

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, $T = 0$.

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

- 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:

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

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 $T = 0$

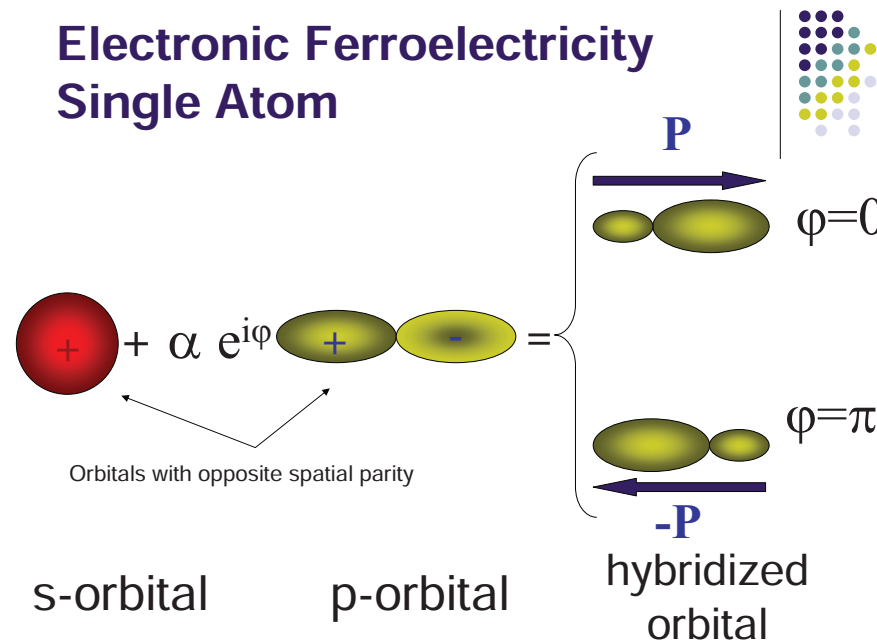
Narrow-band occupancy:

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

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

Electronic Ferroelectricity

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



(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**: $\text{SmB}_6, \text{SmS}, \text{TmSe}, \text{TiSe}_2, \text{TmSe}_{0.45}\text{Te}_{0.55}, \text{LuFe}_2\text{O}_4, \dots$
Typically, a **narrow** band involved. Review: J. Neuenchwander and P. Wachter (1990)
- Suggested relevance for **CMR** manganates and **URu₂Si₂**.
- 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

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

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

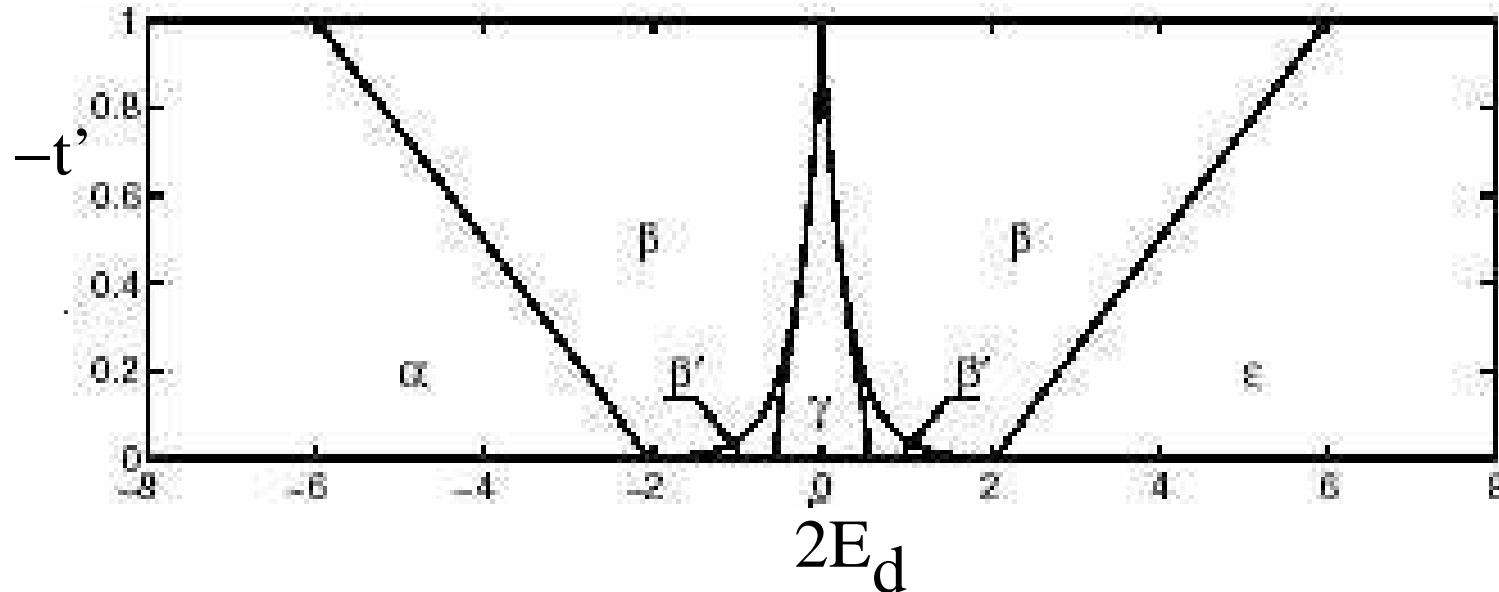
- Different kinds of perturbations break the degeneracy:
 - hopping $t' \ll t$ in the narrow zone.
 - hybridisation: V_0 on-site, V_1 nearest-neighbour (same parity bands), V_2 (opposite parity).

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

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

- 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.

⇒ 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**:

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

- Weak **perturbation**, t' , V_i small:

$$\delta\mathcal{H} = \sum_{\vec{k}} \left\{ t' \epsilon_{\vec{k}} d_{\vec{k}}^\dagger d_{\vec{k}} + V_{\vec{k}} c_{\vec{k}}^\dagger d_{\vec{k}} + V_{\vec{k}}^* d_{\vec{k}}^\dagger c_{\vec{k}} \right\}, \quad V_{\vec{k}} = \begin{cases} V_0 + V_1 \epsilon_{\vec{k}}, & \text{same parity bands,} \\ iV_2 \lambda_{\vec{k}}, & \text{opposite parity,} \end{cases}$$

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

- 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 [...])

- **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 $E_d = 0.4$.

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

- stabilised by the perturbation:

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

–Solid lines: effect of t'

bottom to top: $t' = -0.04, -0.0565, -0.07$

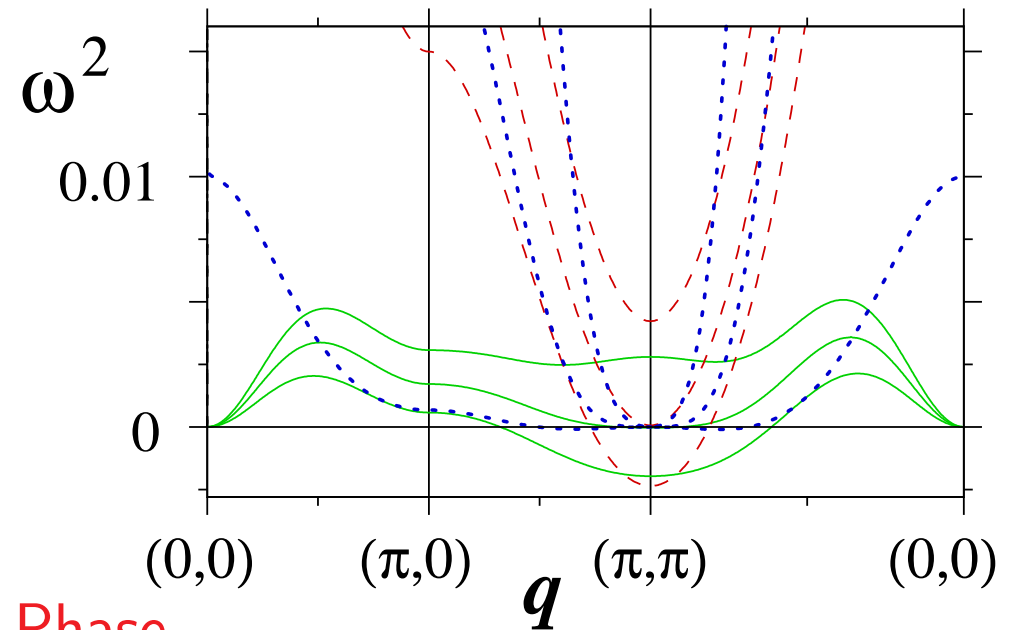
–Dashed lines: effect of V_0

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 $\vec{q} = (0, 0)$

$$d_i \rightarrow d_i \exp(i\varphi) \text{ globally}$$

- Effect of $V_2 \neq 0$: Goldstone mode at $\vec{q} = (\pi, \pi)$

$$d_i \rightarrow d_i \exp(\pm i\varphi) \text{ in a checkerboard pattern, globally}$$

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:** $t'_{cr}(E_d)$

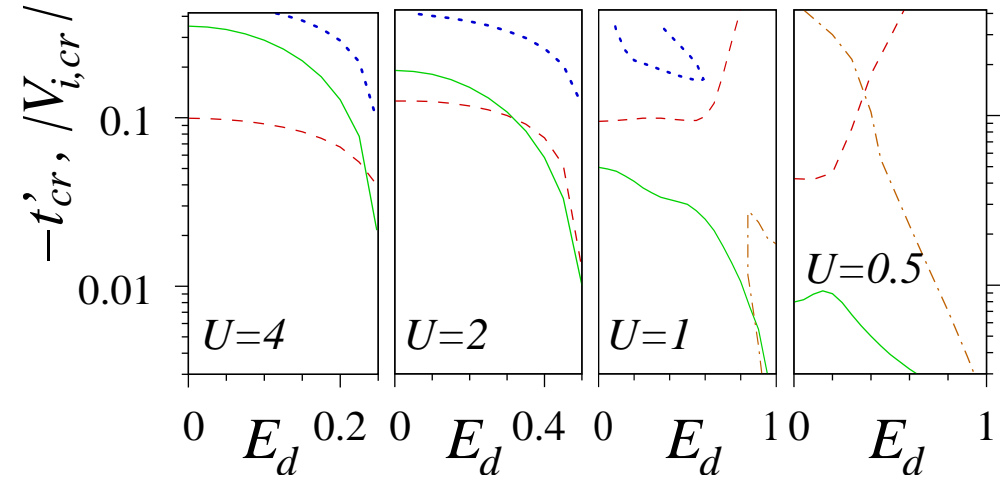
– **Dashed lines:** $V_{0,cr}(E_d)$

– **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**:

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

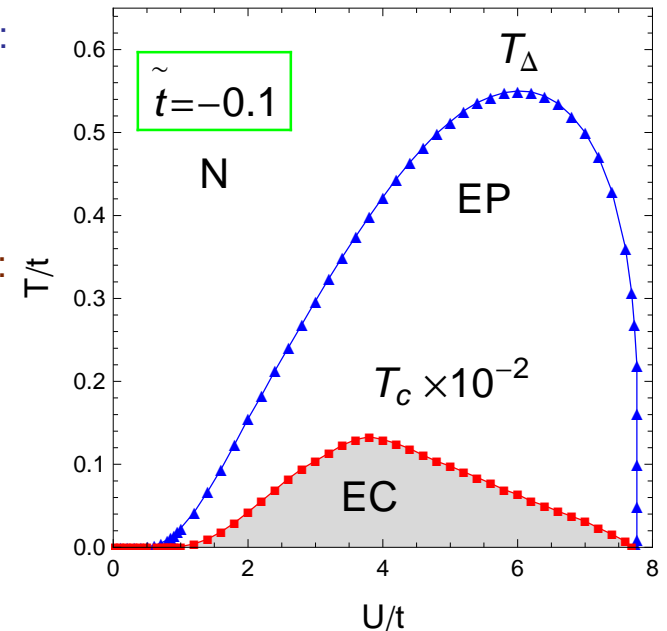
- What happens at $T_C < T < T_\Delta$? **V. A. Apinian & T. K. Kopec, 2013:**

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

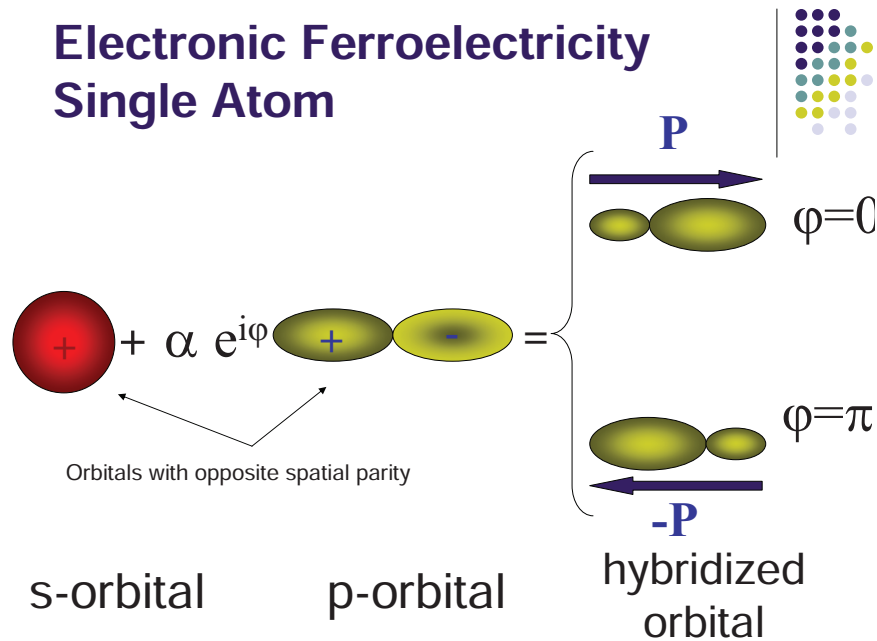
$T < T_C$ – both Δ_i and ϕ_i ordered, **excitonic insulator**.

$T_C < T < T_\Delta$ – 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)

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

- Perturbation terms allowed for **opposite-parity** bands:

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

- **Polarisation density:**

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

$\vec{\mu}$ – 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}_{int} – created by $\vec{P}(r)$

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

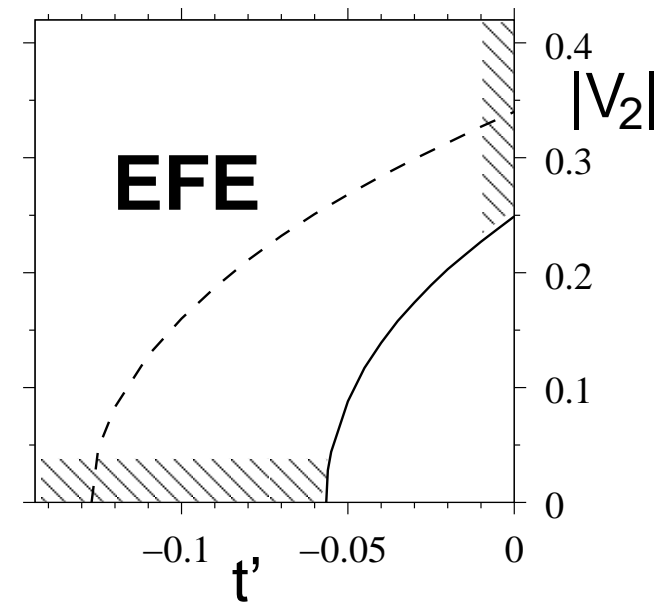
$$\int \vec{E}_{\text{int}} \cdot \vec{D} dV = 0 \Rightarrow -\frac{1}{2} \int_{(\text{sample})} \vec{E}_{\text{int}} \cdot \vec{P} dV = \frac{1}{8\pi} \int_{(\text{entire space})} E_{\text{int}}^2 dV$$

– 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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- ◇ The Israeli Ministry of Absorption.

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** $\Delta = (V_0/N) \sum_{\vec{k}} \langle a_{\vec{k}}^\dagger b_{\vec{k}} \rangle$, **gap equation** for $T = 0$:

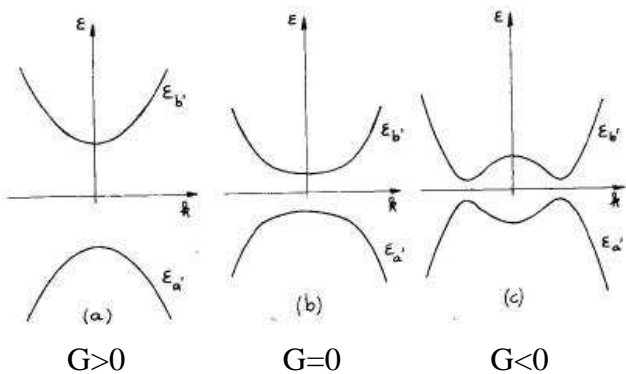
$$1 = \frac{V_0}{2N} \sum_{\vec{k}} \frac{1}{\left\{ \frac{1}{4} [\varepsilon_a(\vec{k}) - \varepsilon_b(\vec{k})]^2 + \Delta^2 \right\}^{1/2}} \Rightarrow \Delta = \Delta(G) \Rightarrow T_c(G) \sim \Delta(G)/k_B$$

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

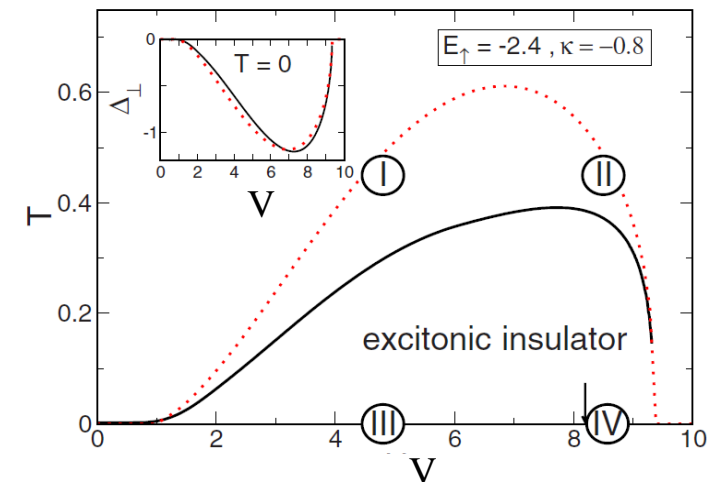
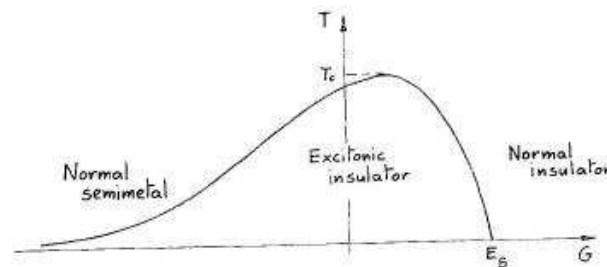
- Hybridised bands:**

“The Snail:”

slave boson MF (**dots** Hartree–Fock):



Critical temperature, $T_c(G)$



\uparrow B. Zenker *et al.* (2010a)