Excitonic insulator phase in the extended Falicov-Kimball model

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We re-examine the three-dimensional spinless Falicov-Kimball model with dispersive f electrons at half-filling, addressing the dispute about the formation of an excitonic condensate. To this end, we work out a slave-boson (SB) functional integral representation of the suchlike extended Falicov-Kimball model (EFKM) that preserves the $SO(2)\otimes U(1)^{\otimes 2}$ invariance of the action. We find a spontaneous pairing of c electrons with f holes, building an excitonic insulator (EI) state at low temperatures, also for the case of initially non-degenerate orbitals.

1 Motivation

Experiment

- although EI was predicted half a century ago a conclusive experimental proof largely failed, actual materials are numbered
- first promising candidate: pressure sensitive $TmSe_{0.45}Te_{0.55}$ analyzed by [Bucher, Steiner and Wachter, PRL 67, 2717 (1991)]

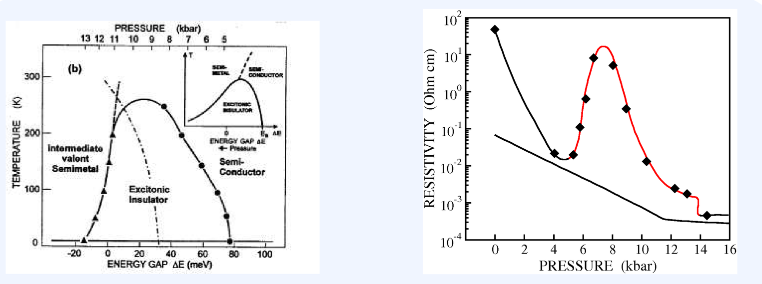

Fig. 1: (a): Phase diagram of $TmSe_{0.45}Te_{0.55}$ deduced by resistivity measurements. Inset: Phase diagram suggested by W. Kohn (b): Resistivity as a function of pressure at 300 K (lower curve) and at 4.2 K (upper curve).

- angle resolved photoemission spectra of Ta_2NiSe_5 reveal evidence for an EI
- in $1T$-$TiSe_2$ the EI phase was invoked as the driving force for the charge-density-wave transition

Theory

- investigation of Falicov-Kimball-type (f-c-electron) models offers a promising route toward the theoretical description of the EI scenario

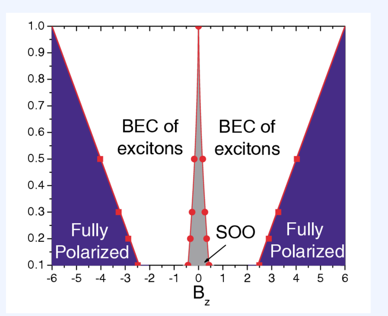

Fig. 2: Ground state phase diagram of the EFKM mapped onto an asymmetric Hubbard model computed with constrained path Monte Carlo by [Batista, Gubernatis, Bonča and Lin, PRL 92, 187601 (2004)].

2 Model

$$H = E_c\sum_i c_i^\dagger c_i + t_c\sum_{\langle i,j\rangle} c_i^\dagger c_j + E_f\sum_i f_i^\dagger f_i + t_f\sum_{\langle i,j\rangle} f_i^\dagger f_j + U\sum_i n_{ic}n_{if}$$

- orbital flavor is represented by a pseudo-spin $\rightsquigarrow$ asymmetric Hubbard model

$$H = \sum_{i\sigma} E_\sigma a_{i\sigma}^\dagger a_{i\sigma} + \sum_{\langle i,j\rangle}\mathbf{a}_i^\dagger\,\underline{t}\,\mathbf{a}_j + U\sum_i n_{i\downarrow}n_{i\uparrow}$$

using a spinor representation

$$\mathbf{a}_i = \begin{pmatrix} a_{i\uparrow}\\ a_{i\downarrow}\end{pmatrix},\quad \mathbf{a}_i^\dagger = (a_{i\uparrow}^\dagger, a_{i\downarrow}^\dagger),\quad \underline{t} = \begin{pmatrix}\kappa & 0\\ 0 & 1\end{pmatrix},$$

with $\kappa = t_f/t_c = t_\uparrow/t_\downarrow$ and $t_c = t_\downarrow = 1$

- EI order parameter:

$$\langle c^\dagger f\rangle \rightsquigarrow \Delta_\perp = \frac{U}{N}\sum_i\langle a_{i\uparrow}^\dagger a_{i\uparrow}\rangle$$

- ground state phase diagram determined within Hartree-Fock agrees in 2D even quantitatively with the Monte Carlo data [Farkašovský, PRB 77, 155130 (2008)]
- scalar slave-boson approach fails to describe an EI phase, when the orbitals are non-degenerate [Brydon, PRB 77, 045109 (2008)] in contrast to Monte Carlo and Hartree-Fock results

3 SO(2)-inv. SB method

[Zenker, Ihle, Bronold, and Fehske, PRB 81, in press (2010); arXiv:0912.2854]

- enlarge Hilbert space (fermions $\rightarrow$ pseudo-fermions and auxiliary bosons) in order to linearize the interaction term

$\lvert 0_i\rangle$	$\rightarrow e_i^\dagger\lvert\mathrm{vac}\rangle$,	empty
$\lvert 2_i\rangle$	$\rightarrow \tilde a_{i\uparrow}^\dagger\tilde a_{i\downarrow}^\dagger d_i^\dagger\lvert\mathrm{vac}\rangle$,	double
$\lvert\sigma_i\rangle$	$\rightarrow \sum_\rho \tilde a_{i\rho}^\dagger p_{i\rho\sigma}^\dagger\lvert\mathrm{vac}\rangle$	single

- unphysical states are eliminated by two sets of local constraints

$$C_i^{(1)} = e_i^\dagger e_i + 2\,\mathrm{Tr}\,\underline{p}_i^\dagger\underline{p}_i + d_i^\dagger d_i - 1 = 0\,,$$
$$\underline{C}_i^{(2)} = \tilde{\mathbf{a}}_i\otimes\tilde{\mathbf{a}}_i^\dagger + 2\underline{p}_i^\dagger\underline{p}_i + d_i^\dagger d_i\,\underline{\tau}_0 - \underline{\tau}_0 = 0$$

- transform Lagrange multipliers into Bose fields, restrict phase transformation to $U(1)^{\otimes 2}$ $\rightsquigarrow SO(2)\otimes U(1)^{\otimes 2}$ invariance of action satisfied
- exploit gauge freedom to remove 3 phases of the 6 (complex) Bose fields in radial gauge
- fermions can be integrated out $\rightsquigarrow$ grand canonical partition function is represented as functional integral over Bose fields only

$$Z = \int D[e]D[p_0]D[p_x^*,p_x]D[p_y^*,p_y]D[p_z]D[d^*,d]\,D[\lambda^{(1)}]D[\lambda_0^{(2)}]D[\vec\lambda^{(2)}]\,\mathrm{e}^{-S}$$

with effective bosonic action ($\underline{p}\leftrightarrow(p_0,\vec p)$)

$$S = \int_0^\beta d\tau\Big\{\sum_i\Big[-\lambda_i^{(1)} + \lambda_i^{(1)}e_i^2 + \sum_\mu(\lambda_i^{(1)} - \lambda_{i0}^{(2)})|p_{i\mu}|^2 - p_{i0}(\vec p_i^* + \vec p_i)\vec\lambda_i^{(2)} - i\vec\lambda_i^{(2)}(\vec p_i^*\times\vec p_i) + (\lambda_i^{(1)} + U - 2\lambda_{i0}^{(2)})|d_i|^2 + p_{iz}^*\partial_\tau p_{ix} + p_{iy}^*\partial_\tau p_{iy} + d_i^*\partial_\tau d_i\Big]\Big\} - \mathrm{Tr}\ln\Big\{-G_{\langle ij\rangle,\rho\rho'}^{-1}(\tau,\tau')\Big\}$$

where inverse Green propagator is given by

$$G_{\langle ij\rangle,\rho\rho'}^{-1}(\tau,\tau') = \Big[\Big(-\partial_\tau + \mu - \lambda_{i0}^{(2)}\Big)\delta_{\rho\rho'} - \frac{E_\uparrow}{2}(\underline{\tau}_0 + \underline{\tau}_z)_{\rho\rho'} - \vec\lambda_i^{(2)}\vec\tau_{\rho\rho'}\Big]\delta_{ij}\,\delta(\tau-\tau') - (\underline{z}_i^*\,\underline{t}\,\underline{z}_j)_{\rho\rho',\tau\tau'}(1-\delta_{ij})$$

- SB representation of the partition function is exact in the case of half filling
- at first level of approximation Bose fields are replaced by their time-averaged values and one looks for an extremum of the bosonized action with respect to the Bose and Lagrange multiplier fields,

$$\phi_{i\alpha} = \left(e_i, p_{i0}, \vec p_i, d_i, \lambda_i^{(1)}, \lambda_{i0}^{(2)}, \vec\lambda_i^{(2)}\right)$$

$$\frac{\partial S}{\partial\phi_{i\alpha}} \overset{!}{=} 0 \quad\rightsquigarrow\quad \bar S = S\Big|_{\phi_{i\alpha}=\bar\phi_{i\alpha}}$$

- physically relevant saddle point gives the lowest free energy per site
- consider an uniform solution of the slave-boson parameters ($\{\bar\phi_{i\alpha}\} = \{\bar\phi_\alpha\}$)
- form of the band renormalization term ('bosonic hopping operator') guarantees the correct free-fermion result

$$z^2 = \frac{2p_0^2d^2}{[1-d^2-\frac{1}{2}(p_0+p)^2][1-d^2-\frac{1}{2}(p_0-p)^2]}$$

- EI order parameter and Hartree shift, respectively, are expressed by slave-boson fields

$$\Delta_\perp = Up_0p_\perp\,,$$
$$\Delta_z = 2Up_0p_z$$

- free energy takes the form

$$f[\phi_\alpha] = \lambda^{(1)}(e^2 + p_0^2 + p^2 + d^2 - 1) - 2\lambda_\perp^{(2)}p_0p_\perp - 2\lambda_z^{(2)}p_0p_z + Ud^2 + \frac{1}{\beta N}\sum_{\vec k\nu}\ln\left[1 - n_{\vec k\nu}\right] + \widetilde\mu n\,,$$

with $\lambda_\perp^{(2)} = \pm\sqrt{(\lambda_x^{(2)})^2 + (\lambda_y^{(2)})^2}$, $p_\perp = \mp\sqrt{p_x^2+p_y^2}$, $p = |\vec p|$, $\widetilde\mu = \mu - \lambda_0^{(2)}$

where

$$n_{\vec k\nu} = [\exp\{\beta(E_{\vec k\nu} - \widetilde\mu)\} + 1]^{-1}$$

holds with the quasiparticle energies ($\nu = \pm$)

$$E_{\vec k\nu} = \frac{1}{2}[E_\uparrow + (\kappa+1)z^2\gamma_{\vec k}] + \nu\sqrt{\frac{1}{4}[E_\uparrow + 2\lambda_z^{(2)} + (\kappa-1)z^2\gamma_{\vec k}]^2 + (\lambda_\perp^{(2)})^2}$$

saddle point equations

$$\lambda_z^{(2)} = \frac{1}{2}\frac{p_z}{p_0}\left(\frac{z^2}{2d^2} - \frac{1}{p_0^2-p^2}\right)z^2I\,,\qquad \lambda_\perp^{(2)} = \frac{1}{2}\frac{p_\perp}{p_0}\left(\frac{z^2}{2d^2} - \frac{1}{p_0^2-p^2}\right)z^2I\,,$$

$$U + 2\lambda_\perp^{(2)}\frac{p_\perp}{p_0} + 2\lambda_z^{(2)}\frac{p_z}{p_0} = \frac{2d^2 - p_0^2 + z^2p^2}{2p_0^2d^2}z^2I\,,$$

$$p_0p_z = \frac{1}{2}\frac{1}{N}\sum_{\vec k\nu}\nu m_{\vec k}n_{\vec k\nu}\,,\qquad p_0p_\perp = \frac{1}{2}\frac{1}{N}\sum_{\vec k\nu}\nu M_{\vec k}n_{\vec k\nu}\,,$$

$$d^2 = \frac{1}{2z^2}\left(z^2(2-p_0^2-p^2) + 2p_0^2 - 2p_0\sqrt{z^2(2-p_0^2-p^2) + z^4p^2 + p_0^2}\right),$$

$$p_0^2 = \frac{1}{2} + \frac{1}{2}\sqrt{(1-z^2)(1-4p_0^2p^2)}\,,$$

with $I = (\kappa+1)\frac{1}{N}\sum_{\vec k\nu}\gamma_{\vec k}n_{\vec k\nu} + (\kappa-1)\frac{1}{N}\sum_{\vec k\nu}\nu m_{\vec k}\gamma_{\vec k}n_{\vec k\nu}\,,$

$$m_{\vec k} = \frac{E_\uparrow + 2\lambda_z^{(2)} + (\kappa-1)z^2\gamma_{\vec k}}{\sqrt{(E_\uparrow + 2\lambda_z^{(2)} + (\kappa-1)z^2\gamma_{\vec k})^2 + (2\lambda_\perp^{(2)})^2}}\,,\qquad M_{\vec k} = \frac{2\lambda_\perp^{(2)}}{\sqrt{(E_\uparrow + 2\lambda_z^{(2)} + (\kappa-1)z^2\gamma_{\vec k})^2 + (2\lambda_\perp^{(2)})^2}}$$

4 Results

4.1 Order parameter

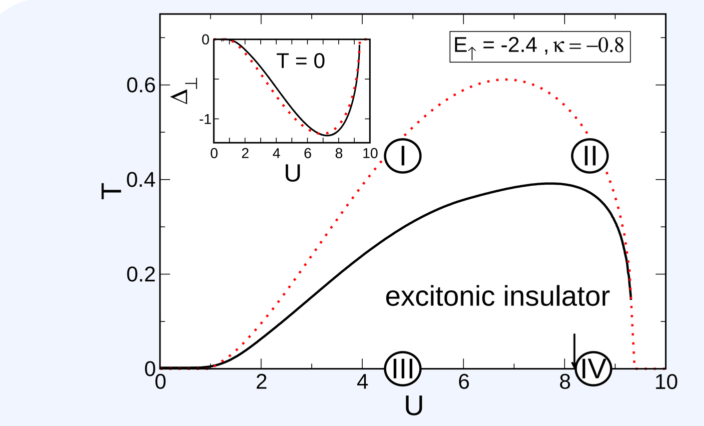

Fig. 3: Stability region of the EI phase in the 3D half-filled EFKM (the arrow marks the critical coupling where the Hartree gap opens). The inset shows the order parameter at zero temperature. Red dotted curves give the Hartree-Fock results for comparison.

- numerical SB semimetal-EI and EI-bandinsulator transition points agree with Hartee-Fock
- $T = 0$ EI order parameter deviates only slightly from the corresponding Hartree-Fock curve
- critical temperature is significantly reduced compared to the Hartree-Fock value

4.2 SB fields

Zero temperature

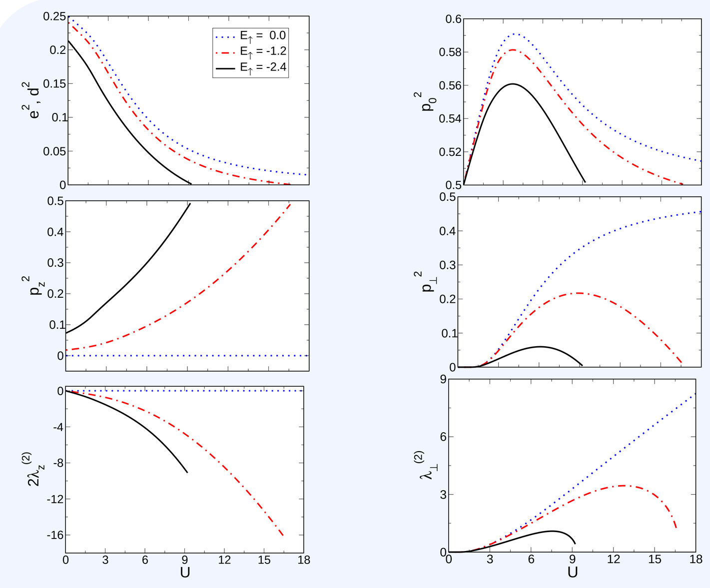

Fig. 4: U-dependence of slave-boson fields and Lagrange parameters for different $E_\uparrow$ at $T = 0$ ($\kappa = -0.8$).

- non-vanishing $p_\perp^2$ and $\lambda_\perp^{(2)}$ indicate EI state
- area of the EI phase is enlarged by reducing the band splitting
- slave-boson band shift $|2\lambda_z^{(2)}|$ increases with increasing U (as Hartree shift)

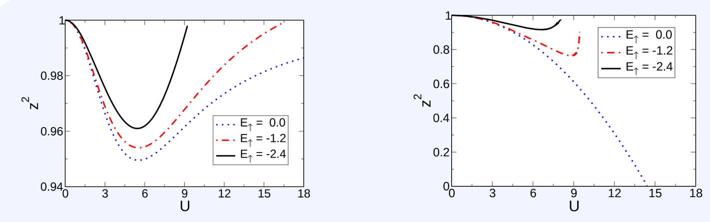

Fig. 5: Band renormalization factors at $T = 0$ within $SO(2)$-invariant (left-hand panel) and scalar (right-hand panel) slave-boson theory. Again, $\kappa = -0.8$.

- small band renormalization, $z^2 \gtrsim 0.95$, explains small deviation of the slave-boson EI order parameter from its Hartree-Fock counterpart
- artificial Brinkmann-Rice transition does not occur within $SO(2)$-invariant SB, but within scalar SB

Finite temperatures

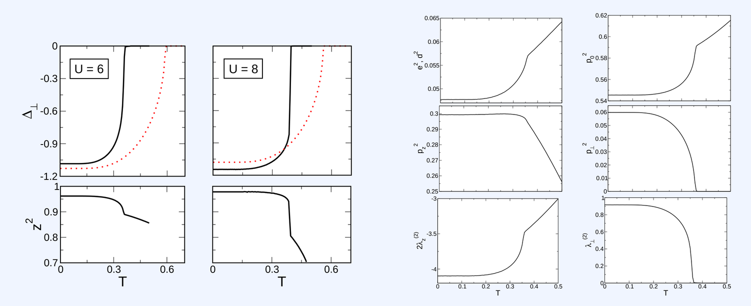

Fig. 7: (a): T-dependence of the EI order parameter $\Delta_\perp$ and of the band renormalization z^2 at fixed Coulomb interaction $U = 6$ (left-hand panels) and $U = 8$ (right-hand panels). Red dotted lines show the corresponding Hartree-Fock data, where $z^2 = 1$. Band-structure parameters are $E_\uparrow = -2.4$ and $\kappa = -0.8$. (b) T-dependence of the various slave-boson fields for $U = 6$, $E_\uparrow = -2.4$, and $\kappa = -0.8$.

- reduction of the critical temperature for the EI-semimetallic/semiconducting phase transition compared to Hartree-Fock
- $p_\perp$ and $\lambda_\perp^{(2)}$ (indicating EI state) are monotonously decreasing functions of the temperature and vanish at critical temperature
- z^2, e^2, d^2, p_0^2, p_z^2, and $2\lambda_z^{(2)}$ exhibit a cusp structure at the critical temperature
- increase of p_0^2 and reduction of p_z^2 indicate a more balanced occupation of f and c electrons

4.3 Partial DOS

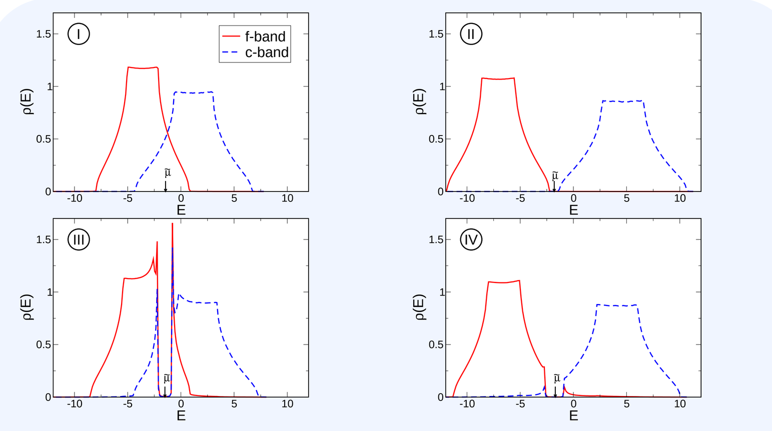

Fig. 8: Partial DOS for f-band (red solid curves) and c-band (blue dashed curves) electrons at the points ($U = 4.8$, $T = 0.45$), (8.5, 0.45), (4.8, 0), (8.5, 0). Band-structure parameters are $E_f = -2.4$ and $t_f = -0.8$.

- high-temperature phase may be viewed as a metal/semimetal (panel I) or a small-gap semiconductor (panel II) in the weak-to-intermediate or strong Coulomb-attraction regime
- at low temperatures, a correlation-induced 'hybridization' gap opens in the weak coupling regime (panel III), the strong enhancement of the DOS at the upper/lower valence/conduction band edges reminds a BCS-like pairing
- zero-temperature DOS in the strong coupling regime (panel IV) evolves from an already gapped high-temperature phase, here, preformed pairs (excitons) may exist and undergo a BEC transition

4.4 Photoemission spectra

[Phan, Becker, and Fehske, arXiv:1003.0765]

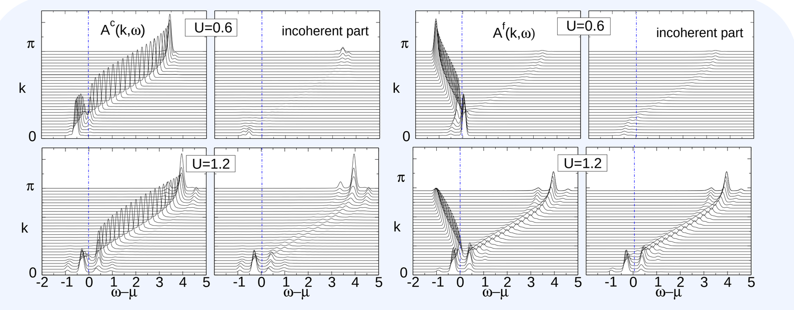

Fig. 9: Wave-number resolved c- (left-hand panels) and f-electron (right-hand panels) single-particle spectral functions for the 1D half-filled EFKM. Band structure parameters are $E_f = -1.7$, $t_f = -0.3$ and $T = 10^{-3}$. The left panels show in each case the total spectra, whereas the right panels give the 'incoherent' contributions only.

- spectral signatures of the BCS-BEC crossover in the 1D EFKM are investigated by the projector-based renormalization method (PRM)
- gap opens, photoemission spectra remains barely the same as in semimetallic state in the weak coupling regime (upper panels)
- significant redistribution of spectral weight from coherent to incoherent part in the strong coupling regime, related to the dissociation of two particle bound states (lower panels)

5 Conclusion

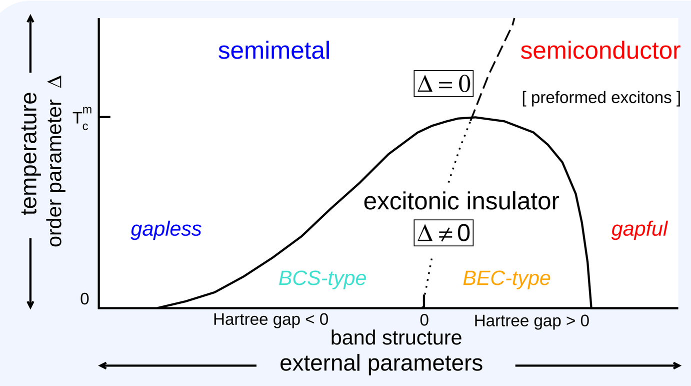

Fig. 10: EI formation and BCS-BEC transition scenario

Our improved slave-boson (PRM) scheme is capable of describing the EI phase in the half-filled EFKM. The agreement of the slave-boson zero-temperature semimetal→EI and EI→bandinsulator transition points with the Hartree-Fock and Monte Carlo values is ascribed to a rather weak band renormalization at $T = 0$. At finite temperature, band renormalization effects due to electronic correlations and particle number fluctuations become important, and, as a result, our slave-boson theory yields significantly lower transition temperatures than Hartree-Fock. The analysis of the f, c spectral functions and (partial) DOS in the EI phase indicate a crossover from a BCS-type condensate to a Bose-Einstein condensate of preformed excitons.

[Mott, Philos. Mag. 6, 287 (1961)]
[Knox, in 'Solid State Physics' (Academic Press, New York, 1963), p. Suppl. 5 p. 100]
[Bronold and Fehske, PRB 74, 165107 (2006)]
[Schneider and Czycholl, EPJ B 64, 43 (2008)]
[Ihle, Pfafferott, Burovski, Bronold, and Fehske, PRB 78, 193103 (2008)]