Quasiparticle dynamics and interactions in non-uniformly polarizable solids

Mona Berciu

University of British Columbia

→ beautiful physics that George Sawatzky has been pursuing for a long time

→ today, example is of iron-pnictides (FeAs) and iron-chalcogenides (FeSe)

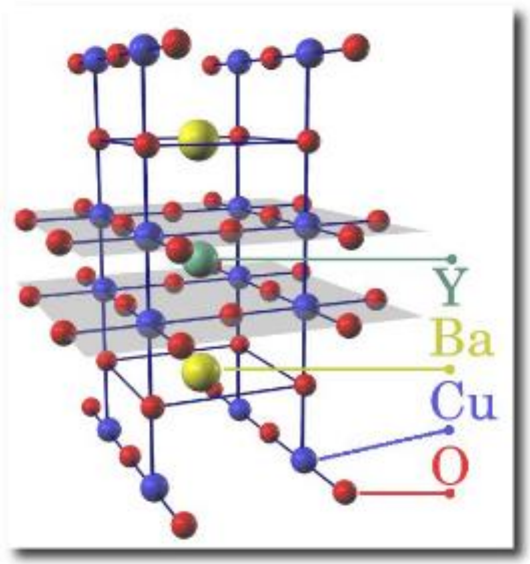
→ references:

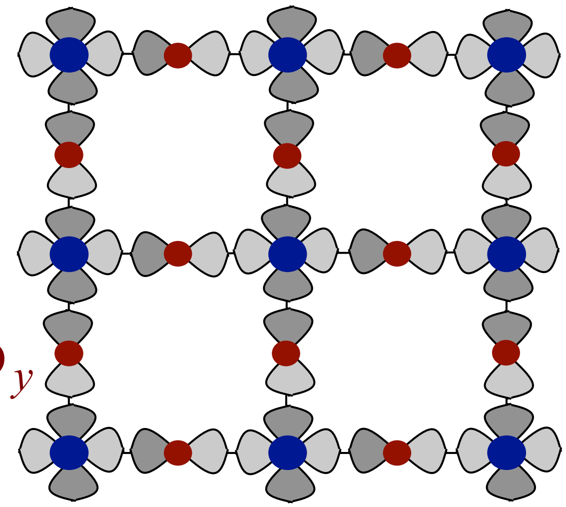
- **G. Sawatzky, I. Elfimov, J. van den Brink and J. Zaanen, EPL 86, 17006 (2009)**
- **M. Berciu, I. Elfimov and G. Sawatzky, PRB 79, 214507 (2009)**

Stewart Blusson Quantum Matter Institute

NSERC CRSNG

~1997: discovery of high-temperature superconducting cuprates

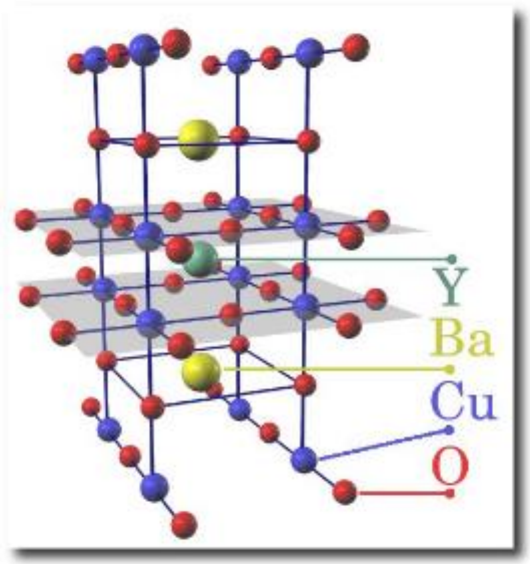


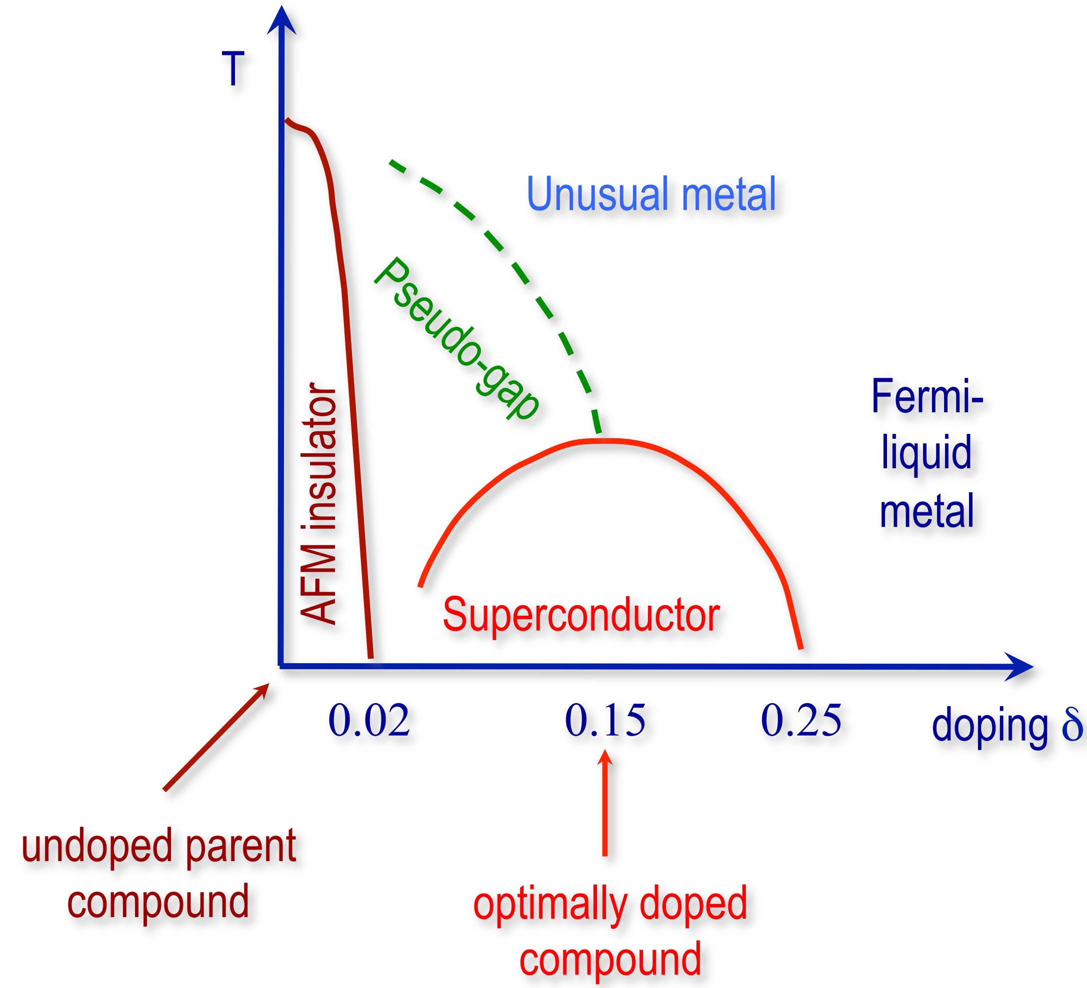


Common elements:

→ planar CuO_2 layers

~1997: discovery of high-temperature superconducting cuprates





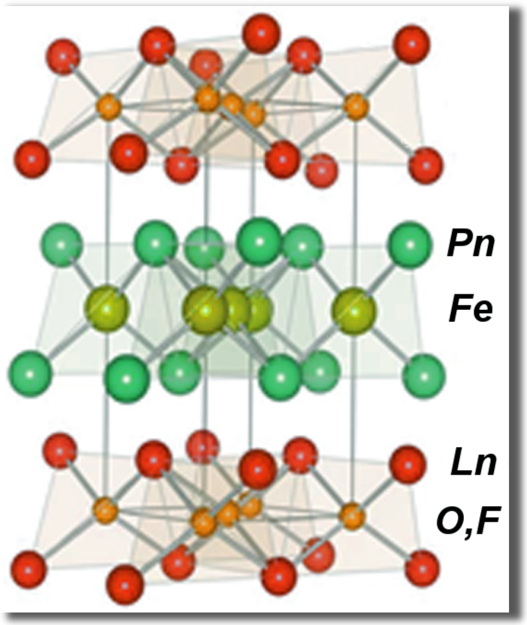
Common elements:

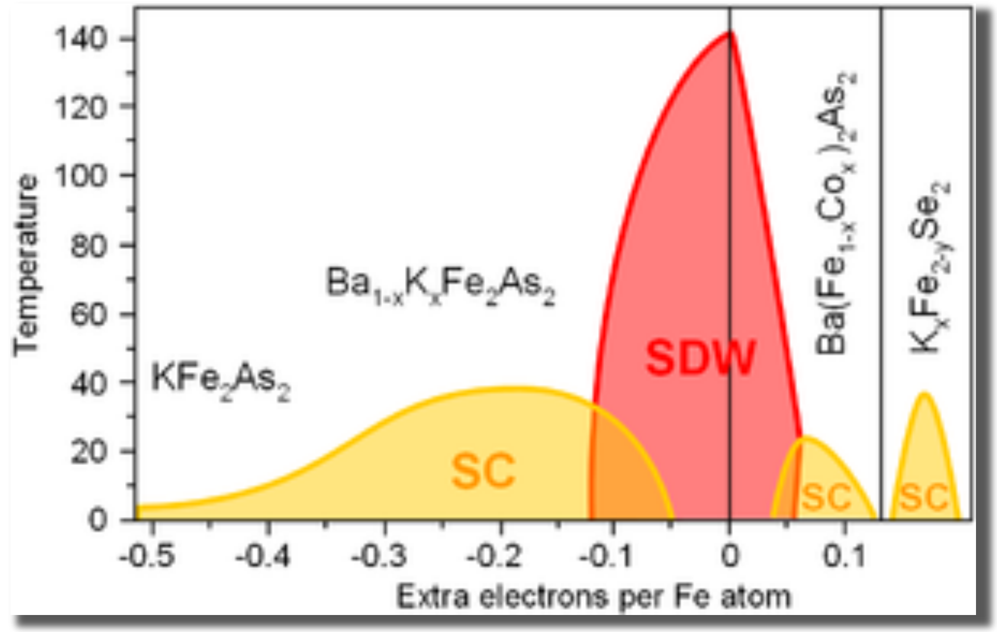
→ planar CuO_2 layers

→ Similar phase diagrams upon doping with holes

→ Mechanism still not understood …

~2008: discovery of high-temperature superconducting iron-pnictides/chalcogenides





Images from wikipedia: https://commons.wikimedia.org/w/index.php?curid=40347835

Common elements:

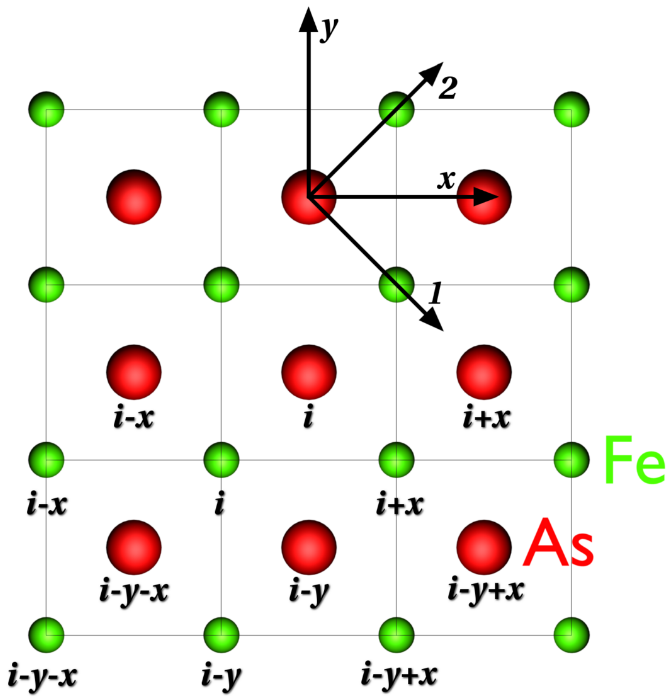
→ (quasi)-planar FePn layers (from now on, anion is taken to be As)

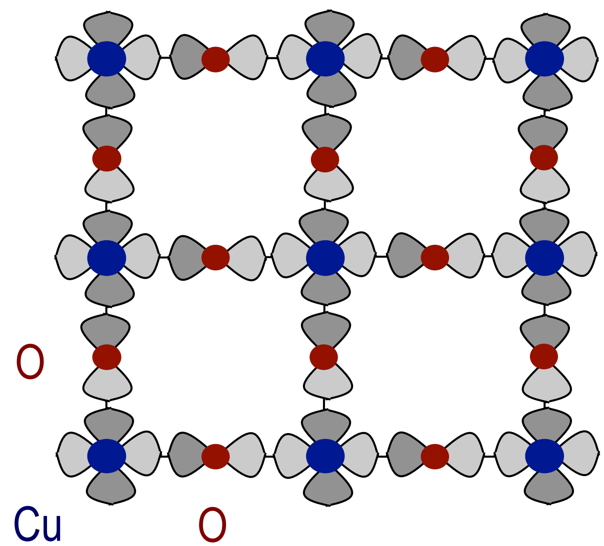
→ Phase diagrams somewhat similar to cuprates

→ Immediate assumption by part of the community: the physics must be the same!

Similarities between CuO_2 and FeAs layers:

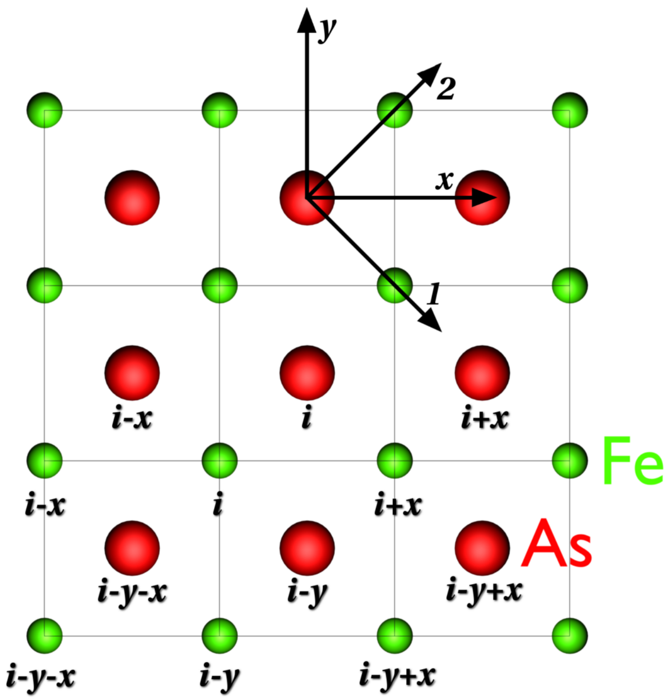
1. undoped parent compound: O 2p and As 4p shells are filled, while Cu $3d^9$ and Fe $3d^6$ → valence electrons are in partially filled 3d shell (hence magnetic properties expected)
2. both Cu and Fe are on planar square lattices
3. both Cu and Fe are in between 4 different anions …



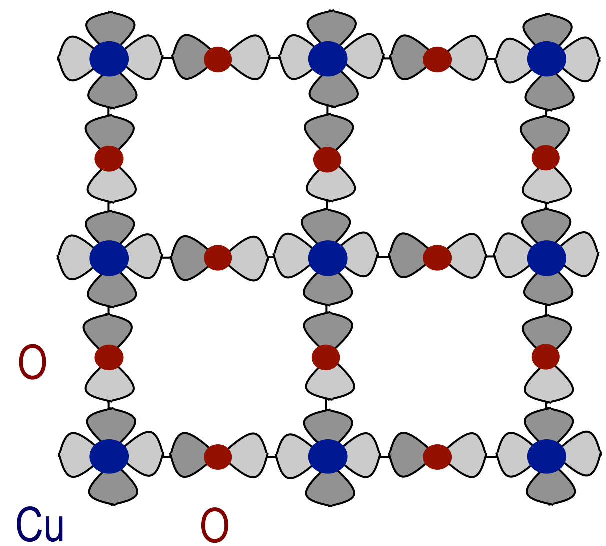


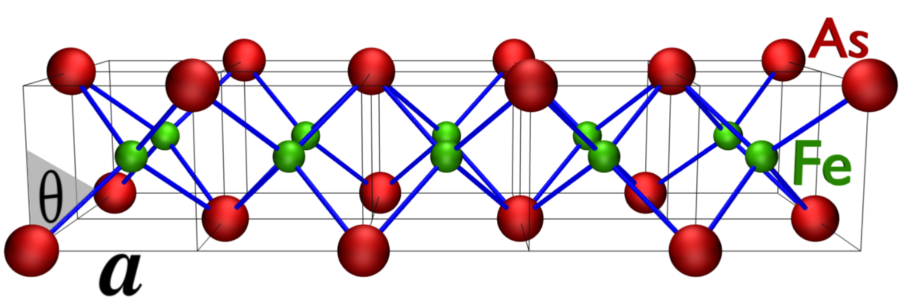
Differences between CuO_2 and FeAs layers:

1. Fe is inside octahedron of 4 As, while Cu is between 4 in-plane O



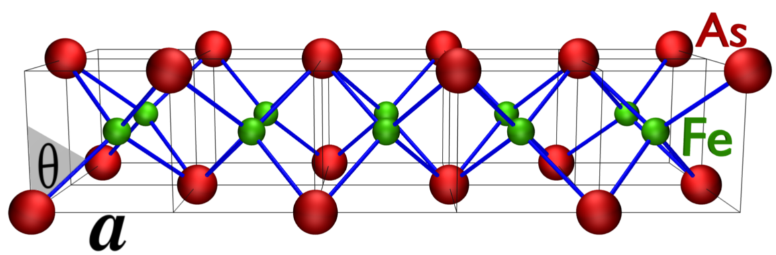
FeAs layer, top view

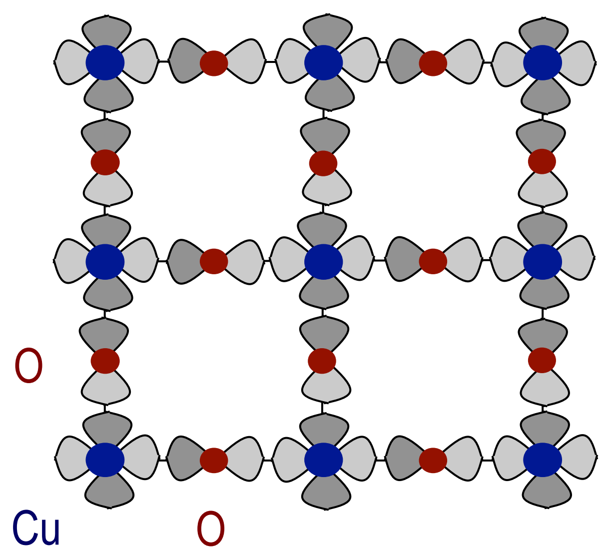


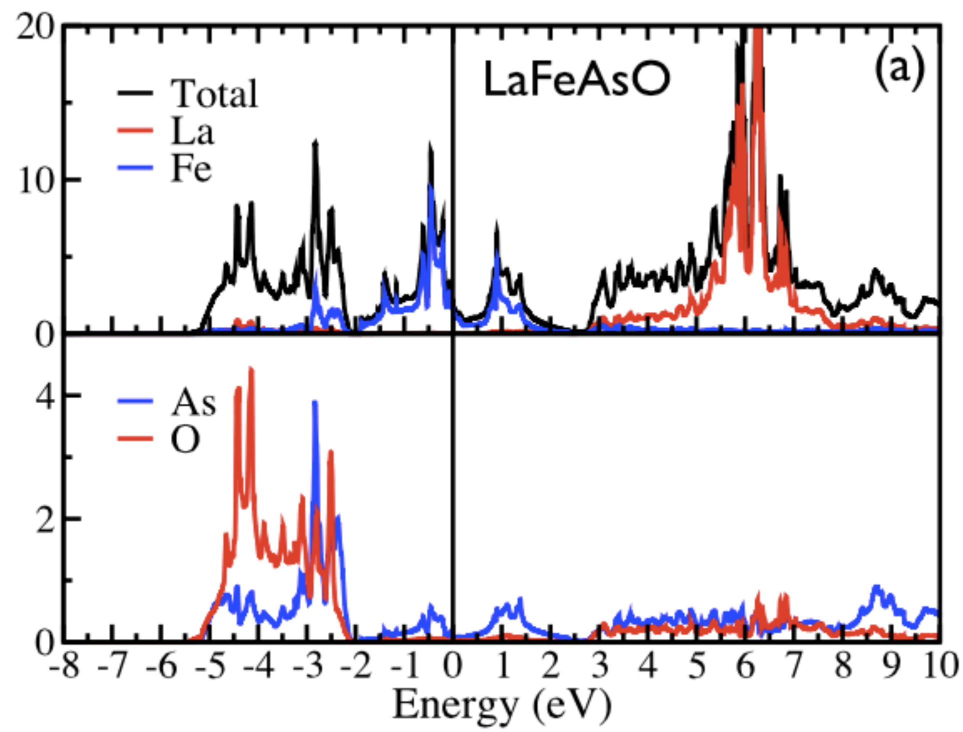


Differences between CuO_2 and FeAs layers:

1. Fe is inside octahedron of 4 As, while Cu is between 4 in-plane O
2. Because of different geometry + much smaller 2p O as compared to 4p As orbitals → while Cu and ligand O2p hybridize strongly, in FeAs all levels close to Fermi energy have almost pure Fe character







G. Sawatzky et al, EPL 86, 17006 (2009)

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1. Fe is inside octahedron of 4 As, while Cu is between 4 in-plane O
2. Because of different geometry + much smaller 2p O as compared to 4p As orbitals → while Cu and ligand O2p hybridize strongly, in FeAs all levels close to Fermi energy have almost pure Fe character.
3. Also: large O-O overlap → wide O 2p band, which hosts the doped holes (charge transfer insulator). Whereas long As-As distance → very narrow band, far from Ef, so the doped carriers go in the Fe 3d states.
4. Cu U_{dd} very large → strongly correlated insulator if undoped, while Fe U_{dd} is rather small → poor metal even when undoped

Q: Why is Fe U_{dd} screened so much more than Cu U_{dd}?

Are the As doing anything at all?!

Q: How to model doped FeAs layer (e on top of the Fe: $3d^6$ As: $4p^6$ in the parent compound)?

A: DFT tells us that all states within 2eV of E_F are of Fe nature, so we can use a Hubbard-like Hamiltonian for 3d Fe placed on square lattice, and argue whether we should use 2 or 5 bands, and whether U large or small, etc.

$$\mathcal{H}_{\mathrm{Fe}} = -\sum_{i,j,\sigma} \left(t_{ij} c^{\dagger}_{i,\sigma} c_{j,\sigma} + h.c. \right) + U_H \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

We take t ~ 0.25eV (DFT), t'= -t/2 or 0, and U_H ~ 10eV because 3d levels have large U

However, little hybridization between Fe with As, and As bands far from Ef

→ we can totally ignore the As so far as electronic properties are concerned, right?

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However, little hybridization between Fe with As, and As bands far from Ef

→ we can totally ignore the As so far as electronic properties are concerned, right?

WRONG!

Q: What do we know about the As ions?

A: full 4p orbitals, large gap to empty 5s orbitals → very “fat” spherical distribution of charge.

Q: What happens when we put extra charges (doped electrons) in its vicinity?

A: The additional electric field will polarize the charge cloud, it will no longer be spherical → dipole moment p created, energy is lowered

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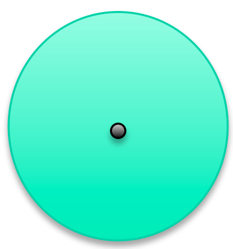
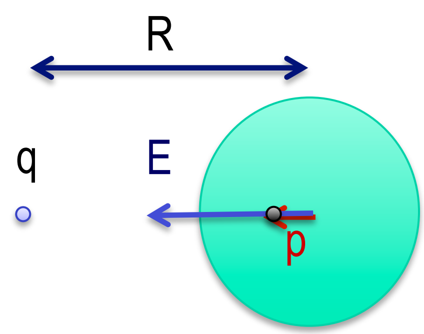
Semi-classical picture:



unperturbed As ion



perturbed As ion

$$\vec{p} = \alpha \vec{E} \qquad \alpha = \text{ polarizability } \propto V$$

$$W \sim -\vec{p}\cdot\vec{E} = -\frac{1}{2}\alpha \vec{E}^2$$

$$\text{Note: } \vec{E} = \frac{q\vec{e}}{R^2} \text{ is determined by}$$

$$\text{lattice geometry}$$

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Quantum picture (very atomistic, but justified given narrow As bands)

Electron from p orbital parallel to E is excited into s orbital,charge distribution is deformed → induced dipole moment

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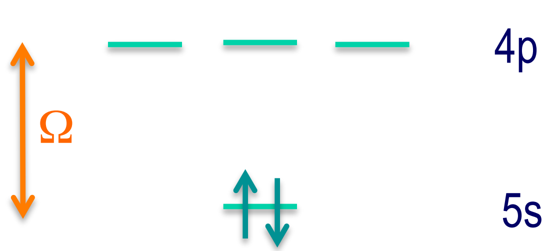
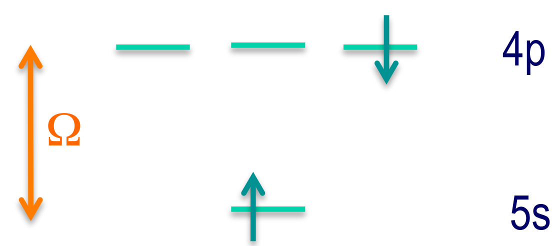
Quantum picture: use hole operators instead!



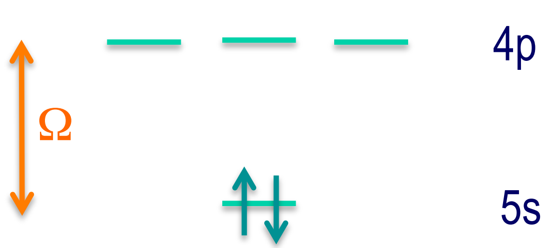
unperturbed As ion



perturbed As ion

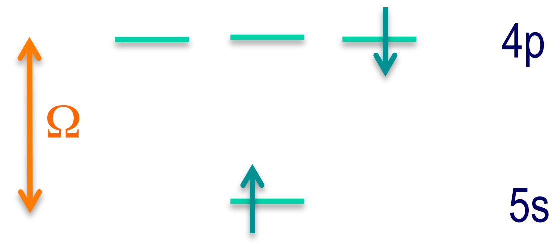
Hole is excited into 4p orbital parallel to E, charge distribution is deformed → induced dipole moment

Quantum picture for As ion:



unperturbed As ion

$$h_{As} = \Omega \sum_{\lambda,\sigma} p^{\dagger}_{\lambda\sigma} p_{\lambda\sigma}$$



perturbed As ion

$$h = h_{As} + h_{q-As}$$

$$h_{q-As} = -\hat{\vec{p}} \cdot \vec{E} = -g \sum_{\sigma} \left(s^{\dagger}_{\sigma} p_{\vec{e}\sigma} + hc \right)$$

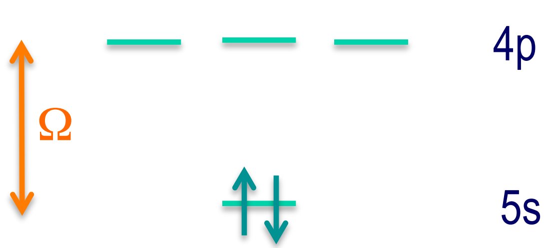
$$\vec{E} = \frac{q\vec{e}}{R^2}$$

$$\hat{\vec{p}} = \sum_{\lambda,\sigma} \langle s | e\vec{r} | p_{\lambda} \rangle \left(s^{\dagger}_{\sigma} p_{\lambda,\sigma} + h.c. \right)$$

$$a_{\mathrm{As}} = \langle s | x | p_x \rangle = \int d\vec{r} \phi^{*}_{s}(\vec{r}) x \phi_{p_x}(\vec{r})$$

$$g = e a_{\mathrm{As}} \frac{q}{R^2}$$

Quantum picture for As ion:



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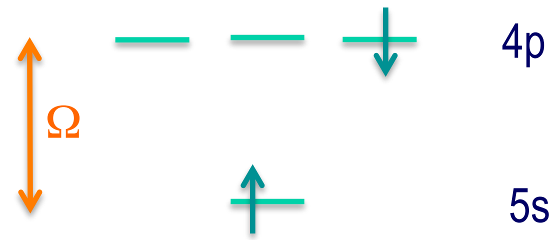

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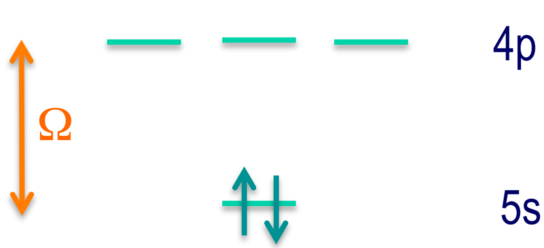
$$g = e a_{\mathrm{As}} \frac{q}{R^2}$$

Parameters:

Ω ~ 6eV (DFT)

g =? ← use “known” α ~ 10A^3

Quantum picture for As ion:



unperturbed As ion

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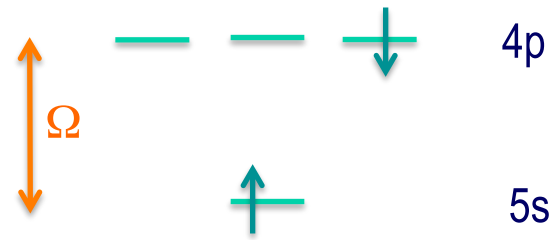


perturbed As ion

$$\hat{h} = \Omega \sum_\sigma p^\dagger_\sigma p_\sigma + g \sum_\sigma (s^\dagger_\sigma p_\sigma + h.c.)$$

$$E_{\text{cloud}} = \Omega - \sqrt{\Omega^2 + 4g^2} \to -\frac{2g^2}{\Omega} = -\frac{1}{2}\alpha \vec{E}^2$$

$$|c\rangle = \gamma^\dagger_\uparrow \gamma^\dagger_\downarrow |0\rangle \qquad \gamma^\dagger_\sigma = \cos\theta s^\dagger_\sigma - \sin\theta p^\dagger_\sigma$$

$$\cos\theta = \sqrt{\frac{1}{2}\left(1 + \frac{\Omega}{\sqrt{\Omega^2 + 4g^2}}\right)}$$

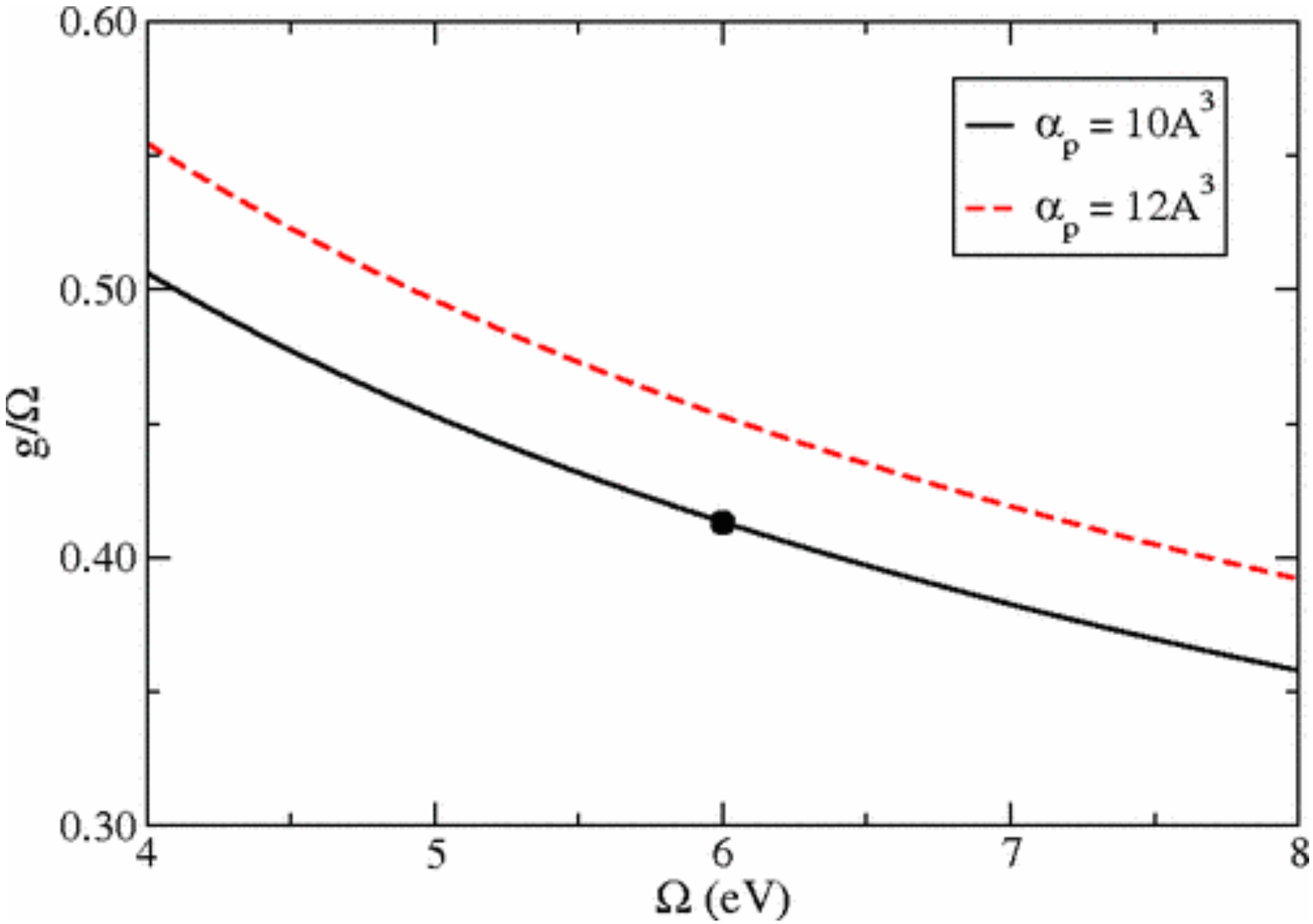
$$\langle p \rangle = \langle c|\vec{e}\cdot\hat{\vec{p}}|c\rangle = \frac{4ea_{\text{As}}g}{\sqrt{\Omega^2 + 4g^2}} \to \alpha E$$

$$g = \sqrt{\frac{\alpha\Omega e^2}{4R^4}}$$

Parameters:

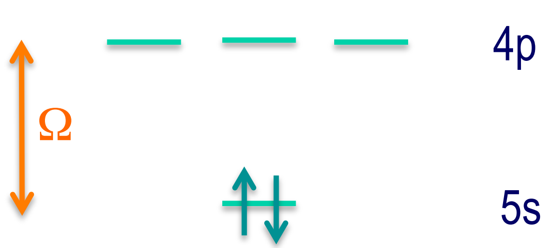
Ω ~ 6eV (DFT)

g ~ 0.4Ω if α~10A^3



0.60
0.50
0.40
0.30
4
5
6
7
8
g/Ω
Ω (eV)
$\alpha_p = 10A^3$
$\alpha_p = 12A^3$

Quantum picture for As ion:



unperturbed As ion

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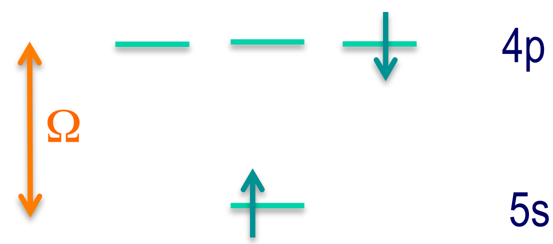

perturbed As ion

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$$g = \sqrt{\frac{\alpha \Omega e^2}{4R^4}}$$

Parameters:

Ω ~ 6eV (DFT)

g ~ 0.4Ω if α~$10A^3$

no longer linear limit

max <p> when $\theta = 45^o$

Model Hamiltonian:

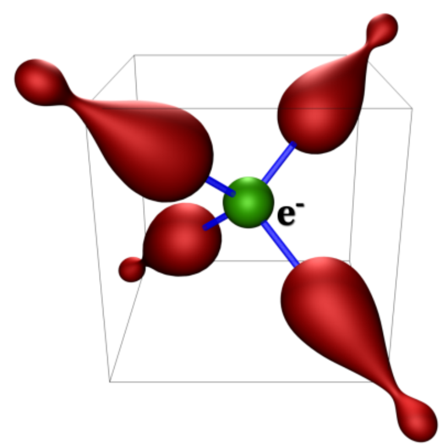
→ use a single band for Fe, for simplicity

$$\mathcal{H}_{\text{Fe}} = \hat{T} + \hat{T}' + U_H \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

$$\mathcal{H}_{\text{As}} = \Omega \sum_{i,\lambda,\sigma} p^\dagger_{i,\lambda,\sigma} p_{i,\lambda,\sigma}$$

→ for simplicity, assume that only the 4 nn As are polarized (E falls off like $1/R^2$)

$$\begin{aligned}\mathcal{H}_{\text{int}} = g \sum_{i,\sigma} \hat{n}_i \Big[& s^\dagger_{i,\sigma}\left(-\sin\theta p_{i,2,\sigma} + \cos\theta p_{i,3,\sigma}\right) \\ & + s^\dagger_{i-x,\sigma}\left(\sin\theta p_{i-x,1,\sigma} + \cos\theta p_{i-x,3,\sigma}\right) \\ & + s^\dagger_{i-x-y,\sigma}\left(\sin\theta p_{i-x-y,2,\sigma} + \cos\theta p_{i-x-y,3,\sigma}\right) \\ & + s^\dagger_{i-y,\sigma}\left(-\sin\theta p_{i-y,1,\sigma} + \cos\theta p_{i-y,3,\sigma}\right) + h.c. \Big]\end{aligned}$$



→ ignore dipole-dipole interactions for As

The consequences of these approximations are discussed later

Single polaron results

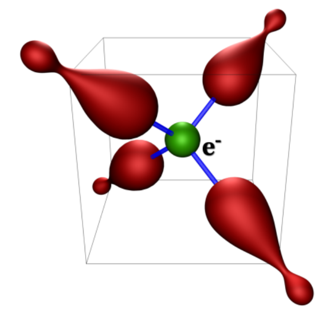
We use first order perturbation theory in T, justified because that energy scale is much smaller than the distance to first set of excited states:

0^{th} order: electron can be at any site, with 4 nn As polarized → electronic polaron

$$|\Phi_i\rangle = c_i^\dagger|i\rangle$$

$$|i\rangle = \prod_\sigma \gamma^\dagger_{i,2,-,\sigma}\gamma^\dagger_{i-x,1,+,\sigma}\gamma^\dagger_{i-x-y,2,+,\sigma}\gamma^\dagger_{i-y,1,-,\sigma} \prod_{|j-i|>1} s^\dagger_{j\sigma}|0\rangle$$

$$E_{P,GS} = 4E_{\rm cloud} = 4\left(\Omega - \sqrt{\Omega^2+4g^2}\right)$$

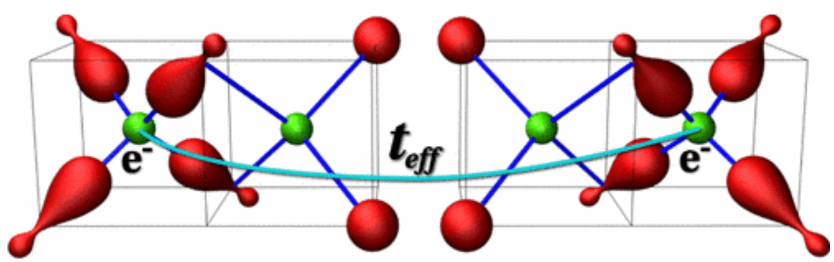


1^{st} order:

$$|\Phi_{\vec{k}}\rangle = \sum_i \frac{e^{i\vec{k}\cdot\vec{R}_i}}{N}|\Phi_i\rangle \qquad E_P(\vec{k}) = E_{P,GS} + \langle\Phi_{\vec{k}}|\hat{T}_{\rm tot}|\Phi_{\vec{k}}\rangle$$

$$E_P(\vec{k}) = E_{P,GS} - 2t_{\rm eff}\left[\cos(k_x a)+\cos(k_y a)\right] - 4t'_{\rm eff}\cos(k_x a)\cos(k_y a)$$

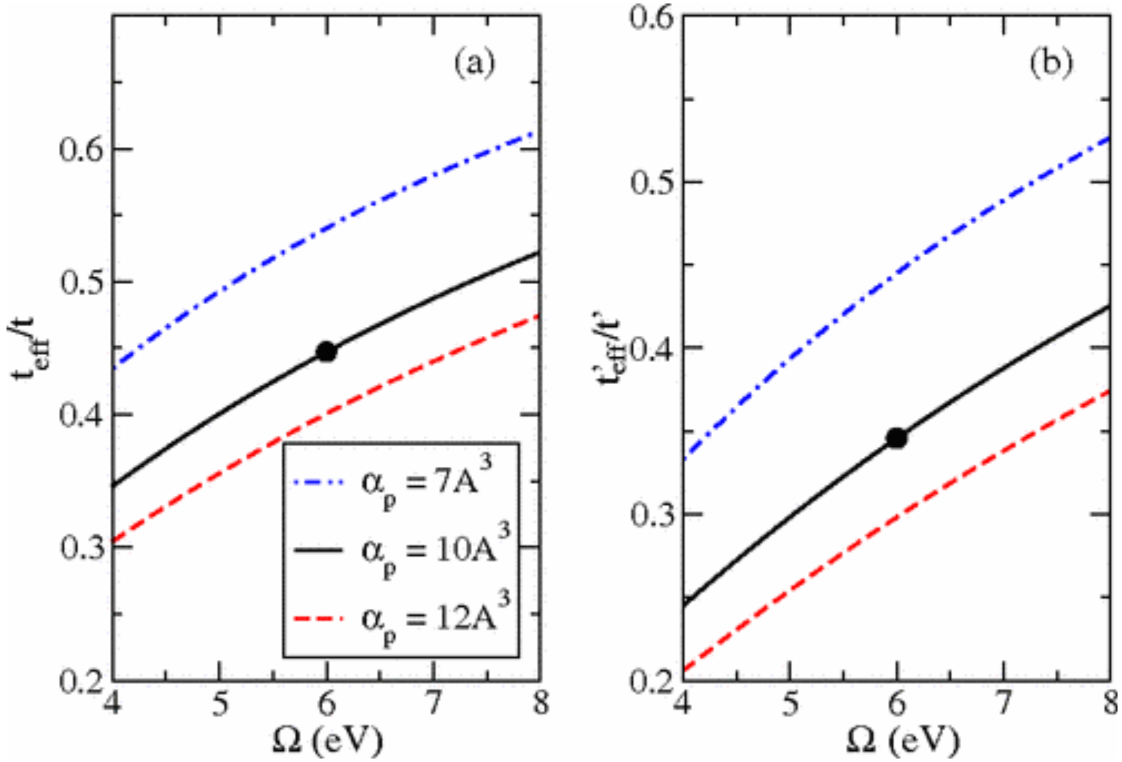
$$t_{\rm eff} = t\langle i|i+x\rangle; \qquad t'_{\rm eff} = t'\langle i|i+x+y\rangle$$



Typical polaronic effect: heavier quasiparticle because of the dressing cloud

Single polaron results (cont'd)

Qp mass enhancement is very moderate, by a factor of 2-3



→ In agreement with ARPES data which shows bands about 2.2 times narrower than DFT predictions, **eg. D. H. Lu et al, Nature 455, 81 (2008)**

→ Note: a multi-band dispersion would be renormalized by precisely the same amount

→ A posteriori justification for perturbation theory: gap to excited states is Ω~ 6eV, while qp bandwidth is ~0.5eV

Bi-polaron results → introduce two carriers and see what happens

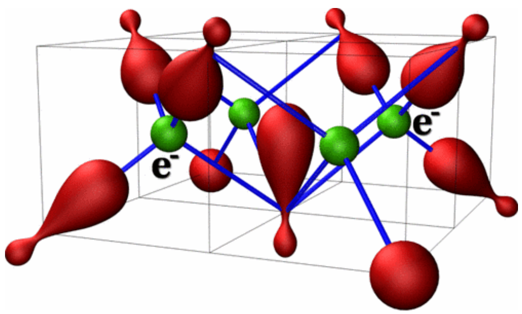
Again, use perturbation theory in T.

To 0th order, eigenstates have localized electrons with various clouds surrounding them:

→ If n > 2, no overlap between clouds:

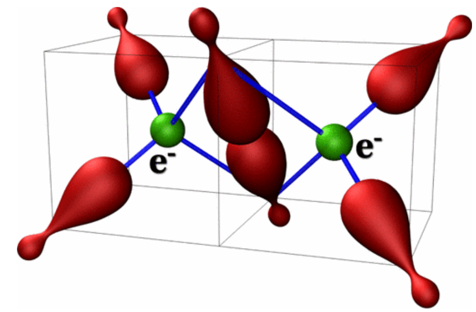
$$E_{BP,n>2} = 2 \times E_{P,GS} = 8\left[\Omega - \sqrt{\Omega^2 + 4g^2}\right]$$

→ If n=2 → one As shared by both clouds



$$U_2 = E_{BP,2} - E_{BP,\infty}$$

If n=1 → two As shared by both clouds



$$U_1 = E_{BP,1} - E_{BP,\infty}$$

→ If n=0 → all 4 As shared by both clouds, but each is polarized more strongly

$$U_0 = U_H + E_{BP,0} - E_{BP,\infty}$$

(of course, bare repulsion can be longer range as well)

Bi-polaron results (cont'd)

→ U_0 screened considerably

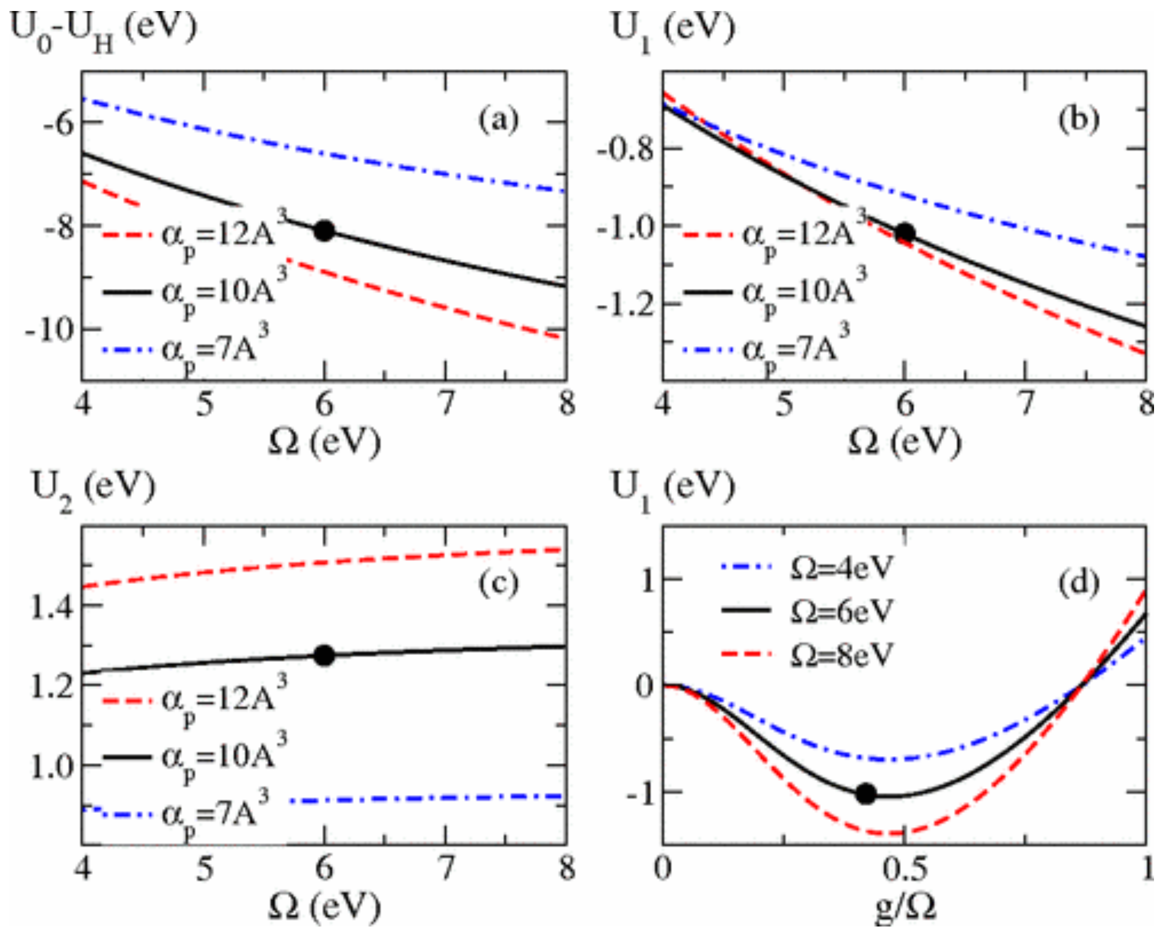
-- typical polaronic behavior

-- explains lack of corrrelations

→ U_1 is attractive!

→ U_2 is repulsive

→ Bound nn pair!!!! (pairing glue?)



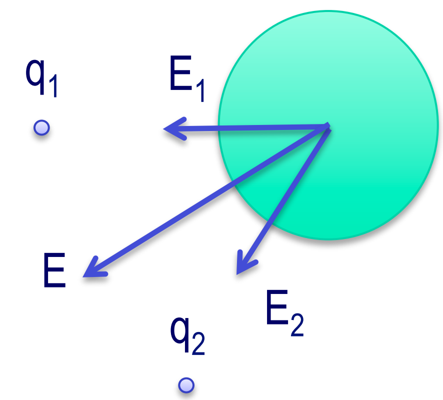
Q: 1. why this non-monotonic behavior of the effective interactions mediated by the clouds (i.e., inhomogeneous polarizable environment)

2. do bipolaron solutions survive if T is turned on, and how heavy are they?

Bi-polaron results (cont'd)

A1: at semi-classical level, energy is lowered by $W = -\frac{1}{2}\alpha\vec{E}^2$ controlled by applied electric field

If an As interacts simultaneously with two charges:



$$W = -\frac{1}{2}\alpha(\vec{E}_1 + \vec{E}_2)^2 = W_1 + W_2 + W_{int}$$

$$W_{int} = -\alpha\vec{E}_1 \bullet \vec{E}_2 = -\alpha E_1 E_2 \cos(\theta_{12})$$

→ The sign of W_{int} is controlled by geometry (lattice structure)! Ih both charges on same site, then θ=0 → always attraction. In FeAs structure, this angle < 90 for nn, > 90 for nnn.

→ Quantum model backs up these results unless g becomes very large and non-linear effects cannot be ignored

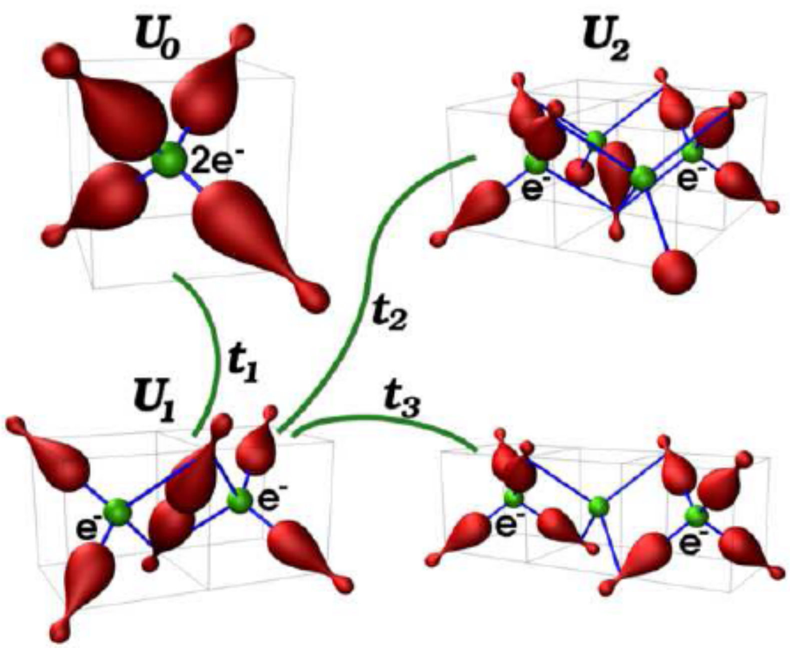
→ One might be able to play some interesting games by properly placing polarizable atoms in suitable locations … and this is a class of materials where this is happening!

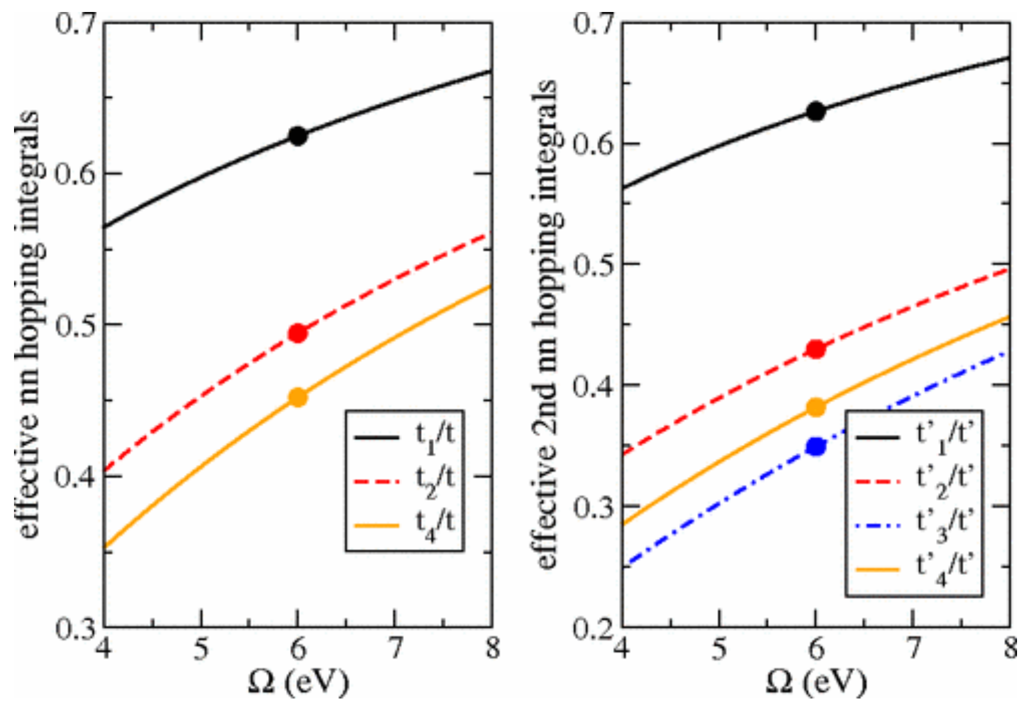
Bi-polaron results (cont'd)

A2: again, do perturbation theory in T. This time, many possible Bloch states!

$$|\vec{k},\vec{\delta}\rangle = \sum_i \frac{e^{i\vec{k}\cdot(\vec{R}_i+\frac{\vec{\delta}}{2})}}{N} s^\dagger_{i,i+\vec{\delta}}|i,i+\vec{\delta}\rangle.$$

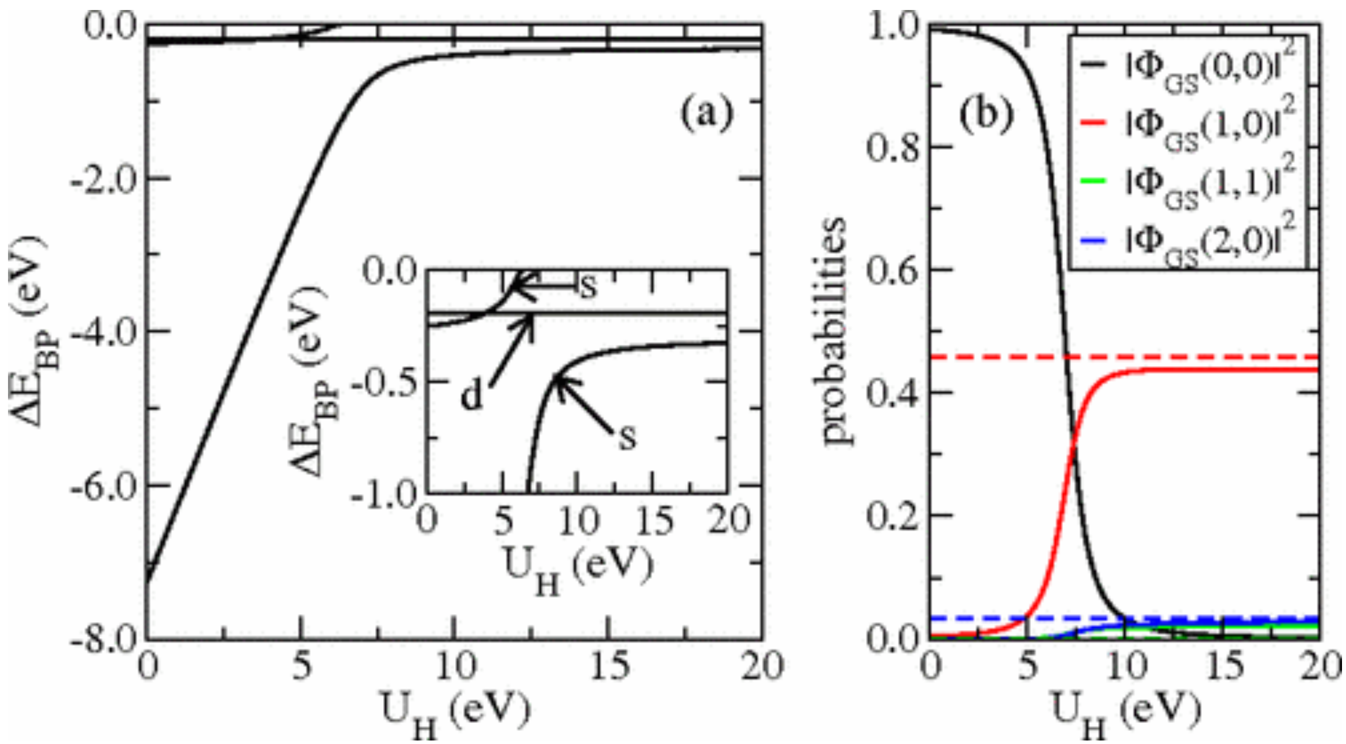
→ Need to compare many effective hoppings beside the effective interactions:





Bi-polaron results (cont'd)

A2: Bipolarons survive for arbitrarily large U_H.



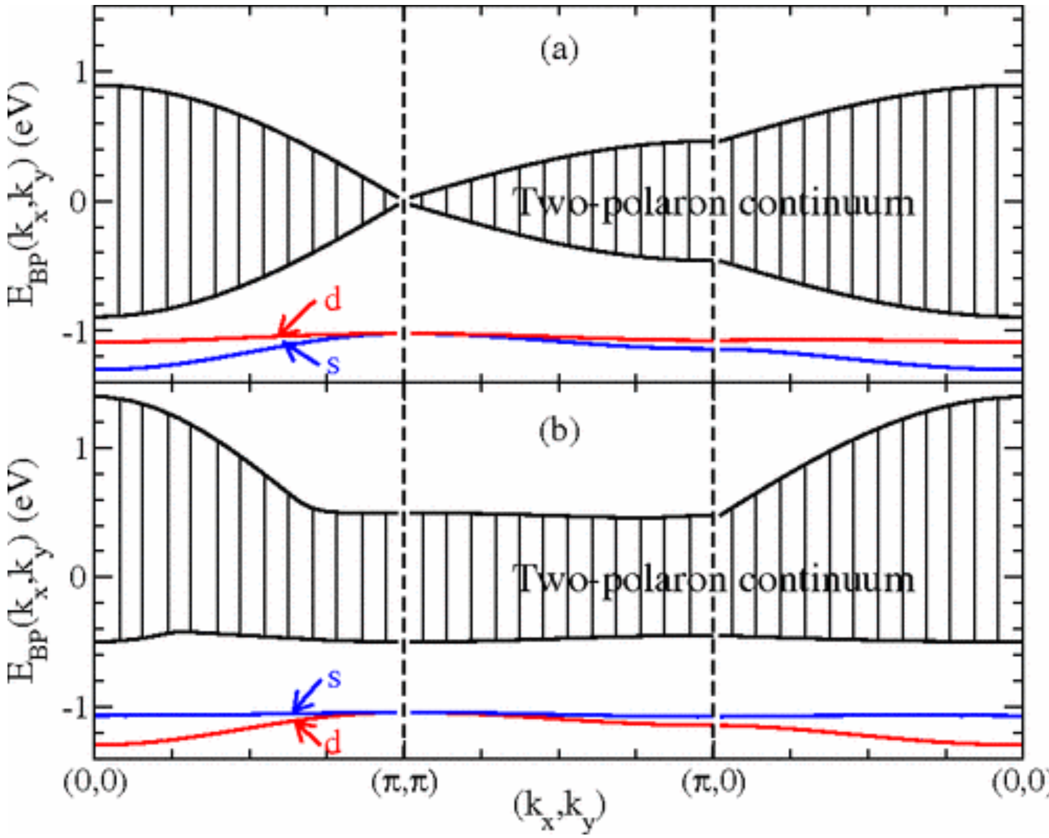
As U_H increases, S0 → S1 crossover. For U_H > 10eV, results independent of its value

Also note higher energy state with d-wave symmetry

These results are for t'=0

Bi-polaron results (cont'd)

A2: Stable bipolarons in the limit $U_H \to \infty$



$\leftarrow t' = 0$

$\leftarrow t' \neq 0$

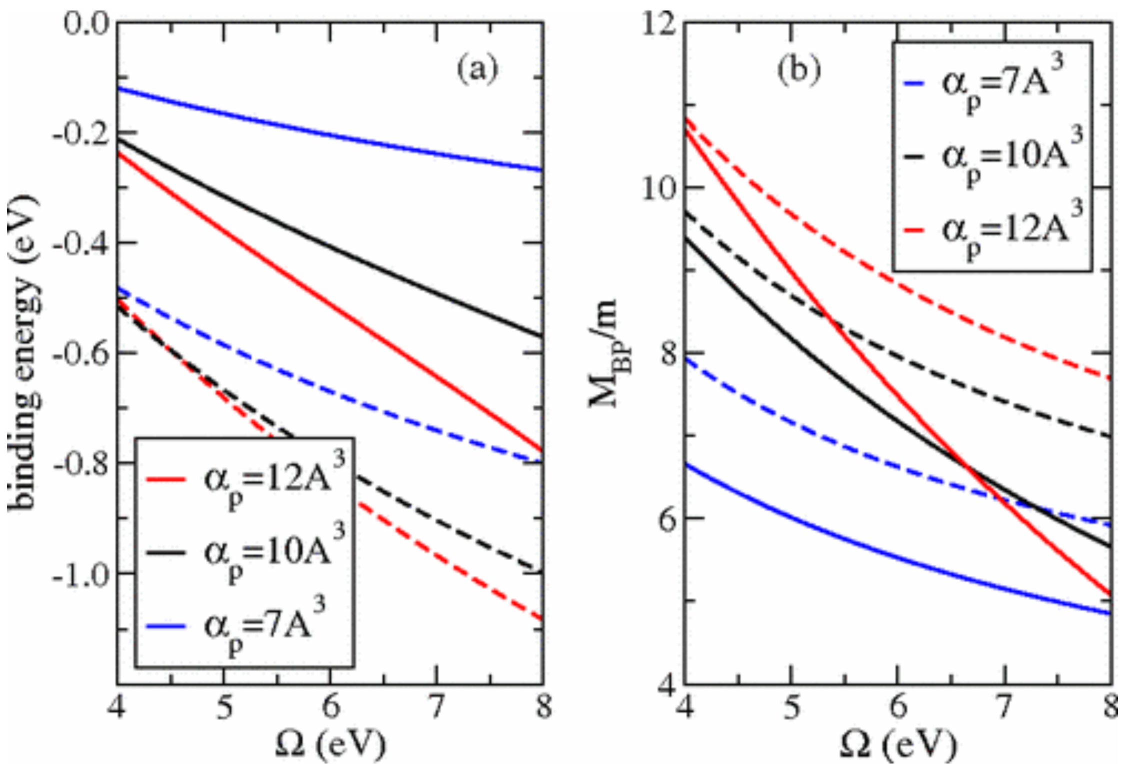
→ d-state pair has primarily nn character → favored by addition of t'

→ Inclusion of t' also increases binding energy considerably

→ Bipolaron has considerable dispersion → (fairly) mobile, light object!

Bi-polaron results (cont'd)

A2: Stable and rather light bipolarons in the limit $U_H \to \infty$



full lines: $t' = 0$

dashed lines: $t' \neq 0$

Discussion:

→ Effective interactions due to polarization clouds can be strongly attractive at longer-range, not just on-site → rather light bipolarons of either s- or d-wave symmetry could form in these materials

→ This would suggest a “pre-formed pair” mechanism closer to BEC, not BCS-like superconductivity

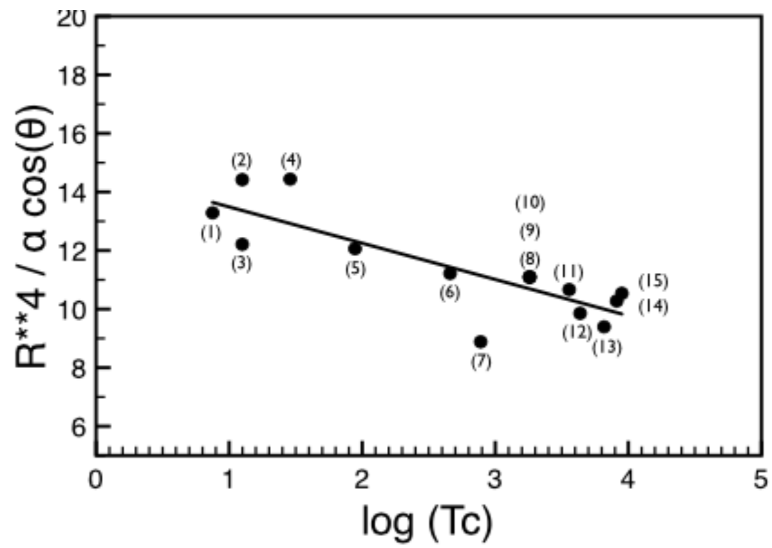
→ But we ignored:

-- multi-band nature: stable bipolarons expected to survive, all else being equal

-- dipole-dipole interaction → would raise total energies, so unfavorable to binding

-- longer-range polarization → favors attraction, favorable to binding (and probably winning)

-- longer-range Coulomb repulsion (longer-range analog of U_H): if large enough will unbind the bipolaron. Still, the effective cloud-mediated attraction may then be a BCS-like glue.



G. Sawatzky et al, EPL 86, 17006 (2009)

→ Correlation between T_c and U_1!

Fig. 4: The superconducting transition temperature T_c of various iron and nickel-pnictide superconductors *vs.* the computed inverse screening energy $E_{scr} = \alpha e^2(\cos\Theta)/R^4$ of the nearest-neighbor Coulomb interaction, suggesting the presence of superconductivity with a higher T_c in materials with larger electronic screening energy E_{scr}. R is Fe-As bond length, α the electronic polarizability ($\alpha_{As} = 10\,Å^3$, $\alpha_P = 9\,Å^3$), and Θ is the Fe-As-Fe bond angle. The solid line is a guide to the eye. The data points represent the following materials: (1) LaONiAs [40]; (2) LaONiP [41]; (3) $BaNi_2P_2$ [42]; (4) LaONiP [43]; (5) LaOFeP [44]; (6) $LaOFe_{0.89}Co_{0.11}As$ [45]; (7) LiFeAs [46]; (8) $LaO_{0.92}F_{0.08}FeAs$ [47]; (9) $LaO_{0.92}F_{0.08}FeAs$ [19]; (10) $LaO_{0.87}F_{0.13}FeAs$ [48]; (11) $CeO_{0.84}F_{0.16}FeAs$ [49]; (12) $Ba_{0.6}K_{0.4}Fe_2As_2$ [9]; (13) $TbO_{0.9}F_{0.1}FeAs$ [50]; (14) $NdO_{0.8}F_{0.2}FeAs$ [51]; (15) $PrO_{0.85}F_{0.15}FeAs$ [52].

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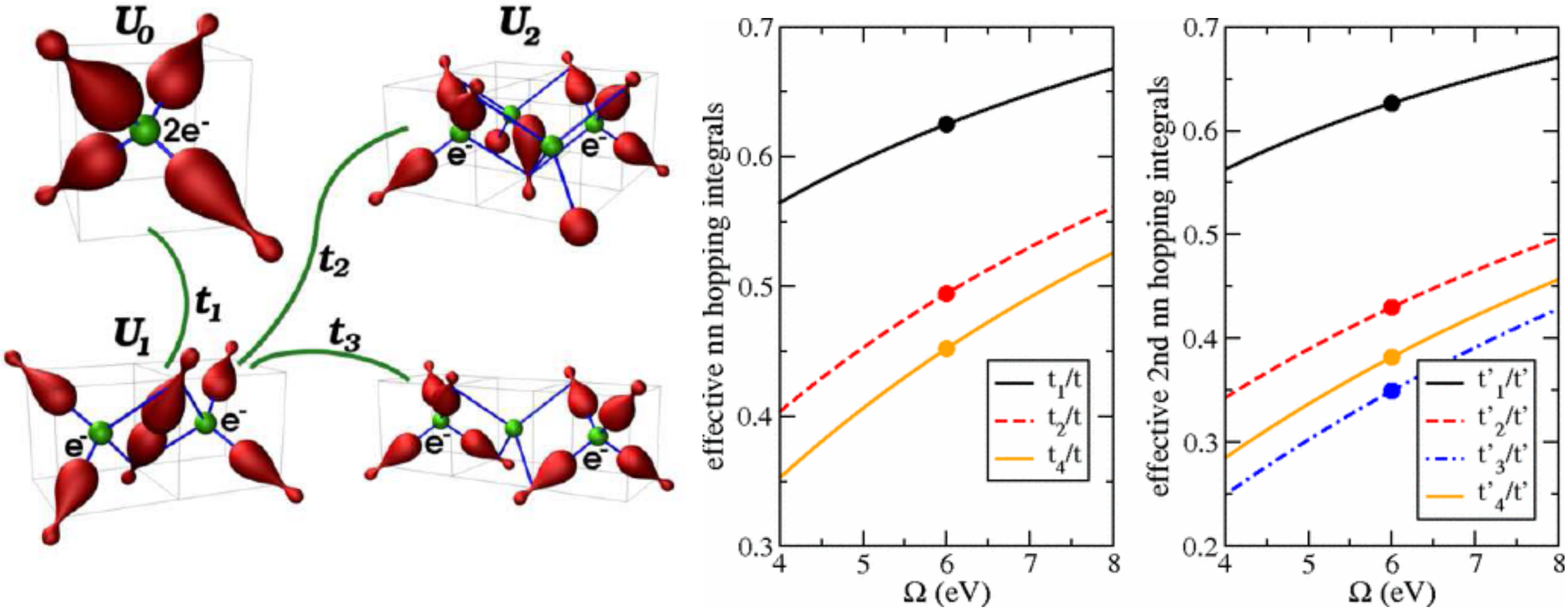

U_0
$2e^-$
U_2
e^-
e^-
t_2
t_1
U_1
t_3
e^-
e^-
e^-
e^-
effective nn hopping integrals
Ω (eV)
t_1/t
t_2/t
t_4/t
effective 2nd nn hopping integrals
Ω (eV)
t'_1/t'
t'_2/t'
t'_3/t'
t'_4/t'
0.7
0.6
0.5
0.4
0.3
0.2
4
5
6
7
8

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But: we provide a microscopic origin (and estimates) for these terms.

→ Similarities with Little and with Allender, Bray and Bardeen models, but there polaronic effects are used to overscreen on-site interaction. This requires very strong coupling → very heavy (bi)polarons

Conclusion:

→ Possible route to engineer size and sign of effective interactions between carriers by placing large, polarizable ions in the right positions

→ Large effects possible, plus non-monotonic dependence of distance

→ Away from linear regime, three- and multiple-particle interactions could be considerable!

→ … lots of possibilities for interesting physics …

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Thank you!