Fermions and bosons : excitons in strongly correlated materials
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1
Introduction: Quantum Matters

Einstein's explanation of the photoelectric effect is often quoted as the birthplace of quantum mechanics, the theory used in this thesis. I think it is misleading to take the thus-proposed quantization, which was already apparent in experiments, as the starting point of the quantum. Instead, in my opinion, we should start with Planck's theory of blackbody radiation. On 14 December 1900 Max Planck presented his idea that electromagnetic radiation can only be emitted in quantized form in order to explain the relation between temperature, frequency and intensity. Without the ability to detect the postulated quantum directly, the idea of quantization is truly revolutionary.

Similarly, the Einstein and Debye theories of the low temperature anomalies in the specific heat of crystalline solids proposed the quantization of vibrational energy. What these theories have in common is that they combine both quantization of energy levels with a large number of degrees of freedom. The in popular culture so frequently mentioned 'uncertainty' associated with quantum mechanics is completely irrelevant for these quantum statistical systems,[1] such as the complex materials studied in this thesis.

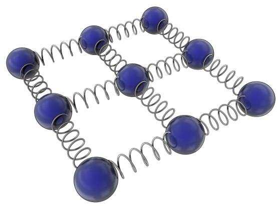

Figure 1.1: A crystal solid can be viewed as atoms connected by springs. In the quantum theory of a solid, the springs can only vibrate at fixed frequencies. This quantization of vibrational modes explains the low-temperature behavior of solids.

[1] More on quantum statistics can be found in Lifshitz and Pitaevskii, 1980, Abrikosov et al., 1965, Mahan, 2000, Wen, 2007 and Coleman, 2013.

When it comes to statistics it is relevant, however, that energy quanta are indistinguishable. Indistinguishable can be best explained by the following coin-flipping example. If I flip two coins elementary probability theory will tell you that the chance of two heads is 25%. However, as Bose famously discovered while making an error during a lecture, if the two coins are fundamentally indistinguishable the probability is 33%. This is the case for **bosons** and from there the Bose-Einstein distribution function

$$n_{BE}(E) = \frac{1}{e^{E/kT} - 1} \tag{1.1}$$

directly follows. Bosons have the unique property that they want to do the same thing as other bosons around them, which can already be inferred from the coin-flipping thought-experiment where the probability of finding the same sides is greatly increased.[2]

[2] Dirac, 1958

This is in stark contrast to electrons in atomic orbitals. There the Pauli exclusion principle dictates that no more than two electrons can be in one orbital. Hence a second species of particles must exist that go by the name of **fermions**, described by the Fermi-Dirac distribution

$$n_{FD}(E) = \frac{1}{e^{E/kT}+1}. \tag{1.2}$$

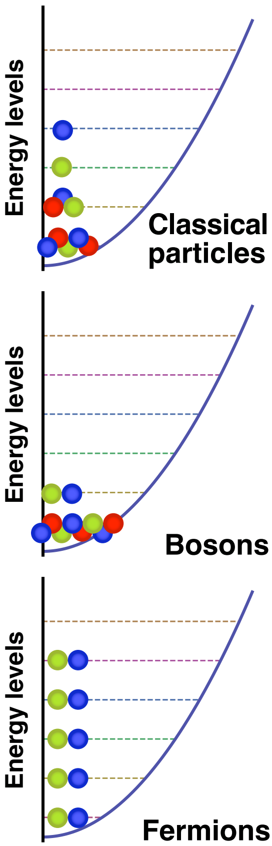

Figure 1.2: An ensemble of particles is distributed among a set of energy levels. Classical particles obey the Maxwell-Boltzmann distribution. Relative to that the conformist bosons tend to cluster together in the lowest energy state. Fermions, on the other hand, have an exclusion principle that limits the number of particles per state.

The Pauli exclusion principle applied to electrons in a crystal immediately suggests that electrons have a very high energy. The lowest energy state of a collection of electrons amounts to filling up energy levels up to the Fermi level E_F, which is typically several electronvolts. Therefore, even at low temperatures the average kinetic energy per electron is huge. In fact, it equals $\frac{3}{5}E_F$ whereas such a kinetic energy per particle in a classical gas is only obtained at a temperature of thousands of Kelvins. Indeed, the Pauli exclusion principle implies that the seemingly boring Fermi sea is in fact a hot boiling active soup of electrons.

The electron band theory that thus originated laid the foundations for our understanding of electrons in metals, insulators and last-but-not-least semiconductors. The latter class of materials have revolutionized our modern world: basically everybody on every corner of the world carries semiconductor technology in her or his pockets. To me this shows that investing in fundamental physics (in this case quantum statistics) leads to practical applications, albeit in a completely different form than Pauli and colleagues would have imagined.

We just saw that there are two quantum species: the bosonic conformist particle and the fermionic individualistic particle, see figure 1.2. However, this is not the whole story: additionally the fermions have a weird property called '**anticommutativity**', which means that creating first particle A and then particle B is equal to 'minus' creating them in the opposite order. In the language of second quantization[3] one explicitly sees the appearance of a minus sign,

[3] 'Second' quantization means that all quantum operators are expressed in terms of creation and annihilation operators of (fermionic or bosonic) particles.

$$c_A^\dagger c_B^\dagger = -c_B^\dagger c_A^\dagger, \tag{1.3}$$

Figure 1.3: The high temperature superconductors are ill understood strongly correlated materials, and they are a prime example of quantum matter. Here the author is shown levitating a piece of superconducting YBCO at the Twente University.

which causes mathematical problems when describing a large set of fermions. Often this anticommutativity, or **fermion sign problem** as it is sometimes called, does not pose a problem, when the quantum mechanical wavefunction of a system can be written as a **product state** of simpler constituents.

The first example of a product state is the non-interacting Fermi gas,

$$|\Phi\rangle_{\mathrm{FG}} = \prod_{\mathbf{k}\sigma}^{\mathbf{k}_F} c^{\dagger}_{\mathbf{k}\sigma}|\mathrm{vac}\rangle \tag{1.4}$$

where $c^{\dagger}_{\mathbf{k}\sigma}$ is the creation operator of an electron with momentum $\mathbf{k}$ and spin σ. The product now runs over all momentum states up to the Fermi momentum. Product states also arise in the case of the formation of some kind of long-range order via spontaneous symmetry breaking. Examples are (anti)ferromagnets, crystalline solids, Bose condensates and superconductors; the latter is described by the BCS wavefunction[4]

[4] Bardeen et al., 1957; and De Gennes, 1999

$$|\Phi\rangle_{\mathrm{BCS}} = \prod_{\mathbf{k}\sigma} \left(u_{\mathbf{k}} + v_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\uparrow} c^{\dagger}_{-\mathbf{k}\downarrow} \right) |\mathrm{vac}\rangle \tag{1.5}$$

with $u_{\mathbf{k}}, v_{\mathbf{k}}$ some momentum dependent parameters. Any quantum system that can be written as a product state can be effectively described as a classical system, corrected with the proper Fermi-Dirac or Bose-Einstein statistics. The ordered product states are called **classical condensates**.[5]

[5] See Anderson, 1984 and Zaanen, 1996.

There is, however, a class of materials that cannot be simply written as a product state, usually due to strong electron-electron interactions[6] and the resulting macroscopic entanglement. For these materials the quantum fermion signs do matter and the

[6] Imada et al., 1998; and Lee et al., 2006

weird non-locality (as Wen, 2007 calls it) of fermions plays an important role. It has become customary to denote all these highly entangled materials '**quantum matter**', since no classical analogue or theory exists.

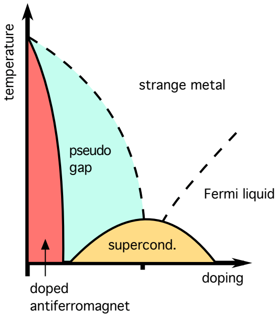

Figure 1.4: Generic phase diagram of a cuprate material such as $La_{2-x}Sr_xCuO_4$. Upon doping away from half-filling the antiferromagnetic order is reduced, and the poorly understood pseudogap and strange metal appear.

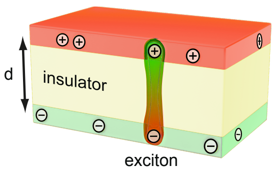

Figure 1.5: A heterostructure consisting of a hole layer and an electron layer separated by an insulator. Interlayer excitons can form as the bound state of an electron and a hole.

Strongly correlated electron systems are an example of quantum matter. While the observation that there exist materials that do not obey band theory was already made in the 50s by Van Vleck, Mott and others, a revived interest in **Mott insulators** sparked after the discovery of high-temperature superconductivity in cuprates (figure 1.3). Embarrassingly enough, there are still no theories that provide a satisfactory explanation of the many phenomena observed in the cuprates, from the fate of the Mott insulating state upon doping, to the pseudogap, the strange metal, the proposed electronic nematicity, etcetera, see figure 1.4. The main reason for this lack of understanding lies in the aforementioned 'weirdness' of fermionic excitations in cuprates.

Whilst doped Mott insulators are little understood, rapid progress is made in materials where the dominant excitations are bosons. The simplest way to make a bosonic system is to couple two electrons together, as is done in the BCS theory of superconductivity. Similarly, one can couple electrons and holes together into **excitons**. While the binding of an electron and a hole into an exciton has the advantage of the much stronger Coulomb attraction, the possible recombination and annihilation of an exciton prevents the practical realization of a so-called exciton condensate. However, if one is able to spatially separate the electrons and holes into separate layers, as shown in figure 1.5, annihilation can be suppressed[7] and an equilibrium density of excitons can be created. Over the last decade such bilayer systems came experimentally within reach, first in quantum Hall bilayers[8] and more recently in systems without magnetic field.[9]

[7] Shevchenko, 1976; and Lozovik and Yudson, 1976

[8] Eisenstein and MacDonald, 2004; and Huang et al., 2012

[9] High et al., 2012

In this thesis I combine the field of strongly correlated materials with the bilayer exciton community. No-one in their right mind would mix those two fields, unless driven by experiments. My theoretical pursuits are therefore rooted in the rapid technological revolution that has occurred in the fabrication of nanomaterials. Using for example pulsed laser deposition (PLD) one can make material 'sandwiches' where the chemical composition of each separate atomic layer is controlled independently. My theoretical research is in close collaboration with the experimental Interfaces in Correlated Electron systems group of Hans Hilgenkamp at the University of Twente, where they possess such technologies.

The ill-understood Mott materials, together with bilayer exciton ideas and the technological nanorevolution form the basis for this research. My main research questions are thus: *Can we make an exciton condensate in a strongly correlated bilayer? And what are the observable properties of such a condensate?* Given these concrete research questions, we hope to understand more about the complex interplay between fermions and bosons in quantum matter.

Outline

The research on excitons in strongly correlated materials addresses the interplay between fermionic and bosonic excitations. Many properties of a system, however, do not depend on the microscopic mechanisms involved, therefore we start with a **phenomenological description** of exciton condensates in chapter 2. Using a Ginzburg-Landau theory we show that an exciton condensate exhibits a flux quantization property.

After that we dive into the microscopic degrees of freedom. The starting point is the **fermionic Hubbard model**, introduced in chapter 3. Weak coupling mean field theory and numerical analysis of the Hubbard model do give us some answers, however, the fermion sign problem plagues the theory.

On the other hand, in the strong coupling limit one can rewrite the fermionic Hubbard model into a purely **bosonic exciton** $t-J$ **model**. In chapter 4 we introduce this new model and discuss its properties. There we find the dynamical frustration of a single exciton in a Mott insulating antiferromagnet. Chapter 5 discusses the properties of the strongly correlated exciton condensate, where we find spin-exciton cooperation. Indeed, the exciton condensate can exist, but only if the kinetic energy of the excitons exceeds their dipole-dipole repulsion. The influence of this dipole-dipole repulsion on possible ordering phenomena is discussed in chapter 6.

Core concepts, like the Hubbard model or strong coupling perturbation theory, are best introduced in the context of the research findings instead of in a separate introductory chapter. Hence, whenever necessary background information will be included in the text or in references in the remaining chapters.

Finally, theoretical research should always be addressing 'actually existing materials'. The main challenge in this regard is to couple down-to-earth computations with insights in general physical concepts. In the **concluding chapter** 7 we put our findings on strongly correlated bilayers into a broader context and relate it to experimental progress, thereby addressing future directions in the theory of quantum matter.