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Ground-state transition in few-electron quantum dots observed by magnetophotoluminescence

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We have investigated electrostatically confined quantum dots containing a few electrons each. In zero magnetic field the photoluminescence (PL) reflects the few-electron density of states of such artificial atoms and we can resolve their energy levels. The magnetic-field dispersion of these quantum dot levels shows anticrossing behavior and sharp energy jumps which we identify as manifestations of a change of ground state in the dots. From our PL measurements we thus determine the size and sign of the electron-electron and final-state interactions within the quantum dots.

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That the electron-electron interaction in quantum dots leads to quantum phase transitions in a magnetic field is a topic that has been extensively discussed in the field of zero-dimensional electron systems.[1–10] The ground state of a small number of correlated electrons confined in a quantum dot is expected to undergo spin and angular momentum transitions as the magnetic field is increased.[1–10] Exact diagonalization calculations have produced rich phase diagrams:[3] In a high magnetic field, the ground state of the electrons in the quantum dot has been shown to jump between a series of incompressible states with ‘‘magic’’ values of the total angular momentum.[4] In the low magnetic field regime, the electrons undergo a sequence of spin-flips and move from the center to the edge of the dot to lower their ground-state energy as the magnetic field is increased.[5,6] Although such a phase transition has previously been experimentally observed by single-electron capacitance spectroscopy,[5] these capacitance experiments can only *indirectly* probe the ground state of the quantum dot via the density of states at the chemical potential.[6] Far-infrared radiation only couples to the center of mass motion and is therefore insensitive to the electron-electron interaction for parabolic confinement.[7,11] Photoluminescence (PL) has previously been used to study electron systems confined in quantum dots,[8,12,13] and is unique in that it probes the entire occupied electronic density of states below the Fermi energy.[8,12] The magnetic field dependence of the PL energy is therefore *directly* related to the total ground-state energy of the interacting electrons confined in these artificial atoms.[2]

In this Brief Report we concentrate on magnetophotoluminescence from GaAs-$\mathrm{Al_{0.3}Ga_{0.7}As}$ electrostatically confined quantum dot arrays, and in particular on the experimental observation of few-electron correlation in a quantum dot. A number of samples, each close to their quantum limit, were investigated. When the two lowest-energy levels are clearly resolved in the PL spectra, we have mapped their dispersion in a magnetic field. These quantum dot states appear to anticross in a finite magnetic field. When only a single quantum dot state is observed, the energy of this lowest-energy level is seen to undergo a sharp downward jump in energy at a particular magnetic field. The magnetic field value at which this occurs changes with the number of electrons per dot. The agreement between these experimental results and theoretical predictions[1,2] identifies this behavior as a manifestation of a change in the few-electron ground state of the quantum dots.

The quantum-dot samples studied were prepared from modulation-doped *n*-type GaAs-$\mathrm{Al_{0.3}Ga_{0.7}As}$ heterojunctions grown by molecular beam epitaxy. In order to increase the PL intensity, a δ-doped layer of Be was grown in the GaAs at a specific distance (either 20 or 25 nm) from the interface.[14] These heterostructures were etched into Hall bars. For the unpatterned gated structure, a 5-nm-thick semitransparent NiCr gate was then evaporated onto this Hall bar. In the case of the quantum dot samples, photoresist was deposited on the top of each Hall bar, and a dot array pattern was fabricated in this photoresist by holographic lithography.[8,11] The periodicity of this dot array, measured by scanning electron microscopy, was 500 nm, and the dot size was $\sim$200 nm. A similar gate to the unpatterned case was evaporated onto this photoresist nanostructure (inset of Fig. 2). The electron system, which lies 80 nm below the surface of the heterostructure, was used as a back contact and was contacted via a Hall bar contact. Thus, by applying a negative bias to the gate, we were able to deplete the electrons between the dots and to vary the number of electrons per dot. PL measurements were carried out at 4.2 K in an optical cryostat with a split-coil magnet. An Ar-ion laser was used to excite electrons above the $\mathrm{Al_{0.3}Ga_{0.7}As}$ band gap in magnetic fields (B) up to 7 T. With excitation at this energy, it has been shown that one can control the concentration of electrons in the electron system.[15] The spectra were taken by a double spectrometer and detected by a cooled GaAs photomultiplier tube. Conductivity measurements of the electron system were taken while a negative gate voltage (V_g) between the gate and the Hall bar contact was gradually applied.

To confirm that all the results from the quantum dot samples described below are solely due to lateral quantum confinement, the behavior of an unpatterned gated two-dimensional electron system (2DES) in a magnetic field was investigated at various gate voltages. Typical 2D Landau

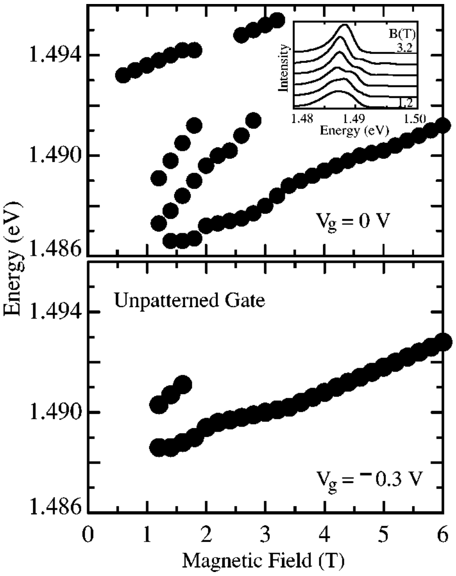

FIG. 1. The PL peak energy as a function of magnetic field for an unpatterned gated 2DES with two different voltages applied to the gate. The inset shows the PL spectra at various magnetic fields for $V_g=0$ V. The spectra have been offset for clarity.

level (LL) behavior in magnetic field (inset of Fig. 1) and clear LL depopulation[16] on application of a negative V_g were observed, as shown in Fig. 1.

The dependence, on V_g, of the zero-magnetic-field PL from sample A with a patterned gate is shown in Fig. 2. At $V_g=0$, a PL peak corresponding to the recombination of electrons from the 2DES with holes bound to Be acceptors is observed. (It should be noted that sample A at $V_g=0$ shows clear 2D LL behavior in a magnetic field.) In Fig. 2 one can see that, at $B=0$, with increasingly negative V_g the low-energy edge of this 2DES PL peak shifts toward the blue in agreement with the behavior previously observed on depletion of 2DESs.[15] As V_g becomes increasingly more negative, the electron system between the dots gradually depletes, isolating the quantum dots from each other.[13] For $V_g<-2$ V a new PL peak at 1.4896 eV can be resolved in the PL spectra, which we attribute to the recombination of electrons from the 0DESs with holes bound to Be acceptors. For sample A only a single quantum dot level is observed.

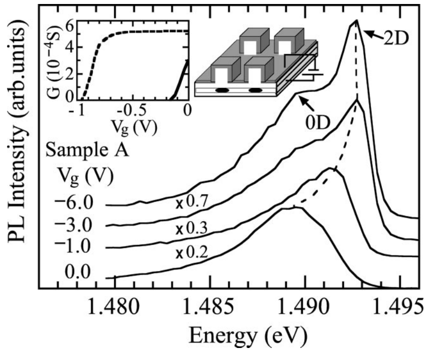

FIG. 2. The PL spectra of sample A at various gate voltages. The dashed line is a guide for the eye and the spectra have been offset for clarity. The left-hand inset shows the gate-voltage dependence of the conductance (G) of the electron system measured in the dark (solid line) and when the sample was illuminated weakly by a laser (dashed line). The right-hand inset shows a schematic of the samples with a patterned gate.

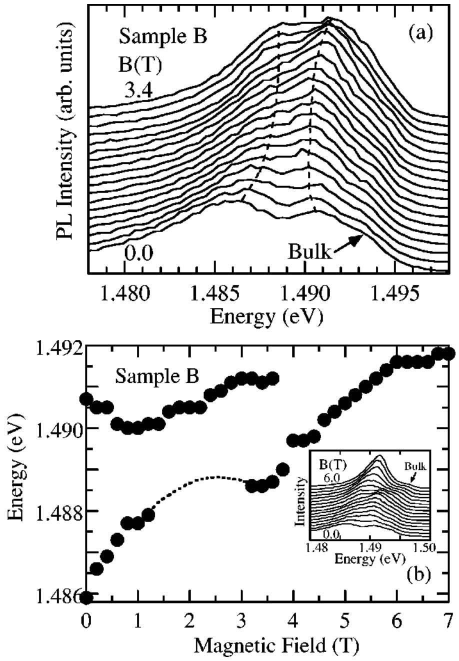

FIG. 3. (a) The PL spectra of quantum dot sample B at low magnetic fields. The spectra have been offset for clarity. The dashed lines are guides for the eye. (b) The PL peak energy a function of magnetic field for quantum dot sample B. The dashed line is a guide for the eye for when the two energy levels become so close that they become difficult to resolve. The inset shows the PL spectra over a greater range of magnetic fields than in (a). The spectra have been offset for clarity.

These PL results are consistent with our conductance measurements on sample A shown in the inset of Fig. 2: At zero gate voltage, the sample shows a larger conductance under weak illumination than in the dark, due to persistent photoconductivity effects. Further illumination with increasing laser power density, results in a decrease of the zero-gate-voltage conductance from this maximum value, due to depletion (caused by the electrons recombining with photoexcited holes).[15] The application of a negative gate voltage gradually decreases the conductance to zero. This we interpret as the electron system gradually changing from a 2DES to an array of interconnected dots, and finally into an array of isolated dots.[13] When the quantum dots become isolated, the conducting channel cuts off completely and the conductivity drops to zero. For quantum dot sample B we observed no V_g dependence: The quantum dot confinement appeared to be produced by the presence of the patterned NiCr film alone. This is in agreement with previous observations, where it was noted that merely depositing films of material on the top surface of such a heterostructure depletes the electron system underneath.[12,13]

Figure 3(a) shows the PL spectra from sample B in zero and finite perpendicular magnetic field. Two PL peaks, due to recombination of electrons in the quantum dots with holes bound to Be acceptors, are observed at 1.486 and 1.491 eV at $B=0$. (There is also a high energy shoulder at 1.493 eV due to radiative recombination in a bulk part of the sample.) Thus the PL appears directly to reflect the few-electron density of states of the quantum dots and we assign these two PL peaks to the two lowest energy levels of the few-electron quantum dot.

In the weak-interaction regime, where the ratio of the effective radius of a parabolic dot to the effective Bohr radius a_B, $\sqrt{(e^2/a_B)/\hbar\omega_0}$ is small, the energy difference between the two lowest states of the quantum dot provides a lower-bound on the quantum dot confinement energy $\hbar\omega_0$.[9] From our quantum-dot-level separation, we thus derive a confinement energy of at least 5 meV in this case. From this confinement energy, we can derive an upper bound on the effective diameter of the confined electrons of sample B, assuming that the confinement potential is parabolic. We thereby deduce that the effective diameter of the quantum dots is less than 30 nm. This is considerably smaller than the size of the defining gate structure ($\sim$200 nm) as one would expect.[17] We can estimate the number of electrons per dot from areal considerations by assuming that the gate only depletes the concentration of electrons between the dots and does not affect the electron density in the dots. For 30 nm diameter dots we calculate that on average that we have at 1.2 electrons per dot for sample A and 1.5 electrons per dot in sample B. It has previously been shown, in similar arrays of electrostatically-confined quantum dots, that all the dots simultaneously contain the same small integer number of electrons.[11]

In Fig. 3(a) at low magnetic field, $B<1.5$ T, the energy of the lower-energy state increases with the magnetic field while the higher-energy level shows a clear negative magnetic field dispersion. These two lowest quantum dot energy levels merge in energy around $B=2$ T and the lower-energy state appears as a shoulder on the higher-energy PL line. Thus the two PL peaks appear to anticross. On further increasing the magnetic field above 3 T the lower-energy level is again resolved. At an even higher magnetic field, the PL intensity of the higher-energy level decreases, indicating depopulation of that level.[16] It eventually disappears from the PL spectra altogether [inset of Fig. 3(b)]. This energy dependence of the two lowest quantum dot levels in magnetic field is plotted in Fig. 3(b). Similar magnetic field behavior has been observed in another similar quantum dot sample.

Figure 4 shows the energy of the lowest quantum dot energy level of sample B as a function of magnetic field at various gate voltages. At $V_g=-3$ V, the PL peak undergoes a sharp jump downward in energy at $B=3.8$ T. This energy jump moves to a lower magnetic field with increasingly negative V_g. The more negative V_g, the fewer electrons per dot. We also observe that this energy jump becomes more pronounced the greater the magnitude of V_g.

We now compare our experimental data of Fig. 3(b) with the energy spectrum of quantum-dot helium calculated by exact diagonalization for parabolic confinement.[1] The calculations predict that the energy of the lowest state of the quantum dot, increases with B at small values of B, and that of the first excited state decreases with B. This is due to B changing the relative importance of the Coulomb, confinement, and kinetic energies. In our experiments for $B<1.5$ T we do indeed observe that the behavior of the two lowest energy levels of our quantum dots is remarkably similar to that predicted by theory. With increasing B, the Coulomb interaction between the electrons is predicted to force these two quantum-dot levels to cross, at which point the ground state of quantum-dot helium changes from a spin-singlet state to a spin-triplet state. At $B\simeq 2$ T we do not observe the predicted level crossing,[1] but instead what appears to be an anticrossing.[18]

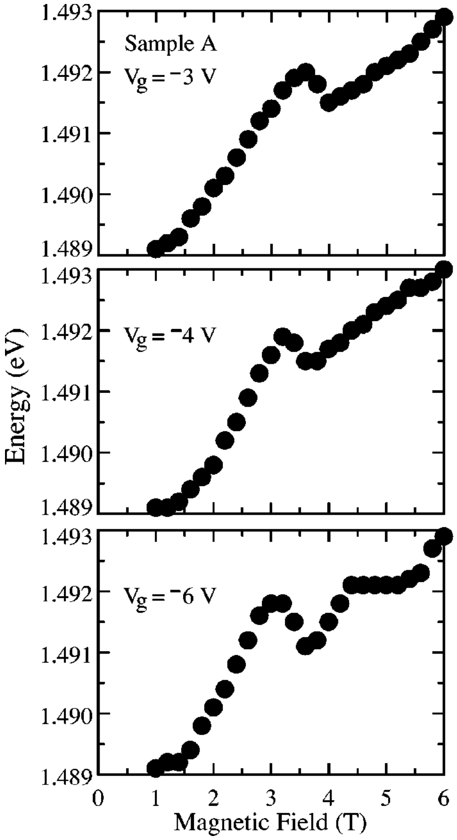

FIG. 4. The PL peak energy as a function of magnetic field for quantum dot sample A at various gate voltages.

It has also been shown theoretically[2] that the mean photon energy of the PL from the quantum dots is uniquely related to the ground state of these artificial atoms. The mean photon energy of the magneto-PL is therefore expected to undergo a series of discontinuous energy jumps as the character of the ground state of the dots changes. Hawrylak and Pfannkuche[2] predicted, for an acceptor far away from the dot as in our heterostructures,[19] that these discrete energy jumps will be downward in energy. Figure 4 shows that what we observe experimentally is a sharp downward jump in the energy of the PL from our dots at a specific magnetic field which depends on V_g. The size of the energy jump is a direct measure of the electron-electron and final-state interactions in the dot.[2] Our measured energy jumps vary between 0.5 meV at $V_g=-3$ V and 0.7 meV at $V_g=-6$ V. Those predicted theoretically vary with the magnetic field but are expected to be in the range 0.26–0.74 meV. Thus the results of Fig. 4 are in remarkably good agreement with the predictions of

Hawrylak and Pfannkuche.[2] The shift of this quantum phase transition to a lower magnetic field with a decreasing number of electrons per dot is again expected,[6,10] and has also been observed by single-electron capacitance spectroscopy.[5]

In summary, we believe these results constitute proof not only of successful magneto-PL measurements from gated quantum dots but also of successful mapping of a magnetically induced quantum phase transition in the quantum dots by PL. PL is a technique that uniquely probes the ground state of the quantum dots directly and thus can measure the size and sign of the electron-electron and final-state interactions within these artificial atoms.

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