Fermions and bosons : excitons in strongly correlated materials
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Phenomenology of exciton bilayers

WITHOUT SPECIFIC KNOWLEDGE of microscopic details one can still describe to an amazing accuracy a large set of physical properties of any system. This is rooted in the vastness of degrees of freedom, so that statistical effects dominate the physics. An effective free energy based on symmetry principles can then be constructed, explaining macroscopic phenomena.

This was initially done by Ginzburg and Landau to describe superconductivity in the early 40s, based on the realisation that the order parameter relevant to superconductivity is a (charged) complex field. In this chapter we will introduce the field of bilayer exciton condensates in section 2.1, and use symmetry arguments to write down a phenomenological theory of these systems. In section 2.2 we deduce its magnetic response and the prediction of flux quantisation.
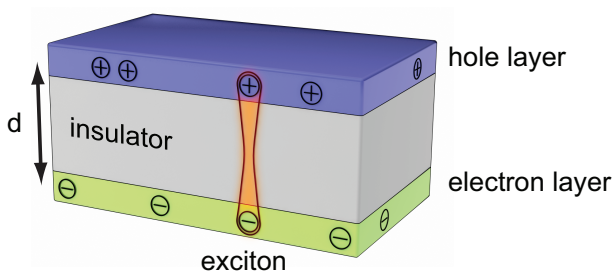

Figure 2.1: Excitons in double layer devices. Double layer devices consist of an electron layer (in green) parallel to a hole layer (blue), separated by an insulating barrier (grey). Electron-hole attraction leads to the formation of excitons. At sufficiently low temperatures the excitons can form a Bose-Einstein condensate.
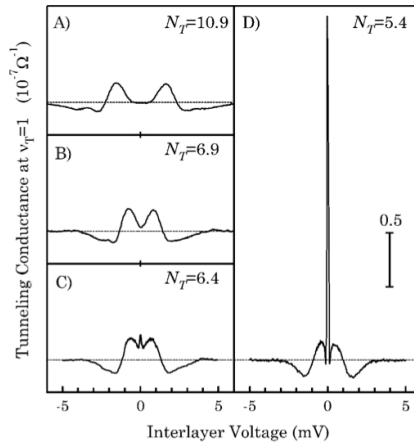
2.1   Bilayer excitons and condensation

In a semiconductor the elementary charged excitations are particles and holes, and naturally these two excitations attract each other via the Coulomb force. A trivial non-charged excitation is therefore composed of both an electron and a hole: the **exciton**.

Directly after the BCS theory of superconductivity it was suggested that excitons, as they are in many regards comparable to Cooper pairs, can also Bose condense and exhibit superfluidity.[1] Whilst the Coulomb force that binds the exciton is indeed orders of magnitude stronger than the phonon glue in Cooper pairs, the major difficulty for exciton condensation lies in their finite lifetime. That is, excitons annihilate when the electron and hole recombine.

[1] Blatt et al., 1962; Keldysh and Kopaev, 1965; and Moskalenko and Snoke, 2000

Nonetheless, exciton annihilation can be suppressed by spatially separating the electrons and holes. This is achieved by constructing a heterostructure where one sandwiches an insulating layer in between a two-dimensional electron gas and a two-dimensional hole gas, see figure 2.1. The Coulomb attraction is, due to its long-range nature, not substantially reduced and therefore electrons and holes can still form bound states. This is called a bilayer exciton, double layer exciton or **interlayer exciton**.

Figure 2.2: Interlayer tunneling in a quantum Hall bilayer as a function of bias voltage, for different magnetic fields. The enhancement of the interlayer tunneling at zero bias is the result of the exciton condensation and is a direct measurement of the order parameter. From Spielman et al., 2000.

At low enough temperatures these interlayer excitons form a superfluid.[2] Unlike many other superfluids, the bilayer nature allows one to directly probe the **superfluid order parameter**. The exciton condensate order parameter reads

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

$$\Delta_{\mathbf{k}} = \langle c^{\dagger}_{\mathbf{k}1\sigma} c_{\mathbf{k}2\sigma} \rangle. \qquad (2.1)$$

where $c^{\dagger}_{\mathbf{k}1\sigma}$ creates an electron in the first layer and $c_{\mathbf{k}2\sigma}$ creates a hole in the second layer. One directly observes that this order parameter amounts to anomalous interlayer tunneling.[3] The onset of condensation is therefore sparked by a dramatic increase of the

[3] Moon et al., 1995; Spielman et al., 2000; and Eisenstein and MacDonald, 2004

interlayer tunneling, as is shown by the experimental results in figure 2.2.

One can question, however, to what extent the system created in this way is really a superfluid. Obviously, since excitons are neutral, there cannot be an electric supercurrent in these systems. However, since the electrons and holes are spatially separated an interlayer exciton current amounts to two opposite but equal countercurrents. By connecting the two layers in series one can measure the resulting **counterflow superfluidity**[4] as is shown in figure 2.3.

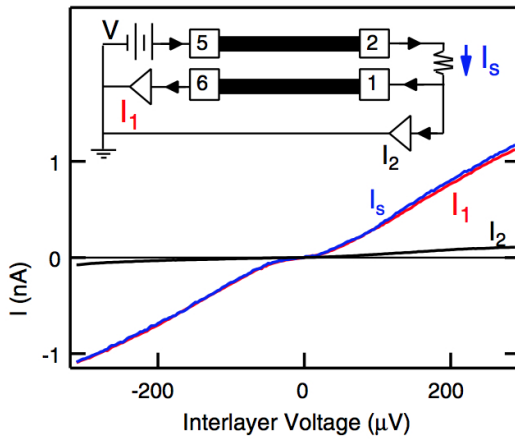[4] Su and MacDonald, 2008; and Finck et al., 2011

Figure 2.3: Near-perfect counterflow is observed in quantum Hall bilayers in a corbino geometry. A current is induced in the top layer (from 5 to 2) and measured in the bottom layer (from 1 to 6). The result $I_s \approx I_1$ suggests that in the bilayer the electric current is carried by superfluid excitons. From Finck et al., 2011.

As of 2013, the field of interlayer excitons is still restricted to a small class of material systems: the quantum Hall bilayers of figures 2.2, 2.3 and Huang et al., 2012; and laser-pumped quantum wells.[5] There are many other possible candidate materials to realise interlayer condensation, such as graphene sheets[6] or topological insulators[7]. In this thesis we consider strongly correlated electron bilayers as candidate materials,[8] the microscopic properties of which will be discussed in the next two chapters.

Besides different materials there are also different experimental probes possible. The current experiments focus mainly on transport properties. One can argue that these do not necessarily prove the existence of an interlayer exciton condensate. To directly probe the coherence associated with the superfluid, we propose a flux quantisation effect.

[5] High et al., 2012

[6] Lozovik and Sokolik, 2008; Zhang and Joglekar, 2008; Dillenschneider and Han, 2008; Kharitonov and Efetov, 2008; and Min et al., 2008

[7] Seradjeh et al., 2009

[8] Pentcheva et al., 2010; and Millis and Schlom, 2010

2.2   *Ginzburg-Landau theory and flux quantisation*

We can find the flux quantisation and related magnetic properties
of an exciton bilayer condensate using **Ginzburg-Landau theory**.
This amounts to constructing a free energy functional $\mathcal{F}[\Psi]$ for
the order parameter field. In the case of interlayer excitons, the
order parameter must describe the bound state of an electron in
one layer and a hole in the other layer, as in equation (2.1). The
exciton is therefore charge neutral, but it does possess an electric
dipole moment. This dipole moment is the starting point for the
derivation of magnetic properties.

As the direction of this electric dipole is fixed, the exciton su-
perfluid is characterized by just a complex scalar order parameter
field $\Psi(\vec{x}) \equiv |\Psi(\vec{x})|e^{i\phi(\vec{x})}$ along a 2D surface, the square of which
gives the superfluid density $\rho(\vec{x}) = |\Psi(\vec{x})|^2$. For a charged su-
perfluid/superconductor with boson charge q electromagnetism
is incorporated by replacing ordinary derivatives with **covariant
derivatives** $\vec{D}$,

$$\hbar\vec{D} = \hbar\vec{\nabla} + iq\vec{A}(\vec{x}) \qquad (2.2)$$

where $\vec{A}(\vec{x})$ is the vector potential. In the charge-neutral exciton
superfluid the electron and hole constituents of an exciton form
an electric dipole $e\vec{d}$ and consequently the covariant derivative
associated with exciton matter must equal[9]

$$\hbar\vec{D} = \hbar\vec{\nabla} + ie\left[\vec{A}(\vec{x}+\vec{d}/2) - \vec{A}(\vec{x}-\vec{d}/2)\right] \qquad (2.3)$$

where the electron is positioned at $\vec{x} - \vec{d}/2$ and the hole at $\vec{x} +
\vec{d}/2$. For small interlayer distance $\vec{d}$ the vector potential can be
expanded in a Taylor series. In addition, since the vector potential
$\vec{A}$ along the 2D superfluid surface is only sourced by in-plane
currents, we can impose that the gradient of the vector potential
component perpendicular to the surface is zero,

$$\left.\vec{\nabla}'\left(\vec{d}\cdot\vec{A}(\vec{x}')\right)\right|_{\vec{x}'=\vec{x}} = 0. \qquad (2.4)$$

This implies that the above vector potential difference can be
written completely in terms of the 'real' magnetic field

$$\vec{A}(\vec{x}+\vec{d}/2) - \vec{A}(\vec{x}-\vec{d}/2) =$$

$$-\vec{d}\times\sum_{k=0}^{\infty}\frac{1}{(2k+1)!}\left(\frac{\vec{d}}{2}\cdot\vec{\nabla}'\right)^{2k}\left.\vec{B}(\vec{x}')\right|_{\vec{x}'=\vec{x}}. \qquad (2.5)$$

Up to first order the exciton covariant derivative turns into

$$\hbar \vec{D} = \hbar \vec{\nabla} - ie\vec{d} \times \vec{B}. \tag{2.6}$$

This is an interesting structure viewed from a theoretical perspective. Equation (2.6) corresponds to the covariant derivatives of a $SU(2)$ gauge theory with gauge fields $A_i^a = \epsilon^{iak} B_k$. Here the $SU(2)$ gauge fields are actually physical fields fixed by Maxwell's equations. Using the above considerations we can write down a general **Ginzburg-Landau free energy**

$$
\begin{aligned}
\mathcal{F}[\Psi] \;=\; & \int d^2 x \left[\alpha|\Psi|^2 + \frac{1}{2}\beta|\Psi|^4 + \frac{\hbar^2}{2m^*}(\nabla|\Psi|)^2 \right. \\
& \left. + \frac{1}{2m^*}\left[\hbar\vec{\nabla}\phi - e\vec{d}\times\vec{B} \right]^2 |\Psi|^2 + d\frac{B^2}{2\mu_0} \right]. \tag{2.7}
\end{aligned}
$$

The parameters α and β can be written formally as a function of the superfluid density and the critical magnetic field B_c. Minimization of the free energy assuming a constant order parameter yields

$$\alpha = -d\frac{B_c^2}{\mu\rho}, \tag{2.8}$$

$$\beta = -\frac{\alpha}{\rho}. \tag{2.9}$$

2.2.1 Electromagnetic response

The direct coupling to physical fields changes the rules drastically as compared to normal superconductors. We define the exciton supercurrent as the standard Noether current[10] $\vec{j} \equiv \frac{\hbar\rho}{m^*}\vec{\nabla}\phi$. Consequently, minimizing the free energy for a fixed applied magnetic field $\vec{B}$ perpendicular to the dipole moment yields the **exciton supercurrent response**

$$\vec{j} \equiv \frac{\hbar\rho}{m^*}\vec{\nabla}\phi = \frac{\rho e}{m^*}\vec{d}\times\vec{B}. \tag{2.10}$$

This result is closely related to spin superfluids[11] where a 'physical field' $SU(2)$ structure arises through spin-orbit coupling.[12] The analogue of Eq. (2.10) is the spin Hall equation[13] $j_j^i = \sigma_s \epsilon^{ijk} E_k \rightarrow \vec{j} = -\sigma\vec{d}_m \times \vec{E}$. We conclude that the spin-superfluid formed from magnetic dipoles is the electromagnetic dual of the exciton (electric dipole) superfluid.

In the double layer system the electric charges forming the exciton dipoles are confined in the separate layers. Hence the exciton supercurrent can be decomposed into the separated electron and hole surface currents. According to Ampère's law, a surface current induces a discontinuity in the magnetic field components parallel to the surface,

$$\Delta \vec{B}(\vec{x}) = \mu_0 \vec{K}(\vec{x}) \times \hat{n}, \tag{2.11}$$

where $\hat{n}$ is the normal vector to the surface and $\vec{K}(\vec{x})$ is an electric surface current density. Consequently, an exciton supercurrent reduces the magnetic field in between the electron and hole layer. The double layer therefore acts as a (non-perfect) **diamagnet** with magnetic susceptibility

$$\chi_m = -\frac{e^2 \rho d \mu_0}{m^*}. \tag{2.12}$$

For typical parameters $\rho = 0.4$ nm^{-2}, $d = 20$ nm and $m^* = 2m_e$, the magnetic susceptibility equals $\chi_m = -10^{-4}$, comparable to what is found in good diamagnets like gold or diamond. In semiconductor quantum wells, the exciton mass is smaller than the free electron mass m_e which enhances the diamagnetic susceptibility even further.[14]

[14] Butov, 2003

2.2.2   Flux quantization

Imposing single-valuedness on the order parameter implies that for any given contour C inside a superfluid $\oint_C \vec{\nabla}\phi \cdot \vec{dl} = \oint_C \vec{j} \cdot \vec{dl} = 2\pi n$ where n is an integer. Therefore, **circular supercurrents must be quantised**, which can be seen by topological defects in the dipolar superfluid.[15] In general, metastability of superflows requires a nontrivial topology of the superfluid.[16] Unlike in other superfluids, the $SU(2)$ structure of dipolar superfluids implies the possibility of more complicated topologies which cannot be obtained by creating defects in the superfluid.

[15] Babaev, 2008; Seradjeh et al., 2008; and Leurs et al., 2008
[16] Leggett, 2006
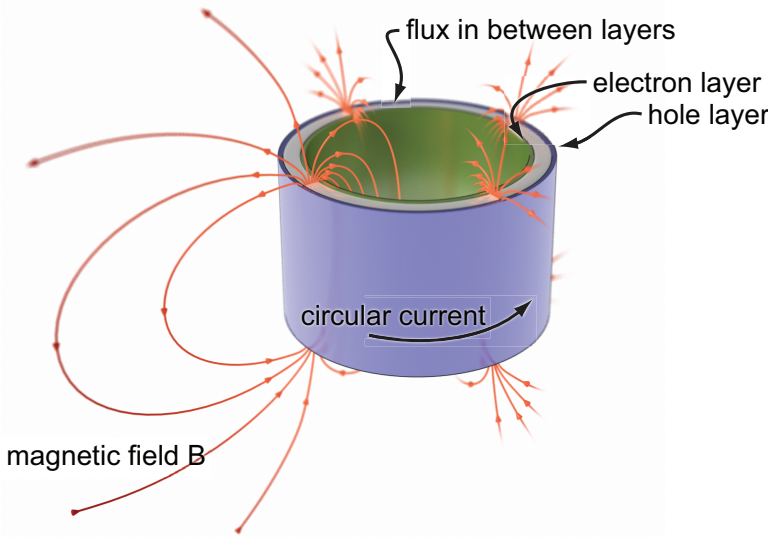
Consider a cylindrical device of radius r consisting of two concentric layers as shown in figure 2.4 with the electric dipole moment $\vec{d}$ of the excitons pointing in the radial direction. For this geometry the current-dependent term in the free energy can be written as

$$\mathcal{F}[\Psi] \sim \oint d\theta \left[\frac{h}{e} \partial_\theta \phi - B_z 2\pi r d \right]^2 \tag{2.13}$$

where $\oint_C d\theta \partial_\theta \phi = 2\pi n$ with n integer valued and B_z is the external magnetic field. Note that the flux going in between the two layers equals up to first order $\Phi = B_z 2\pi r d$. Minimisation of equation (2.13) shows that current quanta can be induced by an axial magnetic field. In the absence of the external field, the current $\vec{j} \sim n$ induces a **magnetic flux** in between the layers, according to Ampère's law (2.11), with a magnitude

$$\Phi = \frac{h}{e}\,\chi_m\, n \equiv \Phi_0\, \chi_m\, n. \qquad (2.14)$$

This is our central result: in the cylindrical double layer geometry, the magnetic flux going in between the sample layers must be **quantised** in units of χ_m times the fundamental flux quantum $\Phi_0 = \frac{h}{e}$. Notice that this flux quantisation effect is quite different from the one realised in superconductors. In the double layer exciton condensate the supercurrent is induced by the magnetic field $\vec{B}$ rather than the gauge field $\vec{A}$ as in the London equation, while the quantised amount of flux equals $d \oint \vec{B} \cdot \vec{dl}$ instead of $\oint \vec{A} \cdot \vec{dl} = \int \int_\Sigma \vec{B} \cdot \vec{d\Sigma}$ for superconductors. In combination these two basic differences add up to an universal expression for the flux quantisation $\Phi = \frac{h}{e^*}\chi_m\, n$ that applies to both superconductors and exciton condensates, where $e^* = -2e$ and $\chi_m = -1$ for superconductors.

Is the strength of the condensate actually sufficient to trap the flux? When the external field is switched off the flux carrying state is metastable and the system can return to the ground state by locally destroying the condensate: the **phase slip**. The condensate can only be destroyed over lengths greater than the Ginzburg-Landau coherence length

$$\xi = \frac{\hbar}{\sqrt{|2m^*\alpha|}} \tag{2.15}$$

and consequently the energy required to break the condensate over a region ξ wide along a cylinder of length z is

$$\delta F_b = \frac{1}{2}\hbar z \left(\frac{d\,\rho}{2m^*\mu}\right)^{1/2} B_c. \tag{2.16}$$

Locally destroying the condensate is only favourable if this energy is lower than the energy stored in the magnetic field, which is $\delta F_m = \frac{B^2}{2\mu}2\pi rd$. We conclude that a **phase slip** will not occur as long as the trapped magnetic flux $\Phi = \Phi_0\chi_m n$ stays below a threshold value,

$$\Phi^2 < \left(\frac{|\chi_m|}{2}\right)^{1/2} \Phi_0 B_c\,rd. \tag{2.17}$$

where B_c is the critical magnetic field. With the typical parameters stated above and $r = 100\ \mu$m, the critical field must exceed 5 nT to trap one flux quantum. Since the critical magnetic field of bilayer superfluids is proposed to lie in the orders of tens of Teslas[17], a phase slip is improbable.

[17] Balatsky et al., 2004

Another possible complication is that annihilation of excitons by tunneling causes the phase to be pinned which introduces a threshold for the formation of stable currents. Microscopic tunneling can be incorporated via an extra term in the Ginzburg-Landau free energy

$$\mathcal{F}_t = -2t \int d^2x \frac{|\Psi|}{L} \cos\phi, \tag{2.18}$$

where L is the in-plane lattice constant and t is a microscopic tunneling energy. This **phase pinning** lowers the energy of the state where no flux is trapped, which introduces a threshold for the trapping of magnetic flux quanta. It is only possible to trap n

magnetic flux quanta if the microscopic tunneling energy t satisfies

$$2t < n^2 \frac{\hbar^2}{2m^*r^2} \sqrt{\rho}L. \qquad (2.19)$$

This corresponds, given the typical parameters mentioned above, to $t < 0.3$ peV (pico-electronvolt) for the first flux quantum.

In order to estimate a value for t, let us imagine that the device is fabricated from copper-oxide layers. The hopping energy in cuprates between two adjacent CuO_2 layers ranges from approximately 10^{-1} eV for LSCO compounds to 10^{-3} eV for Bi-based compounds[18]. Let us now assume that the hopping energy between more distant CuO_2 layers falls off exponentially. A distance $d = 20$ nm between the hole and electron layer corresponds roughly to 30 CuO_2 layers, so that the tunneling energy equals $t \approx e^{-30}10^{-3} = 10^{-16}$ eV.[19] This estimate lies well below the maximum value of t obtained in equation (2.19). However, the precise value of t is highly sample specific and needs to be checked for each separate sample.

The **experimental protocol** to test the flux quantization is as follows: apply an axial magnetic field of magnitude B_{ext} above the critical temperature T_c, and cool the device below T_c such that a circular current quantum is frozen in. The magnitude of the current is determined by the strength of the applied flux: if $\Phi_{ext} < \frac{1}{2}\Phi_0$ no current is induced, for $\frac{1}{2}\Phi_0 < \Phi_{ext} < \frac{3}{2}\Phi_0$ one current quantum is induced, etc. The magnetic field corresponding to $\frac{1}{2}\Phi_0$ is typically $B_{ext} = 0.2$ mT. Upon removing the external magnetic field, a trapped flux equal to $\Phi_0\chi_m n$ remains, corresponding to a field strength of 50 nT. These numbers do not pose a problem of principle for the experimental realisation of such a flux trapping device.

Based on existing technology, one can envision various **practical realisations** of the concentric p-n doped ring geometry, while it is anticipated that further technology developments will create additional opportunities. Using p- and n-doped complex oxide compounds, such as cuprate perovskites, multilayer thin film structures can be fabricated in the desired ring geometry. Using the proven edge-junction technology[20] the structure sketched in figure 2.5 can readily be fabricated, by e.g., pulsed laser deposition and Ar-ion beam etching. As a barrier layer $SrTiO_3$ can be used, with a typical thickness of 10-100 nm, or another insulating oxide

[18] Cooper and Gray, 1994; and Clarke and Strong, 1997

[19] The tunneling energy might be viewed as a transition rate: the lower this energy the less electrons will hop in a given time period.

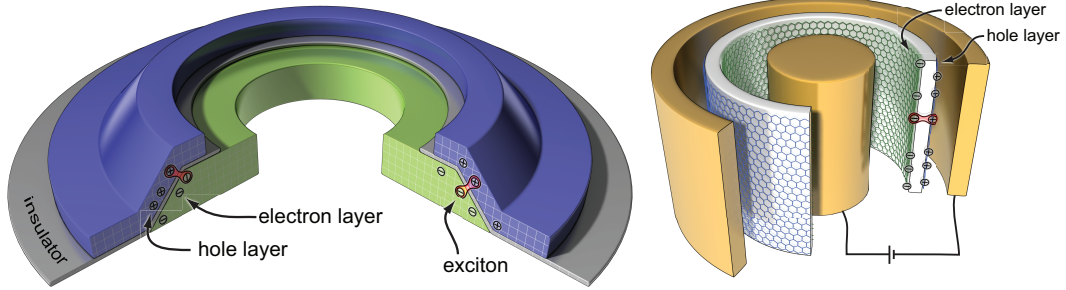[20] Gao et al., 1990; and Hilgenkamp et al., 2003

Figure 2.5: Schematic representations of possible practical realisations of the concentric ring geometry comprised of p- and n-doped layers. Left: Using epitaxially grown complex oxide thin films. Right: Using doubly-gated graphene double layers.

that grows epitaxially on top of the etched base electrode. To guarantee an epitaxial growth of all the layers, the angle α is best kept below about $25°$, but this does not fundamentally alter the physics of the flux quantisation as presented in this chapter.

A second possible practical realisation is based on double-side gated, double layer graphene. Recently, the growth of large area graphene films has been demonstrated on Cu foils, using a high temperature chemical vapor deposition process.[21] Interestingly, a continuous growth was achieved over grain boundaries and surface steps. From this it is feasible to expect that one can also grow a closed graphene tube around a copper cylinder, which would basically be a carbon nanotube with predetermined radius. Covering this with an appropriate epitaxial barrier layer, e.g. 10 nm of Al_2O_3 and a second graphene sheet, which may also be grown by physical or chemical vapor deposition-techniques, would then result in the desired concentric cylinder configuration. Subsequently, the copper can be etched away and the concentric cylinder can be transferred to an appropriate carrier, which can even be made out of plastic.[22] This would straightforwardly allow realizing a doubly gated configuration as depicted in figure 2.5.

[21] Li et al., 2009

[22] Bae et al., 2010