(about) Polarons and Bipolarons

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Thanks: George Sawatzky

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Hadi Ebrahimnejad, Clemens Adolphs and Mirko Moeller

many groups who shared with us their numerical data

NSERC, CIfAR and QMI

Polaron: when a particle (electron, hole, exciton, …) interacts with bosons from its environment (phonons, magnons, electron-hole pairs, etc) it becomes surrounded by a cloud of such excitations; the resulting composite object = dressed quasiparticle = polaron.

Bipolaron: a bound state of two polarons, mediated by exchange of bosons between their excitation clouds

Why interesting?: their properties (effective mass, effective interactions, etc) can be strongly renormalized and they determine the macroscopic properties of the material

Today: only T=0, no-disorder cases with a single polaron/bipolaron in the system (avoids many complications which are very interesting, too). Think insulators with very few carriers.

Plan -- lattice polaron (electron+phonons): detailed discussion of the Holstein polaron → brief discussion of Hubbard-Holstein bipolarons → why you should not assume that this is "all"

-- spin polarons and bipolarons in a ferromagnetic background

-- (if time permits): electronic polarons and bipolarons (model relevant for pnictides).

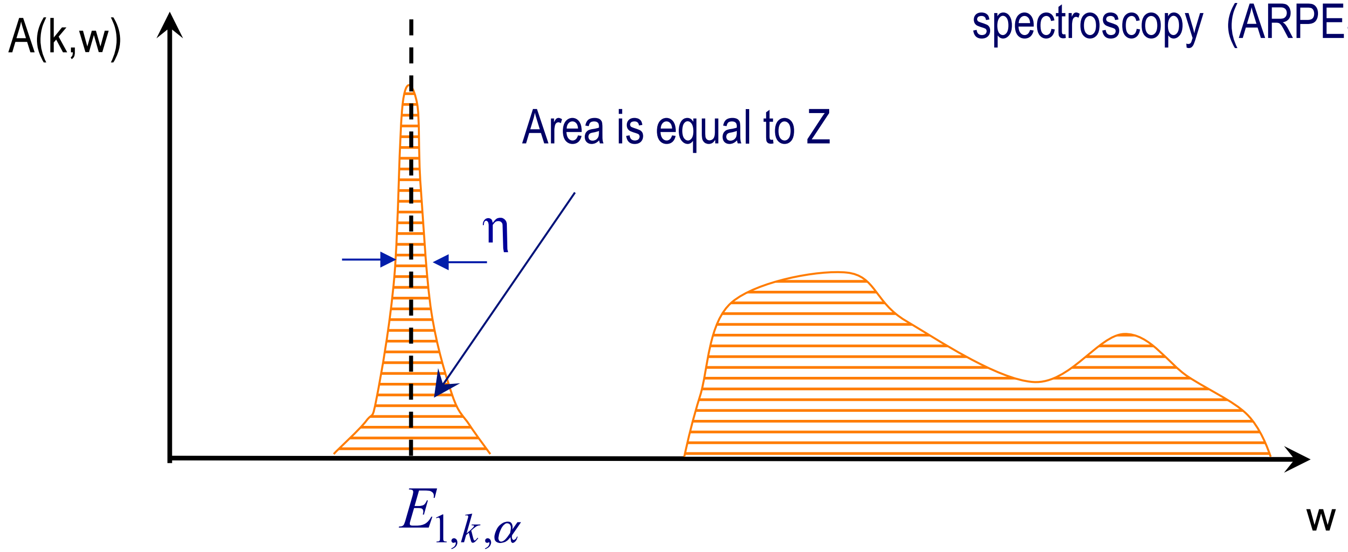
Quantity of interest: the Green's function or propagator→ polaron def's.

$H|1,k,\alpha\rangle = E_{1,k,\alpha}|1,k,\alpha\rangle$ ← eigenenergies and eigenfunctions (1 particle, total momentum k is a good quantum number in a clean system, α a is collection of other quantum numbers)

$$G(k,\omega) = \langle 0 | c_k \frac{1}{\omega - H + i\eta} c_k^+ | 0 \rangle = \sum_\alpha \frac{Z_{1,k,\alpha}}{\omega - E_{1,k,\alpha} + i\eta} \qquad Z_{1,k,\alpha} = \left| \langle 1,k,\alpha | c_k^+ | 0 \rangle \right|^2$$

$$A(k,\omega) = -\frac{1}{\pi}\operatorname{Im} G(k,\omega) = \sum_\alpha Z_{1,k,\alpha}\delta\left(\omega - E_{1,k,\alpha}\right)$$

← = spectral weight, is measured (inverse) angle-resolved photoemission spectroscopy (ARPES) or STM

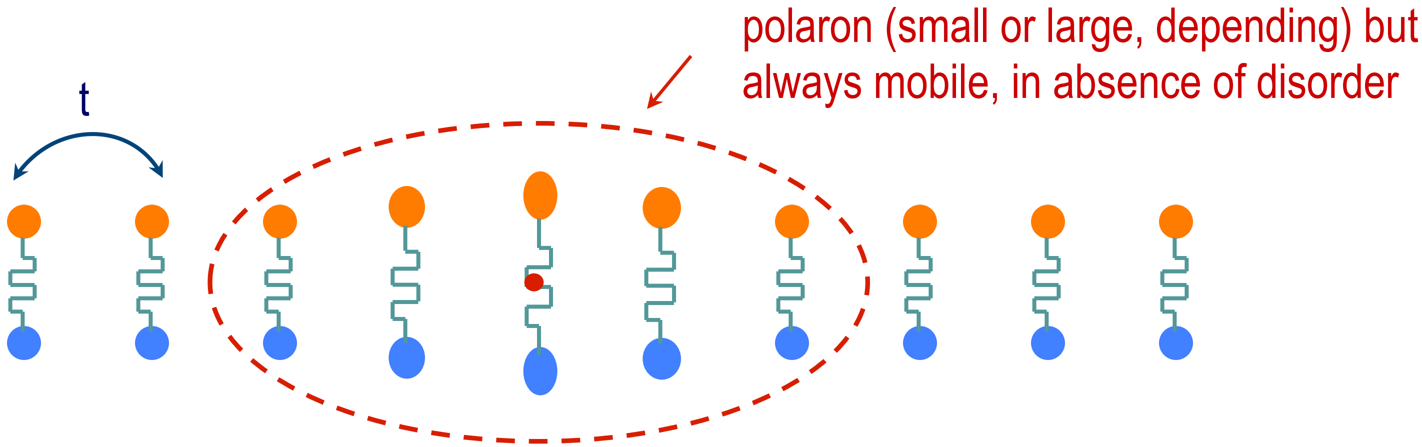


Z = quasiparticle weight → measures how similar is the true wavefunction to a non-interacting (free electron, no bosons) wavefunction

Polaron (lattice polaron) = electron + lattice distortion (phonon cloud) surrounding it

→ very old problem: Landau, 1933;

→ most studied lattice model = Holstein model used to described "molecular crystals" (1959)



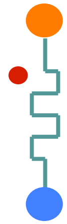
$$H = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^{+} c_{j\sigma} + c_{j\sigma}^{+} c_{i\sigma}) + \Omega \sum_{i} b_i^{+} b_i + g \sum_{i} n_i (b_i^{+} + b_i)$$

3 energy scales: t, W, g → 2 dimensionless parameters $\lambda = g^2/(2dt\Omega)$, Ω/t (d is lattice dimension)

Eigenstates are linear combinations of states with the electron at different sites, surrounded by a lattice distortion (cloud of phonons). Can have any number of phonons anywhere in the system → problem cannot be solved exactly for arbitrary t, g, Ω.

Why this coupling? Consider a single site → analog of the Franck-Condon problem

add extra e



= new equilibrium distance

$$h_0 = \frac{\hat{P}^2}{2M} + \frac{M\Omega^2(\hat{X} - X_0)^2}{2} \to \hbar\Omega\left(b^+ b + \frac{1}{2}\right)$$

$$\begin{cases} b = \sqrt{\dfrac{M\Omega}{2\hbar}}\left(\hat{X} - X_0 + i\dfrac{\hat{P}}{M\Omega}\right) \\ b^+ = \sqrt{\dfrac{M\Omega}{2\hbar}}\left(\hat{X} - X_0 - i\dfrac{\hat{P}}{M\Omega}\right) \end{cases} \to \hat{X} - X_0 \propto (b^+ + b)$$

$$h = h_0 + \hat{n}U(\hat{X}) \approx h_0 + g\hat{n}(\hat{X} - X_0)$$

$$X_0 \to X_0 - \alpha\hat{n}$$

new equilibrium length, determined by how many extra electrons there are

I will assume that the electron-phonon interaction is weak enough that it's ok to keep only linear contribution. Clemens is studying what happens if you go beyond this, see cond-mat/1302.3843 or ask him for details!

Single-site model (if t=0) can be solved exactly:

$$h = \Omega b^\dagger b + g\hat{n}(b^\dagger + b) \to \Omega b^\dagger b + g(b^\dagger + b) = \Omega B^\dagger B - \frac{g^2}{\Omega}$$

n=1 for polaron

$$B = b + \frac{g}{\Omega} \to \left[B, B^\dagger\right] = \left[b, b^\dagger\right] = 1$$

Ground-state is: $\left|GS\right\rangle = c^\dagger \left|-\frac{g}{\Omega}\right\rangle \to E_{GS} = -\frac{g^2}{\Omega}; \left\langle b^\dagger b\right\rangle = \frac{g^2}{\Omega^2}$

where $b\left|-\frac{g}{\Omega}\right\rangle = -\frac{g}{\Omega}\left|-\frac{g}{\Omega}\right\rangle \to B\left|-\frac{g}{\Omega}\right\rangle = 0$

Side-note: coherent states

$$b\left|\alpha\right\rangle = \alpha\left|\alpha\right\rangle \to \left|\alpha\right\rangle = e^{-\frac{|\alpha|^2}{2} + \alpha b^\dagger}\left|0\right\rangle$$

while excited states have energies $E_m = -\frac{g^2}{\Omega} + m\Omega$, m=0,1,..

and polaron eigenfunctions of the general form: $c^\dagger \frac{B^{\dagger m}}{\sqrt{m!}}\left|-\frac{g}{\Omega}\right\rangle$

Green's function can be calculated since eigenspectrum is known → try!

weak coupling $\lambda = \frac{g^2}{2dt\Omega} = 0 \quad (g=0)$

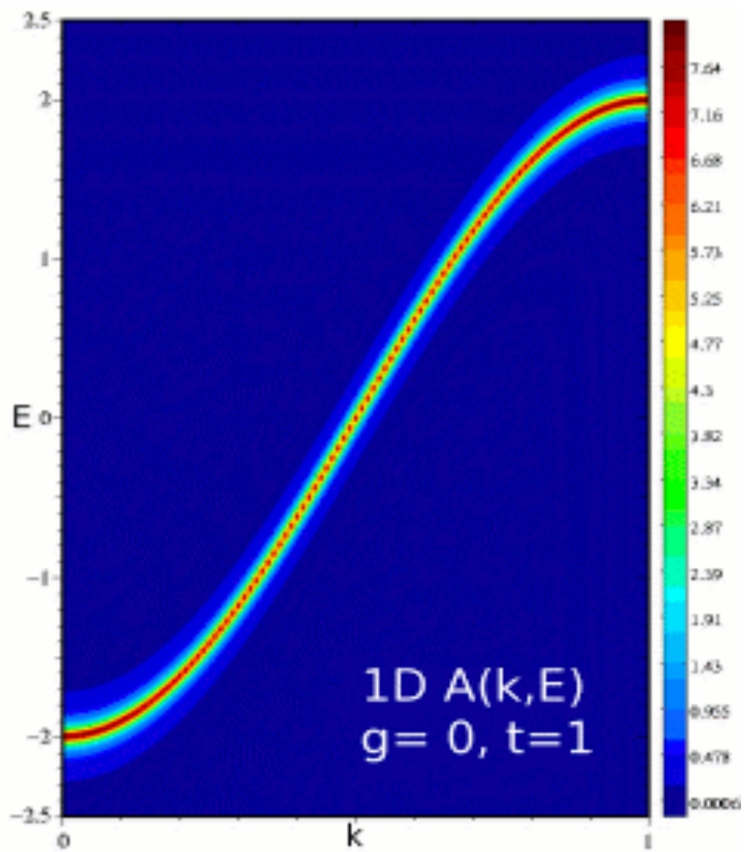
Lang-Firsov impurity limit $\lambda = \frac{g^2}{2dt\Omega} = \infty \quad (t=0)$

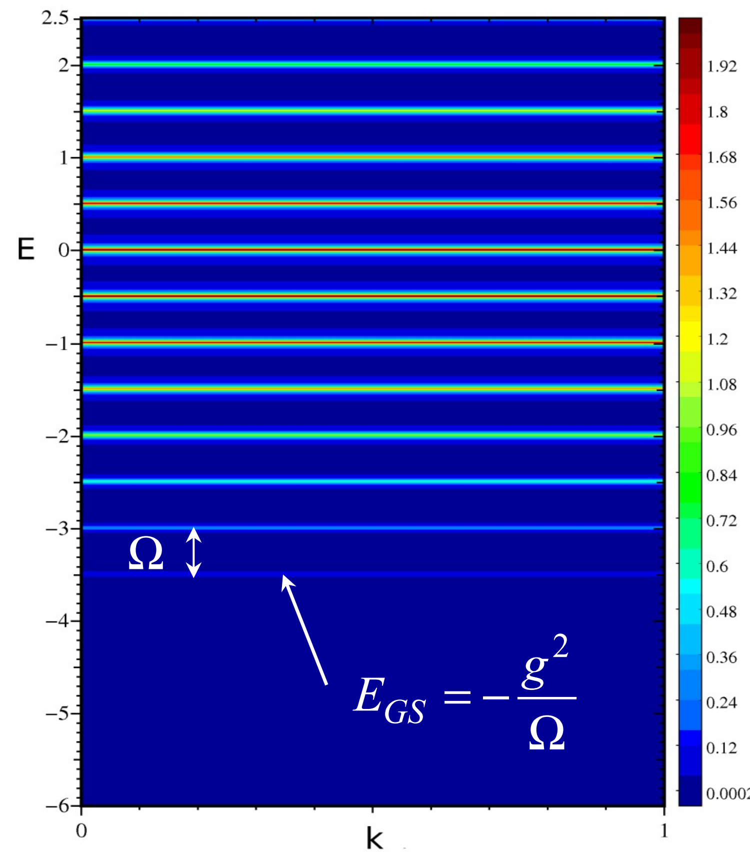
$$G_0(k,\omega) = \frac{1}{\omega - \varepsilon_k + i\eta};$$

$$A_0(k,\omega) = \frac{\eta}{\pi\left[(\omega-\varepsilon_k)^2 + \eta^2\right]} \xrightarrow{\eta\to 0} \delta(\omega - \varepsilon_k)$$

$$G_{LF}(k,\omega) = e^{-\frac{g^2}{\Omega^2}} \sum_{m=0}^{\infty} \frac{1}{m!}\left(\frac{g}{\Omega}\right)^{2m} \frac{1}{\omega + \frac{g^2}{\Omega} - m\Omega + i\eta}$$

$$E_m = -\frac{g^2}{\Omega} + m\Omega$$





How does the spectral weight evolve between these two very different looking limits?

$$H=-t\sum_{<i,j>,\sigma}(c_{i\sigma}^{+}c_{j\sigma}+c_{j\sigma}^{+}c_{i\sigma})+\Omega\sum_i b_i^{+}b_i+g\sum_i n_i(b_i^{+}+b_i)$$

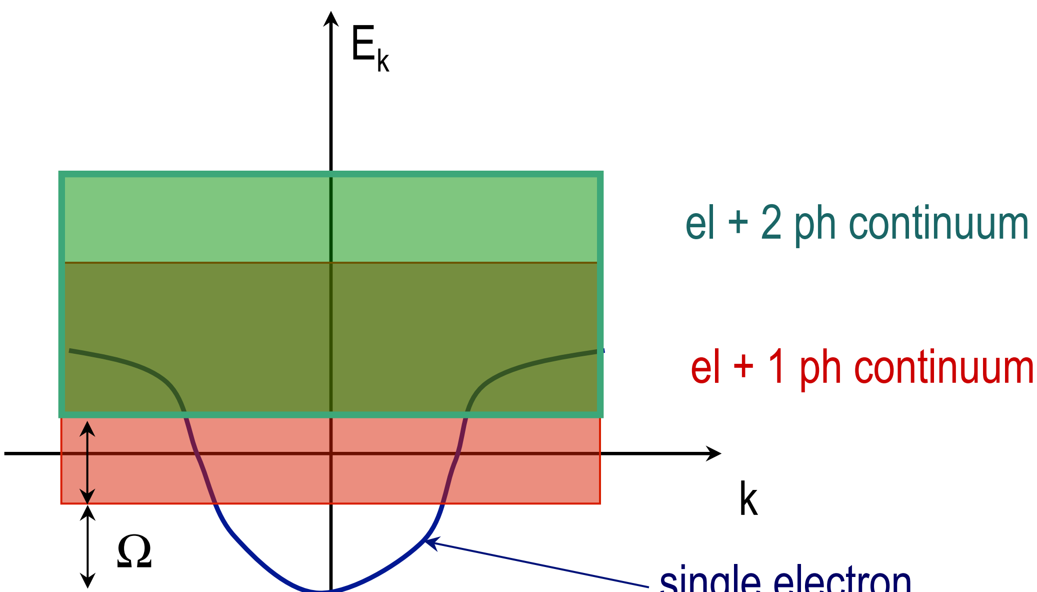
$$=\sum_{\vec{k}}\varepsilon_{\vec{k}}c_{\vec{k}}^{+}c_{\vec{k}}+\Omega\sum_{\vec{q}}b_{\vec{q}}^{+}b_{\vec{q}}+\frac{g}{\sqrt{N}}\sum_{\vec{k},\vec{q}}c_{\vec{k}-\vec{q}}^{+}c_{\vec{k}}\left(b_{\vec{q}}^{+}+b_{-\vec{q}}\right)$$

(spin is irrelevant, N = number of unit cells → infinity, all k,q-sums over Brillouin zone)

Asymptotic behavior:

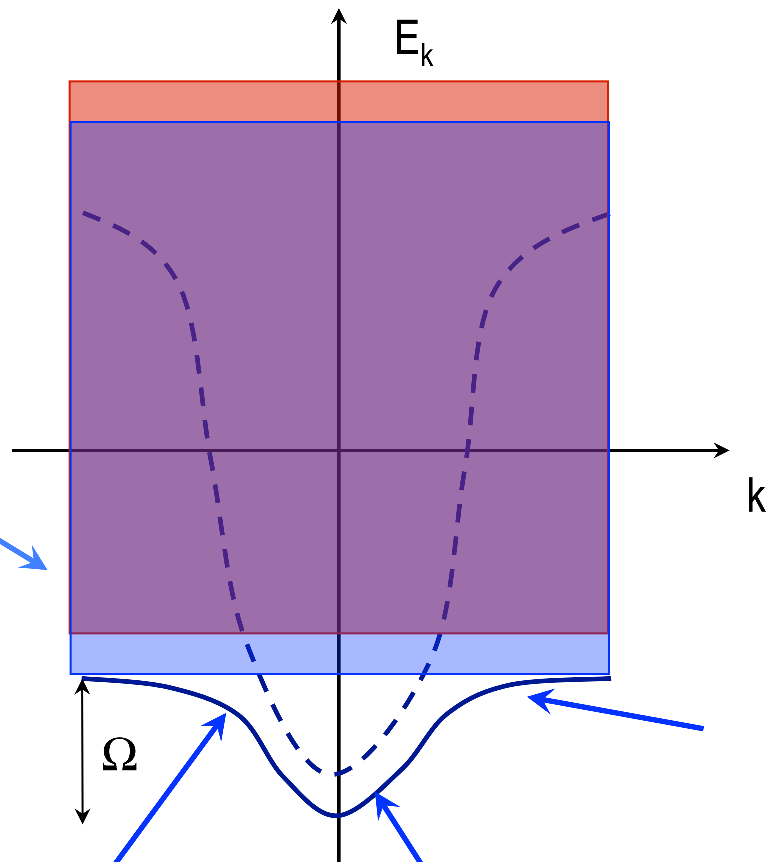
→ zero-coupling limit, g=0 → eigenstates of given k: $c_{\vec{k}}^{+}|0\rangle,\ c_{\vec{k}-\vec{q}}^{+}b_{\vec{q}}^{+}|0\rangle,\ c_{\vec{k}-\vec{q}-\vec{q}'}^{+}b_{\vec{q}}^{+}b_{\vec{q}'}^{+}|0\rangle,\ldots$

with eigenenergies $\varepsilon_{\vec{k}},\ \varepsilon_{\vec{k}-\vec{q}}+\Omega,\ \varepsilon_{\vec{k}-\vec{q}-\vec{q}'}+2\Omega,\ldots.$ where, for example, $\varepsilon_{\vec{k}}=-2t\sum_{i=1}^{d}\cos k_i$



→ weak coupling, g = “small” → low-energy eigenstates of known k: $\left|\psi_{\vec{k}}\right\rangle = c^{+}_{\vec{k}}\left|0\right\rangle + \sum_{\vec{q}} \phi_{\vec{q}}\, c^{+}_{\vec{k}-\vec{q}}\, b^{+}_{\vec{q}}\left|0\right\rangle$

with eigenenergies $E_{\vec{k}} = \varepsilon_{\vec{k}} - \frac{1}{N}\sum_{\vec{q}} \frac{g^2}{\left(\Omega + \varepsilon_{\vec{k}-\vec{q}}\right) - \varepsilon_{\vec{k}}} + \ldots$ (for k< k_{cross})



The phonon(s) can be quite far spatially from the electron → large polaron

polaron + one- phonon continuum (finite lifetimes)

high-k: wavefunction dominated by el+1ph contributions

polaron band (infinitely long-lived quasiparticle, quite different dispersion from electron)

low-k: wavefunction dominated by free electron contribution

In fact, a polaron state exists everywhere in the BZ only in d=1,2. In d>2 and weak coupling, the polaron exists only near the center of the BZ. **G.L. Goodvin and M. Berciu, EuroPhys. Lett. 92, 37006 (2010)**

→ very strong coupling λ>>1 (t→ 0) →small polaron energy is

$$E_k = -\frac{g^2}{\Omega} + e^{-\frac{g^2}{\Omega^2}}\varepsilon_k + \ldots \rightarrow t_{eff} = te^{-\frac{g^2}{\Omega^2}} \rightarrow m_{eff} = me^{\frac{g^2}{\Omega^2}}$$

$$\text{and wavefunction is } |\psi_k\rangle = \sum_i \frac{e^{i\vec{k}\cdot\vec{R}_i}}{\sqrt{N}} c_i^\dagger \left|-\frac{g}{\Omega}\right\rangle_i$$

Again, must have a polaron+one-phonon continuum at $E_{GS} + \Omega$ → details too nasty

Because here polaron dispersion is so flat, there is a polaron state everywhere in the BZ.

→ What happens at intermediate couplings?

- Diagrammatic Quantum Monte Carlo (Prokof' ev, Svistunov and co-workers)

→calculate Green's function in imaginary time

$$G(k,\tau)=\langle 0|c_k e^{-\tau H} c_k^\dagger|0\rangle=\sum_\alpha e^{-\tau E_{1,k,\alpha}}\left|\langle 1,k,\alpha|c_k^\dagger|0\rangle\right|^2 \xrightarrow[\tau\to\infty]{} Z_k e^{-\tau E_k}$$

Basically, use Metropolis algorithm to sample which diagrams to sum, and keep summing numerically until convergence is reached

- Quantum Monte Carlo methods (Kornilovitch in Alexandrov group, Hohenadler in Fehske group, …) → write partition function as path integral, use Trotter to discretize it, then evaluate. Mostly low-energy properties are calculated/shown.
- Exact diagonalization = ED → finite system (still need to truncate Hilbert space) → can get whole spectrum and then build G(k,w)
- Variational methods -> infinite system, smart choice of basis (Trugman group)
- Cluster perturbation theory: ED finite system, then use perturbation in hopping to "sew" finite pieces together → infinite system.
- + 1D, DMRG+DMFT

→…. (lots of work done in these 50+ years, as you may imagine)

Analytical approaches (other than perturbation theory) → calculate self-energy

$$G(k,\omega)=\frac{1}{\omega-\varepsilon_k-\Sigma(k,\omega)+i\eta}$$

Σ(k,w) =  +....

For Holstein polaron, we need to sum to orders well above g^2/Ω^2 to get convergence.

n	1	2	3	4	5	6	7	8
Σ, exact	1	2	10	74	706	8162	110410	1708394
Σ, SCBA	1	1	2	5	14	42	132	429

Traditional approach: find a subclass of diagrams that can be summed, ignore the rest

→ self-consistent Born approximation (SCBA) – sums only non-crossed diagrams (much fewer)

Our group: the momentum average = MA$^{(n)}$ hierarchy of approximations:

Idea: keep ALL self-energy diagrams, but approximate each such that the summation can be carried out analytically – we can show that what we keep is exponentially larger than what we ignore. (Alternative explanation: generate the infinite hierarchy of coupled equations of motion for the propagator, keep all of them instead of factorizing and truncating, but simplify coefficients so that an analytical solution can be found).

MA also has variational meaning:

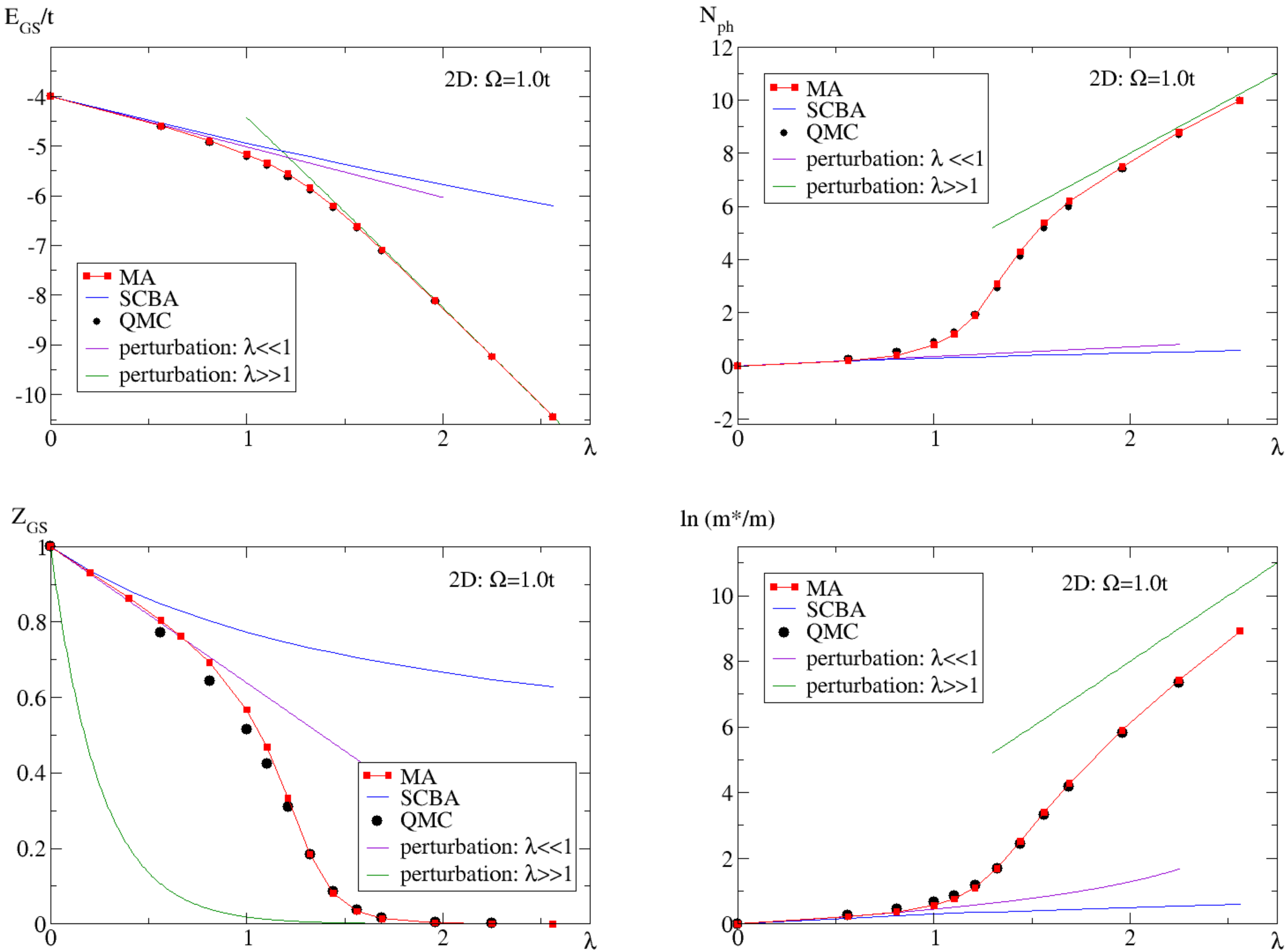
→ only certain kinds of bosonic clouds allowed (**O. S. Barišic, PRL 98, 209701 (2007)**)

→ what is reasonable depends on the model. In the simplest case (Holstein model):

$$\mathrm{MA}^{(0)} \rightarrow c_i^\dagger \left(b_j^\dagger\right)^n |0\rangle, \quad (\forall) i, j, n$$

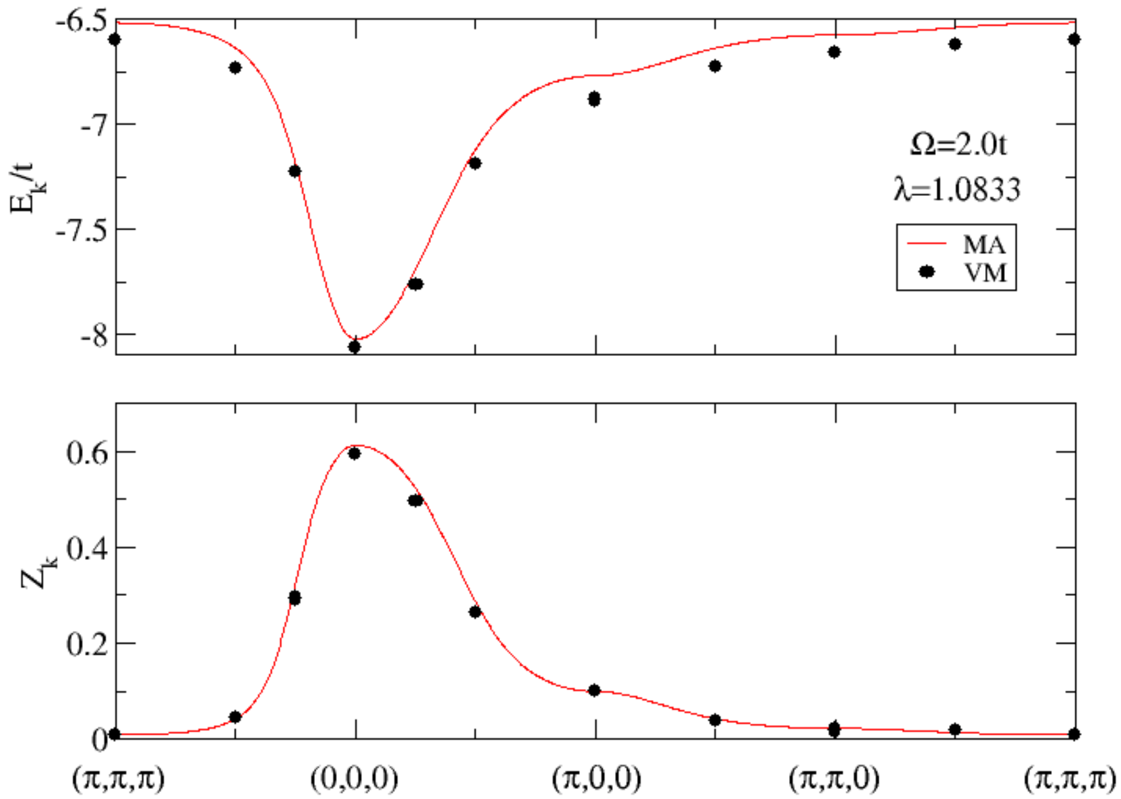
$$\mathrm{MA}^{(1)} \rightarrow c_i^\dagger \left(b_j^\dagger\right)^n b_l^\dagger |0\rangle, \quad (\forall) i, j, l, n$$

(needed to describe polaron + one-boson continuum)



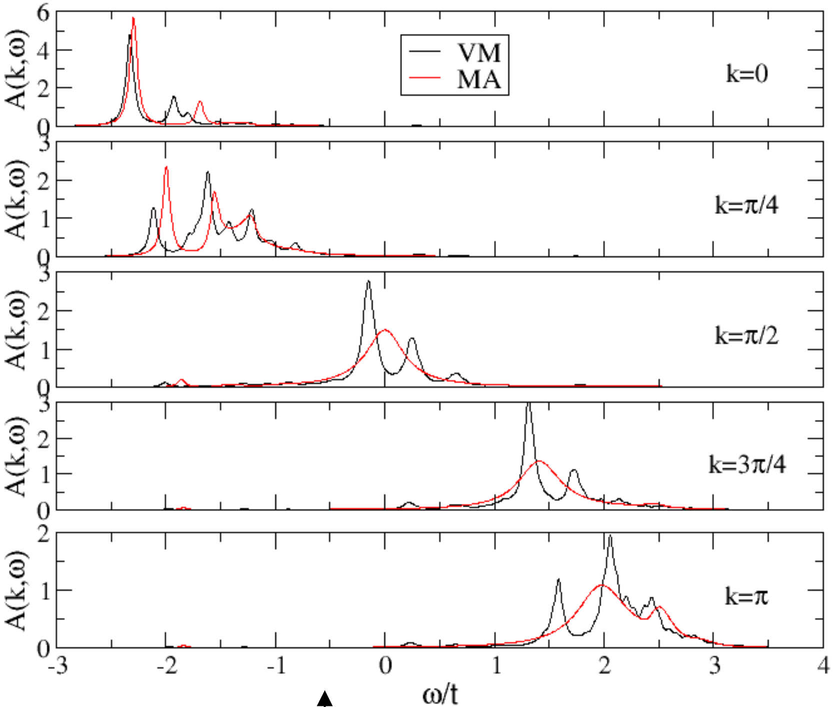
E_{GS}/t
2D: Ω=1.0t
MA
SCBA
QMC
perturbation: λ<<1
perturbation: λ>>1
λ
N_{ph}
2D: Ω=1.0t
MA
SCBA
QMC
perturbation: λ <<1
perturbation: λ>>1
λ
Z_{GS}
2D: Ω=1.0t
MA
SCBA
QMC
perturbation: λ<<1
perturbation: λ>>1
λ
ln (m*/m)
2D: Ω=1.0t
MA
SCBA
QMC
perturbation: λ<<1
perturbation: λ>>1
λ

3D Polaron dispersion



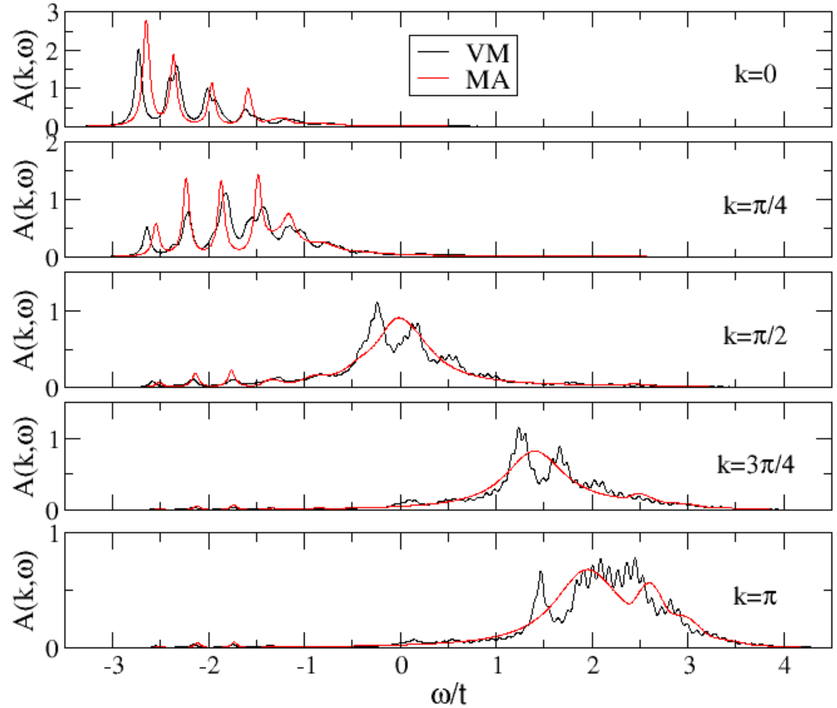
Polaron bandwidth much narrower than 12t! (actually it is below Ω, as discussed)

L. -C. Ku, S. A. Trugman and S. Bonca, Phys. Rev. B 65, 174306 (2002).

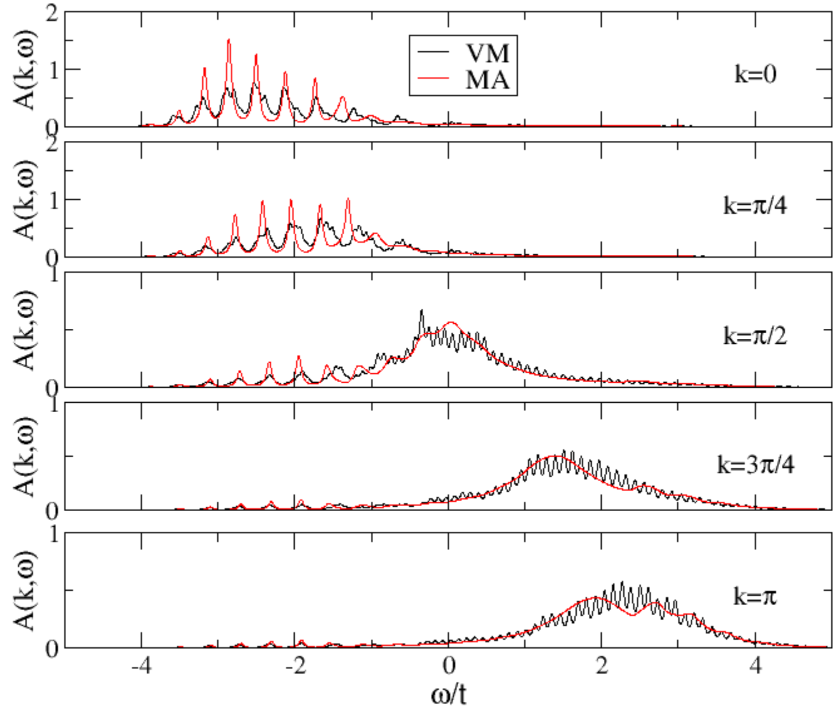


λ = 0.5

λ = 1



λ = 2

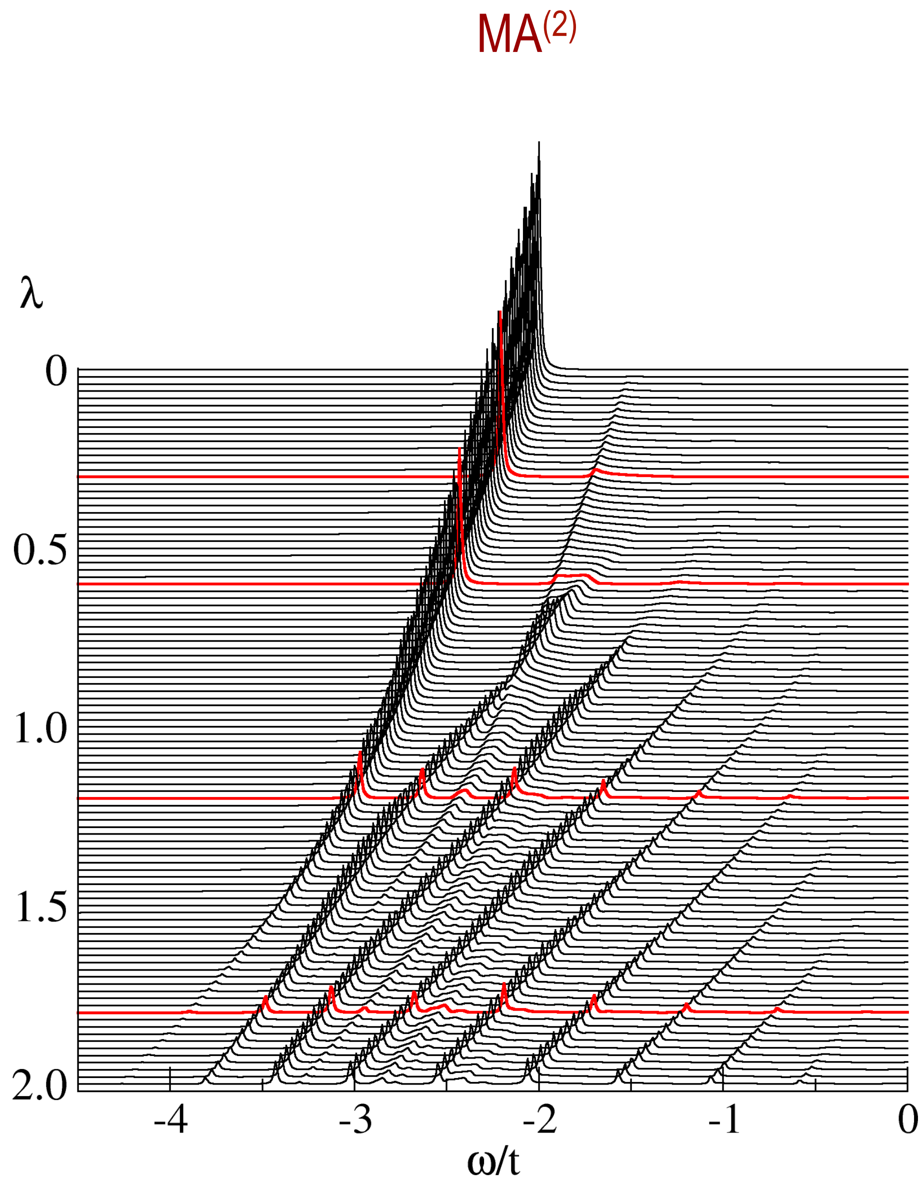


A(k,w) in 1D, Ω =0.4 t

G. De Filippis et al, PRB 72, 014307 (2005)

MA becomes exact for small and for large λ

1D, k=0, Ω=0.5t



How about bipolarons, when are they stable? → look for hints at single site exact results

Remember: single polaron at one site has ground-state energy $-g^2/\Omega$

$$h = \Omega b^\dagger b + g\hat{n}(b^\dagger + b) \rightarrow \Omega b^\dagger b + g(b^\dagger + b) = \Omega B^\dagger B - \frac{g^2}{\Omega}$$

→ two polarons away from each other (unbound) should cost $-2g^2/\Omega$ +…

For a bipolaron (both electrons at the same site):

$$h = \Omega b^\dagger b + g\hat{n}(b^\dagger + b) + U\hat{n}_\uparrow \hat{n}_\downarrow \rightarrow \Omega b^\dagger b + 2g(b^\dagger + b) + U = \Omega \tilde{B}^\dagger \tilde{B} - \frac{4g^2}{\Omega} + U$$

→ ground state for a bipolaron is $-4g^2/\Omega+U$ → $U_{eff}=U-2g^2/\Omega$ (renormalized interaction)

If $U < 2g^2/\Omega$ → onsite bipolaron is energetically favorable (called a S0 small bipolaron)

If $U > 2g^2/\Omega$ → S0 is not energetically favorable; however a bound S1 bipolaron appears if U not too large (this has polarons on neighboring sites, to avoid U, but carriers can take advantage of each other's clouds by hopping back and forth between the two sites) → detailed perturbation theory a bit ugly, see J. Bonca, T. Katrasnik and S. Trugman, PRL 84, 3153 (2000);

If U large enough → unbound polarons are energetically more favorable

More details and also numerical results in PRL 84, 3153 (2000); PRB 69, 245111 (2004); Eur. Phys. J B 85, 111 (2012); PRB 86, 035106 (2012) …

Summary for Holstein polaron/bipolaron behavior:

-- as el-ph coupling increases, polaron becomes heavier and “smaller” (bigger cloud but less spread)

-- bipolarons are favorable if U not too large

(it’s all a bit boring)

Warning: many people believe that this is true for all lattice polarons (up to quantitative differences). This is not true … this “typical polaron behavior” is characteristic for models where the phonons modulated the on-site energy of the particle (so-called g(q) models) but is qualitatively wrong for models where the phonons modulate the hopping of the particle – eg, the SSH (or Peierls) coupling.

Phonon-modulated hopping like in polyacetylene (Su-Schrieffer-Heeger aka Peierls)

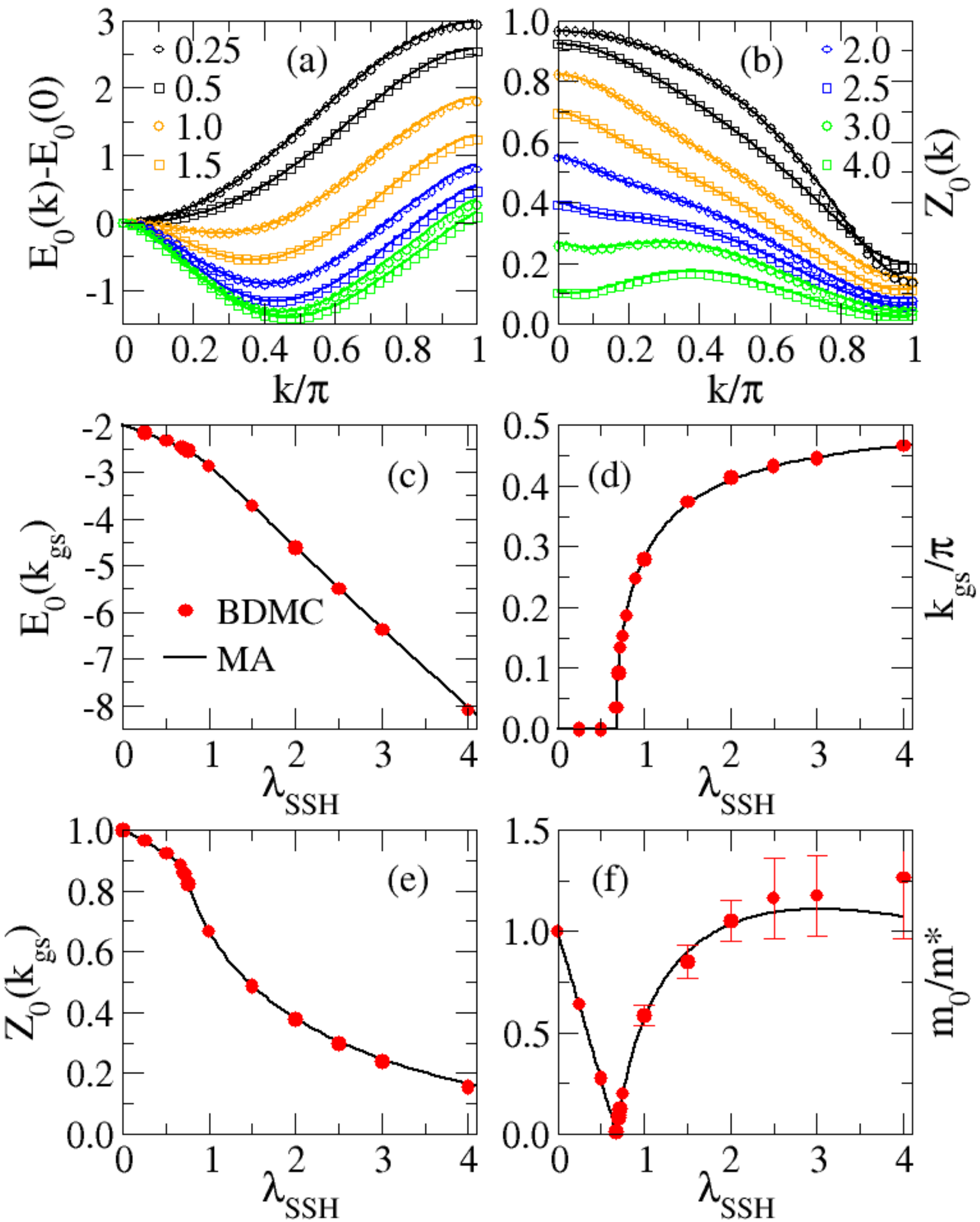
$$t_{i,i+1} \propto e^{-\alpha(R_{i+1}-R_i)} = t_0 e^{-\alpha(u_{i+1}-u_i)} \approx t_0[1-\alpha(u_{i+1}-u_i)]$$

$$u_i \propto b_i^\dagger + b_i$$

$$V_{el-ph} = g\sum_i (c_{i+1}^\dagger c_i + c_i^\dagger c_{i+1})[b_{i+1}^\dagger + b_{i+1} - b_i^\dagger - b_i]$$

→ as particle hops from one site to another, it can either create or annihilate a boson at either the initial or the final site.

"SSH" model in 1D for t=1, Ω=3



Work done with Dominic Marchand and Philip Stamp

Momentum of GS switches from 0 (weak coupling) to finite value (strong coupling)

→ True transition (not crossover) from large to small polaron

→ Such transitions are impossible in models with g(q)

→ above the transition, the polaron is very light even for strong coupling

Also very good agreement with data from G. de Filippis, V. Cataudella and A. Mishchenko and N. Nagaosa

PRL 105, 266605 (2010)

So far as I know, bipolarons have not been studied in such models

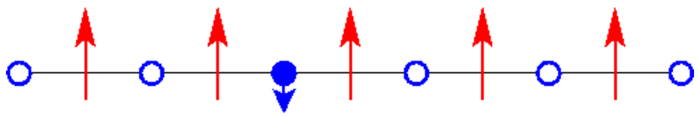
Mixed models → yet more interesting,

see cond-mat/1212.6212 (cold molecules)

Spin-polaron → carrier in a FM lattice of spins S -- these can be solved exactly!

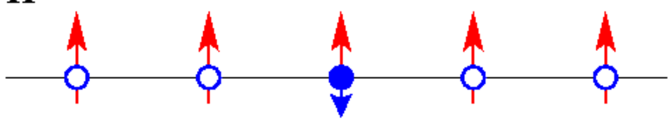
Models: (for simplicity, 1D and background spins S=1/2)

I



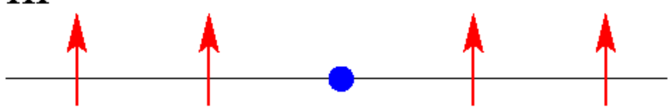
← "parent" model: two sublattices, two bands

II



← effective model: one lattice, two bands

III



← effective model: one lattice, one band

Models I and II: $$H = H_{Hubbard} - J\sum_i \left(\vec{S}_i \cdot \vec{S}_{i+1} - S^2\right) + H_{exc}$$

$$H_{exc}^{(I)} = J_0 \sum_i \vec{s}_{i+\frac{1}{2}} \cdot \left(\vec{S}_i + \vec{S}_{i+1}\right) \quad \text{vs.} \quad H_{exc}^{(II)} = J_0 \sum_i \vec{s}_i \cdot \vec{S}_i$$

Model III: $$H = H_{Hubbard} \rightarrow \text{ projected to proper } \mathrm{S_z} \text{ spin subspace}$$

$$\text{eg. NS-}\frac{1}{2}$$

Single particle sector:

→ spin-up charge carrier: boring (model III cannot treat this) →

$$c^\dagger_{k\uparrow}|FM\rangle, \;\; E_{k\uparrow}=\varepsilon_k+\gamma J_0, \;\; \gamma=\frac{1}{2},\frac{1}{4} \;\; \text{in I, II}$$

→ spin-down charge carrier: interesting for I and II

If we ignore off-diagonal exchange:

$$c^\dagger_{k\downarrow}|FM\rangle, \;\; E_{k\downarrow}=\varepsilon_k-\gamma J_0$$

$$\text{continuum: } c^\dagger_{k-q,\uparrow}S^-_q|FM\rangle, \; E_{k-q,\uparrow}+\Omega_q$$

Off-diagonal exchange hybridizes these → discrete state pushed even lower below the continuum, forming an infinitely-long lived quasiparticle (the spin polaron)

Model III: also has (trivial) quasiparticle:

$$h^\dagger_k|FM\rangle, \;\; \varepsilon_k=-2t\cos(k)$$

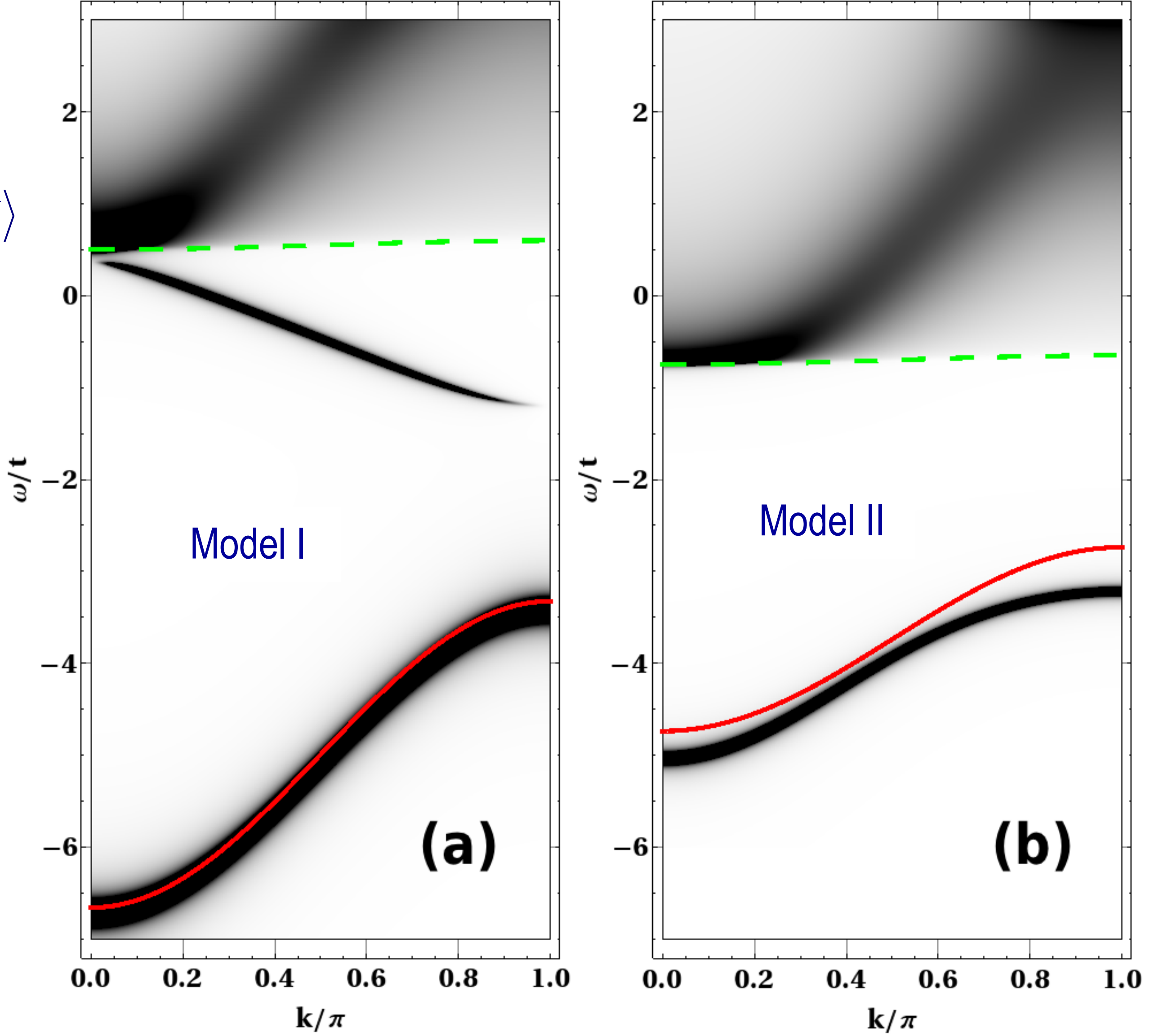
$$A(k,\omega) = -\frac{1}{\pi}\,\mathrm{Im}G\left(k,\omega\right);$$

$$G(k,\omega) = \left\langle FM \middle| c_{k\downarrow}\hat{G}(\omega)c^{\dagger}_{k\downarrow} \middle| FM \right\rangle$$

$t=1$, $J=0.05$, $J_0=5$, $\eta=0.02$

Typical polaron behavior:

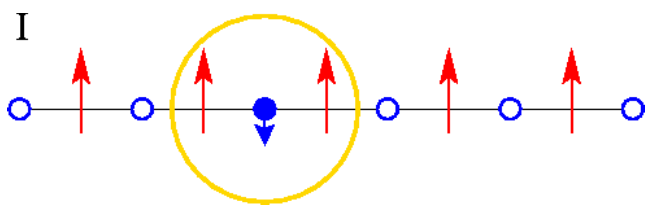
- polaron band below a continuum
- renormalized mass (but only by a finite factor, since it can only bind one magnon)
- Continuum contains unbound states of spin-down carrier + magnon, and disperses with k because magnon's energy depends on k

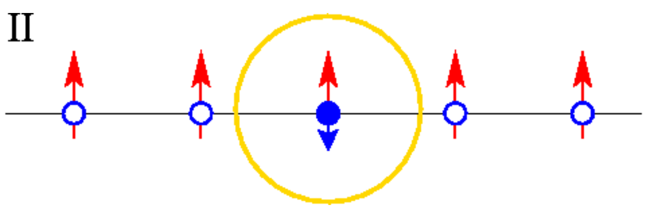


One lattice models: B. S. Shastry and D. C. Mattis, PRB 24, 5340 (1981)

Two sublattice models: M. Berciu and G. A. Sawatzky, PRB 79, 195116 (2009)

What is the nature of the low-energy spin-polaron? Assume $J_0>0$ largest energy scale, do perturbation





$$\left|3sp\right\rangle_{i+\frac{1}{2}} = \left(\sqrt{\frac{2}{3}} c^\dagger_{i+\frac{1}{2},\downarrow} - c^\dagger_{i+\frac{1}{2},\uparrow} \frac{S_i^- + S_{i+1}^-}{\sqrt{6}}\right)\left|FM\right\rangle$$

$$\left|P_I, k\right\rangle = \frac{1}{\sqrt{N}} \sum_i e^{ik(R_i+\frac{a}{2})} \left|3sp\right\rangle_{i+\frac{1}{2}}$$

$$E_I(k) = -J_0 + \frac{5}{6}\varepsilon_k + \frac{1}{6}J$$

$$\left|s\right\rangle_i = \frac{1}{\sqrt{2}}\left(c^\dagger_{i+\frac{1}{2},\downarrow} - c^\dagger_{i+\frac{1}{2},\uparrow} S_i^-\right)\left|FM\right\rangle$$

$$\left|P_{II}, k\right\rangle = \frac{1}{\sqrt{N}} \sum_i e^{ikR_i} \left|s\right\rangle_i$$

$$E_{II}(k) = -\frac{3}{4}J_0 + \frac{1}{2}\varepsilon_k + \frac{1}{2}J$$

$$\left|P_I, k\right\rangle = \frac{1}{\sqrt{N}} \sum_i e^{ikR_i} \frac{1}{\sqrt{2}}\left(d^\dagger_{i,k,\downarrow} - d^\dagger_{i,k,\uparrow} S_i^-\right)\left|FM\right\rangle$$

Zhang-Rice like singlet

$$d_{i,k,\sigma} = \frac{1}{\sqrt{3}}\left(e^{ik\frac{a}{2}} c_{i+\frac{1}{2},\sigma} + e^{-ik\frac{a}{2}} c_{i-\frac{1}{2},\sigma}\right)$$

"on-site" orbital

Two-particle sector:

→ two spin-up charge carriers: boring (model III cannot treat this) →no interactions

→ two spin-down charge carriers: magnon-mediated exchange is not strong enough to lead to bound states → hard to quantify differences.

→ interesting case: inject a spin-up + a spin-down charge carrier (model III cannot treat it)

Calculate two-particle Green's functions and extract two-particle spectrum

(i) in k-space:

$$G(k;q,q';\omega) = \langle k,q | \hat{G}(\omega) | k,q' \rangle$$

$$|k,q\rangle = c^{\dagger}_{\frac{k}{2}+q,\uparrow} c^{\dagger}_{\frac{k}{2}-q,\downarrow} |FM\rangle$$

Note: total momentum k is conserved, but relative momentum q is not!

(ii) in real-space:

$$G(k;n,n';\omega) = \langle k,n | \hat{G}(\omega) | k,n' \rangle$$

$$|k,n\rangle = \frac{1}{\sqrt{N}} \sum_i e^{ik(R_i + \frac{na}{2})} c^{\dagger}_{i,\uparrow} c^{\dagger}_{i+n,\downarrow} |FM\rangle$$

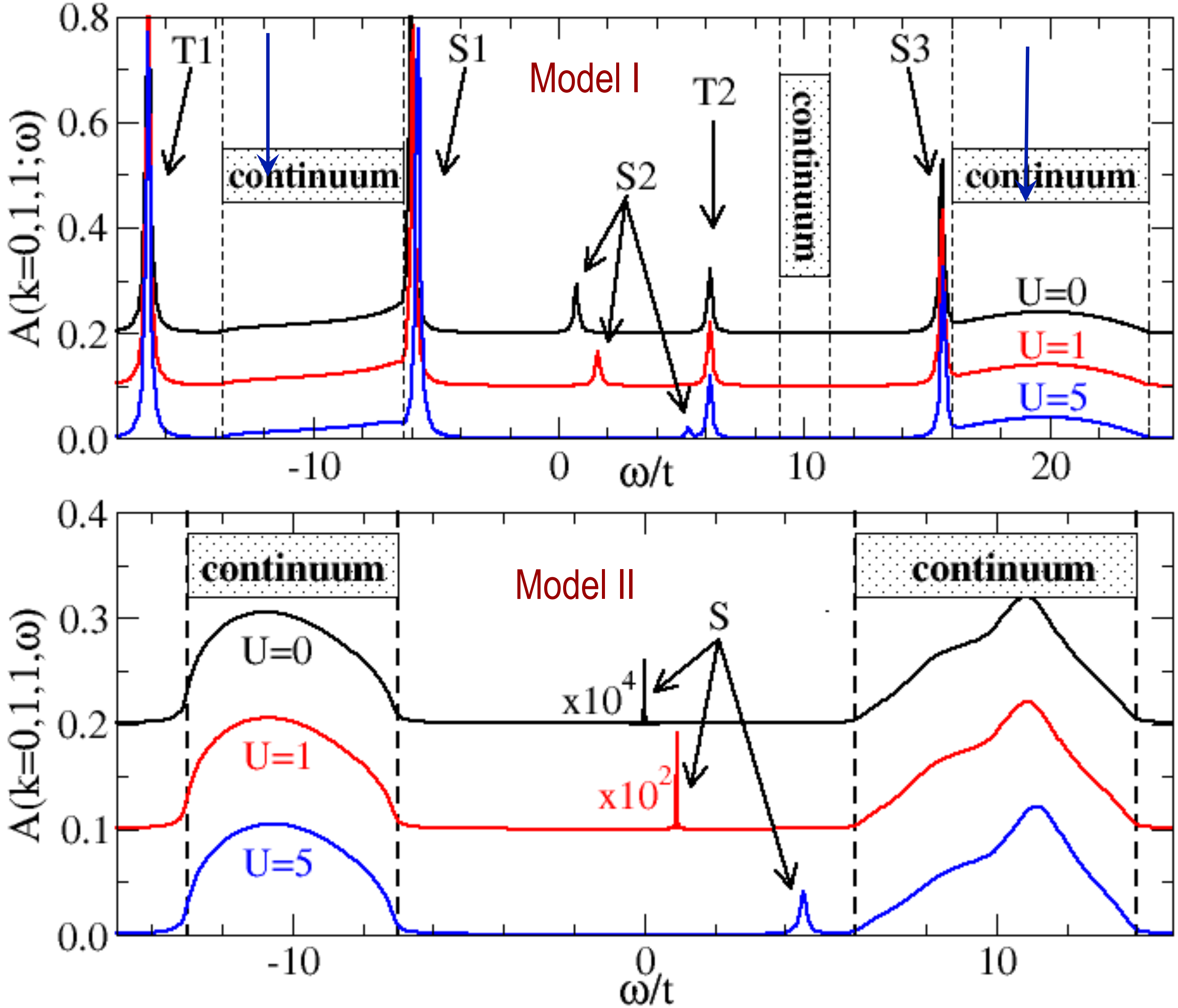
Few particle Green's functions in real space: M. Berciu, PRL 107, 246403 (2011)

polaron + spin-up carrier continuum

two spin-up carriers + magnon continuum

Spectral weights to find the carriers on neighboring sites (n=1)

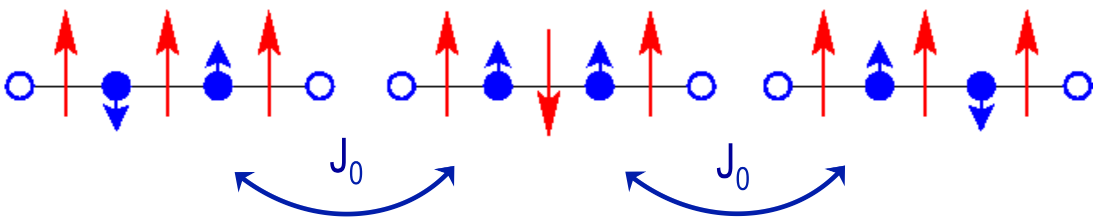
J_0=20, t=1, J=0.05, h=0.1, U=0,1,5



Mirko Moeller, George Sawatzky and Mona Berciu, PRL 108, 216403 (2012); PRB 86, 075128 (2012)

Q: Why is there a low-energy bipolaron in model I, but not in model II?

A: In model I, both carriers can interact with the same spin, simultaneously. The configurations with highest weight contribution to the low-energy bipolaron of model I are:

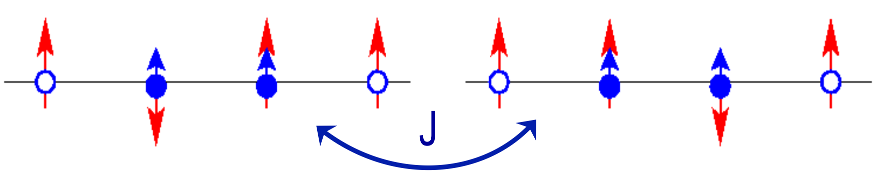


→ Large effective attraction due to magnon exchange (controlled by J_0)

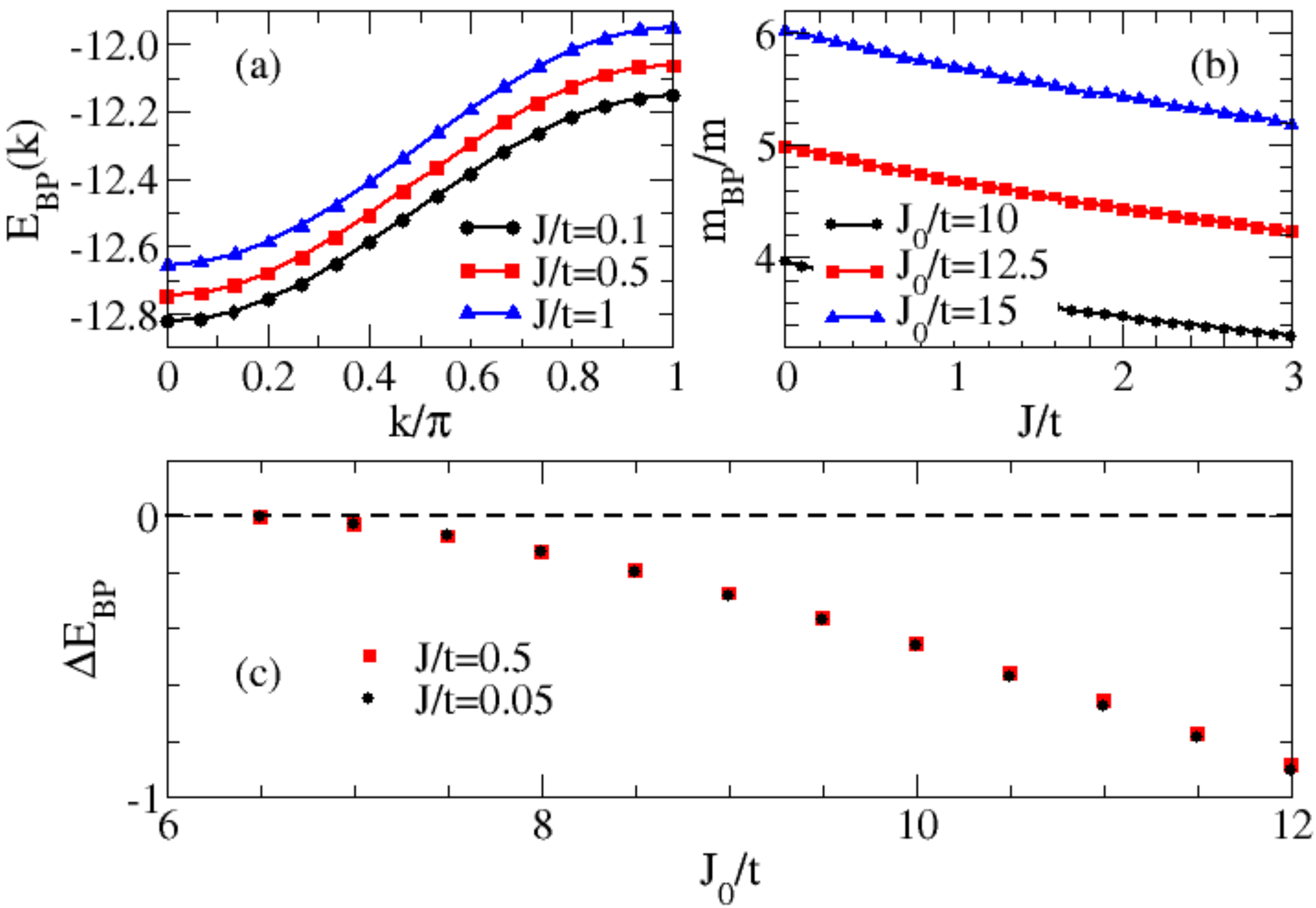
→ also explains insensitivity to U + triplet character at k=0

In model II, if both carriers are on the same site, they form a singlet → no exchange

If carriers are on neighboring sites → magnon exchange is controlled by J, too weak to bind the pair

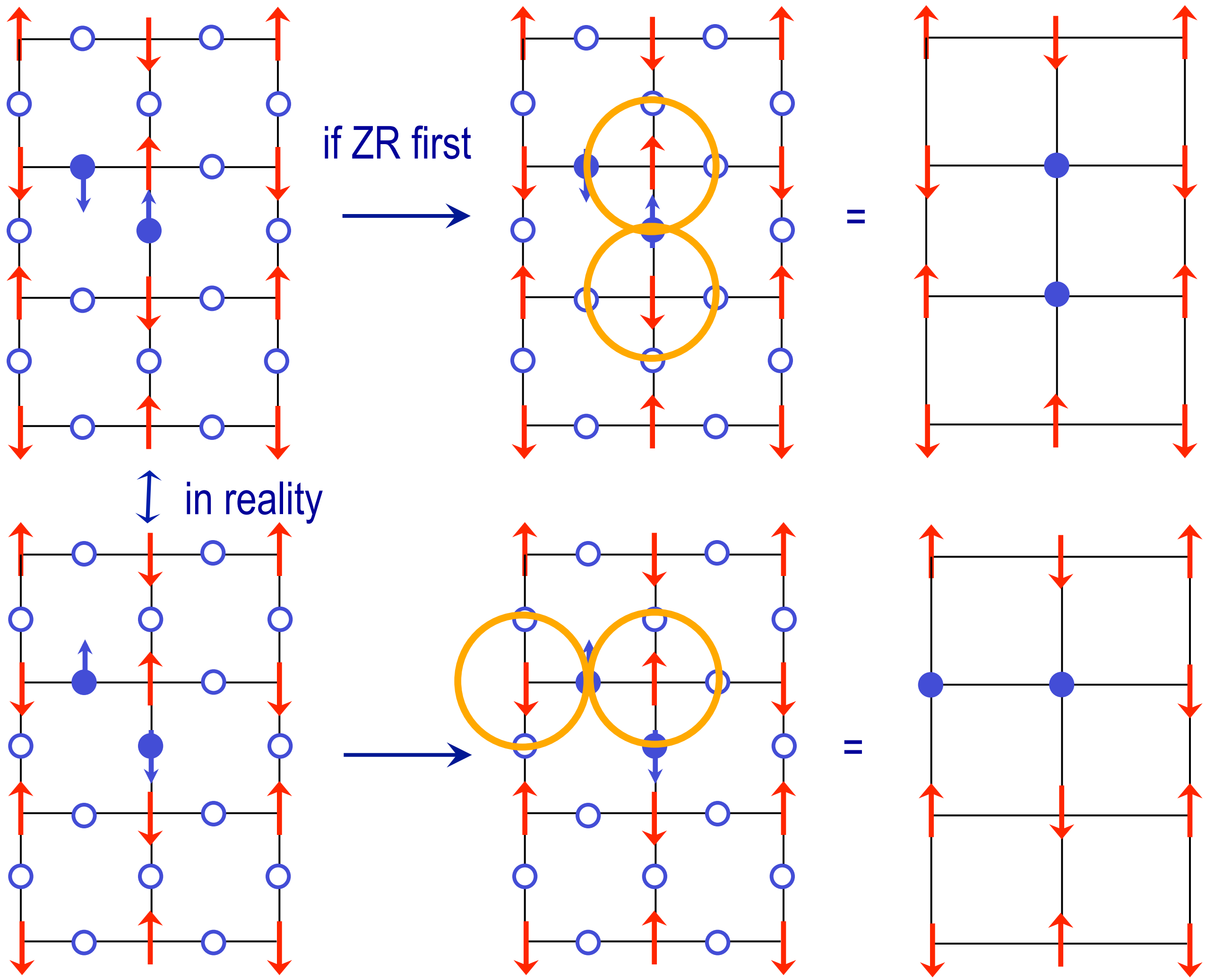


How heavy is the low-energy bipolaron, and when is it stable?



What are the implications for cuprates and other oxides?

"Composite" objects that combine together both charge and spin degrees of freedom may describe accurately the quasiparticle (single particle sector). However, they are likely to severely underestimate the strength of the magnon-mediated interactions.



"glue" of order J, due to minimizing # of broken AFM bonds

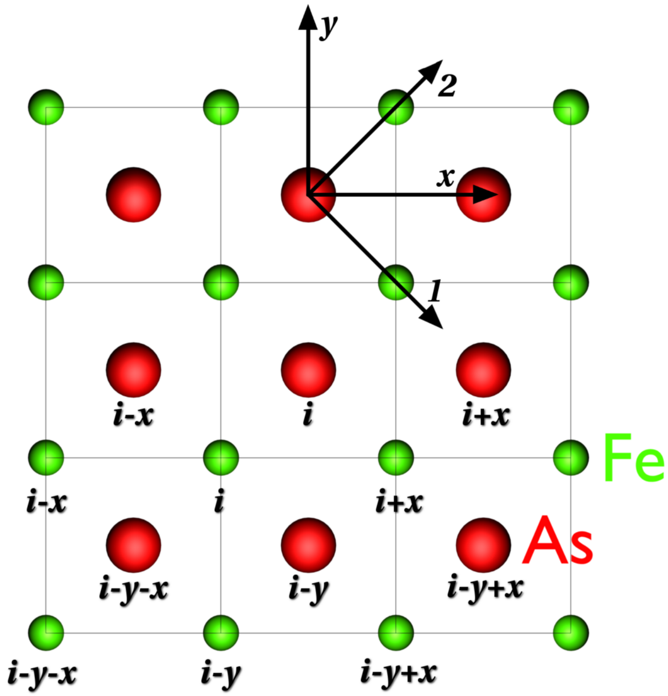
as if funny additional "hopping" of order J_0

We have also done some work to investigate the motion of a hole in an Ising 2D AFM – turns out there are all sorts of interesting things happening there, ask Hadi about them! He can also tell you what happens if disorder is introduced in the system – briefly, we find that the disorder potential is also strongly renormalized in the presence of particle-bosons interactions. Again, this shouldn't be too surprising, since there is no a priori reason for the disorder "seen" by a polaron to be the same as the disorder "seen" by a bare particle. What is surprising is how strong the effect can be, for example it can even turn attractive potentials into repulsive ones and viceversa.

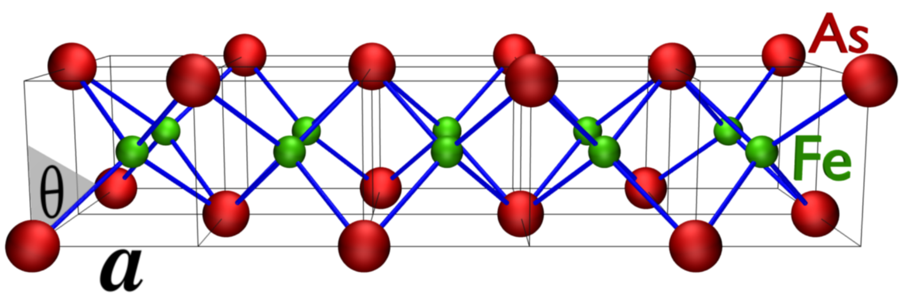
Q: Do we have time to look at electronic polarons?

Electronic polaron → very cool idea that G. Sawatzky has been pursuing for a long time

Let’s use pnictides = a FeAs layer as an example, although the idea is much broader and relevant for all materials (with different degrees of relevance).



← FeAs layer, top view



← FeAs layer, side view: it’s not planar!

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

A: DFT tells us that all states within 1-2eV of E_F are of Fe nature, so we can just use some Hubbard-like Hamiltonian for electrons on a square lattice, and argue whether we should use 2 or 5 bands, and whether U is small or large, etc etc.

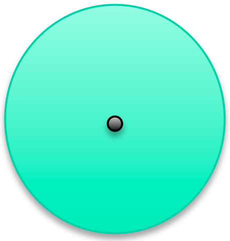
Problem: this implies that As play no role in the physics of these materials. Is that true?

Q: What do we know about the As ions?

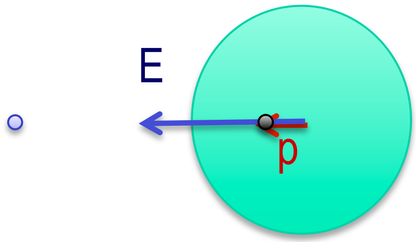
A: have full 4p orbitals, empty 5s orbitals → very "fat" spherical distribution of charge.

Q: What happens when we put extra charges in its vicinity?

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



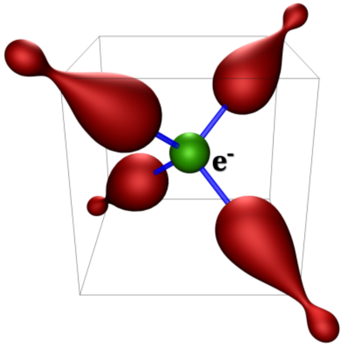
unperturbed As ion



perturbed As ion

$$H_{el-As} = -\hat{\vec{p}} \bullet \vec{E}$$

For simplicity, let's assume that only 4 nn As are polarized (E falls off like $1/R^2$) → each additional charge is surrounded by 4 polarized As whose electrons are now in a superposition of 4p and 5s states → an electronic polaron (particle dressed by electron-hole or exciton excitations). We studied this using perturbation theory in t (known to be the smallest energy in this problem, see PRB 79, 214507 (2009)).



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

$$\mathcal{H}_{\mathrm{As}} = \Omega\sum_{i,\lambda,\sigma} p^\dagger_{i,\lambda,\sigma}p_{i,\lambda,\sigma},$$ ← describes holes (excited states have holes in 4p orbitals)

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

Q: What do we find for a single polaron?

A: Its dispersion is

$$E_P(\vec{k}) = 4(\Omega - \sqrt{\Omega^2 + 4g^2}) - 2t_{eff}(\cos k_x + \cos k_y) - 4t'_{eff}\cos k_x \cos k_y$$

i.e. the effective hoppings (→ effective mass) is renormalized by about ~ 2-3. Again, increasing g will not make this arbitrarily large since there is an upper limit to how polarized the As clouds can be.

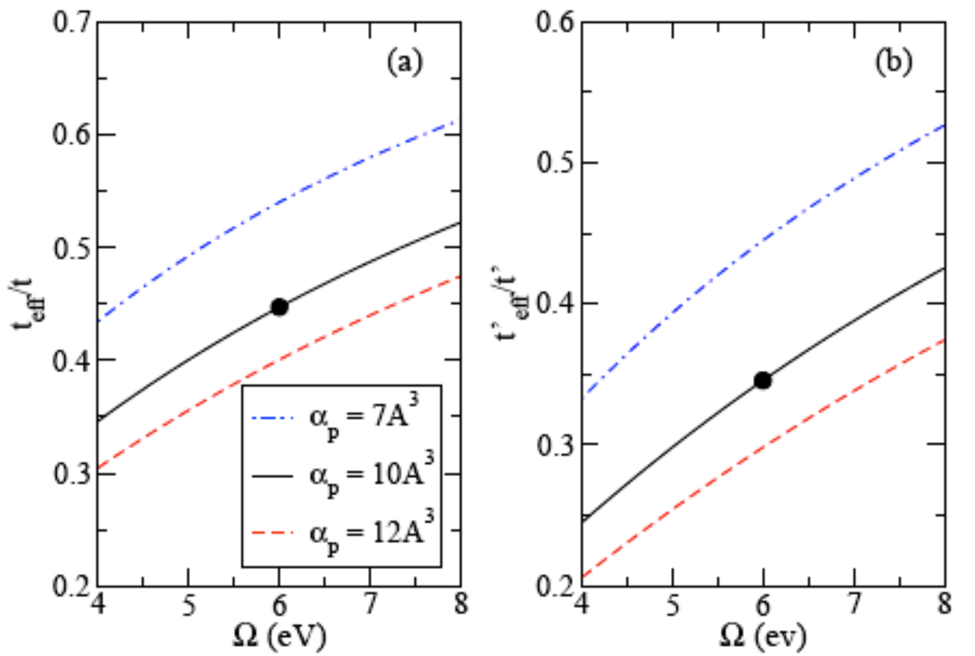


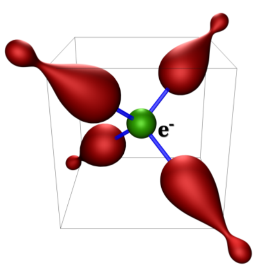
FIG. 3: (a) t_{eff}/t and (b) t'_{eff}/t' vs. Ω, for a polarizability $\alpha_p = 7, 10$ and 12Å^3. The dots show the values used here.

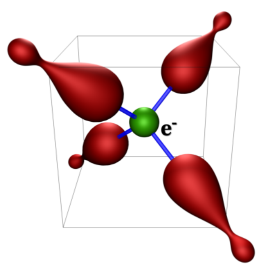
Q: Do we find bipolarons if U_H is really large?

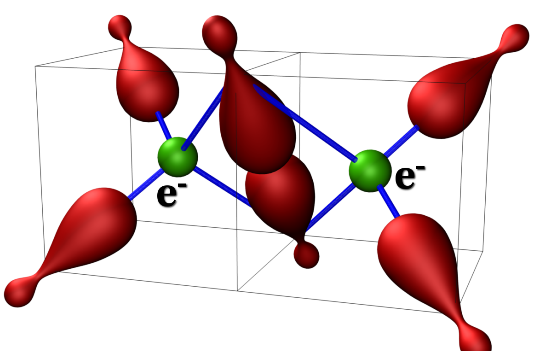
A: Yes! They are S1-like, i.e. with charges on nn Fe, not on the same Fe.

Q: why?

A: “shared” As differently polarized







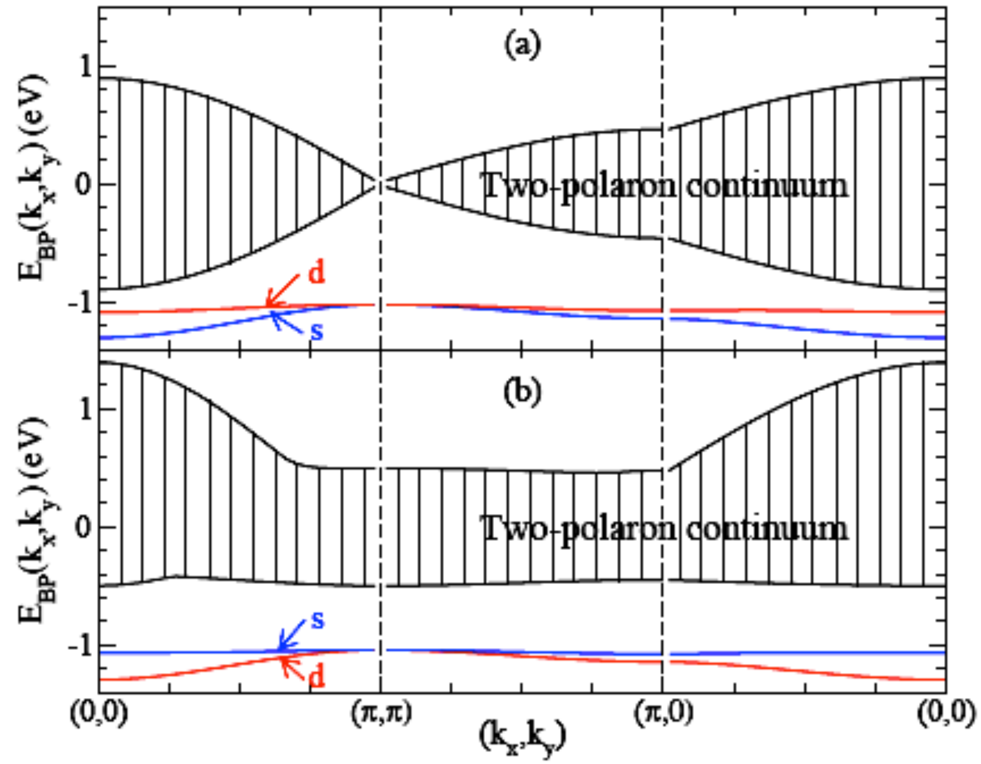


FIG. 7: Dispersion of the two bound bipolaron states along high-symmetry axes in the Brillouin zone, for (a) $t' = 0$ and (b) $t' = -t/2$. The two-polaron continuum is also shown. Parameters are $U_H = 10$ eV, $\alpha_p = 10\AA^3, \Omega = 6$ eV (similar results are found for all $\alpha_p = 7 - 12\AA^3, \Omega = 4 - 8$ eV). The symmetry of the ground state changes from s to d if $t' \neq 0$.

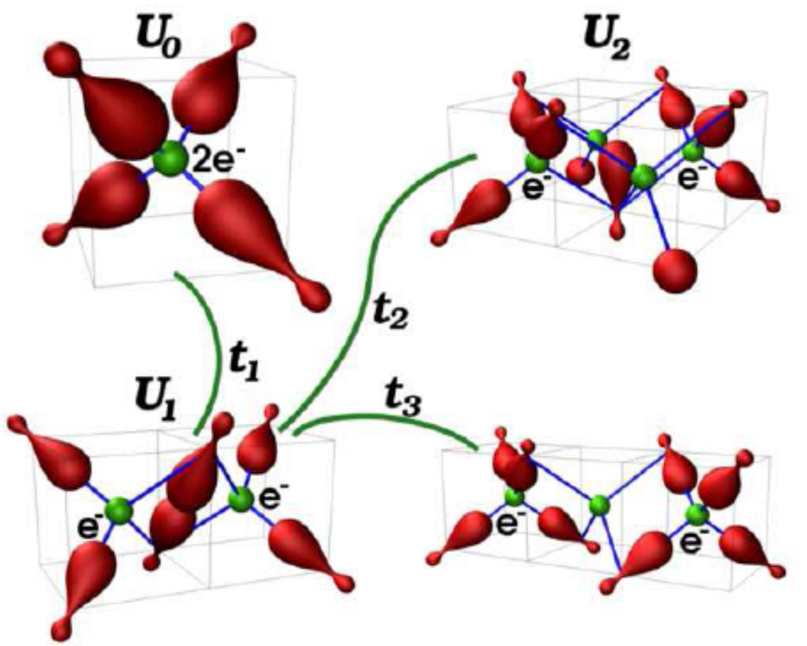


FIG. 4: Sketches of on-site, 1^{st}, 2^{nd} and 3^{rd} nn bipolarons. The first three configurations have interaction energies U_0, U_1 and U_2, respectively. Several of the special effective hopping integrals are also indicated.

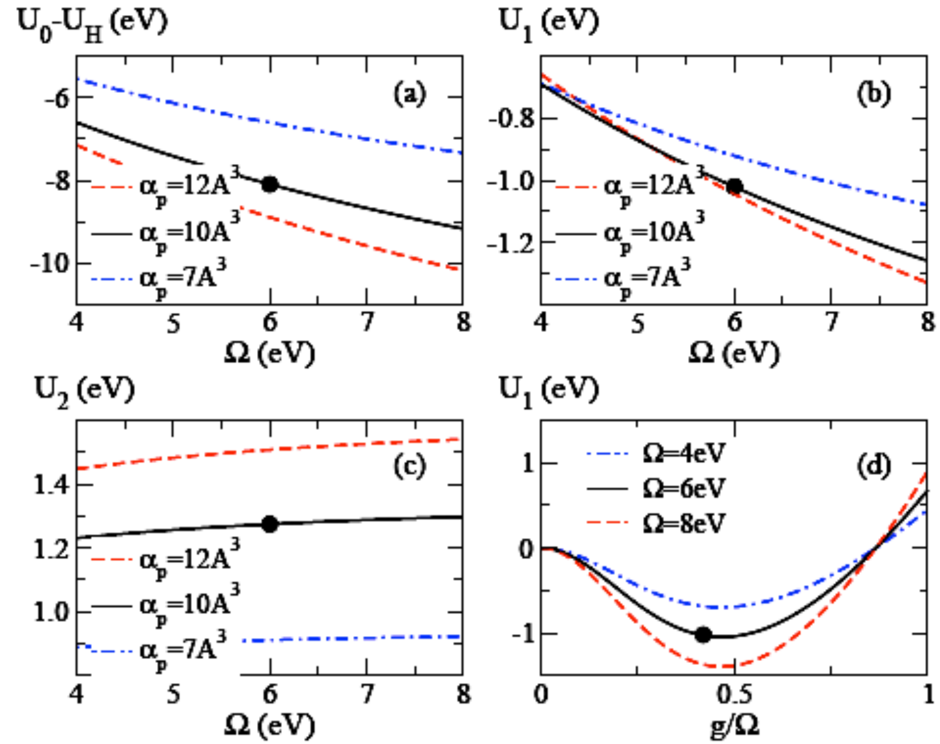
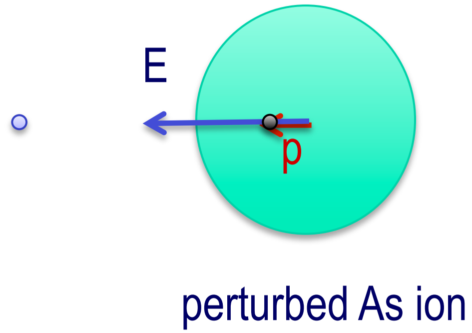


FIG. 5: (a) Renormalization of on-site interaction, $U_0 - U_H$; (b) nn energy U_1 and (c) 2^{nd} nn energy U_2 vs. Ω for various polarizabilities. (d) U_1 vs. g/Ω when $\Omega = 4, 6, 8$eV. The dots show our typical values.

U_0 attractive → typical bipolaron effect. But why is U_1 attractive while U_2 is repulsive?

Simple (semi-classical) explanation: G. A. Sawatzky et al, EPL 86, 17006 (2009)



$$W = -\vec{p} \bullet \vec{E}$$

$$\vec{p} = \alpha \vec{E} \quad (\ \alpha = \text{polarizability})$$

$$\rightarrow W = -\frac{1}{2}\alpha \vec{E}^2$$

If an As interacts simultaneously with two charges:

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

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

The sign of W_{int} is controlled by geometry (lattice structure)! (in FeAs structure, this angle < 90 for nn, > 90 for nnn)

→ One might be able to play some interesting games by properly arranging polarizable atoms in suitable locations …

Some conclusions:

→ Polaronic effects appear in many guises and forms and can have a dramatic influence on the properties of materials

→ Many many many still open questions …