Collective Excitations and Stability of the Excitonic Phase in the Extended Falicov–Kimball model

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- Falicov–Kimball model and the excitonic state
  - Excitonic state as a mean field solution
  - Excitonic insulator in a conventional semi-metal
- Extending the Falicov–Kimball model
  - Degeneracy of the excitonic state
  - Stabilising the excitonic state by the perturbation
- Collective excitations in the Falicov–Kimball model
  - Nature of the instability of the excitonic state at weak perturbation
  - Beyond the leading order: stabilising the spectrum
- Implications for the critical temperature.
  - Phase-disordered state at finite temperature?
- Electronic ferroelectricity: how and when.

Spinless Falicov – Kimball Model and the Excitonic State

- Localised and itinerant fermions, on-site interaction U, half-filling, $T=0$.

$$\mathcal{H} = -\frac{t}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger c_j + c_j^\dagger c_i\right) + E_d\sum_i d_i^\dagger d_i + U\sum_i c_i^\dagger d_i^\dagger d_i c_i\,,$$

- Introduced in 1969 to describe mixed-valence phenomena and metal-insulator transitions.
- Uniform Hartree–Fock mean field solution (A. N. Kocharyan, D. I. Khomskii, 1976; H. J. Leder, 1978):
  - Mixed-valence regime: two partially filled bands.
  - Interaction-induced spontaneous hybridisation $\Delta = \langle c_i^\dagger d_i\rangle$ – excitonic phase.
  - Filled and empty quasiparticle bands:

$$\epsilon_{\vec{k}}^{(1,2)} = \frac{1}{2}\left\{(E_d+Un_c)+(\epsilon_{\vec{k}}+Un_d)\mp\sqrt{[(E_d+Un_c)-(\epsilon_{\vec{k}}+Un_d)]^2+4U|\Delta|^2}\right\}$$

where

$$n_c = 1-n_d\,,\quad \epsilon_{\vec{k}} = -\cos k_x - \cos k_y(-\cos k_z)\,,\quad t=1.$$

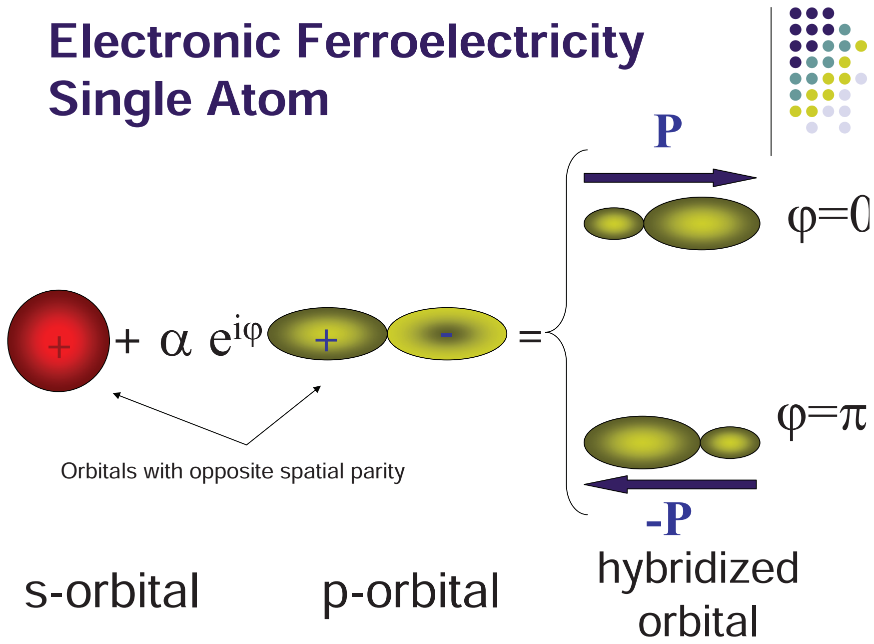
–Gap equation: at $T=0$ Narrow-band occupancy:

$$\Delta = \frac{1}{N}\sum_{\vec{k}}\Delta_{\vec{k}}\,,\quad \Delta_{\vec{k}} \equiv \langle c_{\vec{k}}^\dagger d_{\vec{k}}\rangle = \frac{U\Delta}{\sqrt{\xi_{\vec{k}}^2+4U^2|\Delta|^2}}\,;\qquad n_d = \frac{1}{N}\sum_{\vec{k}} n_{\vec{k}}^d\,,\quad n_{\vec{k}}^d \equiv \langle d_{\vec{k}}^\dagger d_{\vec{k}}\rangle = \frac{1}{2} - \frac{\xi_{\vec{k}}}{2\sqrt{\xi_{\vec{k}}^2+4U^2|\Delta|^2}}.$$

Notation: $\xi_{\vec{k}} = (E_d+Un_c)-(\epsilon_{\vec{k}}+Un_d)$, and $E_{rd} = E_d + Un_c - Un_d$.

Electronic Ferroelectricity

- Ferroelectricity which does not involve the lattice. (T. Portengen *et al.*, 1996)
- Requires opposite-parity bands and $\mathrm{Re}\Delta \neq 0$:



(from a 2006 talk by J. E. Gubernatis)

Experimental Search for Excitonic Insulator or Electronic Ferroelectric

- No generally recognised excitonic insulators at present.
- Large number of candidates: SmB_6, SmS, TmSe, $TiSe_2$, $TmSe_{0.45}Te_{0.55}$, $LuFe_2O_4$,...
  Typically, a narrow band involved. Review: J. Neuenschwander and P. Wachter (1990)
- Suggested relevance for CMR manganates and URu_2Si_2.
- Theory models of specific compounds: T. A. Kaplan, S. D. Mahanti, 1970s; K. A. Kikoin (1993); S. Curnoe, K. A. Kikoin (2000),...

Degeneracy of Excitonic Insulator in the Falicov – Kimball Model

$$\mathcal{H}_0 = -\frac{t}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger c_j + c_j^\dagger c_i\right) + E_d\sum_i d_i^\dagger d_i + U\sum_i c_i^\dagger d_i^\dagger d_i c_i\,,$$

- Local continuous degeneracy of the excitonic state: $d_i \longrightarrow d_i \exp(i\varphi_i)$, phase of $\langle c_i^\dagger d_i\rangle$.

  $\Rightarrow$ Instability of the excitonic state (Elitzur's theorem) (V. Subrahmanyam and M. Barma, 1988)
- An infinitesimal perturbation of $\mathcal{H}_0$ restores excitonic insulator at $T = 0$???

  J. K. Freericks and V. Zlatić, (2003)

Extending the Falicov – Kimball Model

- Different kinds of perturbations break the degeneracy:

  –hopping $t' \ll t$ in the narrow zone.

  –hybridisation: V_0 on-site, V_1 nearest-neighbour (same parity bands), V_2 (opposite parity).

$$\delta\mathcal{H} = -\frac{t'}{2}\sum_{\langle ij\rangle} d_i^\dagger d_j - V_0\sum_i c_i^\dagger d_i - \frac{V_1}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger d_j + c_j^\dagger d_i\right) - \frac{V_2}{2}\sum_{\langle ij\rangle}\left\{(\vec{R}_i - \vec{R}_j)\cdot\vec{\Xi}\right\}\left(c_i^\dagger d_j - c_j^\dagger d_i\right) + \mathrm{H.c.}\,,$$

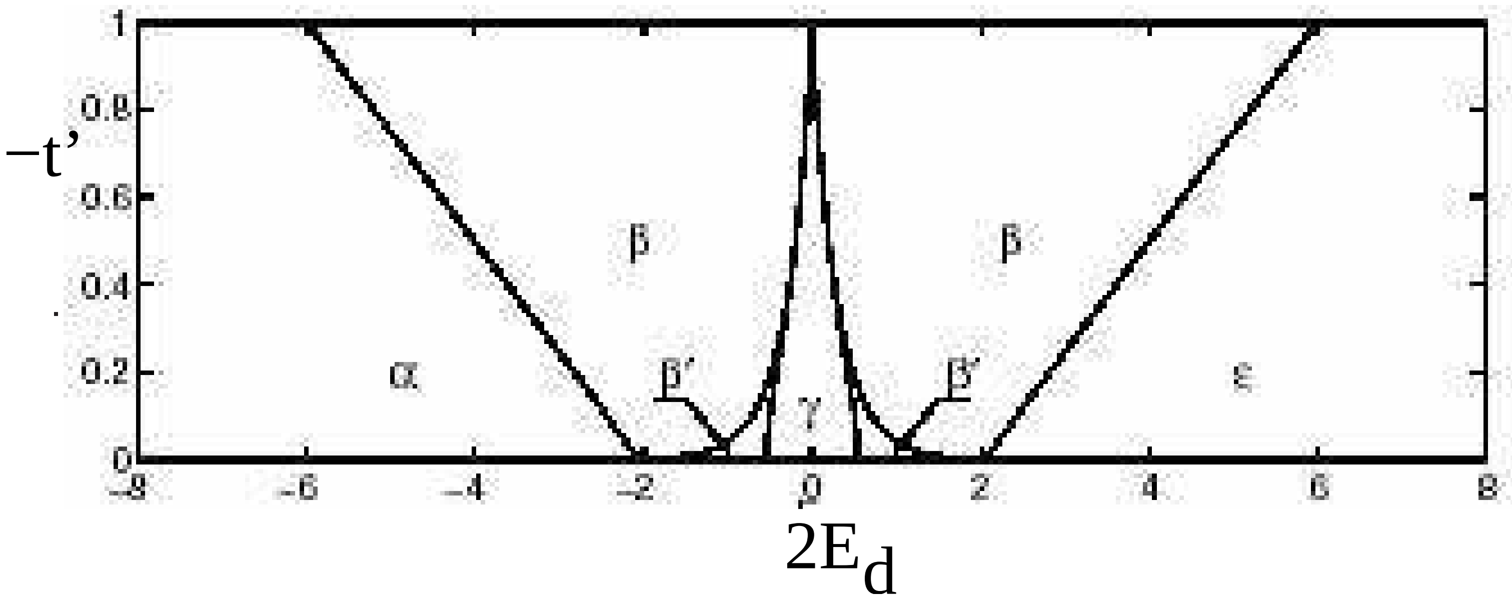
($\vec{R}_i$ is the radius-vector of a site i, $a\vec{\Xi} = \{1, 1(, 1)\}$, lattice period $a \longrightarrow 1$).

- Extended FKM - no longer exactly soluble.

  –literature: mean field, numerical (C. Czycholl, P. Farkašovský, C. D. Batista, P. M. R. Brydon...)

  –mean field solution corresponding to the excitonic phase is still present: $\Delta = \langle c_i^\dagger d_i\rangle \neq 0$.

- Example: small but finite t' stabilises the excitonic solution:



variational mean field
P. Farkašovský (2008)

(2D, $U = 1$, $t' < 0$, $t \equiv 1$)

β - uniform excitonic phase, γ, β' – charge ordered, α, ϵ - no mixed valence

- Stabilisation of the excitonic phase on increasing $|t'|$ via 2nd order phase transition.

$\Rightarrow$ contradicts suggested more exotic behaviour
(the latter expected if the instability of the excitonic phase for pure FKM is due to local degeneracy.)

Collective Excitations in the Excitonic State of the Falicov–Kimball Model

- Hamiltonian of the pure FKM:

$$\mathcal{H}_0 = -\frac{t}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger c_j + c_j^\dagger c_i\right) + E_d\sum_i d_i^\dagger d_i + U\sum_i c_i^\dagger d_i^\dagger d_i c_i \,,$$

- Weak perturbation, t', V_i small:

$$\delta\mathcal{H} = \sum_{\vec{k}}\left\{t'\epsilon_{\vec{k}} d_{\vec{k}}^\dagger d_{\vec{k}} + V_{\vec{k}} c_{\vec{k}}^\dagger d_{\vec{k}} + V_{\vec{k}}^* d_{\vec{k}}^\dagger c_{\vec{k}}\right\} \,, \quad V_{\vec{k}} = \begin{cases} V_0 + V_1\epsilon_{\vec{k}}, & \text{same parity bands,} \\ \mathrm{i}V_2\lambda_{\vec{k}}, & \text{opposite parity,} \end{cases}$$

where $\epsilon_{\vec{k}} = -\sum_{\alpha=1}^{d}\cos k_\alpha$ and $\lambda_{\vec{k}} = -\sum_{\alpha=1}^{d}\sin k_\alpha$.

- Generic particle-hole excitation in the excitonic phase:

$$\mathcal{X}_{\vec{q}} = \frac{1}{\sqrt{N}}\sum_{\vec{k}}\left\{F_+(\vec{k},\vec{q})\, c_{\vec{k}}^\dagger d_{\vec{k}+\vec{q}} + F_-(\vec{k},\vec{q})\, d_{\vec{k}}^\dagger c_{\vec{k}+\vec{q}} + F_c(\vec{k},\vec{q})\, c_{\vec{k}}^\dagger c_{\vec{k}+\vec{q}} + F_d(\vec{k},\vec{q})\, d_{\vec{k}}^\dagger d_{\vec{k}+\vec{q}}\right\} \,.$$

- Eigenstate in the mean field sense:

$$[\mathcal{X}_{\vec{q}}, \mathcal{H} + \delta\mathcal{H}] \stackrel{\text{eff}}{=} \omega_{\vec{q}}\mathcal{X}_{\vec{q}} \,.$$

(Hartree-Fock decoupling in the $[\ldots]$)

- Pure FKM ($t' = V_i = 0$) - degenerate, hence solution, $\mathcal{X}_{\vec{q}}^{(0)} = \frac{1}{\sqrt{N}}\sum_{\vec{k}} d_{\vec{k}}^\dagger d_{\vec{k}+\vec{q}}$, with $\omega_{\vec{q}} \equiv 0$.
- In general, decoupling and collecting terms in the secular equation.

Spectrum in the Pure FKM (no perturbation)

- A branch that vanishes identically throughout BZ:

$$\omega_{\vec{q}} = 0 \text{ for any } \vec{q}$$

- A consequence of local continuous degeneracy

cf. Kagomé antiferromagnet

Leading Order in Perturbation: the Instability

- Weak perturbation $\rightarrow$ Keep leading order in t', $V_{0,1}$ (linear), or V_2 (quadratic), linearise in ω^2,

$$(\omega_{\vec{q}}/U\Delta)^2 \cdot D_\omega(\vec{q}) + M_{11}(\vec{q}) \cdot D_0(\vec{q}) = 0\,.$$

- $\omega_{\vec{q}}^2$ always changes sign in the BZ $\Rightarrow$ instability! $\omega_{\vec{q}}^2 < 0$ for some $\vec{q}$.
- The instability is conventional, due to the presence of a lower-energy state.
  - –Not due to the local continuous degeneracy of the pure FKM.
  - –Not visible in the spectrum unless small perturbation included. (this, due to degeneracy)

Beyond the Linear Order: Stabilising the Spectrum

- Numerically, a very small perturbation stabilises the excitonic phase. Can we see this in our spectrum?
- Technically, leading order instability is due $D_0(\vec{q})$ changing sign in the BZ.
- $D_0(\vec{q})$ is not large numerically.
- The next-order correction $D_0(\vec{q}) \Rightarrow D_1(\vec{q}) + D_0(\vec{q})$
- Stabilisation if $D_1(\vec{q}) + D_0(\vec{q}) < 0$ for all $\vec{q}$.

Spectrum of Excitonic Phase a 2D EFKM, $U = 2$ and $E_d = 0.4$.

$$\mathcal{H} = -\frac{t}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger c_j + c_j^\dagger c_i\right) + E_d\sum_i d_i^\dagger d_i + U\sum_i c_i^\dagger d_i^\dagger d_i c_i\,,$$

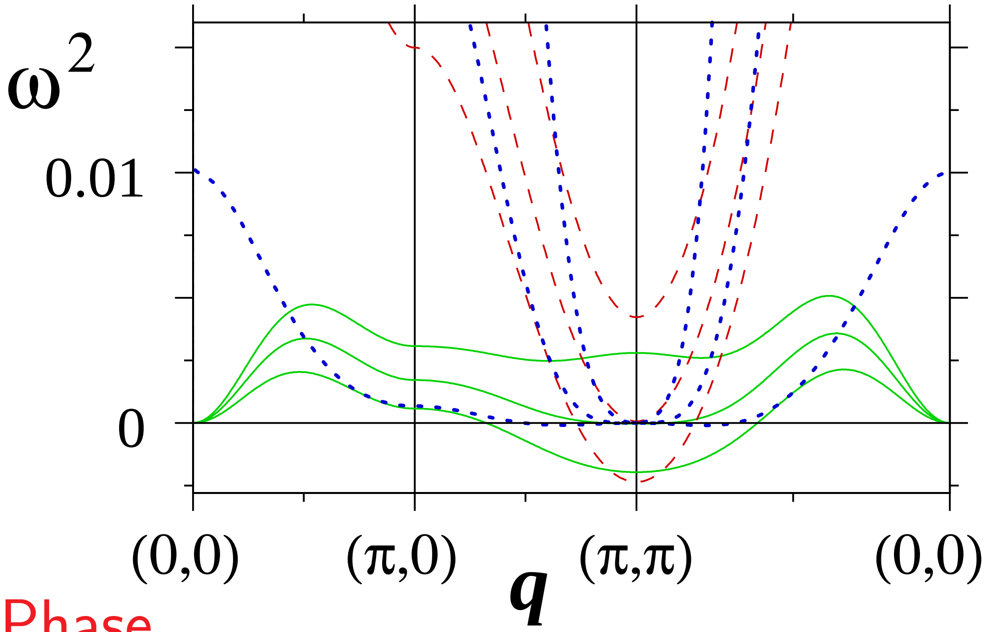
- stabilised by the perturbation:

$$\delta\mathcal{H} = -\frac{t'}{2}\sum_{\langle ij\rangle} d_i^\dagger d_j - V_0\sum_i c_i^\dagger d_i - \frac{V_1}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger d_j + c_j^\dagger d_i\right) - \frac{V_2}{2}\sum_{\langle ij\rangle}\left\{(\vec{R}_i - \vec{R}_j)\cdot\vec{\Xi}\right\}\left(c_i^\dagger d_j - c_j^\dagger d_i\right) + \text{H.c.}\,,$$

–Solid lines: effect of t'
bottom to top: $t' = -0.04, -0.0565, -0.07$

–Dashed lines: effect of V_0
bottom to top: $V_0 = -0.06, -0.074, -0.09$

–Dotted lines: effect of V_2
bottom to top: $V_2 = 0.115, 0.249, 0.35$



- In all cases, $\omega_{\vec{q}}$ within the quasiparticle gap

Surviving Degeneracies of the Excitonic Phase

- Effect of $t' < 0$: Goldstone mode at $\vec{q} = (0,0)$ $\quad d_i \longrightarrow d_i \exp(\mathrm{i}\varphi)$ globally

- Effect of $V_2 \neq 0$: Goldstone mode at $\vec{q} = (\pi,\pi)$ $\quad d_i \longrightarrow d_i \exp(\pm\mathrm{i}\varphi)$ in a checkerboard pattern, globally

Phase Diagram of the Excitonic Insulating State in EFKM

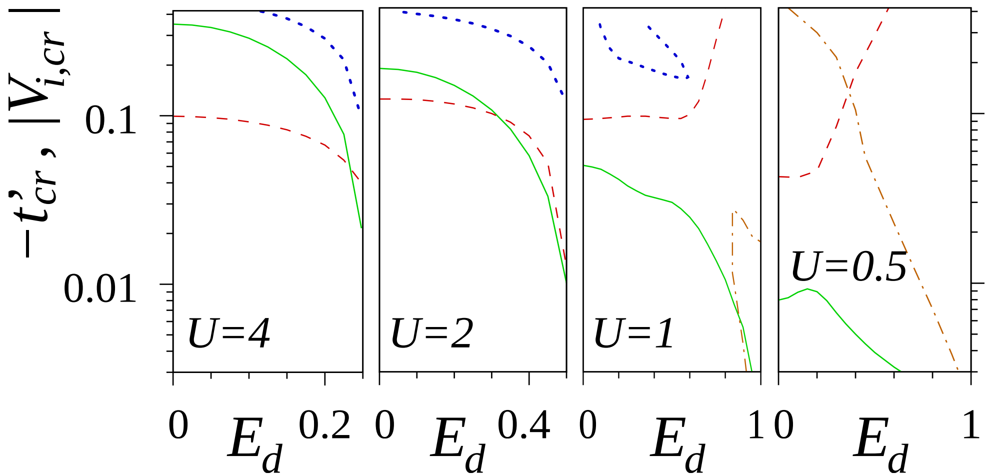
- Critical values of perturbations in a 2D EFKM at $T=0$ for different U:

  –Solid lines: $t'_{cr}(E_d)$

  –Dashed lines: $V_{0,cr}(E_d)$

  –Dashed-dotted lines: $V_{1,cr}(E_d)$

  –Dotted lines: $V_{2,cr}(E_d)$



- $U=0.5$ – weak coupling regime

  $U=4$ – strong coupling

- Expected critical temperature for excitonic insulator in EFKM:

$$T_C \propto \sqrt{|t'|(|t'|-|t'_{cr}|)},\ |V_2|\sqrt{V_2^2-V_{2,cr}^2},\ \sqrt{|V_0|(|V_0|-V_{0,cr})},\ \text{or}\ \sqrt{|V_1|(|V_1|-|V_{1,cr}|)}\ \text{(roughly)}$$

- This is much lower than critical temperature T_Δ from the gap equation:

$$T_\Delta \sim k_B\Delta\,, \qquad \frac{1}{N}\sum_{\vec{k}}\frac{U}{\sqrt{\xi_{\vec{k}}^2+4U^2|\Delta|^2}}=1$$

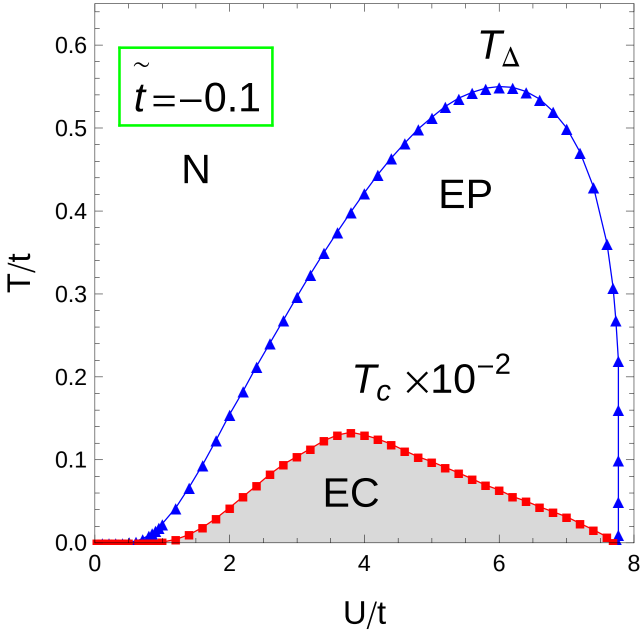
- What happens at $T_C < T < T_\Delta$? V. A. Apinian & T. K. Kopec, 2013:

$$\langle c_i^\dagger d_i\rangle = \Delta_i \exp(\mathrm{i}\phi_i)$$

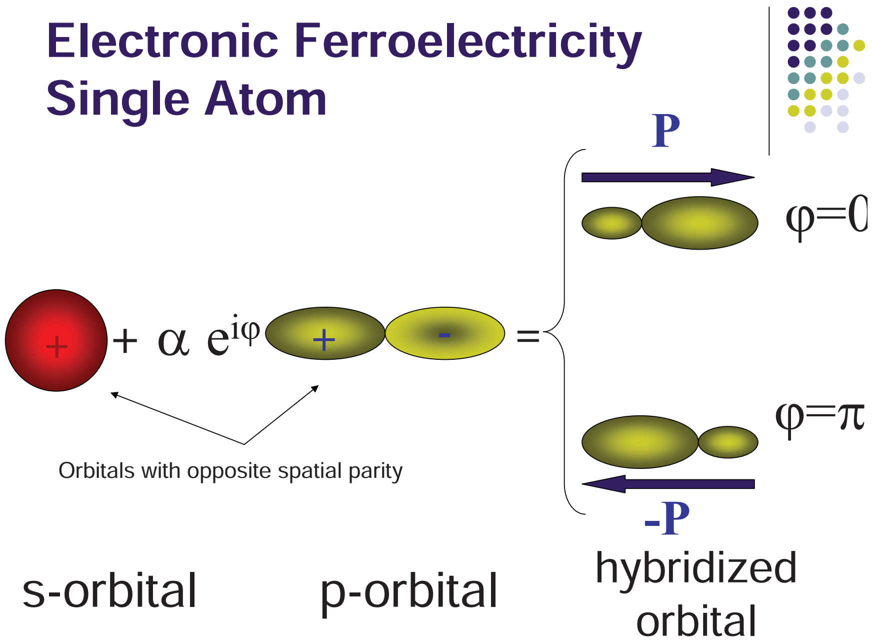
$T<T_C$ – both Δ_i and ϕ_i ordered, excitonic insulator.

$T_C<T<T_\Delta$ – only Δ_i ordered, ϕ_i disordered, novel phase.

$T>T_\Delta$ – all $\Delta_i\equiv 0$, conventional semiconductor or semimetal.



Conditions for Electronic Ferroelectricity at $T = 0$



(from a 2006 talk by J. E. Gubernatis)

$$\mathcal{H} = -\frac{t}{2}\sum_{\langle ij\rangle}\left(c_i^\dagger c_j + c_j^\dagger c_i\right) + E_d\sum_i d_i^\dagger d_i + U\sum_i c_i^\dagger d_i^\dagger d_i c_i\,,$$

- Perturbation terms allowed for opposite-parity bands:

$$\delta\mathcal{H} = -\frac{t'}{2}\sum_{\langle ij\rangle} d_i^\dagger d_j - \frac{V_2}{2}\sum_{\langle ij\rangle}\left\{(\vec{R}_i - \vec{R}_j)\cdot\vec{\Xi}\right\}\left(c_i^\dagger d_j - c_j^\dagger d_i\right) + \mathrm{H.c.}\,,$$

- Polarisation density:

$$\vec{P} = 2\vec{\mu}\mathrm{Re}\left(\frac{1}{N}\sum_{\vec{k}}\langle c_{\vec{k}}^\dagger d_{\vec{k}}\rangle\right)$$

$\vec{\mu}$ – interband element of the dipole moment operator, direction determined by the orbital structure

- Including the electrostatic dipole-dipole interaction:

$$\mathcal{E}_{tot} = \mathcal{H} + \delta\mathcal{H} - \int_{(\text{sample})} \left(\vec{E} \cdot \vec{P} + \frac{1}{2} \vec{E}_{\text{int}} \cdot \vec{P} \right) dV$$

$\vec{E}$ – external, $\vec{E}_{\text{int}}$ – created by $\vec{P}(r)$

- $\vec{E} = 0$, no free charges, hence

$$\int \vec{E}_{\text{int}} \cdot \vec{D} dV = 0 \quad \Rightarrow \quad -\frac{1}{2} \int_{(\text{sample})} \vec{E}_{\text{int}} \cdot \vec{P} dV = \frac{1}{8\pi} \int_{(\text{entire space})} E_{\text{int}}^2 dV$$

– positive quantity, $\propto P^2$ (usual cause of domain formation)

- $-t' > |t'_{\text{cr}}|$ and $V_2 = 0$ – micro degeneracy (global) with respect to the phase of $\langle c_i^\dagger d_i \rangle$

  $\mathcal{H} + \delta\mathcal{H}$ have same value for $\vec{P} = 0$ and $\vec{P} \neq 0$ states

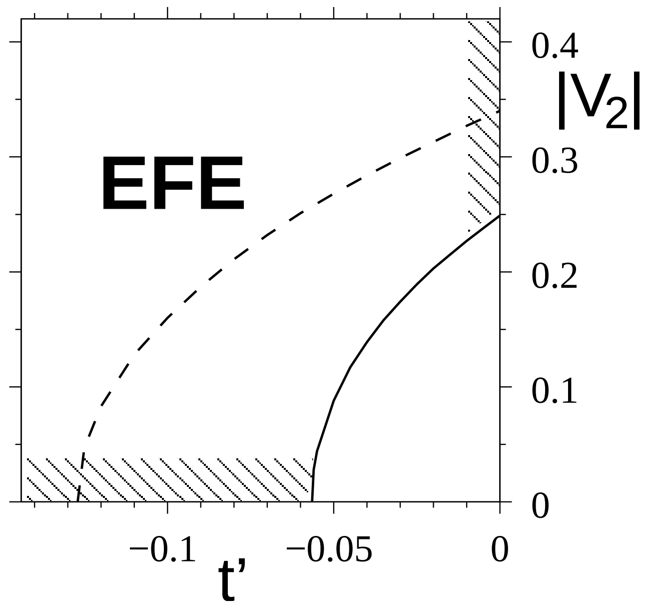
- $\vec{P} = 0$ at $\vec{E} = 0$, no spontaneous polarisation
- $\vec{P}(E)$ linear, $E_{\text{int}} = -E$ –divergent dielectric constant
- $\vec{P}(E)$ saturates for $|\vec{E} \cdot \vec{\mu}/\mu| > 8\pi|\mu||\langle c_i^\dagger d_i \rangle|/3$ (spherical sample).
- $|V_2| > V_{2,\text{cr}}$ and $-t' = 0$ – similar behaviour, $\vec{P} = 0$ at $E = 0$.

  (degeneracy associated with Goldstone at $\vec{q} = (\pi, \pi)$)

- $V_2 \neq 0$, $t' < 0$ – no degeneracy; excitonic insulator stable

  $\Rightarrow$ spontaneous polarisation.

- when either t' or V_2 too small, $\vec{P}$ destroyed by dipole-dipole interaction.



Conclusions

- An insight into the instability of the excitonic phase in the pure FKM.
  - A lower-energy ground state of the pure FKM (not the fluctuations in the degenerate excitonic phase).
  - On an increase of the perturbation, stabilising the excitonic state via the 2nd order phase transition.
- In the literature, T_C of excitonic insulator (in the EFKM or otherwise) is calculated from the gap equation:
  - Transition due to thermal excitations of the electron-hole continuum.
  - Effect of collective excitations on T_C not considered.
- We calculated the low-lying excitation energy in the excitonic state of the extended Falicov–Kimball model.
  - New low energy scale, related to T_C of the excitonic insulator.
  - When the pure FKM limit is approached (narrow band), T_C is strongly suppressed.
  - Expected: 2nd order transition mediated by excitations at T_C.
  - Novel phase-disordered state above T_C?
- The quest for electronic ferroelectricity:
  - Both narrow-band hopping and hybridisation required for a robust spontaneous polarisation.

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Excitonic Insulator in a Conventional Semimetal (Semiconductor)

L. V. Keldysh and Yu. V. Kopaev (1964); the following is after W. Kohn (1967)

- Valence and conduction bands of identical shape, nested Fermi surface:

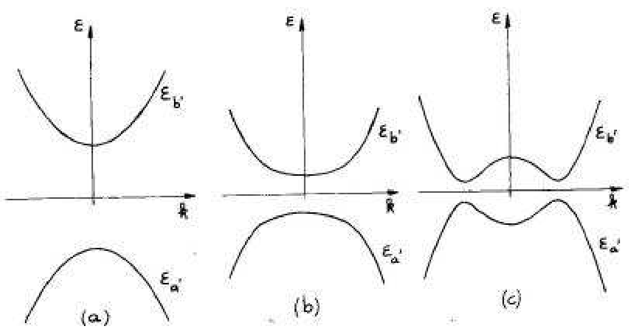
$$\varepsilon_a(\vec{k}) = -\frac{G}{2} - \frac{k^2}{2m_a}\,, \quad \varepsilon_b(\vec{k}) = \frac{G}{2} + \frac{(\vec{k}-\vec{k}_0)^2}{2m_b}$$

- Interaction $V(\vec{q}) \equiv V_0$, gap function $\Delta = (V_0/N)\sum_{\vec{k}}\langle a^\dagger_{\vec{k}} b_{\vec{k}}\rangle$, gap equation for $T=0$:

$$1 = \frac{V_0}{2N}\sum_{\vec{k}} \frac{1}{\{\frac{1}{4}[\varepsilon_a(\vec{k}) - \varepsilon_b(\vec{k})]^2 + \Delta^2\}^{1/2}} \quad \Rightarrow \quad \Delta = \Delta(G) \quad \Rightarrow \quad T_c(G) \sim \Delta(G)/k_B$$

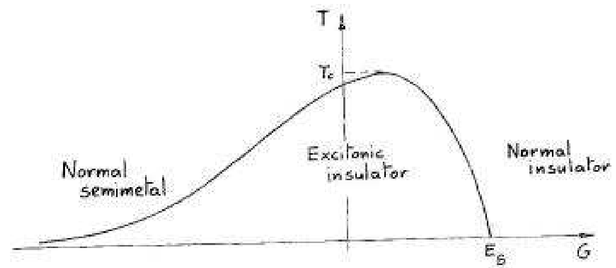
(assuming $\vec{k}_0 = 0$; on the semiconducting side, V is $\vec{q}$-dependent $\rightarrow$ quantitative change)

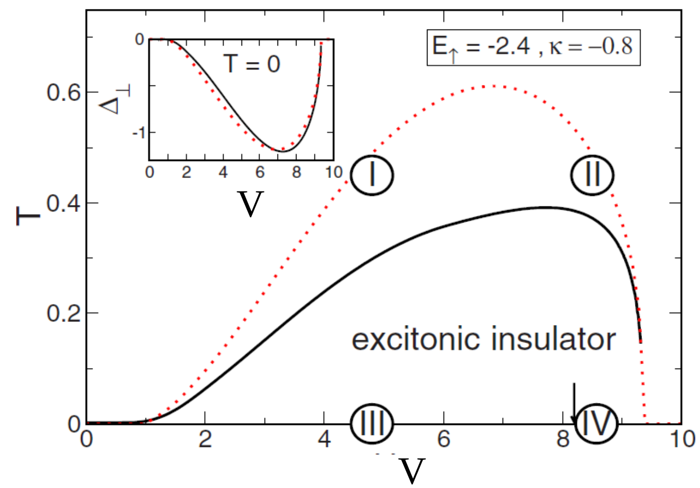
- Hybridised bands: "The Snail:" slave boson MF (dots Hartree–Fock):



G>0 G=0 G<0

Critical temperature, T_c(G)





↑ B. Zenker *et al.* (2010a)