Topical Review

Excitonic condensation in systems of strongly correlated electrons

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Abstract
The idea of exciton condensation in solids was introduced in the 1960s with the analogy of superconductivity in mind. While exciton supercurrents have been realised only in artificial quantum-well structures so far, the application of the concept of excitonic condensation to bulk solids leads to a rich spectrum of thermodynamic phases with diverse physical properties. In this review we discuss recent developments in the theory of exciton condensation in systems described by Hubbard-type models. In particular, we focus on the connections to their various strong-coupling limits that have been studied in other contexts, e.g. cold atoms physics. One of our goals is to provide a ‘dictionary’ that would allow the reader to efficiently combine results obtained in these different fields.

Keywords: excitonic insulator, Hubbard model, exciton condensation

(Some figures may appear in colour only in the online journal)
semi-metals in mind. More recently the ideas of excitonic condensation were applied to strongly correlated systems described by Hubbard-type models. In their strong-coupling limit, these models lead to spin or hard-core boson problems that have been studied in other contexts. The purpose of this topical review is to summarise the recent work on the excitonic condensation in Hubbard-type models, discuss the corresponding phase diagram, and describe connections to various other models such as the Blume–Emmery–Griffiths [6], bosonic i-J, or bi-layer Heisenberg [7] models. In particular, we aim at providing a ‘dictionary’ for the names of mutually corresponding phases found in different models. We only briefly touch the active field of bi-layer systems in section 5.2 and refer the reader to specialised literature. We completely omit another major direction of the exciton-polariton condensation [8].

1.1. Brief history of the weak-coupling theory of excitonic condensation

In 1961 Mott [3] considered metal-insulator transition in a divalent material (even number of electrons per unit cell) associated with continuous opening of a gap, e.g. due to application of external pressure. He argued that the one-electron picture cannot be correct, that a semimetal with a small concentration of electrons and holes will be unstable if electron-hole interaction is taken into account. Knox [4] approached the transition from the insulator side and argued that if the excitonic binding energy overcomes the band gap, then the system becomes unstable. These proposals were put into a formal theory by Keldysh and Kopcha [9], and by des Cloizeaux [10] who developed a weak-coupling Hartree–Fock theory of excitonic condensation analogous to the BCS theory of superconductivity. The role of the pairing glue is played by the inter-band Coulomb interaction, which favours formation of bound electron–hole pairs, excitons. The early theories used so-called dominant term approximation, which consists in keeping only the density–density intra-band and inter-band terms of the Coulomb interaction. This approximation appears well justified for weakly correlated metals or semiconductors where the pair glues arise from the long-range part of the interaction. It leads to a large manifold of degenerate excitonic states. It is due to this degeneracy that the small and so far neglected exchange and pair-hopping interaction terms may play an important role. Halperin and Rice [11] showed that these terms select one of the four following states as the lowest one: charge-density wave, charge-current-density wave, spin-density-wave, and spin-current-density wave.

In the 1970s Volkov and collaborators showed in a series of articles [12–14] that excitonic condensation in slightly off-stoichiometric systems leads to formation of a uniform ferromagnetic state, although the normal state does not exhibit a magnetic instability. The idea of an excitonic ferromagnet was revived at the beginning of 2000s in the context of hexaborides (section 5.1).

2. Spinless fermions

Before discussing the two band Hubbard model (7), we look at its spinless version

$$H_{EFK} = \sum_{(\mathbf{r})} (t_{ab} a_{a}^\dagger r_{b} + t_{ba} b_{b}^\dagger a_{a}) + \text{h.c.} + \sum_{(\mathbf{r})} (V_{ab} b_{b} a_{a}^\dagger + V_{ba} a_{a}^\dagger b_{b}) + \text{h.c.} + \frac{\Delta}{2} \sum_{\mathbf{r}} (n_{a} - n_{b}^c) + \bar{U} \sum_{\mathbf{r}} n_{a} n_{b}^c$$

(1)

which describes electrons of two flavours (a and b) moving on a lattice and interacting via an on-site interaction. Here, $a_{a}^\dagger$ (or $b_{b}^\dagger$) is an operator creating (annihilating) a fermion of flavour a on lattice site $\mathbf{r}$, $n_{a} = a_{a}^\dagger a_{a}$ is the corresponding local density operator, and analogically for b fermions. We use $\psi_{a}(\mathbf{r})$ and $\psi_{b}(\mathbf{r})$ to refer to the density distribution around the lattice sites in orbitals a and b, respectively, when necessary. The sum $\sum_{\mathbf{r}}$ runs over the nearest neighbour (nn) bonds. Model (1) is usually called the extended Falicov–Kimball model (EFKM), where ‘extended’ implies that both $t_{ab}, t_{ba} \neq 0$. In the original Falicov–Kimball model (FKM) [15], $t_{b} = V_{ab} = V_{ba} = 0$, only the a-electrons can propagate while the ‘heavy’ b-electrons are immobile.¹

Three versions of EFKM have been discussed in the literature in the context of excitonic condensation: (i) FKM with $t_{b} = V_{ab} = V_{ba} = 0$; (ii) EFKM without cross-hopping $V_{ab} = V_{ba} = 0$; and (iii) EFKM with cross-hopping $V_{ab}, V_{ba} \neq 0$. Since the conservation of the charge per flavour plays a central role in excitonic condensation we compare models (i) through (iii) from that perspective. Hamiltonian (1) conserves the total charge for any choice of parameters. Since the corresponding $U(1)$ symmetry is not broken in any phase considered here, we do not mention this symmetry explicitly in the text. The FKM (i) conserves the number of the heavy

¹ More traditionally the mobile and heavy electrons are labelled d and f, respectively.
$b$-electron on each site, which gives rise to a local $U(1)$ gauge symmetry. Finite $t_b$ in (ii) removes the local gauge invariance, but preserves independently the number of $a$- and $b$-electrons, which gives rise to a global $U(1)$ symmetry associated with the arbitrariness of the relative phase of the $a$- and $b$-electrons. Finally, finite cross-hopping in (iii) removes this $U(1)$ symmetry. An important special case of (iii) is a system with a site symmetry that prohibits an on-site $a-b$ hybridisation. As an example, we may consider orbitals $a$ and $b$ of different parity, which implies $V_{ab} = - V_{ba}$. In this case, the $U(1)$ symmetry is not removed completely but is reduced to a discrete $\mathbb{Z}_2$ symmetry reflecting the invariance of (1) under the transformation $a_{i} \rightarrow -a_{i}, \ V_{ab} \leftrightarrow V_{ba}$.

On a bipartite lattice there are additional symmetries. Models (ii) with $t_{fb} > 0$ and $t_{bd} < 0$ can be mapped on each other by $a_i \rightarrow (1)^i a_i$. In the EC phase the map turns ferro-EC order ($t_{fb} < 0$) into an antiferro-EC order ($t_{fb} > 0$). This property will be important for understanding the nature of the excitonic insulator phase in FKM ($t_{fb} = 0$), which lies between the ferro and antiferro phases.

2.1. Falicov–Kimball model

The interest in excitonic condensation in FKM started with the work of Port engen et al [16, 17] who studied the FKM model augmented with the hybridisation between the heavy $b$-electrons and light $a$-electrons $V_{ab} = - V_{ba} \neq 0$. Using the self-consistent mean-field (BCS-like) theory they found solutions with spontaneous on-site hybridisation–excitonic condensate. They showed that in the case when the $a$ and $b$ orbitals are of opposite parity the excitonic condensation gives rise to ferroelectric polarisation. Importantly, their excitonic condensate also existed in the limiting case of $V_{ab} = V_{ba} = 0$, suggesting a new ground state of the original FKM. This result was challenged by subsequent theoretical studies [18–21] of FKM in one and infinite dimensions, which found no spontaneous hybridisation. It was pointed out that the method of Portengen and collaborators missed competing ordered states and the possibility of phase separation. Freericks and Zlatić [22] argued that spontaneous hybridisation in FKM breaks the local $U(1)$ gauge symmetry, associated with the phase of the heavy electron, and thus is prohibited by Elitzur’s theorem [23]. The question is, however, quite subtle as can be illustrated, for example, by the excitonic susceptibility in $d = \infty$ FKM, which has logarithmic singularity at $T = 0$ [24]. The insight into the issue can be provided by going into EFKM. The mapping between $t_{bd} > 0$ and $t_{bd} < 0$ models (on bipartite lattice) implies that if there is an (ferro) ordered state in the limit $t_b \rightarrow 0^-$, then there must an antiferro order in the limit $t_b \rightarrow 0^+$, and thus FKM is an unstable point between these two phases.

2.2. Extended Falicov–Kimball model

2.2.1. Strong-coupling limit. An alternative way to EC order in FKM is to make both fermion species mobile, i.e. introducing EFKM. Batista [5] studied EFKM at half-filling in the strong coupling limit $\tilde{U} \gg t_{ab}, t_{ba}$ as an alternative way to electronic ferroelectricity. Introducing pseudo spin variables

$$
\tau_i^\uparrow = a_i^\dagger b_i^\dagger + a_i^\dagger a_i - b_i^\dagger b_i,
\tau_i^\downarrow = a_i^\dagger a_i - b_i^\dagger b_i,
$$

he arrived at a strong-coupling effective Hamiltonian, which for $V_{ab} = V_{ba} = 0$ reads

$$
\mathcal{H}_{\text{eff}} = \frac{1}{2} \sum_{(ij)} (K_i \tau_i^\uparrow \tau_j^\uparrow + K_\downarrow (\tau_i^\uparrow \tau_j^\downarrow + \tau_i^\downarrow \tau_j^\uparrow)) + \frac{\Delta}{2} \sum_i \tau_i^\downarrow. \tag{3}
$$

$\mathcal{H}_{\text{eff}}$ acts in the low-energy space built of states with singly occupied sites only. The coupling constants to the lowest order in $t/\tilde{U}$ read $K_\uparrow = (t_{ab}^2 + t_{ba}^2) / \tilde{U}$ and $K_\downarrow = 2 t_{bd} / \tilde{U}$. Labelling the two local states $| \tau_i \rangle = a_i^\dagger |0\rangle$ and $| \tau_i \rangle = b_i^\dagger |0\rangle$ the structure of the $S = 1/2$ XXZ-model in an external field along the $z$-axis is apparent. Note that while $K_\downarrow$ can be both positive and negative, $K_\uparrow$ is always positive. There is a well-known exact mapping of (3) onto the model of spinless hard-core bosons [26]

$$
\tilde{\mathcal{H}}_{\text{eff}} = \epsilon \sum_i n_i + K_\uparrow \sum_{(ij)} (d_i^\dagger d_j + \text{h.c.}) + 2 K_\downarrow \sum_{(ij)} n_i n_j, \tag{4}
$$

with $\epsilon = \Delta - z K_\| (z$ is the number of nearest neighbours). The operator $d_i^\dagger$ ($d_i$) creates (annihilates) a boson on site $i$, and $n_i = d_i^\dagger d_i$ is the local density operator. The physical states are constrained to those containing zero or one bosons per site. In this language, the $| \tau_i \rangle$ is the local bosonic vacuum and $| \tau_i \rangle = d_i^\dagger | \tau_i \rangle$ is a state with one boson. The transverse spin coupling translates into bosonic hopping, the longitudinal coupling translates into nearest-neighbour repulsion, and the magnetic field translates into bosonic chemical potential. Moving between the spin and boson formulation has been traditionally used to allow convenient treatment of various models [27, 28].

The model (3) and (4) is much studied in the context of cold atoms on optical lattices. Its $T = 0$ phase diagram for a

$^2$Changing sign on one sublattice of the bipartite lattice.
square lattice is shown in figure 2. Besides the trivial phases obtained for large $|\Delta|$, which correspond to orbitals of one flavour being filled and the other being empty (saturated spin polarisation along the $z$-axis), there are two more phases. The solid phase ($z$-axis Néel antiferromagnet) favoured by $K_0$, and the superfluid phase (xy magnetic order) favoured by $K_c$. The solid phase is characterised by checker-board arrangement of sites with occupied $a$ and $b$ orbitals. The solid phase is connected to the ground state of half-filled FKM [29]. The superfluid phase is characterised by a finite expectation value $\langle d_i \rangle$, i.e. it can be described as a condensate of the $d$-bosons. In the language of the EFKM this means a finite $\langle a_i^\dagger b_i \rangle$ expectation value characterising the excitonic condensate. To understand the phase diagram in figure 2 one may start from the familiar point of the Heisenberg antiferromagnet, $\Delta = 0, K_c = K_0$. Upon application of a magnetic field ($-\Delta$) the ordered moments pick an arbitrary perpendicular orientation with a small tilt in the field direction (superfluid). For $|K_c| > K_0$ the in-plane order is obtained without the external field, whereas for $|K_c| < K_0$ a finite field is needed to destroy the Ising (solid) order.

The solid and superfluid phases have quite different properties. The solid phase breaks the discrete translational symmetry. The superfluid phase breaks continuous $U(1)$ symmetry associated with the phase of $d_i$ varying between sublattices. Therefore in two dimensions only solid long-range order exists at finite temperature [30], while the superfluid phase has the form of the Kosterlitz–Thouless phase [31, 32]. The mismatch between the symmetries of solid and superfluid phases implies a first-order transition between them or existence of an intermediate supersolid phase where both the orders are present. The stability of the supersolid phase is a much studied question in the context of another model (4) and its generalisations. Investigations on cubic lattice in two [25] and three [33] dimensions found the first-order transition scenario to be realised. However, a robust supersolid phase was found on the triangular lattice [34–37].

Existence of a supersolid on a triangular lattice is related to the fact that the solid and the superfluid adapt differently to the geometrical frustration. For further reading on hard-core boson we refer the reader to specialised literature.

We conclude this section by considering the effect of cross-hopping. Non-zero cross-hopping $V_{ab}$, $V_{ba}$ in (1) breaks the $U(1)$ symmetry of EFKM and generates additional terms in Hamiltonian (4) [5]. These are of two types. First, the ‘correlated-hopping’ terms of the form $(d_i + d_j)(2n_j - 1)$ and of the order $(V_{ab} n_a - V_{ba} n_b)\tilde{U}$. Summed over the nn sites their contribution can be split into a source field for $d$-bosons, proportional to the mean boson density $(n)$, and a coupling of $d_i + d_j$ to the density fluctuations. With a finite source field the situation is analogous to a ferromagnetic transition in an external magnetic field, i.e. the sharp transition is smeared out and diminishes completely if the external field is comparable or larger than the internal (Weiss) fields. In some symmetries the contributions of individual neighbours to the source fields add up to zero, e.g. in the case of $a$- and $b$-orbitals in (1) of different parities [5]. In this case, phase transition is still possible. However, the second type of terms generated by cross-hopping of the form $d_i d_j$ and $d_i^\dagger d_j^\dagger$ and of the order $V_{ab}V_{ba}\tilde{U}$ reduce the symmetry of the system to $Z_2$. Depending on the sign of this term the order parameter $\langle d_i^\dagger \rangle = \langle a_i^\dagger b_i \rangle$ is either real or imaginary. The meaning of the phase of the order parameter $\langle a_i^\dagger b_i \rangle$ was discussed by Halperin and Rice [11]. In case of real orbitals $\eta_0(r)$ and $\eta_0(r)$, real $\langle a_i^\dagger b_i \rangle$ gives rise to charge density modulation while imaginary $\langle a_i^\dagger b_i \rangle$ gives rise to periodic current pattern. In a model with inversion symmetry about the lattice sites studied elsewhere [5], $V_{ab} = -V_{ba}$ leads to real $\langle a_i^\dagger b_i \rangle$ implying an Ising-like ferroelectric transition. A solution with purely imaginary $\langle a_i^\dagger b_i \rangle$ was reported in $d = 1$ EFKM by Sarasua and Continentino [38] for a somewhat artificial choice of purely imaginary $V_{ab} = V_{ba}$.

2.2.2. Intermediate and weak coupling. Studies using various techniques and in different dimensionalities, reviewed below, lead to the conclusion that the behaviour observed in the strong-coupling limit of EFKM extends to the intermediate coupling and connects to the weak-coupling regime. To be able to compare these results we have to understand how they are interpreted, in particular in $d = 1.2$. Following the Mermin–Wagner theorem [30], the superfluid, which breaks continuous symmetry, is characterised by long-range order at finite $T$ in $d = 3$, by long-range order at $T = 0$ and algebraic correlations at finite $T$ in $d = 2$, and by algebraic correlations at $T = 0$ in $d = 1$. In the Ising-like solid phase a long-range order exists at finite $T$ in $d = 2.3$, while in $d = 1$ there is a long-range order at $T = 0$. The onset of algebraic correlations was used as a criterion to define the transition to the superfluid phase in the $d = 1$, $T = 0$ Monte-Carlo studies [39]. Mean-field techniques however do not distinguish dimensionalities.
and long-range order is obtained even if it does not exist in the exact solution. In these cases one compares the onset of algebraic correlations in more rigorous methods with the onset of mean-field long-range order.

Using the constrained path Monte-Carlo method Batista et al [39] obtained the $T = 0$ phase diagram of the $d = 1,2$ model shown in figure 3. In fact, Farkašovský [40] showed that self-consistent Hartree–Fock method reproduces the Monte-Carlo phase diagram in $d = 2$ remarkably well (figure 3) and extended the study to $d = 3$ and $t_b \ll 1$. A similar HF phase diagram was obtained by Schneider and Czycholl [41] for semieliptic densities of states ($d = \infty$ Bethe lattice). In both studies, the excitonic phase persists in the limit $|t_b| \to 0$ for a certain range of $\Delta$. Due to the symmetry of EFKM under $t_b \leftrightarrow -t_b$, $t_b = 0$ is an unstable fixed point between the ferro and antiferro excitonic phases. Interestingly, for $t_b \sim 0$ Farkašovský [40] also finds a phase where both the solid and excitonic orders are present simultaneously—a supersolid phase. This result has not been confirmed by other studies yet and the supersolid phase was shown to be unstable in the strong-coupling limit [25]. Moreover, existence of a phase with finite $\langle a^\dagger b \rangle$ at $t_b = 0$, which smoothly connects to both $t_b > 0$ and $t_b < 0$ violates the Elitzur’s theorem [23].

2.2.3. BCS-BEC crossover. The similarity of the strong- and weak-coupling phase diagrams leads to an interesting question of how the physics described by the the hard-core bosons (3) evolves into the Hartree–Fock physics of fermions in (1). A similar question is known as the BCS-BEC crossover in the context of superconductivity or crossover between Slater and Heisenberg antiferromagnet. The issue of BCS-BEC crossover in the excitonic phase of half-filled EFKM was studied by several authors using random phase approximation (RPA) [42], slave-boson [43, 44], projective renormalisation [45], variational cluster [46], exact diagonalisation [47], and density-matrix renormalisation group [48] techniques. The different methods provide quite a consistent picture of the phase diagram with the provision that long-wavelength fluctuations of the order parameters are ignored by some of the mean-field methods, which therefore describe the critical phase in $d = 1$, $T = 0$ and the Kosterlitz–Thouless phase in $d = 2$ as phases with true long-range order.

The BCS versus BEC question can be formulated as: are there long-lived excitons above $T_c$ that condense at the transition? The typical $T–U$ phase diagram of half-filled EFKM with exciton condensate is shown in figure 4. Following another work [42] we introduce a boson creation operator $d_q^\dagger = \frac{1}{\sqrt{N}} \sum_{k=0}^{N-1} b_{q+k} b_k$ and the corresponding propagator $\chi^\dagger_q (\omega) = \langle \langle d_{q^\dagger} d_{q^\dagger} \rangle \rangle_{\omega}$. The excitonic transition proceeds differently on the semimetal and semiconductor sides. In semimetal, the particle-hole continuum extends to zero energy in a finite $q$-region of the Brillouin zone. While long-lived excitons may exist in some other parts of the Brillouin zone (figure 5(b)), they are not the lowest energy excitations and the excitonic transition follows the BCS scenario. In semiconductor, the excitonic band in the two-particle spectrum extends throughout the whole Brillouin zone (figures 5(c)–(g)) and lies below the particle-hole continuum. $B_{q}(\omega)$, the coherent part of $\text{Im} \chi^\dagger_{q^\dagger} (\omega)$, can be approximated as

$$B_{q}(\omega) = -nZ_\xi(q) \delta(\omega - \omega_\xi(q)),$$

where $Z_\xi(q)$ is the weight of the excitonic quasiparticle and $\omega_\xi(q)$ is its dispersion. Note that both $Z_\xi(q)$ and $\omega_\xi(q)$ depend on temperature. The number of excitons with crystal-momentum $q$ is given by

$$N_\xi(q) = Z_\xi(q) n_{\text{BE}}(\omega_\xi(q)),$$
Figure 5. Evolution of the exciton quasiparticle weight $Z_X(q)$, obtained with RPA, across the semimetal–semiconductor crossover. On the metallic side (a)–(c) non-zero $Z_X(q)$ exists only in a part of the Brillouin zone. On the semiconductor side (e)–(g), $Z_X(q)$ is finite everywhere in the Brillouin zone and approaches $Z_X(q) = 1$ in the strong-coupling limit (g). (a) Corresponds to a weakly coupled semimetal with no excitonic quasi-particle residues anywhere in the Brillouin zone. (b)–(f) Correspond to the points 1–4 of figure 4. Adapted with permission [42]. Copyright 2012 by the American Physical Society. (a) $\tilde{U} = 0.50, T/T_{\text{max}} = 1.108$. (b) $\tilde{U} = 3.08, T/T_{\text{max}} = 0.679$. (c) $\tilde{U} = 5.07, T/T_{\text{max}} = 1.108$. (e) $\tilde{U} = 5.50, T/T_{\text{max}} = 0.679$. (g) $\tilde{U} = 50.0, T/T_{\text{max}} = 1.108$.

where $n_{\text{BE}}(\omega)$ is the Bose–Einstein (BE) function. The excitonic transition in a semiconductor is connected with the minimum of $\omega_X(q)$ going to zero at $T_c$ and the resulting divergence of $N_X(q)$, figure 6.

The semimetal and semiconductor side of the phase diagram appear to be qualitatively different. It is therefore instructive to see that the evolution from a semimetal to a semiconductor with increasing $\tilde{U}$ is in fact smooth, although they differ by the presence of the excitonic band. The key observation is that the parts of the exciton band that split off the particle-hole continuum carry little spectral weight (c)–(f) and are negligible due to the spatial separation of the excitonic insulator. While the particle-hole gap grows linearly with increasing $\tilde{U}$, the exciton band remains located around $\Delta$ with its width approaching the $t_{ph}/\tilde{U}$ scaling. If the particle-hole gap is sufficiently large, then the excitons are no longer dressed with the particle-hole excitations and thus $Z_X(q) \approx 1$ everywhere in the Brillouin zone. The exciton dynamics in this limit is described by purely bosonic Hamiltonian (4).

3. Two-band Hubbard model ($S = 1/2$ fermions)

The two-band Hubbard model (2BHM) generalises EFKM (1) to include electron spin. In absence of external magnetic field and spin-orbit coupling the one-particle part $H_i$ consists of two identical copies of the corresponding terms in EFKM. The interaction part $H_{\text{int}}$ is richer.

$$H_{\text{BH}} = H_i + H_{\text{int}}$$

$$H_i = \sum_{i,\sigma} (\epsilon_i n_i^{\dagger} n_i + V_i n_i^{\dagger} d_i^{\dagger} d_i + \text{h.c.})$$

$$+ \sum_{i,\sigma} (V_{i,\sigma} b_i^{\dagger} d_i + V_{i,\sigma} d_i^{\dagger} b_i + \text{h.c.})$$

$$+ \frac{\Delta}{2} \sum_{i,\sigma} (n_i^{\dagger} - n_i)$$

$$H_{\text{int}} = U \sum_i (n_i^{\dagger} n_i^{\dagger} n_i n_i + n_i^{\dagger} n_i) + U' \sum_{i,\sigma} n_i^{\dagger} n_i$$

$$- J \sum_{i,\sigma} (n_i^{\dagger} n_i + a_i^{\dagger} a_i b_i b_i^{\dagger} + \text{h.c.})$$

$$+ J' \sum_i (a_i^{\dagger} b_i d_i^{\dagger} b_i + \text{h.c.}),$$

with the notation $\sigma = -\sigma$. There are two general set-ups in which EC in 2BHM have been studied: a lattice of two-orbital atoms and a bi-layer Hubbard model system.

In the bi-layer model the orbitals $a_i$ and $b_i$ are assumed to be spatially well separated and the exchange-interaction is vanishingly small $J' / J \approx 0$. This is equivalent to the dominant-term approximation used extensively for long-range interaction [11]. Similarly the on-site and inter-site inter-layer tunnelling, $V_{a,b} V_{b,a} \approx 0$, is negligible due to the spatial separation of layers.

In two-orbital atoms, the overlap of (orthogonal) $a$ and $b$ orbitals gives rise to a sizeable ferromagnetic Hund’s exchange...
The on-site excitonic condensation is described by the exciton–exciton–electron–hole (EEEH) model, shown in figure 7, and, in particular, to a finite local element \( k \) one obtains a band insulator with symmetric \( 2\text{BHM} \) with \( 3 \text{d} \) bands (semi-elliptic density of states) for \( J = J_U = U/4; U = U - 2J \) (red) and \( J = J = 0; U = U \) (blue) obtained by Werner and Millis [49]. The dotted red line marks the HS–LS crossover. The vertical blue line for \( J = 0 \) corresponds to a model with a six-fold degenerate atomic ground state. \( U \) and \( \Delta \) are expressed in the units of bandwidth \( W \).

\[ J \text{ and pair hopping } J'. \text{ The on-site } a-b \text{ hopping vanishes either by symmetry or can be eliminated by a basis transformation. The cross-hopping } V_{ab}, V_{ba} \text{ is in general non-zero; however, in materials with high symmetry it may vanish as well}^4. \]

### 3.1. Normal state

Before discussing the ordered phases we briefly summarise the basic properties of half-filled 2BHM without broken symmetry. Its physics at strong and intermediate coupling is controlled by competition between the Hund’s coupling \( J \) and the crystal-field splitting \( \Delta \) [49, 50]. Large \( \Delta \) favours the singlet low-spin (LS) state, while large \( J \) favours the triplet high-spin (HS) state (figure 7). For finite \( J \) the phase diagram, shown in figure 7, contains three regions: HS Mott insulator connected to the limit \( U \gg W \), with \( W \) standing for the bare bandwidth, and \( J \gg \Delta \), LS band insulator connected to the limit \( \Delta \gg W, U \) and a metal connected to the non-interacting limit and \( \Delta < W \). The low-energy physics deep in the Mott phase is described by the \( S = 1 \) Heisenberg model with anti-ferromagnetic interaction. The band insulator far away from the phase boundaries is a global singlet separated by a large gap from the excited states. In the vicinity of the HS–LS crossover both LS and HS states have to be taken into account. The physics arising in this parameter region is the subject of subsequent sections.

The physics of the \( J, J' = 0 \) model is different. This setting corresponds to a bi-layer system where one would typically choose \( U > U' \). In this case, the low-energy physics for \( W \ll U \) and \( \Delta \ll (U - U') \) is described by the \( S = 1/2 \) bi-layer Heisenberg model, while for \( \Delta \gg (U - U') \) one obtains a band insulator. The region \( \Delta \approx (U - U') \) is described by the exciton-\( t-J \) model, discussed later. The choice \( U = U' \), shown in figure 7, is anomalous in the sense that the bi-layer Heisenberg region is absent and the region \( \Delta \approx 0 \) corresponds to the exciton-\( t-J \) model with two exciton types.

The low-energy physics of the metallic phase is sensitive to Fermi surface nesting, particularly for weak coupling. Nesting plays an important role in some popular models, e.g. the Fermi surfaces derived from \( a \) and \( b \) bands are perfectly nested on cubic lattice both in \( d = 2 \) and \( d = 3 \).

#### 3.2. Excitonic order parameter

Excitonic condensation is characterised by a spontaneous coherence between the \( a- \) and \( b \)-electrons in (7), i.e. appearance of matrix elements of the form

\[ F_{ii}^{\text{cov}} = \langle a^+_\alpha b_{\beta \sigma} \rangle \quad \text{and} \quad F_{kk}^{\text{cov}} = \langle a^+_k b_{k \sigma} \rangle. \]

These can be generally complicated objects characterised by translational symmetry, internal structure, and spin symmetry.

We consider only ordered phases with single-\( \mathbf{q} \) translational symmetry, \( F_{kk} \sim \delta_{\mathbf{k} + \mathbf{q}} \). On the square lattice we encounter only ferro-EC, \( \mathbf{q} = 0 \), and antiferro-EC, \( \mathbf{q} = (\pi, \pi) \) states, which correspond to uniform \( F_{ii} \) and staggered \( F_{ii} \sim (-1)^{\mathbf{r}} \), respectively.

The internal structure describes the behaviour of the above matrix elements as a function of the reciprocal vector \( \mathbf{k} \) or the relative position \( \mathbf{R} \sim \mathbf{R}_c \). The internal structure reflects the symmetry of the pairing interaction. Isotropic pairing leads to isotropic \( F_{kk} \) and, in particular, to a finite local element \( F_{ii} \). Since this is the case for local Hubbard interaction (7) we can use the local element \( F_{ii} \) as an order parameter for the exciton condensation. The spatial decay of the \( a-b \) coherence can be quantified by a correlation length \( r_{\text{coh}} \) defined as

\[ r_{\text{coh}} = \left( \frac{\sum_k |F_{kk}|^2}{\sum_k |F_{R,R,R}||R|} \right) = \left( \frac{\sum_k |V_k F_{kk}^k|^2}{\sum_k |F_{k}^k|^2} \right), \]

which was studied by several authors for EFKM [46–48] as well as 2BHM [51]. Since the largest contribution to the pairing interaction in these models is on-site the correlation length reflects the relative size of the pairing field to the bandwidth. Weak pairing yields sizeable \( F_{kk} \) only in the vicinity of the Fermi level and one ends with \( r_{\text{coh}} \) over many unit cells (BCS limit). Strong pairing leads to almost constant \( F_{kk} \) and \( r_{\text{coh}} \) limited to a few sites (BEC limit).

Finally, we discuss the spin structure of the EC order parameter, which we divide into the spin-singlet and spin-triplet parts

\[ F_{ii}^{\text{cov}} = \frac{1}{2} (\phi_i^s \delta_{\sigma \sigma} + \phi_i^t \tau_{\sigma \sigma}^s), \]

i.e. singlet \( \phi^s \) and triplet \( \phi^t \) components are defined by

\[ \phi_i^s = \sum_{\sigma} \langle a^+_\alpha b_{\beta \sigma} \rangle \]

\[ \phi_i^t = \sum_{\sigma \sigma'} \langle a^+_\alpha b_{\beta \sigma} \rangle \tau_{\sigma \sigma'} \]

where \( \tau \) are the Pauli matrices and \( ^s \) denotes the complex conjugation. Tensorial character of \( \{ \phi^t, \phi^s \} \) and the fact that their elements are complex numbers allow numerous distinct

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4 For example, for \( xy \) and \( x^2 - y^2 \) orbitals on a square lattice.
phases, as is discussed in what follows. Some of the phases lead to a uniform spin polarisation while others have no ordered spin moments.

3.3. Strong-coupling limit

At half-filling and large \( U, U' \gg t, t_n, V_{ab}, V_{ha}, J, J' \) the charge fluctuations are strongly suppressed. Similar to the strong-coupling treatment of EFKM, the low-energy physics can be described by an effective model built on states containing only doubly occupied sites. The virtual excitations to states with singly and triply occupied sites provide inter-site couplings in the low-energy model. Using Schrieffer–Wolff transformation to the second order one arrives at expressions of the type \( \tilde{r}/U \), known from the analogous transformation from the single-band Hubbard to Heisenberg model. By construction, the low-energy model does not capture one-particle or charge excitations, which take place only at energies of the order \( U \).

The general strong-coupling expressions for the case of symmetric bands \( t, t_n \) were derived by Balents [52]. In the following we discuss several special parameter choices of the strong-coupling model, which are studied in the literature.

3.3.1. Large \( J \)—general formulation

For a sufficiently large Hund’s coupling \( J \), the low-energy Hilbert space can be constructed from the atomic HS and LS states: \( |1\rangle = a^+_s |b^+_s| \text{vac} \), \( |0\rangle = \frac{1}{\sqrt{2}} (a^+_s |b^+_s| \text{vac}) + (-1) = a^+_s |b^+_s| \text{vac} \), and \( |\emptyset\rangle = b^+_s b^+_s |\text{vac} \rangle \), where |vac⟩ is the fermionic vacuum. The local Hilbert space is further reduced if we assume easy axis anisotropy and drop the \( |0\rangle \) state. In this case only the density–density part of the Hund’s interaction contributes\(^5\). In the following, we derive the strong-coupling model for this case to demonstrate the principle. The generalisations are straightforward and can be found in the literature [52, 53]. Similar to (2), the low-energy Hamiltonian can be formulated in terms of pseudospin variables. Following another work [52], we introduce on-site standard-basis operators [54] \( T_i^{(m)} \) with matrix elements in the local basis

\[
\langle m'| T_i^{(m)} |n'\rangle = \delta_{mn} \delta_{m'n'}.
\]  

The effective Hamiltonian reads

\[
\mathcal{H}_{\text{eff}}^{(i)} = \epsilon \sum_{i} T_i^{(s)} + K_1 \sum_{(ij),s} \left( T_i^{(s)} T_j^{(s)} + i \leftrightarrow j \right) + K_0 \sum_{(ij),s} (K_0 + (-1)^{s+j}) T_i^{(s)} T_j^{(s)} + K_1 \sum_{(ij),s} \left( T_i^{(s)} T_j^{(s)} + T_i^{(s)} T_j^{(s)} \right),
\]

where \( s = \pm 1 \) and \( s = -s \). Typical hopping processes contributing to \( \mathcal{H}_{\text{eff}}^{(i)} \) are shown in figure 8. The process (i) lowers the energy of the HS–LS pair on the nn bond relative to the LS–LS pair. Therefore, it lowers the energy \( \epsilon \) of a single HS site on the otherwise LS lattice relative to the single atom value of \( E_{LS} - E_{HS} \) and contributes to a nn repulsion between HS states \( K_0 \). A similar process between two HS states with opposite \( s \) gives rise to the exchange term \( K_0 \).

The process (ii) exchanges the HS and LS states in the nn bond and introduces quantum fluctuations in the model. Neglecting the cross-hopping contributions (see [53] for the general expressions including cross-hopping corrections) the coupling constants read: \( \epsilon = \Delta - 3J - \frac{t^2 + t_n^2}{U} \), \( K_1 = \frac{2t}{U} \), \( K_0 = \frac{t^2 + t_n^2}{U} \), and \( K_0 = \frac{t^2 + t_n^2}{U + J} \), where \( \Delta \) is the number of nearest neighbours. Finally, the process (iii) converts HS–HS pairs with zero total moment into LS–LS pairs and vice versa. This process is possible only with finite cross-hopping, \( K_1 = -2V_{ab} V_{ha} (U + J - \Delta) \). The virtual excitations to states with opposite \( s \) as \( \emptyset \) acts on the vacuum state.

Hamiltonian (13) can be formulated in terms of hard-core bosons. In this picture, \( |\emptyset\rangle \) is identified with the bosonic vacuum and \( |\emptyset\rangle \) with a state containing one boson of type \( s \).

However, practical implementation of the hard-core constraint prohibiting more than one boson per site is complicated. The standard way to treat the constraint is to introduce a new vacuum state \( |\Omega\rangle \) and Schwinger-like bosons: \( |\emptyset\rangle = d^+_s |\Omega\rangle \), \( |\Omega\rangle = h^+_s |\Omega\rangle \). The physical states are required to obey the local constraint

\[
h^+_s |h_i + \sum_s d^+_s d_s = 1.
\]

Rewriting (13) in terms of \( d^- \) and \( h^- \) bosons, using the replacement \( T_i^{s} = d^+_s h^-_i \) and introducing the \( d^- \) number operator

\[
n_i = \sum_s d^+_s d_s = \sum_s T_i^{s},
\]

and the spin operator

\[
S_i^z = \sum_s s d^+_s d_s = \sum_s S_i^{s},
\]

one arrives at

\[
\mathcal{H}_{\text{eff}}^{(i)} = \epsilon \sum_{i} n_i - K_1 \sum_{(ij),s} (d^+_i d_j h_i h_j + \text{h.c.}) + K_0 \sum_{(ij),s} (d^+_i d_j h_i h_j + \text{h.c.}) - K_1 \sum_{(ij),s} (d^+_i d_j h_i h_j + \text{h.c.}).
\]

This allows us to interpret the \( K_0 \) term as nn hopping of \( d \)-bosons, \( K_1 \) as nn repulsion between the \( d \)-bosons, \( K_0 \) as nn spin–spin interaction, and \( K_1 \) as pairwise creation and annihilation of \( d \)-bosons on the nn sites. Besides technical advantages, the introduction of \( h \)-boson for \( |\emptyset\rangle \) treats the LS and HS states on equal footing and thus is well-suited for \( \epsilon < 0 \), where \( |\emptyset\rangle \) is not the atomic ground state and thus cannot be viewed as the vacuum state.

The effective Hamiltonian for the case with \( SU(2) \) spin symmetry has a similar structure but differs by the presence of

\[\text{Figure 8. Typical nn hopping processes that give rise to couplings in the effective Hamiltonian (13), (15) and (17).}\]
a third bosonic type \(|0\rangle = d''(\Omega)\). Introducing a cartesian vector \(d'_i\) with components

\[
\begin{pmatrix}
    d'_x \\
    d'_y \\
    d'_z
\end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix}
    i(d'_{x-} - d'_{y+}) \\
    i(d'_{x+} + d'_{y-}) \\
    \sqrt{2} d'_{z0}
\end{pmatrix},
\]

the effective Hamiltonian can be written in a compact form

\[
\mathcal{H}^{(2)}_{\text{eff}} = \varepsilon \sum_i n_i + K_1 \sum_{\langle ij \rangle} (d_i^\dagger \cdot \mathbf{d}_j) n_i + h.c.
\]

\[
+ K_\parallel \sum_{\langle ij \rangle} n_i n_j + K_0 \sum_{\langle ij \rangle} S_i \cdot S_j
\]

\[
+ K_1 \sum_{\langle ij \rangle} (d_i^\dagger \cdot \mathbf{d}_j + h.c.)
\]

\[
+ K_2 \sum_{\langle ij \rangle} ((d_i^\dagger h_i^\dagger + d_i h_a) \cdot S_j + i \leftrightarrow j).
\]

Here, \(S_i\) are spin \(S = 1\) operators with \(S^z = \frac{\varepsilon}{2}\left( T_{10}^0 + T_{10}^{-1} + T_{10}^{1} + T_{10}^{-10}\right)\). Expressed in vector notation, \(S_i\) takes the form of a cross product

\[
S_i = -i d_i^\dagger \wedge d_i.
\]

Unlike the Ising case (15) where cross-hopping is necessary to generate \(K_1\), in the SU(2) symmetric case (17) \(K_1\) also appears without cross-hopping if the pair-hopping term is finite, \(J' \neq 0\). The last term with coupling constant \(K_2 = V_{ab} d_a + V_{ab} d_b\), which couples the \(d\)-operators to the spin operators, does not have an analogy in the Ising case. The full expressions for the coupling constants can be found elsewhere [53].

Exciton condensation in (17) is characterised by a finite expectation value \((d_i^\dagger h_i)\), related to the spin-triplet order parameter (11) of 2BHM by

\[
(d_i^\dagger h_i) = \phi_i^0 / \sqrt{2}.
\]

Models (15) and (17) for special choices of the coupling constants are known under their own names. We discuss these cases below.

### 3.3.2. Blume–Emmy–Griffiths model

Without cross-hopping and one fermionic species immobile, \(t_{ab}, V_{ab}, V_{ba} = 0\), (15) becomes purely classical, with \(K_a, K_0 = 0\). Known as the Blume–Emmy–Griffiths (BEG) model [6], it was originally introduced to describe mixtures of \(^3\text{He}\) and \(^4\text{He}\). In the standard formulation of the BEG model the discrete index \(s\) is used to describe the local state \(s = \pm 1\) corresponding to \(\pm 1\) and \(s = 0\) is assigned to \(\Omega\). The model is then written as

\[
H_{\text{BEG}} = \varepsilon \sum_i s_i^2 + K_1 \sum_{\langle ij \rangle} s_i^2 s_j^2 + K_0 \sum_{\langle ij \rangle} s_i s_j.
\]

The BEG model found its use in many areas of statistical physics. Despite its simplicity it has a rich phase diagram [55]. The phase diagram of the model on bipartite lattice does not depend on the sign of \(K_0\) as the ferromagnetic \(K_0 > 0\) and antiferromagnetic \(K_0 < 0\) models can be mapped on each other by the transformation \(s_i \rightarrow (-1)^i s_i\). For \(K_0 > |K_0|\) there are only magnetically ordered (AFM) and normal (N) phases. At \(T = 0\) the N phase corresponds to the empty lattice \((n_i) = 0\), while in the AFM phase there is one boson on each site \((n_i) = 1\). A typical phase diagram for \(K_0 > |K_0|\) is shown in figure 9. Besides the magnetically ordered (AFM) phase there is the solid (S) phase (called antiferroquadrupolar [55]) characterised at \(T = 0\) by bosons occupying one sublattice, with the other being empty, \((n_i) = \frac{1}{2}(1 - (-1)^i)\). Note that this phase has a residual spin degeneracy on the occupied sites. This is a consequence of the exchange interaction beyond nn being strictly zero. In a more realistic model one expects the occupied sites to order magnetically at a sufficiently low temperature. Coexistence of the magnetic and solid order (I phase in figure 9) is found at finite temperature in a narrow range of \(\varepsilon\) separating the magnetic and solid phases. For positive \(\varepsilon\) at \(T = 0\), the system is in the vacuum (empty lattice). Interestingly, for moderate \(\varepsilon > 0\) the solid phase is found at an elevated temperature. This reentrant behaviour of the solid phase was found also for finite \(t_{ab}\) in Monte-Carlo simulations of the spinless bosons [25] as well as in DMFT simulations of 2BHM [56]. Generalisation of BEG model to the case with spin-rotational symmetry (17) is straightforward. On the mean-field level it leads to quantitative modification of the phase diagram.

### 3.3.3. Bosonic t-J model

Next, we discuss models (15) and (17) in the parameter range where they describe conserved bosons. We assume that both \(a\) and \(b\) electrons are mobile, generating a finite hopping \(K_j\) in (15) and (17) of the \(d\)-bosons. In addition, we require that \(K_1, K_2 = 0\), which is the case for \(V_{ab}, V_{ba} = 0\). Hamiltonians (15) and (17) describe interacting bosons carrying Ising and Heisenberg spin \(S = 1\), respectively. For suitable parameters, the system may undergo a BE condensation characterised by a complex vector order parameter \(\phi_i^0\) (19). In case of the Ising interaction (15) the vector \(\phi_i^0\) is confined to the \(xy\) plane. The vector character of the order parameter adds additional structure to the condensation of spinless bosons discussed in section 2.2.1.

**Figure 9.** The phase diagram of BEG model for \(K_0/K_0 = 3.5\). Besides the normal phase (N), it contains the solid phase (S), antiferromagnetic phase (AFM) and ferrimagnetic phase (I). The data were taken from another work [55].
The role of $K_0$. The SU(2) symmetric Hamiltonian (17) describes spinful $S = 1$ bosons with infinite on-site repulsion, nn repulsion $K_{0b}$ and nn spin-exchange $K_0$. Hamiltonians of this kind have been studied both theoretically and experimentally for cold atoms in optical traps. Interestingly, in a genuine system of bosons with spin-independent interactions the structure of the exact ground state prohibits the spin-exchange from appearing in any low-energy effective Hamiltonian [57]. As pointed out [57], it can only arise as a low-energy effective description in fermionic system, as in the present case.

BE condensation in the continuum version (17) was studied previously [58, 59]. The sign of the exchange was shown to play a crucial role for the properties of the superfluid phase as it determines the residual symmetry of the BE condensate. Antiferromagnetic exchange ($K_0 > 0$) selects the so-called polar state characterised by $(\phi^\dagger)^a_i \phi^a_i = 0$, whereas ferromagnetic exchange ($K_0 < 0$) selects the ferromagnetic state, where $| (\phi^\dagger)^a_i \phi^a_i |$ is maximised. Different residual symmetries of these states result in different low-energy dynamics and qualitatively different behaviour of topological defects (vortices) [58, 59]. The polar and ferromagnetic phases are found for Ising spins (15) [60], although the topological aspects that determine the low-energy excitations and possible topological defects are different from Heisenberg spins (17).

**Mean-field phase diagram.** The continuum model may be viewed as an effective description of the lattice model at low boson concentrations. At higher boson concentrations other phases, e.g. those present in the BEG phase diagram in figure 9, exist on a lattice and compete with the superfluid. In figure 10 we show the phase diagram of (17) on a square lattice ($z = 4$) obtained with a mean-field decoupling of the pseudospin variables (13) $T_i T_j \approx \langle T_i T_j \rangle + \langle T_i \rangle \langle T_j \rangle - \langle T_i \rangle \langle T_j \rangle$. In absence of condensed bosons, this mean-field theory reduces to that of a slightly modified BEG model (Heisenberg instead of Ising spin $S = 1$) with phase boundaries marked by the red lines. The grey area marks the superfluid (exciton condensate) phase—the different shades of grey correspond to different values of $K_0$. The transition between the normal and the EC phase is continuous, while the transitions to the other ordered phases are first-order as dictated by symmetry. The intermediate phases, such as supersolid (with S and EC orders) or AFM-supersolid (with AFM and EC orders), which would allow continuous transitions, do not exist or are thermodynamically unstable.

At large $e$ the ground state is an empty lattice, a state that is connected to normal Bose gas at elevated temperatures. Upon reduction of $e$ the system undergoes a continuous transition to the superfluid EC phase with $T_c$ determined by $|K_0|$ and $e$. The sign of $K_0$ determines the periodicity of the order parameter: for $K_0<0$ the system goes to uniform ferro-EC state (not to be confused with ferromagnetic EC state) with $\phi_j = \phi$, whereas for $K_0 > 0$ the system goes into antiferro-EC state with $\phi_j = (-1)^j \phi$. The models with $\pm |K_0|$ are connected by the gauge transformation $d_i \to (-1)^j d_i$. The BEG phase boundaries are not affected by $K_0$.

Another parameter that determines the nature of the EC phase is the exchange coupling $K_0$. The phase diagram in figure 10 was obtained for antiferromagnetic $K_0 > 0$, but some of its features remain unchanged when the sign of $K_0$ is flipped. In particular, the BEG phase boundaries are unchanged. This is so because the on-site and interaction terms in the Hamiltonian are invariant under the exchange of spin species on one of the sublattices ($d_{i} \rightarrow d_{j}$), which maps ferromagnetic BEG to the antiferromagnetic BEG model. This transformation works only for Ising spins, but in the mean-field treatment the difference between Ising and Heisenberg spins disappears. The transformation changes the hopping term in (15) and (17) and thus the argument cannot be used for the EC phase. Nevertheless, the mean-field phase boundary between the normal and EC phase does not depend on $K_0$ at all, because it contributes to the free energy on the order $\phi$. However, the first-order phase boundaries depend on $K_0$ and its sign.

**Ferromagnetic EC state.** It is instructive to see where exactly the difference between the $\pm |K_0|$ models comes from. First, we point out that an order parameter of the form $\phi_j = e^{iq R_j} \phi$ implies uniform magnetisation $(S_j) \sim i q h \phi$ irrespective of its periodicity $q$. Second, let us consider the free energy of the FMEC and polar EC states with the same magnitude $|\phi|$ of the order parameter. The on-site, hopping, and interaction terms contribute the same for the two states. However, the exchange energy in the polar EC state is zero while the FMEC state has finite exchange energy. Therefore, like in the continuum models, $K_0 > 0$ leads to a non-magnetic polar EC state, while $K_0 < 0$ leads to FMEC with a finite uniform magnetisation. For the same $|K_0|$, the FMEC/S boundary for $K_0 < 0$ is shifted in favour of the EC phase compared to the polar-EC/S boundary for $K_0 > 0$ because the FMEC energy is lower than the corresponding polar EC energy.

Continuous transition between the FM and FMEC phases is allowed by symmetry. Although we are not aware of an explicit calculation of the FM/FMEC transition for $K_0 < 0$, one can gain insight from the $T = 0$ mean-field wave function, which has the product form [52]
with the coefficients fulfilling $|\tilde{h}|^2 + \sum_i |\tilde{d}_i|^2 = 1$. The magnetisation (18) in terms of the EC order parameter (19) $\phi = \sqrt{2} \hat{d}^\dagger \hat{d}$ is given by

$$\langle \Psi | S_i | \Psi \rangle = -i \hat{d}^\dagger \wedge \hat{d} = \frac{i}{2|\tilde{h}|} \phi^\dagger \wedge \phi. \quad (22)$$

This shows that at a continuous FMEC/FM transition $\tilde{h}$ goes to zero, while the magnetisation smoothly reaches its saturation value.

**Two flavour models.** We are not aware of specific theoretical studies of spinful hard-core bosons (15) and (17) on a lattice. However, models (15) and (17) can be viewed as special cases of more general multi-component boson systems. The bosonic $t$-$J$ model describing mixtures of two boson species with hard-core constraint have been investigated in several studies. This set-up is similar to model (15) with Ising spin and $K_1 = 0$, which conserves both bosonic species separately. In the following, we discuss briefly how and which of the results of these studies can be used to describe (15).

The bosonic $t$-$J$ model is usually formulated in terms of $\tilde{I} = 1/2$ pseudospin: $I_x = d^\dagger_i d_{i+1}$, $I^\dagger_x = d^\dagger_i d_{i-1}$, and $I^\dagger_y = d^\dagger_i d_{i}$, and anisotropic XXZ inter-site exchange interaction. While the definition of the spin operator $S_z$ in (15) coincides with that of $I_z$, the transverse components $I^\dagger$ and $I^{\dagger}$ have no spin counterpart in (15). Therefore, great care is required when interpreting the results of the bosonic $t$-$J$ model in the language of the model (15). In particular, the $z$-axis pseudospin order (often referred simply as an anti-ferromagnetic order) corresponds to a true spin order (15). However, the $xy$ pseudospin order (e.g. $xy$-ferromagnet [62, 63]) does not correspond to a spin order in (15) or its SU(2)-symmetric generalisation (17).

Another difference between the bosonic $t$-$J$ model and (15) concerns the hard-core constraint. In most studies on the bosonic $t$-$J$ model simultaneous presence of two bosons of different types on the same site are allowed either explicitly (the hard-core constraint applies only to particles of different species) or implicitly (double occupancy is allowed in the hard-core constraint and the absence of the pseudospin exchange. However, the total hard-core constraint and the absence of the $xy$ exchange (15) are viewed as kinematic constraints\(^8\). This calls for care when using the phase diagrams of the bosonic $t$-$J$ model since the existence of some phases, e.g. the counter-superfluid in one study [63], depends crucially on the $xy$ exchange and therefore such phases are not present in (15).

In the following we discuss two types of studies on the bosonic $t$-$J$ model, which can be translated to provide information about (15). The first concerns the transition between AFM and SF phases and the possibility of intermediate AFM-supersolid phase. Boninsegni [64] and Boninsegni and Prokof'ev [65] studied the bosonic $t$-$J$, model, corresponding to (15) for $K_4 = -K_0 < 0$ using Monte-Carlo simulations. Since the solid phase is absent for $K_0 < |K_0|$ [55], one may expect AFM-SF transition for moderate $K_0$ (for strong $K_0$ the BEG model predicts first-order transition between the Mott AFM and vacuum states at low temperatures)(.) The Monte-Carlo calculations find a first-order AFM-SF transition with no indication of AFM-supersolid characterised by coexistence of the AFM and SF order parameters.

Numerical calculations with repulsive $K_0$ ($K_0 = K_1 > 0$), motivated by observation of the supersolid in the spinless case, have been reported on a triangular lattice recently [66, 67]. Although double occupancy by different bosonic types was allowed and the lattice differs from the square lattice, the basic features common to figure 10 were found in the phase diagram for large on-site (inter-species) repulsion. In particular, the sequence of $T = 0$ phases ’empty lattice-SF-S’-AFM’ with decreasing $\epsilon$ can be expected.

3.3.4. Bi-layer Heisenberg model.** Another special case of the SU(2)-symmetric model (17) studied in the literature corresponds to bi-layer Heisenberg model

$$H_{bi-H} = \sum_{i} S^a_{im} \cdot S^b_{im} + J_1 \sum_{i} S^a_{im} \cdot S^b_{im} - \mathbf{h} \cdot \sum_{m} S_{im}. \quad (23)$$

with $S = 1/2$ spin operators $S_{im}$, $i$ being the site index and $m = a,b$ a layer index. The model arises as the large $U$ limit of the half-filled bi-layer Hubbard model (7), which describes two identical layers, indexed by the orbital index $m$, coupled on inter-layer rungs. This situation corresponds to the parameter set $U \gg |t| = |t_b|$, $\{\Delta, U', J'\} = 0$, $J < 0$ in (7), where the anti-ferromagnetic coupling on the rung $J' = J$ may arise from inter-layer tunnelling. Integrating out the charge fluctuations one arrives at (23) with $J_1 = 4t^2_{ba}/U$. Model (17) is obtained by going to the singlet-triplet basis, which diagonalises the rung exchange (local) part of (23). Introducing a map

$$S^a_{im,b} = \frac{1}{2} (\pm t^i_{a} s^i_{b} \pm t^i_{b} s^i_{a} - i t^i_{a} \wedge t^i_{b}) \quad (24)$$

one arrives at [7, 68]

$$H_{bi-H} = \frac{J_1}{4} \sum_{i} (t^i_{a} \cdot t^i_{a} - 3s^i_{a} s^i_{a})$$

$$+ \frac{J_0}{2} \sum_{i} (t^i_{a} \cdot t^i_{b} s^i_{a} + h.c.)$$

$$+ \frac{J_0}{2} \sum_{i} (-i)(t^i_{a} \wedge t^i_{a} \cdot t^i_{a} \wedge t^i_{a})$$

$$+ \frac{J_0}{2} \sum_{i} (t^i_{a} \cdot t^i_{b} s^i_{a} + h.c.)$$

$$+ \sum_i \mathbf{h} \cdot (t^i_{a} \wedge t^i_{a}), \quad (25)$$

where $s_i$ and $t_i = (t_{i,x}, t_{i,y}, t_{i,z})$ are bosonic operators and the physical subspace fulfills the local constraint $s^i_{a} s^i_{a} + t^i_{a} \cdot t^i_{a} = 1$. Hamiltonian (25) is equivalent to (17) with parameters $K_1 = K_2 = 0$, $K_4 = K_0 = J_0/2$ and $e = J_1$. The main difference of (23) to the bosonic $t$-$J$ model consists in

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\(^8\)The $xy$ exchange processes in (15) arise in fourth-order expansion in $t_a t_b$ of 2BHM.
the presence of non-zero $K_1$, which leads to the number of $d$-bosons being a non-conserved quantity. While the system undergoes transitions [68] to phases characterised by non-zero $\phi_i = \langle t_S \psi_i \rangle$, the phase of $\phi_i$ is not arbitrary and thus the transitions cannot be viewed as Bose–Einstein condensation of spinful bosons. In zero magnetic field $h = 0$ the ordered phases are characterised by real $\phi_i$ which breaks the SU(2) symmetry of (23) and corresponds to a Néel state with ordered moments $\langle S_{ab} \rangle = \pm \phi_i$ (figure 11). Non-zero magnetic field $h \neq 0$ reduces the symmetry of (23) to $U(1)$. The vector $\phi_i$ gets oriented perpendicular to $h$ and acquires an imaginary part perpendicular to the real one. The real part of $\phi_i$ describes the AFM order, while $i\phi_i$ describes the FM component along $h$. Breaking of the $U(1)$ symmetry can be described as BE condensation of spinless bosons. This is particularly easy to see for $J_1 \gg J_0$ because close to the transition $h \approx h_0$ and model (17) reduces to the Hamiltonian of spinless bosons for the type polarised along the field direction.

BE condensation in quantum magnets has been an active area of research and the reader is referred to a recent review [69] for further reading and references. In general, the basic difference between BE condensation in spin systems and excitonic condensation discussed here consists in the microscopic origin of the $U(1)$ symmetry broken by the condensate. In spin models, it is the spin rotation in the $xy$-plane of systems with uniaxial anisotropy and condensation refers to some kind of in-plane magnetic order. The $U(1)$ symmetry in excitonic systems is more abstract and refers to the arbitrariness of the relative phase between $a$ and $b$ orbitals in (7).

3.3.5. Exciton t-J model. The strong-coupling limit of 2BHM with anti-ferromagnetic Hund’s exchange was studied by Rademaker and collaborators [70–74]. They started from the antiferromagnetic bi-layer Heisenberg model (25) and considered doping one layer by holes and the other by the same amount of electrons. This is equivalent to introducing the crystal-field $\Delta$ in (7) while keeping the total electron concentration at half-filling. The anti-ferromagnetic Hund’s coupling $J < 0$ (corresponding to inter-layer exchange) was assumed to originate from small, but finite, inter-layer tunnelling. Starting from the undoped Heisenberg limit with on-site states $|m\rangle = i^{m}/(\Omega)$, and $|0\rangle = s/|\Omega\rangle = 1/2 (a_i^{\dagger} a_j^{\dagger} - a_j^{\dagger} a_i^{\dagger})|\text{vac}\rangle$, other authors [70–74] introduce a bound exciton state $|\Omega\rangle = h^{|\Omega\rangle}$ to the system. The new Hamiltonian, which is a special case of that discussed previously [52], reads

$$
H = H_{\text{hi-1l}} + \mu \sum_i h_i h_i + V \sum_i h_i h_i h_i h_i + t_{\text{ex}} \sum_i (t^{\dagger}_i h_i h_i + s^{\dagger}_i s_i h_i h_i + \text{h.c.}),
$$

(26)

with the hard-core constraint $h_i h_i + s_i s_i + t^{\dagger}_i \cdot t_i = 1$. The exciton site energy $\mu$ controls the exciton concentration $\rho$. The spin-singlet bosons $h$ and $s$ appear symmetrically in (26), except for the term $t^{\dagger}_i h_i h_i$, which does not have a $t^{\dagger}_i h_i h_i$ counterpart. The exciton hopping $t_{\text{ex}}$ and repulsion $V$ play roles analogous to $K_a$ and $K_b$, respectively, in (15, 17).

Note that the notion of exciton and vacuum is exchanged with respect to that introduced in section 3.3.1. Unlike the unambiguous vacuum state of real bosons, the meaning of vacuum for hard-core bosons is ambiguous, similar to the notion of electron and hole for fermions. The condensation of hard-core boson generally means that the system is in a quantum-mechanical superposition of the vacuum and one-particle states, whatever their definition is.

Rademaker et al [70, 71] studied the propagation of a single $|\Omega\rangle$ exciton in the model with antiferromagnetic coupling $J_a, J_1 > 0$. The propagation of exciton strongly depends on the ratio of the Hund’s (inter-layer) and intra-layer exchange $J_a J_b$. For $J_1 \gg J_0$ the ground state of undoped system is a product of local singlets $|0\rangle$ and the exciton can propagate as a free particle forming a band with the width $2t_{\text{ex}}$. In the opposite limit, $J_1 \ll J_0$ the system consists of weakly coupled AFM layers. It is well known from the fermionic $t - J$ model that the motion of a hole in the AFM background is strongly inhibited since a moving hole disturbs the AFM order. This physics is also reflected in the motion of an exciton. For $t_{\text{ex}} \ll J_0$ exciton propagation is severely limited and the exciton bandwidth is reduced to the order $t_{\text{ex}}^2 / J_0$. In the opposite limit $t_{\text{ex}} \gg J_0$ the exciton can explore neighbouring sites over larger distances, which gives rise to the typical incoherent string-state spectrum [70, 71, 74].

Exciton condensation in the excitonic $t-J$ model was studied previously [72, 74]. The general features of the mean-field phase diagram, shown in figure 12, are similar to the case of antiferromagnetic Hund’s exchange in figure 10 and can be traced back to their ‘common ancestor’ in the XXZ model. In particular, there are three basic phases: AFM, solid and the EC superfluid and first-order AFM/solid, solid/ superfluid, and AFM/superfluid transitions. We can compare these in figures 12 and 10 keeping in mind that increasing $\epsilon$ in figure 10 corresponds to increasing $\rho$ in figure 12. In both models the $T = 0$ solid (S) phase is absent for large exciton hopping $K_a (t_{\text{ex}})$. At intermediate hopping $K_a (t_{\text{ex}})$, the AFM-EC-S-EC sequence of phases is found in both models, while...
at small hopping there is a direct transition between the AFM and S phases. The small positive $J_z$ selects the spin-singlet condensate, which is characterised by spontaneous coherence between the [00] and [00] states. The mean-field ground state can be written as $|\Psi\rangle = \Pi_{k}(\sqrt{\rho} h^0_k + \sqrt{1 - \rho} s^0_k)(\Omega)$. An interesting consequence of the singlet condensation is an enhanced propagation of spin excitations (triplons). Authors [72, 74] have observed that the triplet excitations in the condensate of mobile excitons $t_{\text{ex}} \gg J_0$ propagate faster than in the quantum paramagnet. Surprisingly, they found the effective tripolon hopping scales with the density of the condensate $\rho_{\text{SF}} = \sqrt{\rho(1 - \rho)}$ rather than the exciton density $\rho$.

3.4. Weak coupling

In the weak-coupling (BCS) limit the formation of excitons and their condensation take place at the same temperature. The physics of the system in this limit can be described by approaches such as Hartree–Fock approximation and RPA. The key feature of the weak-coupling theories is that EC instability is driven by nesting between the Fermi surface sheets formed by the valence and conduction bands. The lack of perfect nesting in real materials is likely one of the reasons why EC is rarely found in nature. The weak-coupling theory of the excitonic condensation in systems with equal concentration of holes and electrons was developed in the 1960s [9, 10] and summarised in the review articles of Halperin and Rice [11, 75]. The pairing glue considered in the weakly-coupled semimetals or semiconductors comes from the long-range part of the Coulomb interaction. The exchange part of the long-range Coulomb interaction is small, i.e. similar to the choice $J, J' = 0$ in (7), and one has to consider both spin-singlet and spin-triplet pairing. Halperin and Rice [11] classified the possible excitonic condensates in systems with single Fermi surface sheet per band into the charge-density wave (real singlet), charge-current-density wave (imaginary singlet), spin-density wave (real triplet), and spin-current-density wave (imaginary triplet) type. In case there are multiple Fermi surface sheets related by point-group symmetries, e.g. as in hexaborides, a more complex symmetry classification is necessary [76, 77]. The classification elsewhere [11] includes only the non-magnetic polar EC states. In the mid-1970s Volkov and collaborators showed that in doped material with an unequal number of electrons and holes the ferromagnetic EC state develops characterised by simultaneous presence of finite singlet and triplet components [12–14].

We briefly review the mean-field (Hartree–Fock) theory of (7) and consider the simplest case of uniform EC order ($t_{\text{ex}} < 0$). The mean-field Hamiltonian that allows polar as well as ferromagnetic EC order reads

$$H = \sum_{k,\sigma} (\epsilon_k \delta_{\sigma\sigma'} - h_\sigma \cdot \tau_{\sigma\sigma'}) \phi^\dagger_k \phi_{k'} + \sum_{k,\sigma} (\epsilon_k \delta_{\sigma\sigma'} - h_\sigma \cdot \tau_{\sigma\sigma'}) \phi^\dagger_k \phi_{k'} - \sum_{k,\sigma} ((\Delta_{\sigma\sigma'} + \Delta_{\sigma'} \cdot \tau_{\sigma\sigma'}) \phi^\dagger_k \phi_{k'} + \text{h.c.}) .$$

(27)

Here, $a_{k\sigma}, b_{k\sigma}$ are Fourier transforms of $a_{\alpha}, b_{\alpha}$, respectively. The crystal-field splitting $\Delta$ as well as the spin-independent part of the self-energy are absorbed in band dispersions $\epsilon_k, \epsilon_{k'}$. In the following discussion we assume $a$ to form the conduction band and $b$ to form the valence band. The Weiss fields $h_\sigma, h_\sigma$, $\Delta_\sigma, \Delta_{\sigma'}$ are given by

$$h_\sigma = h + \frac{U}{2N} \sum_{k,\sigma'} (\phi^\dagger_k \phi_{k'}) \tau_{\sigma\sigma'}$$

$$\Delta_\sigma = \frac{U'}{2N} \sum_{k,\sigma'} (\phi^\dagger_k \phi_{k'}) \delta_{\sigma\sigma'}$$

$$\Delta_{\sigma'} = \frac{U'}{2N} \sum_{k,\sigma'} (\phi^\dagger_k \phi_{k'}) \tau_{\sigma\sigma'} .$$

(28)

where $h$ is the external magnetic field (acting on spin only). The field $h_\sigma$ is defined as $h_\sigma$ with the orbital flavour replaced. Note that for model (7) with local interactions $(\Delta_\sigma, \Delta_{\sigma'}) = U'(\phi_\sigma, \phi_{\sigma'})$, where $\phi_\sigma, \phi_{\sigma'}$ are the local order parameters. The generalisation to models with non-local interaction, which leads to $k$-dependent Weiss fields, is straightforward and can be found in the literature.

Bascones et al [78] studied model (27) as a function of doping and external field $h$ at $T = 0$. The phase diagram, shown in figure 13, contains four phases: the normal phase (N), polar excitonic insulator (EI) phase, and two metallic FMEC phases called NC and COL [78]. To illuminate the nature of these phases it is helpful to introduce $\Delta_{\sigma\sigma'} = \frac{1}{2}((\Delta_{\sigma\sigma'} + \Delta_{\sigma'} \cdot \tau_{\sigma\sigma'})$. In the zero-field EI phase, the singlet and triplet EC orders are degenerate. A finite field $h$, assuming $h = h,2h$ and $h > 0$ in the following, lifts the degeneracy. The undoped system selects a triplet state with $\Delta_{\uparrow\uparrow} = \Delta_{\downarrow\downarrow} = 0$ and $|\Delta_{\uparrow\downarrow}| > |\Delta_{\downarrow\uparrow}|$. For sufficiently large $h$, the system enters the EI2 phase of figure 13.
bands. The order parameter in the NC phase is purely and predicted by Kozlov and Maksimov [81] and RPA. They observed acoustic-like modes with linear dispersion at small \( q \). The normal/solid phase boundary qualitatively with a quadratic dispersion at small \( \mathbf{q} \). They reported [56] observation of the solid phase for the case of strongly asymmetric bands \( |a| \gg |b| \). The normal/solid phase boundary qualitatively agrees with strong-coupling BEG model. In particular, the re-entrant transition as a function of temperature for large \( \Delta \) has been found. In another study [53], an unbiased linear response approach was applied to study excitonic condensation in 2BHM at half-filling. Recently, dynamical cluster approximation was also applied to study excitonic condensation [94].

\[
\Delta_{\text{NC}} = 0.0
\]

Upon doping two distinct FMEC phases are found. In the NC the only non-zero element of the order parameter is \( \Delta_{\text{NC}} \neq 0 \). The EI2 and NC phases are distinguished by presence of a h-dependent gap between the uncondensed \( a \) and \( b \) bands. The order parameter in the NC phase is purely spin-triplet and the phase coincides with the FMEC phase of \( S = 1 \) bosons in the strong-coupling limit. In the COL phase, the only non-zero element of \( \Delta_{\text{COL}} \) is \( \Delta_{\text{COL}} \neq 0 \). In this phase, as predicted by Volkov et al [13], the singlet and triplet order parameters mix with equal weight (\( \Delta_s = -\Delta_t \)).

The \( \mathbf{h} = 0 \) spin-wave spectrum of the COL state for a general chemical potential \( \mu \) is characterised by two gapless modes with a quadratic dispersion at small \( |q| \) and an additional soft but gapped mode [78]. At a special value of \( \mu \) the soft mode becomes gapless and the spectrum has one quadratic and two linear modes. The mean-field phase diagram and magnetic excitations of the polar EI phase in the undoped model were studied by Brydon and Timm [79] and Zocher et al [80] using RPA. They observed acoustic-like modes with linear dispersion at small \( |q| \) predicted by Kozlov and Maksimov [81] and Jérôme et al [82].

Moving away from the strong-coupling limit the separation of energy scales of exciton formation and exciton condensation is progressively less well defined and eventually these scales are not separated at all. As in the spinless case of EFKM, EC exists also at weak-coupling and one can follow the BEC-BCS crossover as the interaction strength is lowered. The weak-coupling methods, such as Hartree–Fock approximation and RPA, were applied to study the EC in its early days and are summarised in the review article of Halperin and Rice [11, 75].

3.5. Intermediate coupling

Investigations of systems with intermediate-coupling strength are notoriously difficult due to the lack of small parameters. The general approaches to this problem include numerical simulations of finite systems such as exact diagonalisation or quantum Monte-Carlo (QMC) methods, large-N expansions, and embedded impurity or embedded cluster methods such as dynamical mean-field theory [83] (DMFT), variational cluster approximation [84], dynamical cluster approximation, or cluster DMFT [85]. A major obstacle in simulation of ordering phenomena with finite-system methods is the necessity of scaling analysis, i.e. the cluster size must be large enough to show the ‘diverging’ correlation length. Monte-Carlo simulations on large clusters are available for many bosonic and spin systems, but usually not for fermions. Rademaker et al [73] applied the determinant Monte-Carlo method to a bi-layer Hubbard model and were able to demonstrate an enhanced response to the excitonic pairing field but could not reach temperatures below \( T_c \). Besides the Green’s function QMC methods for calculation of correlation functions, wave function QMC approaches can be applied to variational search for ground states. While we are not aware of variational QMC studies of excitonic condensation in Hubbard-type lattice models, variational QMC has been used to study the corresponding continuum problem [86].

The DMFT methods have been very valuable for investigation of Hubbard model and its multi-band generalisations in the past two decades. However, most applications of DMFT so far focused on one-particle quantities and normal (paramagnetic) phase. With an exception of a multi-band study [87], linear response calculations needed to identify instabilities have been so far limited to simple one- [88] and two-band models [53, 89, 90]. Recent DMFT investigations of ordered phases in models as simple as 2BHM uncovered a rich physics. Capone et al [91, 92], Koga and Werner [93], and Vanhala et al [94] found various forms of superconductivity in 2BHM and also applied to study excitonic condensation [94].

3.5.1. Half filling

Kuneš and collaborators studied 2BHM (7) in the vicinity of spin-state transition (see also figure 7) using DMFT [53, 56, 60, 95] for 2BHM (7) with \( \Delta_{ab}, \Delta_{ba} ^>, \Delta_{bc} ^<, J = 0 \) and density–density interaction. They reported [56] observation of the solid phase for the case of strongly asymmetric bands \( |a| \gg |b| \). The normal/solid phase boundary qualitatively agrees with strong-coupling BEG model. In particular, the re-entrant transition as a function of temperature for large \( \Delta \) has been found. In another study [53], an unbiased linear response DMFT approach [96] was used to probe stability of the normal phase. Two types of instabilities were found, an instability toward the solid and an instability toward the excitonic condensate. This situation resembles EFKM. Indeed, the physics behind formation of the solid and the superfluid phases in the strong-coupling limits of EFKM and 2BHM is similar. As in EFKM, the excitonic instability in 2BHM extends to the weak-coupling limit. This is not so for the solid phase. In the weak-coupling limit, the instability toward solid either does not exist at all or is weaker than the antiferromagnetic instability.

The basic physical properties of the excitonic phase were studied [95]. In figure 14, we show a typical \( T \)-dependence of the magnitude of the order parameter \( \langle |\phi| \rangle \). The system selected...
for the system constrained to the normal \( c_{1/2} \) between was kept fixed. Data taken at various \( A_{\text{aa}} \) (eV \( \Delta \)). The \( \chi_s \) along is consistent with the mean-field \( \chi_{\text{mean-field}} \) and HS states and opens a gap of the order spin gap. In the polar EC phase, the Weiss field mixes the LS – Weiss susceptibility and a vanishing of the This leads to Curie – Weiss behaviour, the susceptibility of the polar \( \chi_s \) shown in figure 16. While the normal-phase susceptibility reflects the formal analogy to an \( s \)-wave superconductor discussed in section 2.

This analogy, however, does not extend to the electromagnetic properties of the condensate. The neutral exciton condensate does not contribute to the charge transport, which is facilitated by the quasi-particle excitations. In figure 15 we show the optical conductivity and \( \rho_c \) resistivity at various temperatures. The opening of the charge gap leads to an optical gap and exponential increase of the resistivity below \( T_c \).

Finally, we discuss the spin susceptibility \( \chi_s^Z \), which is shown in figure 16. While the normal-phase susceptibility exhibits Curie–Weiss behaviour, the susceptibility of the polar EC phase appears \( T \)-independent. This observation holds within the numerical accuracy for the uniform susceptibility and approximately also for the local susceptibility. This behaviour can be understood from a single atom picture. In the normal phase, the system is locally in a statistical mixture of the LS ground state and thermally populated HS multiplet. This leads to Curie–Weiss susceptibility and a vanishing of the spin gap. In the polar EC phase, the Weiss field mixes the LS and HS states and opens a gap of the order \( U' \phi / 2 \gg T \) between the local ground state and the excited states. The local ground state is a superposition of the form \( \phi_0 + \beta(\uparrow\downarrow + \downarrow\uparrow) \). The spin susceptibility has \( T \)-independent van Vleck character and a finite spin gap appears (see figure 16).

Kaneko et al \[98\] used variational cluster approximation to study the spin-triplet EC in 2BHM without Hund’s coupling (\( J, J' = 0 \)) and with symmetric bands \( t_a = t_b \) for a broad range of interaction parameters. Similar to the strong-coupling phase diagram in figure 10, other authors \[98\] found continuous transition between the EC and normal state (band insulator), and a first-order transition between the EC and AFM phases as shown in figure 17. Kaneko and Ohta \[51\] extended the study to include the Hund’s coupling, which confirmed that \( J < 0 \) favours the spin-singlet charge-density-wave state, while \( J > 0 \) selects the spin-triplet spin-density-wave state \[11, 99\].

3.5.2. Doping. The weak-coupling theory of doped excitonic insulator was developed by Volkov and collaborators \[12–14\] for systems without Hund’s coupling. They showed that such systems tend to develop ferromagnetic order due to simultaneous appearance of the spin-singlet and spin-triplet order. In section 3.4, we have reviewed the application of the weak-coupling approach by Bascones et al \[78\]. Besides finding Volkov’s ferromagnetic phase (COL) the authors \[78\] also found a spin-triplet phase (NC) induced by an external magnetic field. The possibility of the triplet ferromagnetism was also discussed by Balents \[52\] who considered the effect of doping in a strong coupling limit on a qualitative level.
At higher doping levels and lower temperatures the system enters the FMEC phase with finite magnetisation \( m \sim i(\psi \times \phi) \neq 0 \). The transition between polar and ferromagnetic phases proceeds via an intermediate phase (blue), which is distinguished from the FMEC only in the absence of cross-hopping and pair-hopping [60]. The FMEC phase is equivalent to the COL phase [78] (figure 13). Similar to the \( T = 0 \) phase diagram [78], first-order transitions and phase separation are found at low temperatures. It is not clear from the numerical data whether a \( T = 0 \) polar phase exists at finite doping. At the moment we can only speculate that the antiferromagnetic nn coupling provides a means to stabilise it. A peculiar feature of the FMEC is the \( T \)-dependence of the magnetisation in the vicinity of the continuous transition to the normal state, which follows the linear \( 1 - T/T_c \) dependence. This behaviour is a consequence of the quadratic dependence on the EC order parameter \( m \sim i(\psi \times \phi) \).

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Finally, we briefly discuss the effect of finite cross-hopping and/or pair-hopping, which break the charge conservation per orbital flavour. As a result the phase factor in the polar EC phase is fixed and the system selects either the spin-density-wave or the spin-charge-density-wave order. It is also possible that both types of order are realised in parts of the phase diagram. Another consequence of finite cross-hopping/pair-hopping will be the absence of a continuous normal to FMEC transition. The FMEC state is selected by terms of the order \( \psi^4 \) in the Ginzburg–Landau functional and a continuous transition is only possible if the second-order terms do not depend on the phase of \( \psi \). Finite cross-hopping/pair-hopping removes this degeneracy. For weak cross-hopping one can expect a small wedge of polar EC phase separating the normal and FMEC phases. The first-order normal/FMEC transitions are still possible.

4. Multi-orbital Hubbard model

Little has been done in regarding the generalisation of the physics of section 3 to systems with more than two orbitals per atom. Kuneš and Augustinský [95] used the Hatree–Fock (LDA+U) approach to study EC in quasi-cubic perovskite with nominally six electrons in the \( d \) shell. Let us demonstrate the new features on five-orbital model describing the \( d \)-orbital atoms on a cubic lattice. The basic setting of such system is similar to the half-filled two-orbital model. The crystal-field splits the \( d \) orbitals into three-fold degenerate \( t_{2g} \) and two-fold degenerate \( e_g \) states. With six electrons per atom the lower \( t_{2g} \) levels are filled and the upper \( e_g \) levels are empty. An exciton is formed by moving an electron from a \( t_{2g} \) orbital to an \( e_g \) orbital on the same atom. Unlike 2BHM with only one possible orbital structure of an exciton, in the \( d \)-atom there are six possible orbital combinations. The cubic symmetry distinguishes the orbital symmetries of an exciton into two three-dimensional irreducible representations \( T_{1g} \) and \( T_{2g} \). Considering the geometry of these two excitons [95] one can infer that \( T_{1g} \) excitons are more tightly bound and substantially more mobile that the \( T_{2g} \) ones. Therefore only the \( T_{1g} \) excitons need to be considered as candidates for condensation. The three-fold orbital degeneracy of the \( T_{1g} \)-excitons adds to the \( S = 1 \) spin degeneracy, making the order parameter a more complex object. It can be arranged to a tensorial form of a \( 3 \times 3 \) matrix \( \phi^{\alpha}_{\beta} \), where the \( \alpha \) indexes the spin and \( \beta \) indexes the orbital components. \( \phi^{\alpha}_{\beta} \) thus transforms as a vector under the SO(3) rotations in the spin space and as a pseudo
vector ($T_{1g}$ representation) under the discrete symmetry operations of the cubic point group. The explicit expression for $\varphi_{q}$ in terms of the on-site occupation matrix can be found in the supplementary material of another study [95]. The structure of the order parameter resembles of superfluid $^3$He [100] and one can use the group theoretical methods applied there to analyse the possible EC phases [101]. A more complex structure of the order parameter allows for existence of numerous distinct phases.

Another important difference of 2BHM concerns the hard-core constraint imposed in the excitons in the strong coupling limit. While in the case of 2BHM there cannot be more than one exciton on a given atom, in the five-orbital model the hard-core constraint is less restrictive. There cannot be more than one exciton of a given orbital flavour on atom, but it is possible that two excitons with different orbital flavours meet.

In the covalent terminology a single exciton on an atom represents the intermediate state $S = 1$, while the $S = 2$ high-spin state can be viewed as a bi-exciton. The fact that the site energy of the high-spin state is lower than that of the intermediate-spin state [102], it is then expressed as an attraction between excitons of different orbital flavour and parallel spins. Bosonic models with infinite intra-species but finite inter-species interaction have been studied for two species of (spinless) bosons [62, 63, 66, 67] and shown to exhibit phases that are not allowed with inter-species hard-core constraint.

5. Materials

5.1. Bulk materials

There have been numerous proposals of materials to exhibit excitonic condensation, but very few realisations of EC were actually documented. The early candidates for excitonic condensation, which followed the weak-coupling picture of proximity to semimetal–semiconductor transition, included the group V semimetals (Bi, Sb, As) and divalent metals (Ca, Sr, Yb), possibly under pressure [82]. However, the search for signatures of EC in these materials was not successful.

Wachter and collaborators [103–106] reported observations of EC in TmSe$_{0.35}$Te$_{0.65}$ under pressure. While they did not see a sharp thermodynamic transition, they argued that the observed anomalies are consistent with a ‘transition’ in the weak (pairing) field. Wakisaka et al [107] interpreted their photoemission data on Ta$_2$NiSe$_5$ in terms of exciton condensation [98, 108, 109]. Also, the lattice distortion and photoemission spectra of a layer compound $\Gamma^7$-TiSe$_2$ have been interpreted in terms of charge-density-wave type exciton condensation and described with the weak-coupling theory [110–113].

While the previous examples involved cases of spin-singlet condensation, in the following we discuss excitonic magnetism. In 1999 Young et al [114] reported observations of 600 K ferromagnetism in La$_{0.005}$Ca$_{0.995}$B$_6$. Ferromagnetism in a slightly doped semiconductor lead several groups to generalise the weak-coupling theory of excitonic ferromagnetism pioneered by Volkov and collaborators [12–14] to the case of multiple Fermi surface sheets [76, 77, 115–117]. Nevertheless, subsequent investigations showed that the band gap in parent compound CaB$_6$ is $\approx$1 eV [118] and thus inconsistent with the EC scenario. It is now generally accepted that the ferromagnetism in La$_{1-x}$Ca$_x$B$_6$ arises from defects rather than doping.

Another group of materials where the EC concept found its use is the iron pnictides [119, 120]. The physics of these materials is governed by nesting between several Fermi surface sheets formed by bands of different orbital characters [121]. Several groups studied a simplified two-orbital model [79, 122–124] and its multi-orbital extensions [125, 126] using weak-coupling approaches, and observed a spin-density-wave order with a periodicity given by the nesting vector. Arising from nesting between bands that mix several orbital characters, the corresponding Weiss field in general couples all possible orbital combinations. However, since the nested patches of the Fermi surface have different dominant orbital characters a large part of the condensation energy comes from orbital off-diagonal pairing, which produces local magnetic multipoles but no local moments (polar EC state). The generally present, but small, orbital diagonal contributions then give rise to the apparently small ordered moments. A first principles calculation of the ordered state supporting this picture was done by Crinicchio et al [127].

Kuneš and Augustinský [53] proposed that a transition observed in some materials of Pr$_{1-x}$CoO$_3$ (PCCO) family [128–131] can be understood as an excitonic condensation. Materials from this family exhibit a phase transition with $T_c$ as high as 130 K, which is characterised by sharp peak in the specific heat, transition from high-$T$ metal to a low-$T$ insulator, disappearance of Co local moment response, and simultaneous Pr$^{3+} \rightarrow$ Pr$^{4+}$ valence transition. A puzzling feature of the low temperature phase is the breaking of time reversal symmetry (in absence of ordered moments) evidenced by the Schottky anomaly associated with splitting of the Pr$^{4+}$ Kramer’s ground state. The transition to a spin-density-wave EC state provides a comprehensive explanation of these observations and the EC ground state is obtained with Hartree–Fock-type LDA+U calculations.

Another class of the materials with potential to exhibit the exciton condensation was proposed by Khalifullin [132, 133]. He considered a strong-coupling model of a $d$-electron material with cubic crystal field, strong spin-orbit coupling, and an average $d$ occupancy of four electrons per atom, a scenario possibly realised in materials with Re$^{3+}$, Ru$^{4+}$, Os$^{5+}$, or Ir$^{5+}$ ions. The large crystal field restricts the low energy physics to the space spanned by $t_{2g}$ states. Now the spin-orbit coupling plays the role of $\Delta$ in (7) competing with the Hund’s coupling $J$. Sufficiently strong spin–orbit coupling renders the single-ion ground state a singlet $\tilde{S} = 0^+$ and the first excited state a triplet $\tilde{S} = 1$. The perturbative treatment of $nn$ hopping results in the model similar to (17). There is, however, one important difference between (17) and the Khalifullin’s model formulated in terms of pseudospin $\mathbf{S}$. The real spin $\mathbf{S}$ is decoupled from the lattice and thus (17) is invariant under the SO(3) spin–spin

This corresponds to the state with the four $J_{d\downarrow} = 3/2$ one-particle states filled and the $J_{d\downarrow} = 1/2$ states empty.
rotations, particularly the hopping $K_c$ is spin-independent. The pseudospin $\hat{S}$ represents a spin-orbital object, which is coupled to the lattice and thus the model cannot be invariant under continuous pseudospin rotations. This is reflected in the hopping amplitudes being $\hat{S}$-dependent, i.e. the hopping containing terms known from Kitaev model [134].

Finally, the possibility of exciton condensation in layered cuprates [135] and oxide heterostructures [136] was discussed by several authors but has not been realised so far.

5.2. Bi-layer structures

A major direction in the research of exciton condensation are bi-layer structures [137]. The basic idea is that a bi-layer with negligible inter-layer tunnelling provides a system where two orbital flavours are independently conserved with high accuracy. Exciton condensation takes place under suitable conditions including small inter-layer distance, so that inter-layer electron–electron interaction is sufficiently strong, has large enough intra-layer electron mobility so that sufficiently high $T_c$ can be achieved, and a matching doping such that the electron concentration in one layer closely matches the hole concentration in the other layer. Two types of structures have been studied: 2D quantum well systems in a perpendicular magnetic field. The idea here is to use the formation of Landau levels and, in particular, the dependence of the number of quantum states per Landau level on the magnetic field as a means to achieve the desired electron and holes concentrations. The exciton condensation in these systems was evidenced by enhanced inter-layer tunnelling [138] or vanishing Hall conductivity [139]. For a review of experimental challenges probing exciton condensation in bilayers we refer the reader to specialised literature [140].

Another bi-layer system that has been intensely studied is the system of two graphene layers separated with a dielectric where EC was predicted to take place at room temperature [141]. The important and much debated issue here is the screening of the inter-layer interaction [142]. The system therefore cannot be described with a simple Hubbard type model for the strongly correlated electrons. For a review of the physics of electron–electron interaction in graphene structures we refer the reader to the specialised literature [143].

6. Outlook

Analogy to superconductivity has been the traditional driving force of the field of exciton condensation. In particular, there was a considerable effort in realisation of supercurrents driven by the gradient of the condensate phase—a hallmark of superfluidity. There are two important conditions to realise a supercurrent. The first is the phase invariance (conduction/valence charge conservation) in the normal phase. The second is the ability to contact separately the electron and the hole parts of the exciton. These conditions can be met in bi-layer systems where the electrons and hole are spatially separated while maintaining sufficiently strong interaction. However, we find it unlikely that similar situations can be realised in bulk materials. Already the first condition is bound to be violated by various one- and two-particle terms in the Hamiltonian as discussed previously. Instead of a superfluid with an arbitrary phase, the system selects either the charge(spin)-density-wave or the charge(spin)-current-density-wave state [11] with a fixed phase. Omitting possible special points in the phase diagram where the symmetry may be enhanced, e.g. close to a second-order boundary between these phases, the system will not exhibit a superfluid behaviour.

The main value of the concept of exciton condensation in bulk materials lies, in our opinion, in providing a comprehensive picture of potentially complex phase diagrams and understanding long-range orders that are not easy to detect because they do not lead to charge(spin)-density modulations on an inter-atomic scale. Moreover, the geometrical form of the order parameter in the EC system may not be as intuitive as, for example, magnetisation in systems with local moments. For example, exciton condensation in $s$-$p$ systems leads to formation of electric dipoles [5] or condensation in a system close to the spin-state transition gives rise to a magnetic multipole order [53]. There are other examples of orbital-off-diagonal orders, e.g. orbital currents in cuprates [144–146], nematic order in iron pnictides [147–149], and hidden order in URu$_2$Si$_2$ [150], where we can only speculate that they may be viewed as a special example of the EC physics. It should be clear from this discussion that the concept of exciton condensation extended beyond the strict superfluid state is not sharply defined and, in particular, the weak-coupling borderline between exciton condensation and ‘just a’ Fermi surface instability is quite fuzzy.

We see numerous open questions and possible directions of further investigation. In the two-band model, realisation of the spin-current-density-wave phase and in general the phase diagram in the presence of cross-hopping are to be explored. The Weiss field in the spin-current-density-wave state can be viewed as a spontaneous spin-orbit coupling—a field that breaks the spin-rotational symmetry but preserves the time reversal symmetry. Interaction between the exciton condensate and the lattice has attracted attention recently [151]. The multi-band systems with multiple orbital flavours of excitons are completely unexplored to our knowledge. A model that can shed some new light on the long-standing problem of perovskite cobaltites is necessary.

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