

# CMSN



# Newsletter

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## *In this issue:*

### *Research Highlight*

- Evidence for weak electronic correlations in the iron pnictides

### *Conferences & Workshops*

- Dynamics and Cohesion CRT Fall Coordination Meeting

### *CMSN News – Call for New CMSN Proposals*

### *CMSN Information*



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# Research Highlight

In this issue of the *CMSN Newsletter*, we feature a research highlight from the Resonant Inelastic X-Ray Scattering CRT: “Evidence for weak electronic correlations in the iron pnictides.”

## Evidence for weak electronic correlations in the iron pnictides

*RESONANT INELASTIC X-RAY SCATTERING CRT*

W. L. YANG, A. P. SORINI, C-C. CHEN, *ET AL.*<sup>1</sup>

**Summary:** Using x-ray absorption (XAS) and resonant inelastic x-ray scattering (RIXS), charge dynamics at and near the iron *L* edges is investigated in iron-pnictide materials and contrasted to that measured in other iron compounds. It is shown that the XAS and RIXS spectra for 122 and 1111 iron pnictides are each qualitatively similar to iron metal. Cluster diagonalization, multiplet, and density-functional calculations show that Coulomb correlations are much smaller than in the cuprates, highlighting the role of iron metallicity and strong covalency in these materials. The best agreement with experiment is obtained using a Hubbard *U* of less than about 2 electron volts and Hund’s *J* of about 0.8 electron volts.

In 2008, high-temperature superconductivity was discovered [1,2] in doped iron-pnictide compounds. This discovery has understandably generated tremendous interest throughout the physics community. In addition, differing predictions between various theoretical models of high-temperature superconducting materials have provoked a good deal of debate regarding the physics of iron pnictides. In this paper we present a comprehensive study of XAS measurements and resonant inelastic x-ray scattering experiments using a wide range of theoretical and computational techniques.

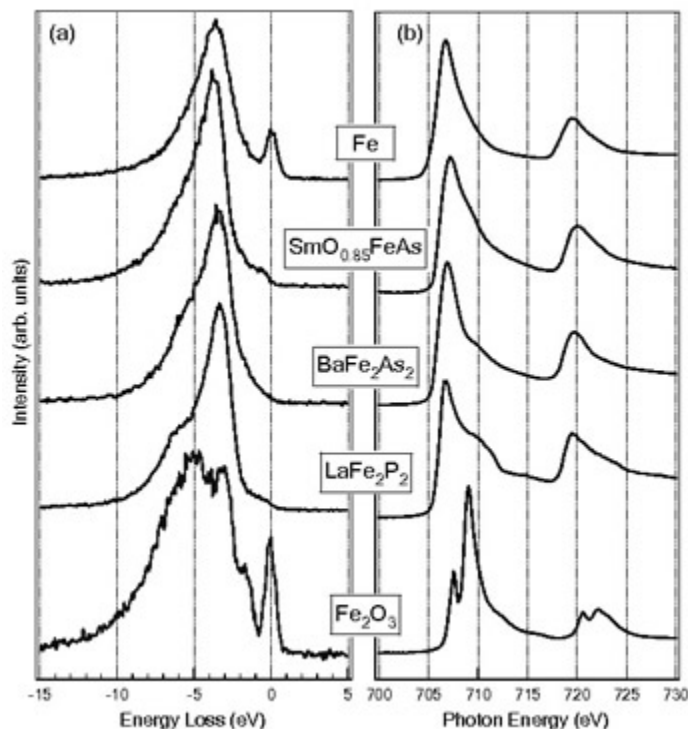
At first glance, the iron-pnictide family of superconductors appears to be somewhat similar to the well-known cuprate family of high-temperature superconductors. For example, both compounds contain layered structures, which can be approximated as two dimensional. However, a closer examination of these materials reveals many interesting differences. Even if one considers just the lattice structure of these materials, one realizes that the iron-pnictide layers are far less “two dimensional” than copper-oxide layers in the cuprates; each iron atom is surrounded by an arsenic tetrahedron. In addition, the superconductivity of the iron pnictides is more three dimensional than the cuprates.

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<sup>1</sup> Adapted from full article (editor's choice ) in PRB (<http://link.aps.org/doi/10.1103/PhysRevB.80.014508>) with accompanying Viewpoint commentary by Zlatko Tesanovic (<http://physics.aps.org/articles/v2/60>).

Another difference between the pnictides and the cuprates, which has generated much debate and disagreement, is that of electronic correlations. The parent compounds (both cuprates and pnictides must be doped to superconductivity) of the cuprate superconductors are well known to be strongly correlated Mott insulators; in the absence of strong short-range Coulomb repulsion (large “Hubbard  $U$ ”) these materials would be metals. On the other hand, the iron-pnictide parent compounds are metallic, which already indicates that Coulomb correlations in iron pnictides may be weaker than cuprates. Thus a key question that needs to be addressed is whether or not the iron pnictides are strongly correlated like the cuprates. Since density-functional-theory (DFT) calculations have indicated that the electron-phonon interaction is too weak to account for high transition temperatures [3], the strength of the Coulomb correlations would give some account for the pairing strength possible in an electronic-based pairing mechanism [4].

In this paper we present a comprehensive study of XAS measurements and RIXS at the iron  $L_2/L_3$  edges in a variety of iron-based materials, including the superconducting 1111 iron pnictide  $\text{SmO}_{0.85}\text{FeAs}$  and the undoped 122 pnictides  $\text{BaFe}_2\text{As}_2$  and  $\text{LaFe}_2\text{P}_2$ . It is shown that the XAS spectra of iron pnictides look qualitatively, and in some cases quantitatively, similar to iron metal and show no features resembling the multiple peak structures seen in iron insulators such as hematite and other iron oxides. A resonance study of x-ray emission across the  $L_2$  and  $L_3$  edges demonstrates that the RIXS spectra is dominated by fluorescence, with no observance of discernible excitonic or satellite peaks. A summary plot of experimental data, which compares the XAS and RIXS spectra of five different iron-containing materials, is shown in Fig. (1).



*Fig. 1. Comparison of RIXS data at 708 eV excitation energy. All iron pnictides and iron metal show only fluorescent peaks same as the nonresonant spectrum,*

while  $Fe_2O_3$  displays multiple peaks as the signature of  $dd$  excitations. (b) Comparison of XAS data on the same five samples.  $Fe_2O_3$  again displays very different line shapes.

In addition, we present calculations using three separate models, which specifically include and account for the role of the core hole in x-ray absorption and emission processes. These calculations were performed using quantum cluster, multiplet, and DFT-based methods and highlight the roles of iron metallicity, iron-arsenic covalency, and local Coulomb and Hund's couplings. DFT calculations using FEFF [5] give quantitative agreement with XAS measurements and align absorption peaks to Fe dDOS above the Fermi level. Cluster calculations of XAS support the role of strong iron-arsenic hybridization involving arsenic states below the Fermi level, setting an upper bound of  $U \approx 2$  eV (which should be compared to the iron bandwidth of about 4 eV). This indicates that iron-pnictide materials are weakly correlated and that the physics is governed largely by Fe metallicity.

The three main theoretical approaches that we have used to analyze the data incorporate electronic correlation effects due to electron-electron interactions to different degrees. First, in order to see explicitly how correlations affect the XAS profile, we performed a model many-body calculation based on the exact diagonalization technique, with a multi-orbital Hubbard model as the effective Hamiltonian. This approach has been successfully applied to understand the correlated physics in materials such as the high- $T_c$  cuprate parent compounds [6,7]. Some of the results we obtained, and which were used to limit the value of the Hubbard  $U$ , are shown in Fig. (2).

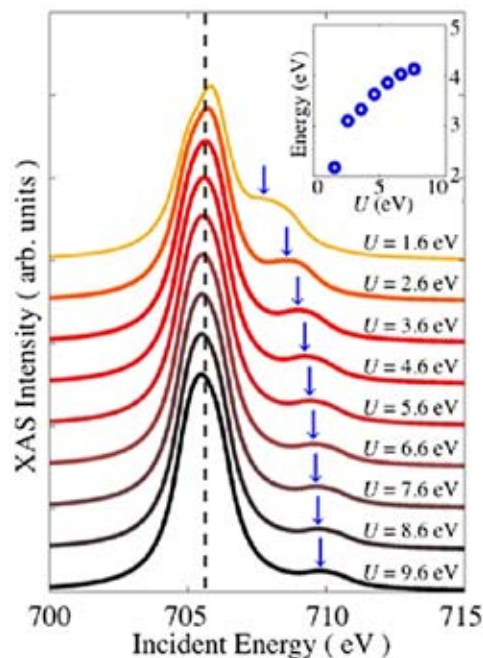


Fig. 2. Fe-pnictide  $L_3$ -edge XAS spectra obtained from small cluster diagonalization for a fixed  $J(e_g)=0.8$  eV and various  $U$ . A strong Coulomb repulsion tends to suppress the XAS shoulder peak intensity. The arrows sketch the energy separation of the dominant peak and its shoulder. The inset is a plot for the peak/shoulder energy separation versus the on-site repulsion  $U$  from which a naïve upper bound of  $U \sim 2$  eV is drawn.

Next, we performed Dirac multiplet calculations of the XAS for the specific case of a  $\text{BaFe}_2\text{As}_2$  crystal field. These results are shown in Fig. (3).

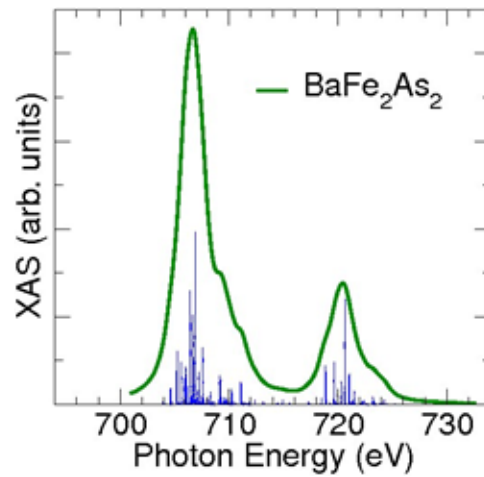


Fig. 3. X-ray absorption spectrum: iron L-edge multiplet calculation for  $\text{BaFe}_2\text{As}_2$ .

Finally, as a material-dependent first-principles approach to XAS simulations, we performed a DFT-based calculation using both the FEFF code [5] and the computer code WIEN2k [8]. These results are shown in Figs. (4) and (5).

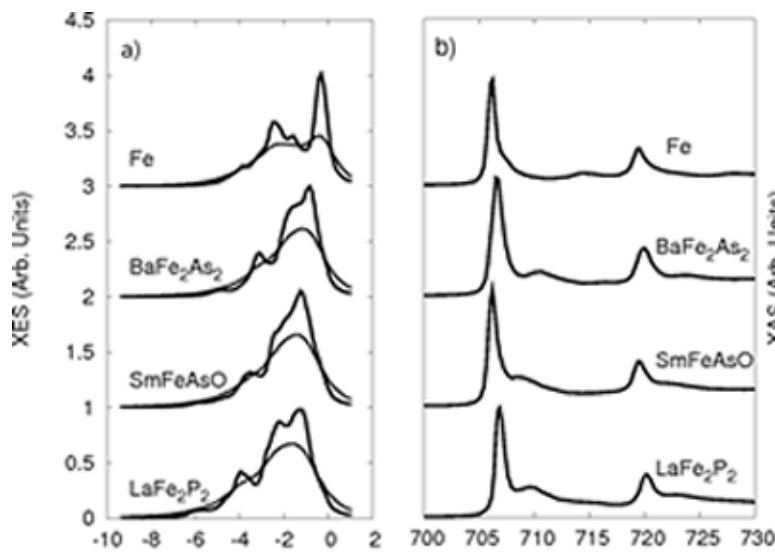


Fig. 4. (a) The  $\text{Fe } L_3$  edge XES signal calculated using FEFF for four different metallic iron-containing materials. (b) The  $\text{Fe } L_{2,3}$  edge XAS calculated using FEFF.

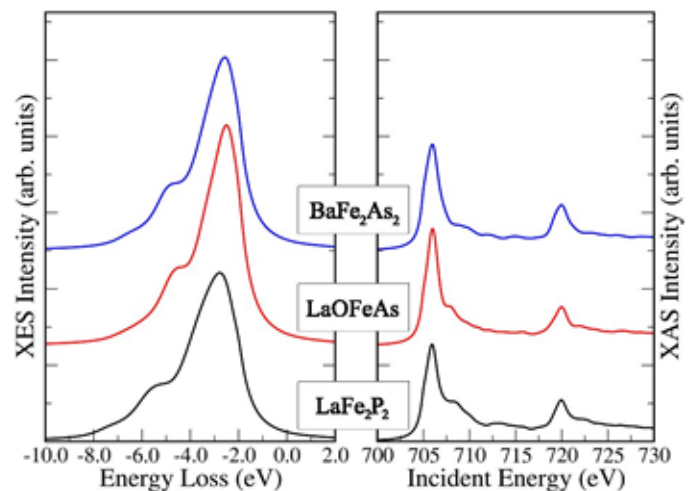


Fig. 5. The iron  $L_{2,3}$  edge XAS and XES, calculated using WIEN2K, for three different iron-containing materials.

From the above considerations we can learn a number of things. The first general observation is that the experimental data for the iron pnictides is qualitatively similar to other metallic iron materials and it is significantly different from large-gap iron-based insulators such as hematite. If the pnictides were very localized, one would expect that the essential features of XAS and RIXS spectra could be readily captured in a small iron-arsenic cluster. Our exact diagonalization computation of the absorption spectra for such a cluster in the localized, strong-coupling limit (large Hubbard  $U$ ) clearly shows the appearance of a high-energy peak well separated from the main absorption line at the  $L_3$  edge. In the experimental data this peak is absent—or rather appears as a shoulder of the main XAS line. From a comparison of the energy position of this shoulder in the data and the cluster simulation, we extract an upper limit of the Hubbard  $U$  of about 2 eV—substantially smaller than the iron  $3d$  bandwidth. We find the Hund's rule  $J$  is about 0.8 eV. Coulomb correlations are thus much weaker than in the cuprates. This result is confirmed by our multiplet calculation, which is a local approach that has the advantage of including the spin orbit and Coulomb interactions related to the core hole. The calculated  $L_3$ - $L_2$  edge energy splittings and intensity ratios agree with the experimental data.

The inference that the Hubbard  $U$  is small and the  $3d$  electrons are weakly correlated suggests a comparison of the spectroscopic data with the results of independent-particle *ab initio* calculations. Such approaches are complicated by the fact that a core hole is present in the final state of XAS and the intermediate state of RIXS. The Coulomb interaction of the core hole with the valence electrons can, in principle, be strong and such an electronic correlation effect has a profound influence on XAS and RIXS spectra. The FEFF code treats the effects of the core-hole potential with a high level of accuracy. The spectra computed with FEFF confirm the presence of the XAS shoulder and agree with the experimental XAS and RIXS data very well. We have used this agreement to further filter out the core-hole-induced correlations. When calculating the spectra with plane-wave-based WIEN2K code—which includes the proper dipole transition matrix elements but lacks the final-state XAS core-hole potential—we observe that the simulated spectra basically do not change. We conclude that in the iron pnictides not only the Hubbard  $U$  but also the core-hole potential is therefore heavily screened [9]. The electronic correlations that the core hole induces are thus weak and consequently the spectra can safely be interpreted in terms of single-particle densities of states and the appropriate dipole transition matrix elements. The present spectroscopic data and its theoretical description thus emphasize the role of strong covalency and iron metallicity in the iron pnictides.

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## *CMSN News*

Proposals for forming new CMSN Collaborative Research Teams (CRTs) are being accepted currently for funding in Fiscal Year 2010. Please check the CMSN web site ([www.phys.washington.edu/~cmsn/proposals](http://www.phys.washington.edu/~cmsn/proposals)) or the DOE-BES web site ([http://www.science.doe.gov/bes/dms/Research\\_Programs/tcmp.htm](http://www.science.doe.gov/bes/dms/Research_Programs/tcmp.htm)) for further information.

## *Conferences and Workshops*

- Dynamics and Cohesion of Materials Interfaces and Confined Phases Under Stress CRT Final Coordination Meeting, George Mason University, Fairfax, VA., September 16 & 17, 2009.

Please consult the CMSN website for updated information on conferences and workshops, at <http://www.phys.washington.edu/~cmsn/>.

# CMSN Information

CMSN's teams, oversight, and administration are listed below. Further information can be found at <http://www.phys.washington.edu/~cmsn>.

## Collaborative Research Teams

Dynamics and Cohesion of Materials Interfaces and Confined Phases Under Stress

Predictive Capability for Strongly Correlated Systems

Multiscale Simulation of Thermo-mechanical Processes in Irradiated Fission-reactor Materials

Resonant Inelastic X-Ray Scattering

Predictive Modeling of the Growth and Properties of Energy-Relevant Thin Films and Nanostructures

## Leaders

Mark Asta (University of California at Davis), Alain Karma (Northeastern), and Anthony Rollett (Carnegie-Mellon)

Richard Scalettar and Warren Pickett (UC-Davis)

Dieter Wolf (INL) and Simon Phillpot (University of Florida)

Bob Markiewicz (Northeastern University), Jim Freericks (Georgetown University), Michel van Veenendaal (Northern Illinois University and Argonne National Laboratory), and Arun Bansil (Northeastern University)

Kai-Ming Ho (University of Iowa and Ames Laboratory) and Zhenyu Zhang (University of Tennessee and Oak Ridge National Laboratory)

# CMSN Oversight

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