_M(\Omega_0, U) - \Pi''_M(\Omega_0, U = 0)) \]

Hardening just for large values of $U$ when e-ph coupling goes via $g_{pd}$

The softening is related to the spectral weight redistribution in a multiorbital system from higher energies ($\Omega > \Omega_0$) to lower energies ($\Omega < \Omega_0$) when entering into the magnetic state.

Softening of the phonon frequency in the magnetic state.

Narrowing or broadening appear depending on parameters

BNL, New York 2013
Summary of our work

• The electron-phonon coupling is via the dependence of the Slater-Koster parameters ($\alpha$, $pd\sigma$, $pd\pi$) on the As position.
• Magnetism is included at the mean-field Hartree-Fock level.
• The Raman response is evaluated in the paramagnetic and in the ($\pi$, 0) magnetic states using the proper generalization of the charge-phonon theory.

• A finite Raman intensity can be observed in the magnetic state in the B1g but not in the B2g polarization and it is a consequence of the coupling of the phonons to the anisotropic ($\pi$,0) magnetic state.
• Softening and hardening are possible.
• For a quantitative comparison more work is needed.

Outlook

• It is possible that in the nematic state there is a signal in $B_{1g}$.
• In the double stripe magnetic state of FeTe, the out-of-plane Te-phonon will be different from zero in the B2g polarization geometry, instead B1g symmetry.

It would be interesting to explore these possibilities experimentally.
Thank you!
Going beyond....

- The interplay between magnetism and As-heigh should be treated self-consistently
- The electron-phonon coupling could be go through the interactions
- Magnetism beyound mean-field.
Anisotropy in transport:

- Anisotropy in transport (Chu et al. PRB’10; Science’10, Tanatar et al. PRB’11) and in optical conductivity (Dusza et al, EPL’11, Review: I.R. Fisher et al. Rep. Prog. Phys.’11, Nakakima et al. ‘11), etc.

Chu et al. Science’10

Unexpected because:
- FM bond is shorter than AF one
- AF is naively associated with gaps and FM with metallicity
- Scattering rate is larger in the AF direction

BNL, New York 2013
Magnetism: $\left(\pi, 0\right)$ mean field phase diagram: metallic region

BNL, New York 2013
Drude weight ratio

\[ \frac{D_a}{D_b} = \frac{\sum_{k,n} \nu_a^2(k, n) g(k, n) \delta(\epsilon_n(k) - E_F)}{\sum_{k,n} \nu_b^2(k, n) g(k, n) \delta(\epsilon_n(k) - E_F)} \]

We also calculate the Drude ratio with the Kubo formula and get the same result.

We assume the scattering rate is isotropic.
$D_a/D_b > 1$ consistent with experimental $r_a/r_b < 1$
Is given in regions with low magnetic moment

**BNL, New York 2013**
Experimental signs of anisotropy and orbital ordering are anticorrelated, B. Valenzuela, E. Bascones, M.J. Calderón, PRL 105, 207202 (2010)

\[ \frac{D_a}{D_b} > 1 \] consistent with experimental \( \frac{r_a}{r_b} < 1 \)

\[ n_{yz} < n_{zx} \]
Magnetic reconstruction as origin of the conductivity anisotropy

Anisotropy linked to topology and morphology of the Fermi Surface. Experimental anisotropy in general for low moment.

Sensitivity of the anisotropy to the angle $\alpha$

In the squashed tetrahedron the region with $D_x/D_y>1$ has reduced very much and it is increased for the elongated tetrahedron.
The effect of magnetism is not just at the Fermi surface but it is seen at all energies in the band.

U=2.2, J=0.07U

BNL, New York 2013
Orbital reorganization in the Hartree-Fock phase diagram

Resembles the orbital reorganization found in the strong coupling limit

Tight-binding for five orbitals: angle dependence of the hoppings

Hoppings related by symmetry and calculated with four fitting parameters

Experimental range

MJ Calderon, B.V, E Bascones PRB’09

BNL, New York 2013
Hartree-Fock phase diagram. Sensitivity to Crystal field

Correlated metal

Nesting driven \((\pi,0)\)
Reasoning from the strong coupling point of view to understand the LM

The low moment phase is stabilized because crossed hoppings are as big as direct hoppings and also very anisotropic: Release frustration

\[ Q = (\pi, 0) \]

\[
\begin{align*}
t^y_{yz,x^2-y^2} &= 0 \\
t^x_{xy,yz} &= 0
\end{align*}
\]
Orbital ordering in the band structure for $U=2.2$ eV and $J=0.07U$
Phase separation

JH/U = 0.22
In the mean field description there is a DS phase charge modulated instead of FM. But strong coupling analysis also points to DS instability at high $J_H$. 


BNL, New York 2013