# Studying metal clusters by Quantum Monte Carlo method



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## A problem with Ag clusters

# 2D to 3D structural transition in noble metal clusters



Schmidt, M., et al., JCP 2003. 118: p. 10956-10962. Weis, P., et al., CPL 2002. p. 355-364.

## Method: Fixed-node diffusion quantum Monte-Carlo

- An accurate and practical method for medium-large system.
- Time cost is in order of O (N<sup>3</sup>+εN<sup>4</sup>), compared to O(N<sup>7</sup>) for CCSD(T).
- Trial wavefunction: Jastrow-Slater wavefunction.

$$\Psi(\mathbf{R}) = \exp\left[J\right] \sum_{n} c_n D_n^{\dagger} D_n^{\downarrow}$$

where  $D(\mathbf{X})$  is Slater determinant,  $J(\mathbf{X})$  is Jastrow factor, describing the correlation. In this work,  $D(\mathbf{X})$  is determined by Hartree-Fock, using an effective core potential (ECP). We use Our results will be compared with multireference doubleexcitation configuration-interaction (MRD-CI), PBE, B3LYP and coupled cluster (CCSD(T)).

#### **Computational details**

- Code: CASINO
- Both effective core potential (ECP) (Li, Na, Ag) and all-electron (AE) (Na) DMC were performed to calculate the energies of clusters.
- Single determinant and fixed node approximation were used, two-body interactions, were included in Jastrow factor (minimizing variance).
- For ECP-DMC, Trail-Needs Hartree-Fock pseudopotentials are generated for Li Na, and Ag (using Opium). We used a time step of 0.01 a.u. for Li and Na, and 0.005 a.u. for Ag, after testing timestep errors. The single-particle orbitals were obtained by DFT plane wave calculations using the Quantum-Espresso package (with HF pseudopotential) and re-expanded in terms of B splines in real space. The number of walkers used is 2,000 for Li and Na, and 15,000 for Ag.
- For AE-DMC, two sets of time steps, 0.001 and 0.0005 a.u. (for Na), were used. Time step needs to be much shorter for AE-DMC than ECP-DMC. The single-particle orbitals were from Hartree-Fock calculations using Gaussian package, using a basis set of cc-pvTZ.
- The population of walkers ranges from 60,000 to 80,000, depending on the size of the clusters.

## Lithium clusters



S. E. Wheeler and K. W. Sattelmeyer, P. v. R. Schleyer, and H. F. Schaefer III, J. Chem. Phys. 120, 4683 (2004). C. H. Wu, J. Phys. Chem. 87, 1534 (1983) J.Koutecky and P. Fantucci, Chem. Rev. 86, 539 (1986)

#### Binding energies (eV) per atom

	ECP-DMC	CCSD(T) <sup>1</sup>	MRD-CI <sup>2</sup>	Expt. <sup>3</sup>
$Li_2(D_{\infty h}, {}^1\Sigma_g^+)$	0.488 (0.00016)	0.496	0.45	0.55
Li <sub>3</sub> ( C <sub>2v</sub> , <sup>2</sup> B <sub>2</sub> )	0.511 (0.00014)	0.535	0.45	0.60
Li <sub>4</sub> ( D <sub>2h</sub> , <sup>1</sup> A <sub>g</sub> )	0.676 (0.00018)	0.723	0.63	0.84

(....) Statistical error

1. S. E. Wheeler and K. W. Sattelmeyer, P. v. R. Schleyer, and H. F. Schaefer III, J. Chem. Phys. 120, 4683 (2004).

2. V. Bonai-Koutecký, P. Fantucci, and J. Koutecký, Chem. Rev. (Washington, D.C.) 91, 1035 (1991).

3. C. H. Wu, J. Phys. Chem. 87, 1534 (1983).

## **Sodium Clusters**



V. Bonai-Koutecký, P. Fantucci, and J. Koutecký, Phys. Rev. B, 37, 4396, 1988

## **Sodium Clusters**



#### Na clusters: binding energies (per atom)



I. A. Solov'yov, A. V. Solov'yov, and W. Greiner, Phys. Rev. A 65, 053203 (2002).

Calculated energies (Hartree) and binding energies per atom (kcal/mol) of small sodium clusters.

	Energy (CI	Energy (PBE	Binding	Binding
	geometries)	optimized	energy (CI	energy (PBE
		geometries)	geometries)	optimized
				geometries)
Na <sub>1</sub>	-0.182103	-0.182103		
	(0.000002)	(0.00002)		
Na <sub>2</sub> ( $D_{\infty h}$ , $^{1}\Sigma_{g}$	-0.39078	-0.39027	8.337 (0.006)	8.177 (0.008)
+)	(0.00002)	(0.00003)		
$Na_3 (C_{2v}, {}^2B_2)$	-0.58262	-0.58337	7.596 (0.010)	7.752 (0.006)
	(0.00005)	(0.00003)		
$Na_4 (C_{2v}, {}^2B_2)$	-0.79670	-0.79744	10.713	10.829 (0.007)
	(0.00003)	(0.00004)	(0.005)	
$Na_4 (C_{2v}, {}^{3}A_2)$	-0.79457	-0.79720	10.38 (0.010)	10.791 (0.004)
	(0.00006)	(0.00003)		
$Na_5 (C_{2v}, {}^2A_1)$	-1.00026	-1.00197	11.26 (0.01)	11.478 (0.005)
	(0.00007)	(0.00003)		
$Na_5 (C_{2y}, {}^2B_1)$	-0.99354	-0.99782	10.42 (0.002)	10.957 (0.005)
	(0.00001)	(0.00003)		
$Na_6 (D_{3h},$	-1.21750	-1.21829	13.061	13.143 (0.005)
$^{1}A_{1})$	(0.00001)	(0.00003)	(0.002)	
$Na_{6}(C_{5v}^{-1}A_{1})$	-1.21910	-1.21891	13.23 (0.002)	13.210 (0.005)
	(0.00001)	(0.00003)		
$Na_7 (D_{5v},$	-1.4283	-1.43643	13.77 (0.018)	14.497 (0.003)
$^{2}A_{2})$	(0.0001)	(0.00002)		

## All electron DMC

• Trial wavefunction comes from all electron calculation (Hartree-Fock).



# Comparison of Na<sub>n</sub> (2,3,4) binding energies from different methods



Binding energy (in kcal per mol per atom) for  $Na_n$  (n = 2, 3, 4) obtained by CCSD(T), AE-DMC, ECP-DMC, and MRD-CI.

	MRD-CI	ECP-DMC	AE-DMC timestep = 0.001 a.u.	AE-DMC timestep = 0.0005 a.u.	CCSD(T)
Na2	6.4	8.337 (0.006)	8.30 (0.22)	8.121 (0.227)	8.539
Na3	6.0	7.596 (0.010)	7.01 (0.16)	7.350 (0.208)	7.789
Na4	8.95	10.713 (0.005)	9.55 (0.16)	10.960 (0.201)	10.930

## Tests for Ag and Ag<sub>2</sub>

- Effective core potential: HF
- Time step: 0.005 a.u.
- Number of walkers: 50000.

	Energy (a.u.)	Binding Energy (eV)
Ag <sub>1</sub>	-37.75191 (0.00013)	
Ag <sub>2</sub> (d=2.50 Å)	-75.45132 (0.00034)	-0.714 (0.008)
(d=2.53 Å)	-75.45807 (0.00030)	-0.623 (0.008)
(d=2.56 Å)	-75.43712 (0.00043)	-0.908 (0.009)
(d=2.60 Å)	-75.52150 (0.00033)	0.240 (0.008)

Negative binding energy (in red): NO binding. Experiment data: 2.53 Å; binding energy 0.83 eV per atom. GGA result: 2.60 Å and 0.81 eV per atom.



## Conclusions

- Binding energies of lithium clusters obtained by single determinant DMC are in good agreement with CCSD(T) and experimental results.
- For sodium clusters, DMC gives higher binding energies than MRD-CI and DFT.
   MDD CI regulate (literature) energy to be under bound

MRD-CI results (literature) appear to be under-bound.

- There is a big problem with Ag<sub>2</sub>, pseudo-potential? fixednode? any ideas?
- But for our problem with Ag<sub>7</sub>+, both TPSS and HSE show 3D ground state! HSE06:J. Paier, M. Marsman, K. Hummer, G. Kresse, I. C. Gerber, and J. G.Angyan, Journal of Chemical Physics \*125\* (2006).J. Heyd, G. E. Scuseria, and M. Ernzerhof, Journal of Chemical Physics\*124\* (2006).TPSS:J. Tao, J.P. Perdew, V.N. Staroverov, G.E. Scuseria, Phys. Rev. Lett.\*91\*, 146401(2003)

## First-Principles calculations of iron-pnictides

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# Outline

- Brief review: FeAs based high-T<sub>c</sub> superconducting materials
- Structure, electronic structure and magnetism of 1111 and Ba122
- Constructing projector augmented wave (PAW) potentials
- Constructing model Hamiltonian: down folding
- Co-doping: Scattering Center, three dimensionality

## First family of ferropnictide SC



- a) Y. Kamihara et.al., Tokyo, JACS
- b) b) X.H. Chen, et.al., Beijing,arXiv: 0803.3790
- c) Zhi-An Ren, Beijing, arXiv: 0803.4283
- d) Zhi-An Ren, Beijing, arXiv: 0804.2053.

LOFFA under pressure: T<sub>c</sub>=43K (Takahashi et al Nature 453 376 (2008))

Rare earths

SmF <sub>x</sub> O <sub>1-x</sub> FeAs x~0.2 d)	Tc=55K, cm/ 0803.3603 a=3.933A, c=8.4287A
PrF <sub>x</sub> O <sub>1-x</sub> FeAs c)	Tc=52K, cm/ 0803.4283 a=3.985A, c=8.595A
CeF <sub>x</sub> O <sub>1-x</sub> FeAs b)	Tc=41 K, cm/ 0803.3790 a=3.996A, c=8.648A
LaF <sub>x</sub> O <sub>1-x</sub> FeAs a)	Tc=26 K, JACS-2008 a=4.036A, c=8.739 A
La <sub>1-x</sub> Sr <sub>x</sub> OFeAs	Tc=25K, cm/ 0803.3021, a=4.035A, c = 8.771A
57 58 59 60 60 La Ce Pr Nd 138.90 140.11 140.90 144.24	62 63 <b>Pm</b> Sm Eu (145) 150.36 151.96

also:

# Magnetic order coupled to structural phase transition



Superconducting state is non-magnetic! Suppression of  $\mu \ll$  Superconductivity ?

## 1111 vs. 122 vs. 111 vs. 11 materials



## Features of doped 122 materials

#### • T<sub>c</sub> up to 38 K

- good crystals which cleave well—Angle resolved photoemission spectroscopy (ARPES), STM
- dope with K or Co, or apply pressure to suppress spin-density wave  $\rightarrow$  superconductivity
- Magnetic order tied to structural transition, possible coexistence with superconductivity

properties are more 3D than 1111 materials



#### Calculation Details

- •PWSCF, Ultrasoft Pseudopotential (also VASP, PAW)
- •PBE exchange-correlation functional
- •8x8x4 Monkhorst-Pack grid (16x16x8 for DOS)
- •40 Ry  $E_{cut}$  for plane-wave, 400 Ry  $E_{cut}$  for core charges
- •GGA+U in VASP: U=2.0~5.0, J=0.89 for Fe

•Fully relaxed structure (optimized lattice constant, optimized internal coordinates)

•Fitting of band structure to model Hamiltonians using Maximally Localized Wannier Functions

#### LOFA: Optimized Structure

	NM	CB- AFM	SDW- AFM
а	3.9899(4.0327)	4.0200	(5.6826)
b	3.9899(4.0327)	4.0200	(5.7104)
С	8.6119(8.7411)	8.7394	(8.7196)
<b>Z<sub>La</sub></b>	0.1418(0.1413)	0.1382	(0.1417)
Z <sub>As</sub>	0.6408(0.6517)	0.6492	(0.6513)

in parenthesis: Experimental results CB-AFM: ~2.3  $\mu_B$ , SDW-AFM: ~2.5  $\mu_B$ , CB-AFM is 84 meV/Fe lower than NM state, SDW-AFM state is 109 meV/ Fe lower than NM state.

Cao, Hirschfeld, and Cheng, PRB77, 220506R(2008)
 First prediction of FM ground state

Also first calculation Singh and Du PRL(2008), PM state



#### LOFA: Band Structures





a) CB-AFM state, b) NM state.



Red lines: DFT results; Blue lines: fitted TB-Hamiltonian using maximally localized Wannier functions (MLWFs) MLWFs: 10 Fe-3d orbitals



(a)NM-unrelaxed

Gao, Hirschfeld, and Cheng, PRB77, 220506R(2008)

#### LOFA: Two-Dimensionality



Fermi surface constructed using tight-binding H from 10-orbital MLWF

### Strongly-correlated system? GGA+U



Mott transition @ ~ U=3eV,  $E_g$ =1 eV @ U=4.5 eV

But no ARPES experiment shows broadening of spectrum lines

Not a strongly-correlated system in conventional sense (such as BSCCO) => everybody ••• But, then what?

# Constructing PAW potential (for PWSCF) Constructing effective Hamiltonian



•Problem with USSP: Severe overestimation of Fe magnetic moment (~2  $\mu_B$  from uspp, ~1  $\mu_B$  from FLAMP, ~0.7  $\mu_B$ from experiments), magnetic stabilization energy

•Problem with FLAPW: too expensive to perform full structural optimization while full relaxation is preferred to study pressure induced phase transition and doping induced quantum

•Problem with  $Ba_1Fe_2As_2$ : DFT calculation with full relaxation fails to predict experimental structure, short c-axis (by over 10%).

### PAW for $Ba_1Fe_2As_2$

	PAV	V	USPP		
atom	valence	Rcut	valence	Rcut	
Fe	3s3p3d4s4p	2.0 (2.1 <sup>d</sup> )	3d4s4p	2.2 (2.3 <sup>d</sup> )	
Ва	5s5p5d	2.7(2.8 <sup>s</sup> )	5s5p6s	2.5(2.1 <sup>p</sup> )	
As	4s4p3d	2.1	4s4p	2.4(2.1 <sup>p</sup> )	

Atomic tests: at least 4 different configurations

Crystal tests (pure crystal and crystal oxides): magnetism, equilibrium lattice constant, bulk modulus

P. E. Blochl, "Projector Augmented-Wave Method," Physical Review B 50 (24), 17953-17979 (1994).

### Compared with FLAPW results

		SDW-	AFM	CB-AFM		
		unrelaxed	opt-z	unrelaxed	opt-z	
m <sub>Fe</sub> (μ <sub>B</sub> )	FLAPW	1.75	0.70	1.60	N/A	
	PAW	1.75	0.60	1.54	N/A	
	USPP	2.12	1.04	1.90	0.58	
	FLAPW	92	N/R	41	N/A	
ΔE (meV)	PAW	65	<1	19	N/A	
	USPP	119	6.9	60	<1	

FLAPW results taken from D. J. Singh, *Phys. Rev. B* **78**, 094511 (2008), where the SDW-AFM/CB-AFM crystal structure was assumed to be the same as NM crystal structure.

"unrelaxed" refers to experimental structure, "opt-z" refers to fixed lattice constant with relaxed internal coordinates.



NM state band structure FLAPW (WIEN2k): reproduces D. J. Singh, *Phys. Rev. B* 78, 094511 (2008)

#### Ba<sub>1</sub>Fe<sub>2</sub>As<sub>2</sub>: Structure relaxation

	NN	1	SDW-AFM		
	opt-z	full-opt	opt-z	full-opt	
а	3.963	3.873(3.968)	5.615	(5.697)	
b	3.963	3.873(3.968)	5.574	(5.594)	
С	13.02	12.14(12.49)	12.94	(12.76)	
Z <sub>As</sub>	0.341(0.344)	0.347(0.346)	0.341(0.352)	(0.3514)	
m <sub>Fe</sub> (μ <sub>B</sub> )	0.0	0.0	1.62(2.14)	(2.19)	
ΔE (meV)	0.0	0.0	105(83)	(68)	

"opt-z": optimized  $z_{As}$  with fixed crystal lattice constants "full-opt": optimized  $z_{As}$  and optimized crystal lattice constants

results: LDA (PBE)

### Ba<sub>1</sub>Fe<sub>2</sub>As<sub>2</sub>: Electronic Structure



#### $Ba_1Fe_2As_2$ : Fermi surface, nesting, etc

k<sub>y</sub>

 $\bigcirc$ 

 $k_z = 0.0$ 

k<sub>x</sub>



(a)NM-unrelaxed







k<sub>z</sub>=0.5

∕k<sub>x</sub>

k<sub>y</sub>

(b)NM-full-opt

NM-unrelaxed

SDW-opt

### Remarks on Ba<sub>1</sub>Fe<sub>2</sub>As<sub>2</sub>

SDW-AFM ground state of undoped parental iron-pnictide compound
Structure sensitive electronic structure in NM state, structure not so sensitive electronic structure in SDW-AFM state

•Neither LDA or GGA can describe NM state well, but LDA cannot even get SDW-AFM ground state

•Recent calculations (by Y. Wu): Neither TPSS nor HSE helps.



•Phonon calculations in NM state are not reliable

•Experimental geometry must be preserved to obtain correct electronic structure in NM state

•The apparent correlation between magnetism and crystal structure: magnetophonon coupling as electron pairing mechanism.

•Can QMC help?

$$\begin{array}{l} & \text{Down folding} \\ H_0 = \sum_{\mathbf{k}\sigma \ mn} \left( \xi_{mn}(\mathbf{k}) + \epsilon_m \delta_{mn} \right) d_{m\sigma}^{\dagger}(\mathbf{k}) d_{n\sigma}(\mathbf{k}), \\ H_{int} = U \sum_{i_s} n_{i,s\uparrow} n_{i_s\downarrow} + \frac{V}{2} \sum_{i_s,i\neq s} n_{i_s} n_{i_t} - \frac{J}{2} \sum_{i_s,i\neq s} \tilde{s}_i \cdot \tilde{s}_{i_t} + \frac{J'}{2} \sum_{i_s,i\neq s} \sum_{\sigma} c_{i_s\sigma}^{\dagger} c_$$

The colors correspond to  $d_{xz}$  red ,  $d_{yz}$  green ,  $d_{xy}$  blue ,  $dx^2-y^2$  orange , and  $d3z^2-r^2$ magenta . All energies are measured from the Fermi energy  $E_F$ =10.86eV

S. Graser, et al. PRB81, 214503(2010)

Due to the high degeneracy of the two As positions in NM state, it is convenient to look at an effective unit cell with only 1 Fe and 1 As atom.

10 orbitals => 5 orbitals



N: number of Fe lattice sites,  $\beta=1/T$ ; n,m: the fermionic, and bosonic Matsubara frequencies in the imaginary time formalism; s, t, p, and q: indices of Fe 3d orbitals 3D susceptibility the momentum sum runs over  $k_x$ ,  $k_y$ , and  $k_z$  2D:  $k_z$  is kept fixed and  $q_z=0$ .

$$G_{sp}(\mathbf{k}, i\omega_n) = \sum_{\mu} \frac{a_{\mu}^{s}(\mathbf{k})a_{\mu}^{p*}(\mathbf{k})}{i\omega_n - E_{\mu}(\mathbf{k})} \qquad a_{\mu}^{s}(\mathbf{k}) = \langle s \mid \mu \mathbf{k} \rangle \qquad S: \text{ orbital} \\ \mu: \text{ band} \\ k: \text{ k-value} \end{cases}$$

$$\chi_{qtsp}(\mathbf{q}, \omega) = -\frac{1}{N} \sum_{\mathbf{k}, \mu\nu} \frac{a_{\mu}^{p}(\mathbf{k})a_{\mu}^{t*}(\mathbf{k})a_{\nu}^{q}(\mathbf{k}+\mathbf{q})a_{\nu}^{s*}(\mathbf{k}+\mathbf{q})}{\omega + E_{\nu}(\mathbf{k}+\mathbf{q}) - E_{\mu}(\mathbf{k}) + i0^{+}} \qquad \times \{f[E_{\nu}(\mathbf{k}+\mathbf{q})] - f[E_{\mu}(\mathbf{k})]\} \qquad \text{K.Kubo, PRB 75, 224509 (2007)}$$

$$(\chi_1^{\text{RPA}})_{stpq} = \chi_{stpq} + (\chi_1^{\text{RPA}})_{stuv} (U^s)_{uvwz} \chi_{wzpq}$$

# Spin-fluctuation theory and iron-pnictides

- The theory sums a few classes of diagrams where electrons scatter by local Hubbard interactions and Hund's rule exchange.
- All the interactions are repulsive, but they generically produce a pairing interaction (magnetic susceptibility) which is peaked at π,0 in the 1-Fe zone.
- Gaining pairing energy (in the case of a repulsive interaction) by creating a pair state which changes sign when k-k' = q (where the interaction is sharply peaked).
- In the iron pnictides, the q = π,0 connects two nearly nested Fermi surfaces (this is the origin of the peak at π,0), => a sign change in Δ between the hole pockets and electron pockets enables the system to pair even though the sign of the interaction is repulsive (first proposed by Mazin for an isotropic sign changing s-wave state)
- Our contribution: the sign change is there but the k-dependence is strong. These effects (in addition to zeroth order sign changing s-wave state) come from the orbital character of the Fermi surface and the intraband Coulomb interaction. S. Graser, et al. PRB81, 214503(2010)





# Calculation details

- DFT / PBE Quantum Espresso
- Plane wave+ultra-soft pseudo-potential
- Energy cut-off 40 Ry
- Density cut-off 400 Ry
- Maximally localized Wannier functions → constructing tight-binding H<sub>ij</sub> and Fermi surfaces
- PM state: a=3.9625, c=13.0168 Å l4/mmm, tetragonal
- Undoped SDW: a= 5.6146, b = 5.5742 and c = 12.9453 Å Fmmm orthorombic
- Doped x=0.16, for both 40/80 atoms with 8/16 Fe atoms. Relaxation for 40/8.

# Notes on Co-doping

- Directly replaces the Fe atom (Fe: 26; Co:27)
- Co is similar to Fe (compared to alkali), but the inplane doping possibly introduces a strong scattering potential
- Co is ferromagnetic, may have strong influence on the SDW state of 122 materials
- Co-doping suppresses and splits the magnetic and structural phase transitions P. C. Canfield et al., preprint (2009), arXiv:

0904.3134.

• Co doping corresponds to a homogenous coexistence of PM&SDW states X.F. Wang (2008), arXiv:0811.2920 .... Y. Laplace et al, reprint (2009) arXiv:0906.2125

## PM vs. SDW States

at Γ.





Undoped PM band structure along high-

symmetry lines: electron pockets at M and hole

DOS for BaFe2As2 in the undoped PM and undoped SDW states. The Fermi levels for both systems are aligned at 0.

- SDW: spin strip along 110 direction, 0.55 eV/primitive lower than PM state
- PM state has high DOS at the Fermi energy

k<sub>y</sub> (π,π) k<sub>x</sub>



## Co-doping the SDW state: $Ba(Fe_{1-x}Co_x)_2As_2$



Left: Majority of displacements occurs near the dopant site, the Co atom pushes away the Fe atoms that lie along the line of collinear spins in the SDW state; the Co atom attracts the nearest As atoms, causing the distance to decrease by 0.03 Å.





Doping induce a small decrease in DOS at the Fermi level, the two DOS are similar, is rigid-band model valid?

## Band structure of the SDW state





Undoped SDW band structure along high symmetry lines. Spin up- and down-spin are the same. No electron and hole pockets.

Doped SDW band structure along high-symmetry lines. Black (green) are the majority (minority) spin. A number of crossings have appeared from X to M

- Co breaks the degeneracy between up and down electrons
- The net spin of the system increases from 0 to 0.46  $u_B$ /cell.
- Co induce a strong non-local effect, decreasing the magnetization over the entire plane, and enhancing the spin polarization in the undoped plane.



- Very clear resonances in the Co signal at -800 and +200 meV.
- The 200 meV peak agrees quantitatively with the bias used to image Co atoms on the surface of lightly Co-doped Ca-122 in STM recently

(T.-M. Chuang et al, preprint (2009), submitted)



- The collinear SDW state couples very strongly to the Co and drives a large anisotropy in the effective potential
- Charge channel feature localized within ~ 1 Å, spin  $\rightarrow$  non-local
- Within the size of our unit cell, no significant decay of spin potential is observed → may be responsible for the destruction of magnetic order

# Fourier Transform



charge spin Shell structure is better seen in the 2-D plot

# Effective impurity Hamiltonian

$$U_c^m(\vec{R}) = \sum_{\sigma} \langle \phi_{l=2}^m(\vec{r} - \vec{R}) | \Delta V_\sigma(\vec{r}) | \phi_{l=2}^m(\vec{r} - \vec{R}) \rangle,$$

V: KS potential for spin up and down

 $U^m_s(\vec{R}) = \sum_{\sigma} \langle \ \phi^m_{l=2}(\vec{r}-\vec{R}) | \sigma \Delta V_{\sigma}(\vec{r}) | \phi^m_{l=2}(\vec{r}-\vec{R}) \rangle, \label{eq:User}$ 

 $\Delta V$ : difference between V spin up and down

		Impurity site Same-spin neighbo		pin neighbor	Opposite-spin neight		
m	Orbitals	$U^m_c$	$U^m_s$	$U_c^m$	$U_s^m$	$U_c^m$	$U^m_s$
0	$d_{z^{2}}$	1.29	-0.331	0.572	0.709	-0.634	-0.639
$\pm 1$	$d_{xz}, d_{yz}$	1.52	-0.378	0.672	0.848	-0.805	-0.733
$\pm 2$	$d_{x^2-y^2}, d_{xy}$	1.87	-0.495	0.882	1.099	-0.956	-0.965

Useful for phenomenological modelers (all in eV)

(more work for Peter)



# Local DOS



Plane cuts of the local Fermi level DOS (see Eq. 3). Red (gray) balls indicate Fe ion in the spin down (up) state. The Co dopant is indicated blue, circled where visible.

- (A) Local DOS increases along the line of majority spin and decreases along the minority, commensurate with SDW, for both doped and undoped plane
- (B) and (C) show a weak contrast compared to (A).
- Strips in (A)-(B) may be visible by STM



Red: spin-down Fe, grey: spin up Fe atom, LDOS scaled by max(in-plane ldos)

- LDOS show 122 systems having more states between 2 FeAs layers than 1111 system Origin: As atoms are out of phase in 122 materials
- Co doping reduces the interlayer states  $\rightarrow$  increases anisotropy

Summary on Co-doping 122

- Co is found in position of Fe
- Co dopes the FeAs plane
- Co induces a stripe-like modulation
- Intermediate strength, non-magnetic scattering potential within the range of ~ 1 Angstrom
- 3D characteristic compared to 1111 systems

# Goal: Designing new high T<sub>c</sub> materials

Thank you!