Quantum Hall regime in emission spectra of single self-organized InP/GaInP quantum dots

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Abstract. We used high spatial resolution near-field scanning optical microscopy (NSOM) to measure low-temperature (10K) emission spectra of single InP/GaInP QDs (lateral size ~100 nm) at magnetic fields up to 10 T. A multi-electronic structure having energy splitting 3-5 meV, resulting from charging of the dot up to 20 electrons, has been observed for these QDs. We measured a strong variation of the near-field emission intensity on a length scale ~30 nm related to Wigner localization. Magnetic-field-induced shifts of the multi-shell peaks for some dots follow remarkably well the energy levels of a Fock-Darwin Hamiltonian, which allow the observation of a Wigner-molecular-droplet transition.

A Wigner phase is a strongly correlated state of an electron system, in which electrons occupy separate sites forming a regular lattice. The possibility of crystallization of an electron gas at densities below a certain critical value (n_s) was predicted by Wigner in 1934 [1]. Experimentally such crystallization has been observed in two dimensional electron systems on the surface of liquid He [2], in a GaAs/GaAlAs heterojunction [3] and in Si [4].

The electrons confined in traps having volume >1/ n_s form Wigner Molecules (WMs). Wigner localization (WL) of electrons in such traps formed by interface fluctuations is responsible for the quantum Hall effect in high mobility semiconductor heterostructures [5,6]. The WL regime has been realized in single electron transistors (SETs) using GaInAs/AlGaAs quantum dots (QDs) nanofabricated from modulation doped quantum well structures [7]. Coulomb blockade measurements [7] and theoretical analysis [8] have shown that a WM in such SETs can reveal a rich set of electron arrangements and spin states, which are controlled by the number of electrons present, and by an applied magnetic field.

In the present contribution we report the observation of the signatures related to WL in emission spectra of single self-organized InP/GaInP QDs measured using high-spatial-resolution near-field scanning optical microscope (NSOM). To the best of our knowledge our results are the first observation of WL in QDs using emission spectra [9], which opens the way to study WMs by nanoptical methods.

1. Experimental details

InP/GaInP QD structures having GaInP cap layer thickness from 0 to 60 nm were grown by Metal-Organic Chemical Vapor Deposition epitaxy as described in [10, 11]. The dot density (1-5*10⁹ cm⁻²) and their sizes (base ~100 nm, height ~15 nm) were estimated using transmission electron microscopy (TEM) for capped samples (see below) and atomic force microscopy (AFM) for uncapped samples.

The details of the NSOM experiments were described elsewhere [12]. We measure NSOM images and spectra in the collection-illumination mode. Emission was excited by the 488 nm line of an Ar ion laser and dispersed using a 300 or 1200 gr/mm grating in a 270 mm focal length monochromator. The spectra were measured using a nitrogen-cooled CCD detector and 1200 gr/mm

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grating. Monochromatic NSOM images were measured using a 300 gr/mm grating and a GaAs photomultiplier working in the photon-counting regime with an accumulation time of 20 ms per pixel. Excitation power density was ~20-100 and 0.1-1 W/cm² for 300 and 10 K, respectively.

2. Single dot spectroscopy and imaging

Fig.1a-f shows the results of the NSOM measurements of InP/GaInP QD structures (a, d-f) together with TEM images (b - cross section and c - plan view). Room temperature NSOM images (spatial resolution ~150 nm) presented in the inserts in Fig.1a show a set of bright spots having density ~20 μm⁻² (see also the TEM image in Fig.1c) and size ~200 nm related to single QDs. Clearly-resolved single QD images are observed for 5 and 20 nm capped QDs as well as for the uncapped QDs. For a 60 nm cap the single dot image contrast strongly decreases, which demonstrates the expected reduction of spatial resolution due to increased tip-dot separation. It is important to note the strong emission intensity for the uncapped QDs at room temperature. We believe that this is the first observation of such strong emission from uncapped QDs.

The spectra of single InP QDs at room temperature (Fig.1a) consists of a single band having wavelength in the range of 710-790 nm (1.75-1.56 eV) and halfwidth γ ~60 nm (~150 meV). At 10K the emission spectra of a single InP QD results from the filling of several electron shells (see Fig.1d), which occurs as a result of residual doping (~10¹⁶ cm⁻³) of the GaInP matrix [13]. For most of the dots three peaks (s, p and d shells) having width γ ~3 meV and energy splitting 3-5 meV are observed, indicating charging of the QD by 7-16 electrons. For some dots a fine structure of the shells consisting of a several ultra-narrow lines with γ = 0.2 meV is revealed as was shown in detail in ref [12]. Spectra of three selected QDs denoted by QD1, QD2 and QD3 in Fig.1d show a progressive decrease of the shell energy splitting ΔE from ~5 to ~4 and to ~3 meV, which is directly related to a change of the confinement potential, i.e. “effective” dot sizes, from ~90 to ~100 and to ~115 nm; this is in good agreement with TEM measurements (see Fig.1b and c). For the ~100 nm dot, one can expect WL of electrons [8] and thus each of these InP/GaInP QDs represents a natural WM.

In Fig.1e and f we present high-spatial-resolution monochromatic 10K-NSOM images taken for the same area (size 500x500 nm²) and detection wavelength (690 nm) in two separate scans. In addition to the bright resonant QD denoted Qa, two weaker non-resonant QDs denoted Qb and Qc...
appear in the images. The image of Qa has size ~120 nm and it reveals a strong (up to 50%) intensity fluctuation on a length scale ~30 nm, which is of the order of the single electron separation for a WL, as can be seen from the possible configuration of an ten-electron-WM [8], indicated by small circles superimposed on the Qa image. We determined that the photon counting detection noise only partly contributes to the spatial intensity fluctuation in Fig.1e and f (note that photons are counted only for 20 ms in monochromatic imaging); this provides evidence that the observed spatial variation of the near-field intensity is related to a WL. Indeed, the measurements of the spectra using a CCD with accumulation time 1 s revealed similar spatial fluctuations of the emission intensity. Spectral measurements also show that the intensity variations are accompanied by spectral diffusion of a few meV. We found that intensity and spectral fluctuations are suppressed by decreasing spatial resolution (near-field defocusing). Spectral diffusion indicates the effects of rearrangement of the charge distribution inside and/or around the QDs under near-field excitation; this will require further investigations, which are in progress.

3. Magnetic field spectra

The multi-shell structure observed in Fig.1d makes it possible to observe a magnetic-field-induced phase transition of a WM using magneto-NSOM spectroscopy (see Fig.2a-d), which is similar to that observed in Coulomb blockade measurements using nano-lithographically-defined GaInAs/AlGaAs QDs in a SET [7]. In Fig.2a and b we show the emission spectra versus magnetic field of two InP/GaInP QDs denoted QD2 and QD4 having $\Delta E=4.5$ and 3.5 meV, respectively, and in Fig.2c and d we compare the magnetic-field-induced shifts of the shell peaks for these QDs with the energy levels of the Fock-Darwin (FD) Hamiltonian. From Fig.2b and d one can see that for QD4 having larger size (i.e. smaller $\Delta E$, see also Fig.1d) the f-shell peak is observed, which indicates the accumulation up to 20 electrons. Here the FD levels poorly describe the experimentally-observed shifts and no distinct assignments of the shell peaks at high fields can be done.

In contrast, for QD2 (Fig.2a and c) the observed magnetic-field-induced shell shifts follow the FD energy levels remarkably well. Here two magnetic-field induced transitions are observed. The first transition occurs at $B=2T$. It arises from optical pumping of the nuclear spins [14] inducing an internal magnetic field ($B_{\text{int}}$) of 2T which is anti-parallel to the external field. The existence of such a field was established using measurements of circular polarization [15]. In the unpolarized spectra presented in Fig.2a $B_{\text{int}}$ is responsible for the shift to lower energy of the s- and d-shell peaks at $B<2$. For $B>2$

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**Figure 2.** Near-field spectra at 10 K representing shell structure (peaks s-g) of single InP QDs at magnetic fields 0, 1,...,10T (a and b); energy shift of emission peaks of these QDs versus magnetic field (solid circles) together with calculated Fock-Darwin energy levels (solid curves) and Landau levels ($\nu=0$ and 1) dashed lines (c and d). Peaks * in (a and b) are contributions of neighbouring QDs. In c and d the abscissa is the net internal magnetic field.
In conclusion, we used high-spatial-resolution NSOM to demonstrate the effect of Wigner localization of electrons (quantum Hall regime) in the emission spectra of self-organized InP/GaInP QDs. Our results open the way to study such localization by nano-optical methods.

4. References
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