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Fermions and bosons : excitons in strongly correlated materials

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Conclusions and outlook

Our study of excitons in strongly correlated bilayers has yielded several experimentally testable predictions.

- Using phenomenological Ginzburg-Landau theory we predict that an exciton condensate must exhibit **flux quantization**.[1] [1] Section 2.2.
- The fermionic Hubbard model can describe strongly correlated bilayers. An unreliable mean field theory predicts room-temperature superfluidity.[2] The numerical **Determinant Quantum Monte Carlo** suggests that exciton condensation might occur around 15-20% doping, but the applicability of this method is severely limited by the sign problem.[3] [2] Section 3.2. [3] Section 3.3.
- We derive a low energy bosonic model[4] called the exciton $t-J$ model. For most parts of the phase diagram there is **phase separation** between the superfluid, the exciton solid and the antiferromagnet.[5] [4] Section 4.1. [5] Section 5.2.
- In the limit of low exciton density, there is **frustration** between moving excitons and the antiferromagnetic background leading to Ising confinement. This can be seen in optical experiments of for example undoped YBCO bilayers.[6] [6] Section 4.2.
- Exciton condensation within the $t-J$ model exists at large exciton kinetic energies. There the magnetic triplet excitations 'borrow' kinetic energy from the exciton which is visible in a **large triplon bandwidth**, proportional to the superfluid density.[7] [7] Section 5.1.
- The long-range dipolar interaction might cause the formation of **complex ordered phases**, such as generalized Wigner crystals or stripe phases.[8] [8] Chapter 6.

Despite these several predictions, the theory related to excitons in strongly correlated bilayers is **far ahead of the experimental progress**. This poses a limitation on further theoretical progress, since questions must always be driven by experiments. Nevertheless, there are a few interesting open theoretical questions which are worth mentioning.

Most of our predictions were obtained in the strong coupling limit, where the electron and hole are tightly bound into a boson. It is then natural to ask what happens at intermediate exciton coupling. Most likely the excitons will be spatially **broadened**, reintroducing the complicated fermion sign structure. The inclusion of finite temperatures puts forward the issue of **dissociation** of the excitons into separate holes and electrons. This can be viewed as the extreme limit of spatial broadening. The broadening and formation of excitons is certainly the most interesting open problem regarding the cuprate bilayers, but the fermion sign problem stands in the way of simple answers.

Another possible theoretical direction is to study **exciton-mediated superconductivity**. Superconductivity in the BCS sense[9] requires a bosonic glue to form Cooper pairs, and excitons could play this role.[10] Though this proposal is quite old, there are as of yet no known exciton-mediated superconductors. Condensation of the excitons themselves, especially in the case of imbalanced electron-hole densities, could increase the probability of electron-electron pairing. Whether exciton-mediated superconductivity is truly possible is still an open debate, especially within the cuprate family.

[9] See section 3.2.

[10] Allender et al., 1973; and Inkson and Anderson, 1973

Besides the two major theoretical proposals one can extend the analysis of this thesis to **similar systems**. For example, we can consider different interlayer couplings. In 214 systems such as $La_{2-x}Sr_xCuO_4$ the copper atoms in nearest neighbor layers do not lie directly above each other. Consequently, instead of an interlayer antiferromagnetic coupling one finds a Dzyaloshinskii-Moriya interaction. In addition to different interlayer couplings we could also study different lattice structures, such as the hexagonal lattice.[11] The choice of different lattices and interactions, however, should be guided by actually existing materials that are expected to have these properties.

[11] Meng et al., 2010

Let us therefore discuss the **experimental progress** on cuprate bilayers. The main practical difficulty lies in the fabrication of both

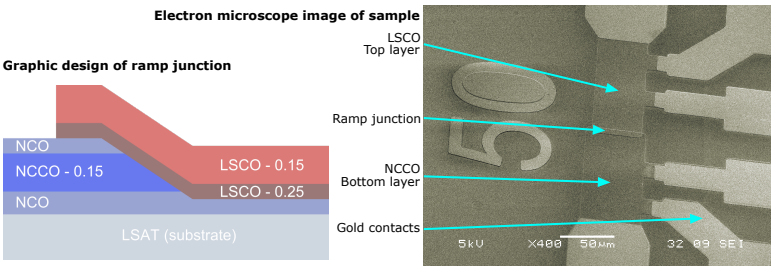

Figure 7.1: In Twente ramp contacts combining p and n-type cuprates were constructed. On the left the design is shown. On the right an electron microscope image of the actual sample is shown. Both layers are visible, and the junction in between them.

p and n-type cuprates in a single sample. Marcel Hoek, Francesco Coneri and Hans Hilgenkamp at the University of Twente are currently making heterostructures of the hole-doped $La_{2-x}Sr_xCuO_4$ (LSCO) and electron-doped $Nd_{2-x}Ce_x$ CuO_4 (NCCO), see figure 3.2 for their crystal structure. This is done by **pulsed laser deposition** (PLD). This technique involves focussing a high power laser on a target in a vacuum chamber, which results in a plasma plume of the target material. The plume deposits on a substrate as a thin film. By tuning the amount of laser pulses, one can construct heterostructures one unit cell layer at a time. The problem is, however, that p- and n-type cuprates need different growth conditions in the PLD process. To obtain superconducting LSCO one needs to anneal the substrate in oxygen, whereas NCCO usually requires annealing in vacuum. The Twente group has managed to successfully create NCCO layers under the growth conditions of LSCO.

Subsequently they have fabricated p/n heterostructures with both LSCO and NCCO in a single sample. The strategy thereby is to first deposit NCCO layers and on top of that an insulator such as $SrTiO_3$. A ramp edge is etched and on top of that a LSCO layer is deposited. This results in a **ramp contact** as shown in figure 7.1.

The p/n contacts are of extreme importance within semiconductor technology. Such p/n junctions consisting of Mott insulators have been little studied.[12] One might for example wonder whether a Josephson current between n and p-type superconductors is possible. Back-of-the-envelope theory predicts that p/n-Josephson junctions might behave qualitatively different from normal Josephson junctions. Transport measurements on these p/n ramp contacts are on the way. At the same time RIXS measurements similar to the one proposed in section 5.1 are started on

[12] Manousakis, 2010

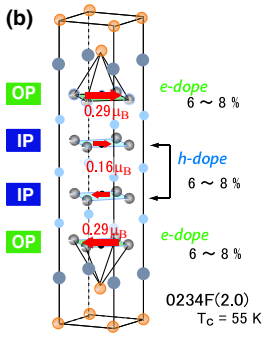

Figure 7.2: In the four-layer cuprate Fo234 selfdoping yields electron- and hole-type layers close to each other. From Shimizu et al., 2007.

[13] Chen et al., 2006

[14] Shimizu et al., 2007

[15] Hotta et al., 2007

[16] See the introductory chapter 1.

p/n heterostructures.

Still, the experimental progress in LSCO/NCCO heterostructures is far away from the p/n bilayers studied in this thesis. Probably the closest connection with experiment this thesis has, lies in cuprates that themselves are bi- or multilayered. In section 4.2 we showed that we expect the dynamical frustration to occur also in the undoped **bilayered YBCO**, for which experiments are underway.

Another Mott compound, **$Ba_2Ca_3Cu_4O_8F_2$ or Fo234**, shows the unique property of selfdoping.[13] In Fo234 the CuO_2 layers come in groups of four. The outer two layers are electron doped and the inner two layers are hole doped (see figure 7.2). One might wonder whether the physics of the exciton $t-J$ model is already at work. Following the phase diagram of figure 5.8, one might expect that Fo234 should exhibit microscopic phase separation between antiferromagnetism and exciton superfluidity. Whereas the latter is not observed (nor excluded), NMR studies[14] clearly show the coexistence of superconductivity and antiferromagnetism. A study of the magnetic excitations, following the work of chapter 4, would further elucidate the interlayer properties in Fo234.

Finally, we mention the novel area of **interface conductance in oxide insulators**, which entails intriguing prospects to realize closely coupled p- and n-type conductors. An example has been provided by Pentcheva et al., 2010 for the case of 2 unit cells of $LaAlO_3$ and 1 unit cell of $SrTiO_3$ grown epitaxially on a TiO_2-terminated $SrTiO_3$ substrate. This research-area also extends to interfaces with Mott insulator compounds such as $LaVO_3/SrTiO_3$.[15]

Despite the considerable distance between theory and experiment, the exciton $t-J$ model gives room for many interesting theoretical advances. Microscopic phase separation, frustration, strongly correlated physics: these are effects that are usually associated with fermions. Now that we find such complicated phenomena in a purely bosonic setting one can investigate the relevance of fermion signs in quantum matter.

7.1 *Propositions on quantum matter*

In this thesis we studied properties of a strongly correlated bilayer, a material that is commonly categorized as 'quantum matter'.[16]

Following the wide variety of quantum theories (phenomenologically, fermionic and bosonic) that were presented in this thesis we are now in the position to state some open questions regarding the research field of quantum matter.

At first glance the 'quantum' distinguishes itself from classical phenomena only through the concept of **superposition**. However, not all superpositions necessarily exclude a classical description. Any macroscopic object such as a coffee mug or an airplane is in a superposition of many of its momentum eigenstates - nonetheless they are clearly classical objects.

One must therefore be more precise in separating the classical from the quantum, by which we now imply superpositions that cannot be untwined into classical objects. Those states are called **entangled**, and the simplest example of an entangled state is two electrons in a singlet state,

$$|\Psi\rangle = \frac{1}{\sqrt{2}} \left(|\uparrow_1 \otimes \downarrow_2\rangle - |\downarrow_1 \otimes \uparrow_2\rangle \right). \tag{7.1}$$

This state has no classical analogue, as is shown by the famous Einstein-Podolsky-Rosen experiment.[17]

[17] Einstein et al., 1935; and Aspect et al., 1982

Observe that the singlet state (7.1) describes two **indistinguishable particles**: we cannot say which of the two particles is in the spin up state, and which is in the spin down state. In fact, there is a close connection between indistinguishability and entanglement. The requirement of indistinguishability implies that the wavefunction of a collection of quantum particles is highly entangled. In the case of fermions the wavefunctions can be written as a **Slater determinant**, thus correctly incorporating the fermion minus signs. For example, three indistinguishable fermions occupying states A, B and C are described by the wavefunction

$$\begin{aligned} |\Psi\rangle &= \frac{1}{\sqrt{6}} \left(|A_1 B_2 C_3\rangle - |A_1 C_2 B_3\rangle + |B_1 C_2 A_3\rangle \right. \\ &\quad \left. - |B_1 A_2 C_3\rangle + |C_1 A_2 B_3\rangle - |C_1 B_2 A_3\rangle \right) \qquad (7.2) \\ &= \frac{1}{\sqrt{3!}} \begin{vmatrix} |A_1\rangle & |B_1\rangle & |C_1\rangle \\ |A_2\rangle & |B_2\rangle & |C_2\rangle \\ |A_3\rangle & |B_3\rangle & |C_3\rangle \end{vmatrix}. \qquad (7.3) \end{aligned}$$

It would be a grave misnomer, however, to classify this state as a quantum material. By introducing anticommuting creation operators, a procedure known as **second quantization**, this state

is simply written as

$$|\Psi\rangle = c_A^\dagger c_B^\dagger c_C^\dagger |0\rangle. \tag{7.4}$$

What we have just described is the free (classical) Fermi gas, which is not a quantum matter at all. Since basically all macroscopic collections of quantum particles are entangled in the Einstein-Podolsky-Rosen sense, we need yet again a different way to understand the difference between classical and quantum matter.

Therefore we will call the many-particle extension of state (7.4) an **antisymmetrized product state**,[18] since the wavefunction can be fully untwined into separate single-particle wavefunctions. Entanglement is now limited to the antisymmetrization required by the indistinguishability. The distinction thus introduced, between states that can be written as (anti)symmetrized product state and those who cannot, truly captures the difference between classical and quantum states.

[18] For many-boson systems this will become a symmetrized product state.

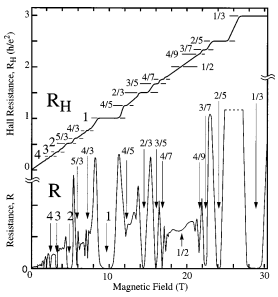

Figure 7.3: A two-dimensional electron gas in a perpendicular magnetic field exhibits plateaus in its Hall resistance. This effect, known as the quantum Hall effect, occurs either at integer or fractional filling fractions. Image from the Nobel Lecture of Stormer, 1999.

A beautiful example of the latter is the Laughlin wavefunction[19] that describes the $\nu = \frac{1}{3}$ **fractional quantum Hall effect** (FQHE),

$$\psi(z_1 \dots z_N) = \prod_{j<k} (z_j - z_k)^3 e^{-\frac{1}{4}\sum_\ell |z_\ell|^2} \tag{7.5}$$

where $z_j = x_j + iy_j$ is the complex coordinate of the jth electron. The single-particle states of an electron in a magnetic field are, in the lowest Landau level, of the form $\psi(z) \sim z^m e^{-\frac{1}{4}|z|^2}$. The construction of an antisymmetrized product state out of these single-particle states yields

$$\psi(z_1 \dots z_N) = \prod_{j<k} (z_j - z_k) e^{-\frac{1}{4}\sum_\ell |z_\ell|^2}, \tag{7.6}$$

the wavefunction of the $\nu = 1$ integer quantum Hall state. The Laughlin state, however, can only be expressed as a **superposition of antisymmetrized product states**. It is therefore considered to be a true quantum liquid.

[19] Laughlin, 1983

The concept of superposing different antisymmetrized states can be taken further, starting with the singlet or **valence bond**[20]

$$|\psi\rangle = \frac{1}{\sqrt{2}} \left(c_{i\uparrow}^\dagger c_{j\downarrow}^\dagger - c_{i\downarrow}^\dagger c_{j\uparrow}^\dagger \right) |0\rangle. \tag{7.7}$$

The quantum paramagnetic phase of the bilayer Heisenberg model consists of such singlets on each interlayer rung,[21] and is therefore

[20] In first quantization language this state is given by $|\psi\rangle = 4^{-1/2}(|i\uparrow\rangle_1 |j\downarrow\rangle_2 - |j\downarrow\rangle_1 |i\uparrow\rangle_2 - |i\downarrow\rangle_1 |j\uparrow\rangle_2 + |j\uparrow\rangle_1 |i\downarrow\rangle_2)$.

[21] See the phase diagram, figure 4.2, and equations (4.2) and (4.37).

called a valence bond solid. A macroscopically entangled extension is the **resonating valence bond** (RVB) state, which consists of a superposition of all possible pairings of two particles into a singlet state. When high T_c superconductivity was discovered, Anderson proposed that the cuprates can in fact be described by a long-range RVB state.[22]

[22] Anderson, 1973; and Anderson, 1987

But is it truly a long-range quantum entangled state? The long-range RVB state on a square lattice has a finite overlap with the **Néel state** describing antiferromagnetism,[23]

[23] Liang et al., 1988

$$|\Psi\rangle_{\mathrm{AF}} = |\uparrow_1 \downarrow_2 \uparrow_3 \cdots\rangle = \prod_{i\in A} c^\dagger_{i\uparrow} \prod_{j\in B} c^\dagger_{j\downarrow}|0\rangle \tag{7.8}$$

where the lattice is broken into two sublattices A and B. The Néel state, however, is a clearcut example of a product state. So is the ground state of the Heisenberg model (4.10) a quantum state or a classical state?

The antiferromagnetic long-range RVB state of Liang et al., 1988 is the currently best known approximation to the ground state of the Heisenberg model. It satisfies the rule designed by Marshall, 1955, who proved that the ground state of the Heisenberg model can be written as a superposition of spin configuration states $\mathcal{C}$,

$$\begin{aligned} |\Psi\rangle_0 &= w_1|\uparrow_1 \downarrow_2 \uparrow_3 \cdots\rangle + w_2|\downarrow_1 \uparrow_2 \uparrow_3 \cdots\rangle + \ldots & (7.9)\\ &\equiv \sum_{\mathcal{C}} w_{\mathcal{C}}|\mathcal{C}\rangle & (7.10)\end{aligned}$$

where the sign of each weight $w_{\mathcal{C}}$ is determined by the number of up spins on the A sublattice,

$$w_{\mathcal{C}} = (-1)^{N^\uparrow_A}|w_{\mathcal{C}}|. \tag{7.11}$$

With the **sign structure** thus imprinted into the set of basis kets, the Heisenberg model on a square lattice at half-filling can be described purely in terms of positive-definite weights. Similarly, in the high-temperature expansion or Suzuki-Trotter decomposition[24] of $e^{-\beta H}$ all statistical weights and/or matrix elements are positive definite.

[24] See section 3.3.

On a frustrated lattice such as the **triangular lattice**, however, a short-range RVB state may be the lowest in energy.[25] This might indeed constitute a realization of a quantum material. Another obvious candidate is the ground state of the Hubbard model at finite doping.[26]

[25] Anderson, 1973

[26] See section 3.1.

Summarizing, we defined quantum materials as a superposition of different antisymmetrized product states. However, once the sign structure of this superposition is known one can relate the apparent quantum state to a classical product state, as is shown for the long-range RVB and Néel state. The main question therefore remains whether there exist *macroscopic quantum (long-range entangled) states which cannot be reduced to classical (antisymmetrized product) states*. In terms of the fermion sign problem language we should ask whether there are superposition states with an **irreducible sign structure**.

In the case of the exciton $t-J$ model presented in this thesis, we implicitly employed this reduction. Even though the model has an as of yet unknown sign structure,[27] variational mean field theory[28] yields a product state that has finite overlap with the true ground state. Similarly the recent experimental detection of 'current loop order' in cuprates[29] suggests that in the end a quantum sign-full strongly correlated material might still be represented by a classical product state.

[27] See section 4.1.4.

[28] See section 5.2.

[29] Varma, 1997; Shekhter et al., 2013; and Zaanen, 2013

Once the correct product state is found, the question arises what the **quantum corrections** should be. A systematic way to answer this is by means of spin wave theory: equation (4.63) in section 4.2.1 shows how the ground state of the Heisenberg model can be approximated by a superposition $|\psi_0\rangle$ of the Néel state $|G\rangle$ and its quantum corrections. Many authors call these corrections **quantum fluctuations**, but I perceive this as a misleading picture. *There is nothing fluctuating about a superposition.* For example, saying that the singlet state (7.1) is fluctuating between $|\uparrow_1\downarrow_2\rangle$ and $|\downarrow_1\uparrow_2\rangle$ fundamentally misunderstands the quantum nature of that state.[30]

[30] Note that the spin liquid community uses the words 'fluctuation' and 'superposition' interchangeably, see the review by Balents, 2010.

How a macroscopic superposition should be understood is therefore an open question. Experimental realization of superpositions of classically distinct configurations is being pursued by several research groups. A superposition of two opposite circular supercurrents in a SQUID is a prime example thereof.[31] Following the phase diagram of section 5.2 another suggestion is to consider **first order quantum phase transitions**. At such a transition, there are two macroscopically fundamentally different states with the same energy which can therefore be put in a superposition. The possibility of a superposition is competing with the more classical first order effects of phase separation and the idea that the system can actually be fluctuating between the two phases.[32] *First*

[31] Friedman et al., 2000; and van der Wal et al., 2000

[32] As for example in supercooled water, where numerics suggest 'phase flipping' between the high and low density liquid. Kesselring et al., 2012.

order quantum phase transitions are therefore ideal candidate systems to elucidate the notions of quantum superpositions and fluctuations.

Yet another way to combine two different phases of matter is as a **statistical mixture** of states. This is done within the thermal density matrix formalism. In general, a quantum system at temperature T is believed to be described by the density matrix $\rho = e^{-H/k_B T}$. However, in the case of **spontaneous symmetry breaking** ρ is at best ill-defined in the thermodynamic limit and at worst incorrect: a magnet is never in a mixed state of its different possible magnetization directions. The fact that an infinitesimal symmetry breaking field radically changes the density matrix implies that ρ in zero field is singular. On top of that, the thermal density matrix misses interesting superposition effects such as in the aforementioned SQUID experiments.[33] Therefore, *a good understanding of quantum matter at finite temperature, specifically regarding the role of macroscopic superpositions and entanglement, requires a novel approach beyond the thermal density matrix.*

[33] Which can be viewed as a magnet in a superposition of two opposite magnetizations.

In this thesis we have considered the existence of fermions as fundamental. A completely different approach to quantum matter discards this notion of fermions as being fundamental entities. Just like phonons[34] are emergent quantized particles, fermions could be **emergent**. Examples are the emergence of fermions in string-net condensates[35] or in complex weighted networks.[36] Close to the Mott state, the fermions might not even act as fermions due to their localization constraint, leading to novel statistical effects.[37] In one-dimensional systems effects such as spin-charge separation of fermions are well understood. Nevertheless, *in higher dimensions the breakdown or emergence of fermionic behavior deserves more research attention*.

[34] Phonons are quantized lattice vibrations.

[35] Wen, 2007

[36] Garlaschelli and Loffredo, 2009

[37] Zaanen and Overbosch, 2011

Let us conclude that the mysteries of quantum mechanics become increasingly relevant in the understanding of actually existing materials, such as cuprates. This presents the opportunity to study basic quantum phenomena without the need of building billion-dollar accelerators or satellites. Instead, the greatest mysteries of modern condensed matter can be held in one's hand. And hopefully, just like the specific heat anomaly experiments paved the way for the development of quantum statistical theory, the current stream of ill-understood experimental results will lead to new fundamental insights into the laws of nature.