Optical readout of charge and spin in a self-assembled quantum dot in a strong magnetic field

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Abstract – We present a theory and experiment demonstrating the optical readout of charge and spin in a single InAs/GaAs self-assembled quantum dot. By applying a magnetic field we create the filling-factor–2 quantum Hall singlet phase of the charged exciton. Increasing or decreasing the magnetic field leads to electronic spin-flip transitions and increasing spin polarization. The increasing total spin of electrons appears as a manifold of closely spaced emission lines, while spin flips appear as discontinuities of the emission lines. The number of multiplets and discontinuities measures the number of carriers and their spin. We present a complete analysis of the emission spectrum of a single quantum dot with \(N = 4\) electrons and a single hole, calculated and measured in magnetic fields up to 23 tesla.

The electronic properties of quantum dots and their potential applications in nano-electronics, nano-spintronics, nano-optics and quantum information processing depend critically on the number of confined carriers [1]. This number can be tuned with metallic gates or by optical means [2,3]. In lateral gated quantum dots, the electron number can be determined by attaching leads and counting the number of spin flips in the Coulomb blockade spectra [4]. In a self-assembled quantum dot (SAD) [5] electrons can be counted optically by adding a single hole to the confined electrons and observing the emission spectrum of the resulting charged exciton. This can be achieved by tuning the number of electrons in the SAD with an electrostatic potential created by a gate, and comparing the emission spectra obtained for different charge states of the system [6]. Here we demonstrate that the determination of charge is also possible for a fixed number of electrons, without a need for attaching gates. We achieve this by detecting the optical signatures of electronic spin flips in an external magnetic field. The spin-flip transitions require very high magnetic fields to resolve the Landau-level structure of single-particle states [7]. The optical measurement of the electron number is nontrivial as the optical transitions involve only electrons from the lowest occupied state and are, at first sight, an insensitive probe of the number of carriers \(N\) [6,8–10]. The proposed readout scheme is demonstrated on the example of the emission spectra of the triply charged exciton \(X^{3−}\), composed of four electrons and one valence hole, in magnetic fields of up to 23 T. We show that the magnetic field builds up the total electron spin through spin flips. Different total spin states are characterized by different emission spectra, including spin multiplets due to exchange interaction with the valence hole. This is analogous to an exciton interacting with the spin of a magnetic ion localized in a quantum dot [11], except that here the total spin of electrons is engineered by the magnetic field.

The single-particle energies of an electron and a hole confined in a SAD and in a magnetic field \(B\) parallel to the SAD’s growth direction are well approximated by those of a pair of harmonic oscillators [7]

\[
e^{β}(n, m, σ) = Ω^β_c(n + 1/2) + Ω^β_h(m + 1/2) + g_β μ_B B σ_β,
\]

where \(n, m = 0, 1, \ldots\), \(β = e/h\) denotes the particle type, \(g_β\) is the effective Landé factor, \(μ_B\) is the Bohr magneton, and \(σ_β\) is the \(z\) component of the spin (\(σ_e = ±\frac{1}{2}, \sigma_h = ±\frac{3}{2}\)). The oscillator energies \(Ω^β_c = Ω^β_h = Ω^β_{c,h}\), where \(Ω^β_{c,h} = eB/m^∗_β\) is the cyclotron energy, \(m^∗_β\) is the effective mass, \(Ω^β_{c,h}\) and \(\Omega^β_{0,β}\) is the energy
operators for an electron and one hole. In the language of the creation (annihilation) operators for an electron $c^\dagger_{i\sigma}$ ($c_{i\sigma}$), and a hole $h^+_i$ ($h^-_i$), the Hamiltonian of interacting carriers is

$$\hat{H} = \sum_{i,\sigma} \varepsilon_i^e c^\dagger_{i\sigma} c_{i\sigma} + \frac{1}{2} \sum_{i,j,k,l,\sigma,\sigma'} \langle ij|V_{ee}|kl\rangle c^\dagger_{i\sigma} c_{j\sigma'} c^\dagger_{k\sigma'} c_{l\sigma} + \sum_{i,\sigma} \varepsilon_i^h h^+_i c^\dagger_{i\sigma} h^+_i c_{i\sigma} + \sum_{i,j} \sum_{\sigma_1,\sigma_2,\sigma_3,\sigma_4} \langle \sigma_1,\sigma_2|V_{eh}|\sigma_3,\sigma_4\rangle c^\dagger_{i\sigma_1} c^\dagger_{j\sigma_3} h^+_i h^+_j c_{i\sigma_4} c_{j\sigma_2}$$

where the composite indices $i, j, k, l$ enumerate the single-particle states, i.e., $|i\rangle = |nm\rangle$, and $\langle ij|V_{ee}|kl\rangle$ and $\langle ij|V_{eh}|kl\rangle$ are respectively the electron-electron (e-e) and electron-hole (e-h) Coulomb matrix elements, proportional to $V_0 = \sqrt{\frac{\alpha}{\ell^4}}$, with $\ell = \frac{1}{\sqrt{\pi\hbar}}$ being the electronic oscillator length [12]. The term $V_{eh}$ describes only the direct e-h attraction, while the last term in eq. (1) describes the e-h exchange. For orbitals $i, j$, in the basis of electron and hole spin states $|↓\uparrow\rangle, |↑\downarrow\rangle, |↑↑\rangle, |↓↓\rangle$, $H_{X}^i$ is approximated by [9]

$$H_{X}^i = \frac{\Delta_i^e}{2} \begin{pmatrix} \Delta_i^e & \Delta_i^h & 0 & 0 \\ \Delta_i^h & \Delta_i^h & 0 & 0 \\ 0 & 0 & -\Delta_i^h & -\Delta_i^e \\ 0 & 0 & 0 & \Delta_i^e \end{pmatrix}. \tag{2}$$

The exchange matrix elements $\Delta_i^e$ are calculated as $\Delta_i^e \approx \Delta_0^e \int d\mathbf{r} \phi_i^e(\mathbf{r})^2 \phi_i^e(\mathbf{r})^2$, with $\phi_i^e$, $\phi_i^h$ being the electron and hole orbitals, and $\Delta_0^e$ material-dependent parameters. This allows to express the elements $\Delta_i^e$ in terms of the fundamental elements $\Delta_0^e$, with $\Delta_0^e = 400 \mu eV$, $\Delta_0^0 = 180 \mu eV$, and $\Delta_2^0 = 90 \mu eV$ [9].

In the first step of our computational procedure we consider the Hamiltonian (1) without the electron-hole exchange term. In this case the eigenenergies and eigenstates of the charged exciton are calculated in the configuration-interaction approach in the basis of all electron-hole configurations classified by total angular momentum and total spin $S_z$. In order to conserve the total angular momentum, the valence hole couples many electronic configurations and correlations are important, but many conclusions can be drawn from a simplified analysis given below.

Further, we account for the electron-hole exchange term $H_{X}^i$. In general, this term couples the exciton states with different total angular momenta and spins $S_z$. Since the characteristic energy scale introduced by this term is very small, we include this interaction in a perturbative manner, building the many-particle basis out of states with different total spins $S_z$, but with the same orbital angular momentum.

In the third step we use the Fermi’s Golden Rule $I(\omega) = \sum_f \rho_f \delta(E_f - E_i - \omega)$ to calculate the emission spectrum $I(\omega)$ from the initial state $|N, i\rangle$ of the charged exciton with energy $E_i$ to all final states $|N-1, f\rangle$ of $N-1$ electrons with energy $E_f$, remaining in the system. Here $P^f = \sum_f \langle c^\dagger_i h^+_{j\beta} + c^\dagger_j h^+_{\beta}\rangle$ is the interband polarization operator [13].

We start our qualitative analysis by mapping out the phase diagram of the system of $N$ electrons and one hole as a function of the magnetic field. We start with the $\nu = 2$ integer quantum Hall state $|\nu = 2\rangle_{2M} = \left(\prod_{m=0}^{M-1} c^\dagger_{0m1} c^\dagger_{0m1}\right) h^+_{000} |0\rangle$ as a reference configuration of the charged exciton with $N = 2M$ electrons. This configuration is a ground state of the system over a finite range of magnetic fields. At high magnetic fields it is unstable against the edge spin flips. The first spin-flip configuration $|SF\rangle = c^\dagger_{0M1} c^\dagger_{0M-1} |\nu = 2\rangle_{2M}$ is an edge spin-flip state, in which one electron spin-up was removed from the highest occupied orbital, and placed, with spin down, on the next, unoccupied LLL orbital. At low magnetic fields the $|\nu = 2\rangle$ configuration is unstable against a center spin flip. The spin-triplet configuration $|CF\rangle = c^\dagger_{0M1} c^\dagger_{0M-1} |\nu = 2\rangle_{2M}$ stabilizes as a result of a spin-flip transfer of the electron from the edge of the droplet to the center orbital [10], belonging to the second LL. In a similar fashion we seek the ground-state configurations and energies of the charged exciton as a function of the magnetic field. In fig. 1 we plot the lowest energies for $N = 1$ (a neutral exciton $X^0$) to $N = 8$ (an exciton with seven charges $X^-7$) with respect to the reference energy $E_{0=2}(N)$ (traces for different values of $N$ have been shifted apart for clarity). The horizontal plateau marks the region of magnetic fields where the $|\nu = 2\rangle_N$ configuration is the ground state of the system. As we move from this plateau towards the higher magnetic fields, we see a series of cusps marking the spin flips [14]. Counting the number of spin flips allows us to read out the charge of the SAD with accuracy of $\pm 1$ electron. For example, we find two spin flips for both $N = 4$ and $N = 5$. The two electron numbers can be further distinguished by analysing the details of the emission spectrum, as discussed below.

In fig. 1 the first spin flip appears for all charged excitons, while the second spin flip is first seen for the triply charged exciton $X3^-$ ($N = 4$). The $X3^-$ complex, whose energy is marked with the thick line, is also the first one to exhibit the center spin flip at low fields, and this is why we focus on it in the rest of this work.

Note that the phase diagram shown in fig. 1 is characteristic for self-assembled dots with large kinetic energy quantization (here we take $\Omega_0^0 = 20 \text{ meV}$ and $\Omega_0^0 = 4 \text{ meV}$).
Optical readout of charge and spin in a self-assembled quantum dot in a strong magnetic field

Fig. 1: Energies of the charged excitons composed of $N = 1$ to 8 electrons and one hole as a function of the cyclotron energy. In all cases the reference energy is that of the respective $\nu = 2$ configuration. The shade of the area under each trace corresponds to the total spin of the system, with darker areas denoting higher spins.

The magnetic-field evolution reported for dots with smaller confinement energies reveals a richer phase diagram, with a spin oscillation preceding the second spin-flip transition [15]. This behavior is due to the electronic correlations, and was reported also for lateral gated dots confining electrons only [16].

Figure 2(a) shows the magnetic-field evolution of the ground-state energy of $X^{-3}$, and the configurations dominant in the corresponding eigenstates (diagrams at the top of the figure). The first one is the $|CF\rangle$, the second is the $|\nu = 2\rangle$ reference state, the third is the $|1SF\rangle$ and the fourth is the second spin-flip $|2SF\rangle$ configuration.

Let us now account for the electron-hole exchange Hamiltonian $\hat{H}_X$ in a perturbative manner. This Hamiltonian couples the members of the total electronic spin manifold and the spin of the valence hole. For example, for the state $|1SF\rangle$ all possible spin configurations of the valence hole and the unpaired electrons can be classified into two families: $\{|\downarrow\uparrow\uparrow\rangle, |\downarrow\uparrow\downarrow\rangle, |\uparrow\uparrow\uparrow\rangle\}$ and $\{|\uparrow\downarrow\downarrow\rangle, |\downarrow\uparrow\downarrow\rangle, |\downarrow\downarrow\downarrow\rangle\}$. Both families are described by the same e-h exchange Hamiltonian matrix:

$$H_X = \frac{1}{2} \begin{pmatrix} \Delta_{0}^{01} + \Delta_{0}^{02} & (\Delta_{0}^{01} + \Delta_{0}^{02})/\sqrt{2} & 0 \\ (\Delta_{1}^{01} + \Delta_{1}^{02})/\sqrt{2} & 0 & (\Delta_{1}^{01} + \Delta_{1}^{02})/\sqrt{2} \\ 0 & (\Delta_{2}^{01} + \Delta_{2}^{02})/\sqrt{2} & -(\Delta_{0}^{01} + \Delta_{0}^{02}) \end{pmatrix}.$$ (3)

We diagonalize this matrix numerically to obtain three exchange-split levels originating from the three members of the electronic spin $S = 1$ manifold. Their energies are added to the energy $E_{1SF}$. Upon the inclusion of the Zeeman effect, each level further splits into two, reflecting the differences in spin alignments of the two families. A similar analysis can be carried out for the $CF$ and second spin-flip configuration $|2SF\rangle$. Since in the $|2SF\rangle$ state the electrons have total spin $S = 2$, we obtain families of five exchange-split levels, derived from five members of the electronic spin $S = 2$ manifold: $S_z = 0, \pm 1, \pm 2$. 

Fig. 2: (a) Energy of the triply charged exciton as a function of the magnetic field. Diagrams show the initial- and final-state configurations in each regime, while the vertical dashed lines mark the spin flips. (b) The calculated emission spectrum as a function of the cyclotron energy for $g_e = g_h = 0$ and with the electron-hole exchange treated perturbatively.
Let us now turn to the analysis of the emission spectrum of the X3− complex. The dominant final-state configurations corresponding to each phase are depicted in the lower part of fig. 2(a), while the resulting calculated emission spectra are shown in fig. 2(b).

In the recombination process, the low-magnetic field configuration \(|CF\rangle\) couples to two final states of the three electrons, \(S = \frac{3}{2}\) (shown as a spin-polarized state) at high energy and \(S = \frac{1}{2}\) at low energy. Each line is, in general, further split by the Zeeman energy, reflecting the two possible spin alignments of the pair of recombining carriers, and by the gaps arising from the e-h exchange interaction in the initial state. In fig. 2(b) we only account for the latter, setting \(g_e = g_h = 0\).

The \(|\nu = 2\rangle\) configuration is an electronic spin singlet, not affected by the e-h exchange. This phase can combine to two final states, composed of configurations shown in the bottom part of fig. 2(a), second diagram from the left. Thus, we again obtain two emission lines. For the initial \(|\nu = 2\rangle\) configuration containing the hole spin-down the emission spectrum is the same, except for the splitting of the lines introduced by the Zeeman terms.

In the \(|1SF\rangle\) phase, the optically active final configuration shown in the bottom part of fig. 2(a), third diagram from the left, is mixed with three additional configurations. Two of them are created by changing the position of the spin-up electron, while the fourth one is an Auger type excitation. These configurations can be grouped into one \(S = \frac{3}{2}\) state (high-energy emission line) and a group of three \(S = \frac{1}{2}\) states visible in fig. 2(b) at lower energy. Due to the triplet character of the initial state, each of these lines shows a fine structure of three maxima.

Finally, the only possible final-state configuration left after the recombination from the \(|2SF\rangle\) state is the spin-polarized configuration shown in the bottom part of fig. 2(a) in the first diagram from the right. This is an \(S = \frac{3}{2}\) configuration. Here, the inclusion of the other members of the \(|2SF\rangle\) multiplet results in several additional final states, whose energies are larger than \(E_{1SF}\).

In addition, each line will become split due to Zeeman and fine-structure effects. Since the \(S = 2\) multiplet is composed of five configurations, the fine structure of the emission line is composed of five closely-spaced maxima.

At the magnetic fields corresponding to spin flips in X3−, the energies of the adjacent phases are equal, but the energies of the corresponding final states differ. As a result, the emission spectrum, shown in fig. 2(b), exhibits characteristic discontinuities at the critical magnetic fields. The number of discontinuities past the filling-factor=2 droplet equals half the number of confined electrons. Also, the total electronic spin of the system can be identified in each phase from the multiplicity of the corresponding emission line, because the different total spin couples differently to the hole via the e-h exchange.

Let us now identify the signatures of the spin flips in the emission spectrum of a single, mesa-selected InAs/GaAs SAD in magnetic fields up to 23 T. The structure of the sample and the details of the experiment are described elsewhere [17]. The excess confined charges originated from the n+ doping of the substrate, and their population was tuned by the energy of the photoexciting laser. Figure 3(a) shows the measured photoluminescence (PL) spectrum which we identify as that of the X3− complex. In this spectrum, a single, bright maximum, detected at very low magnetic fields at about 1.315 meV, exhibits a discontinuity in the vicinity of 1 T. As the field increases, we follow the single PL line up to \(B \sim 8\) T, where it splits into two maxima. It is not clear whether this splitting is due to the Zeeman effect (not seen in any other part of the spectrum), or to emission from an excited state. Absence of the Zeeman splitting at low fields has been reported in PL measurements on similar samples [7]. The complicated character of the magnetic field evolution of the system in the region 5–12 T may also be due to the emission from different charge states of the dot or the existence.
have calculated the emission spectra of the model parameters $\Omega$ quantuminformationprocessing. in nano-electronics, nano-spintronics, nano-optics and ofSADswiththenumberofcarriers,withapplications Thisopensthepossibilityoftuningelectronicproperties tinuitiesintheemissionline. Thetotalelectronspincan numberofspinflips, which can be identified as discontinuitiesat $B=12T$ to $14T$ the spectrum broadens and appears to undergo a discontinuous shift to higher energies. At very high magnetic fields the trace consists of three closely spaced lines. 

We identify the low-field discontinuity of the trace as the signature of the spin-flip transition between the $|CF\rangle$ and the $|\nu=2\rangle$ phases of the $X3^-\,$$, and the discontinuity at $B=14T$ as the onset of the first spin-flip $|1SF\rangle$ configuration. To confirm this interpretation, we have calculated the emission spectra of the $X3^-\,$$ for model parameters $\Omega_0^{\text{in}}=20\text{meV}$, $\Omega_0^{\text{out}}=4\text{meV}$, $g_e=-3$ and $g_h=1$. The electronic $g$ factor was chosen to reproduce the critical magnetic field corresponding to the first spin flip, while the chosen value of the hole $g$ factor suppresses the Zeeman splitting of the electron-hole pair undergoing the recombination. The calculated low-field transition energies are shown in fig. 3(b). The maximum originating from the $|CF\rangle$ configuration is split into three peaks by the e-h exchange. However, the resulting energy gaps are too small to be resolved in the experiment, where only a single, broad maximum is seen. The discontinuity at $B=1.3T$ marks the onset of the $|\nu=2\rangle$ phase. The transition energies calculated in the high-field range are shown in fig. 3(c). Starting with the emission from the $|\nu=2\rangle$ ground state, at $B=16T$ we find an upward discontinuity corresponding to the first spin flip. At this point, the trace acquires a three-line character — an optical signature of an electronic triplet, split by the e-h exchange. This three-line spectrum agrees well with the experimental trace.

To emphasize the fine structure seen in the emission spectra, insets to fig. 3(a) show the calculated and measured emission traces for several magnetic fields. At $B=4T$ the ground state of the $X3^-\,$$ is the spin-singlet $|\nu=2\rangle$ configuration, unaffected by the e-h exchange, resulting in the appearance of a single emission line. At $B=21.5T$ the $|1SF\rangle$ configuration is the ground state. The e-h exchange splits the three-fold degenerate manifold of this triplet into three maxima, as seen in the calculations and experimental data. At even higher magnetic fields a second spin flip is expected, resulting in five maxima characteristic for the $S=2$ manifold. This magnetic field, however, is not reached in this experiment.

In conclusion, we have demonstrated that one can optically determine the number of carriers in a SAD by analyzing the emission spectra of the system at high magnetic fields. The number of carriers determines the number of spin flips, which can be identified as discontinuities in the emission line. The total electron spin can be detected from the multiplicity of the emission lines. This opens the possibility of tuning electronic properties of SADs with the number of carriers, with applications in nano-electronics, nano-spintronics, nano-optics and quantum information processing.
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