Zeeman Bifurcation of Quantum-Dot Spectra

W. Hansen, T. P. Smith, III, K. Y. Lee, J. A. Brum,[a] C. M. Knoedler, J. M. Hong, and D. P. Kern
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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We observe the magnetic-field–induced bifurcation of quantum levels into surface states and bulklike Landau states. The disruption of the electric field quantization by a magnetic field is most dramatic for electrons bound in two dimensions perpendicular to the magnetic field. The interplay between competing spatial and magnetic quantization mechanisms results in a pronounced and complex level splitting. The observed splitting of zero-dimensional energy levels depends critically on the size of the quantum dots, and can be explained with a calculated single-particle energy spectrum.

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For more than half a century confined electron systems in a magnetic field have been investigated theoretically in terms of their influence on the Landau diamagnetism of free electrons.[1-3] Investigations of surface states in confined electron systems have been revived more recently in order to explain the quantized Hall effect.[4,5] Their skipping orbit nature is also demonstrated by transport measurements with ballistic point contacts.[6,7] The influence of edge states on the properties of the electron system is expected to increase with decreasing system size and even more dramatically with decreasing dimensionality. In an electron system that is free to move in only one dimension perpendicular to the magnetic field each electric subband transforms into a hybrid band when the magnetic length $l_B=(\hbar/eB)^{1/2}$ becomes comparable to the width of the electron system. With increasing magnetic field the energy separation between adjacent hybrid bands approaches the cyclotron energy and the density of states at the bottom of each hybrid band increases, so that the bands become Landau-level-like at high magnetic fields. There is a continuous transition and a one-to-one correspondence between the electric subband structure at zero-magnetic field and Landau-level-like hybrid bands at high-magnetic field.[8-10]

In contrast a far more complex behavior is predicted for zero-dimensional (0D) systems.[1-3] At zero magnetic field the discrete energy levels are each occupied by two electrons except for degeneracies that depend on the symmetry of the confinement. With increasing magnetic field this degeneracy is lifted and hybrid levels originating from the same zero-field energy level join different Landau levels at high magnetic field. In general, there is no one-to-one correspondence between energy levels at zero magnetic field and Landau levels at high magnetic fields.

The splitting of 0D energy levels at low magnetic field is similar to the normal Zeeman splitting of electronic states in atoms. However, in atomic physics the magnetic field is usually a weak perturbation of the Coulomb confinement. To observe Landau-level-like behavior of atomic electrons either the magnetic field must be several orders of magnitude larger than those experimentally realizable today[11] or the atoms must be highly excited.[12] Because of the low effective mass and the high dielectric constant the hydrogenic states of shallow donors in semiconductors transform into the Landau limit at lower magnetic fields.[13] Unlike in the case of semiconductor impurities, the electrons in the microstructured heterojunctions studied here are confined by an artificially imposed potential that differs drastically from the Coulomb potential. Furthermore, the number of bound electrons can be varied systematically. The extent of the confinement potential in such devices is of the order of 100 nm.[14,15] Thus at low magnetic fields all states can be considered surface states, while at magnetic fields of about 10 T (where the magnetic length is ~10 nm), almost all states are Landau-level-like.

We have observed the magnetic field rearrangement of energy levels in quantum dots for the first time. The density of states of the discrete energy levels in the quantum dots is measured with capacitance spectroscopy as a function of the gate voltage and a magnetic field applied perpendicular to the heterojunction interface. The samples are modulation-doped GaAs-AlGaAs heterojunctions. The electron system at the GaAs-AlGaAs interface is so strongly confined in the direction normal to the interface that the system is in the extreme quantum limit with respect to motion in this direction. The AlGaAs layer of the heterostructure is covered with a matrix of many ($\sim 10^5$) 30-nm-thick squares of undoped GaAs with widths of 200, 300, or 400 nm. Minimal variation of the square size is achieved by lithography with a high-resolution vector-scan electron-beam writer. From process parameters such as resist resolution, beam diameter, and accuracy of positioning of the exposing electron beam we estimate the size variation of the fabricated dots to be less than 5% of the diameter. The matrix is covered with a metal gate, so that lateral confinement of the electron system is provided by the band bending beneath the Schottky barrier. The electron system resides below the GaAs squares, where the front gate has a larger separation from the heterojunction interface. Back contact is provided by a doped substrate separated

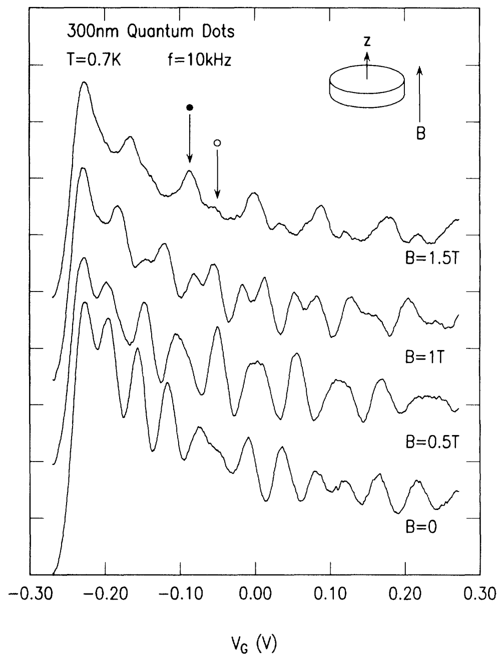

FIG. 1. Gate voltage derivative of the capacitance of a sample with 300-nm squares at four different magnetic fields. The orientation of the magnetic field with respect to the geometry of the confinement is indicated in the inset. The data were recorded at $T=700$ mK.

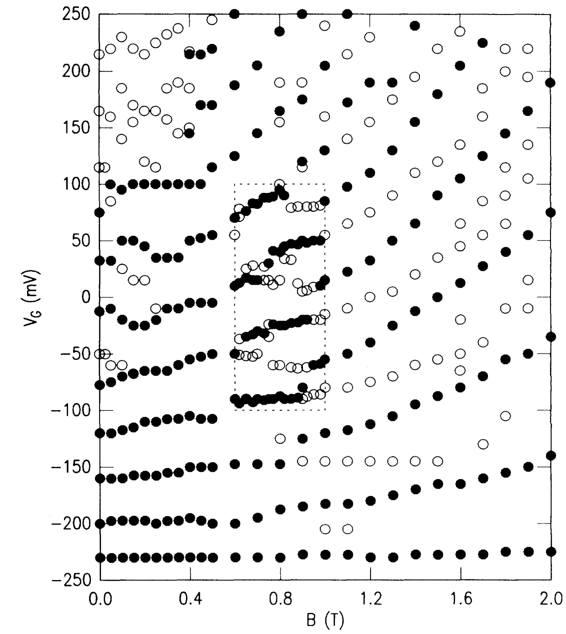

FIG. 2. Gate voltage position of the maxima in the gate voltage derivative of the capacitance vs the magnetic field. The data presented in Figs. 1 to 3 were taken on the same sample. Filled circles indicate strong maxima (cf. Fig. 1). The dashed square indicates the gate voltages and magnetic field range of Fig. 3.

from the heterojunction interface by an undoped 80-nm-thick GaAs layer. The gate voltage derivative dC/dV_G is measured with phase lock-in techniques at modulation voltages between 5 and 10 mV and frequencies of about 10 kHz.

Typical traces of the gate voltage derivative of the capacitance versus the gate voltage at four different magnetic fields are shown in Fig. 1. The magnetic field is applied in the direction of strong confinement normal to the sample surface (z direction). At gate voltages below $V_G=-270$ mV there are no electrons in the potential well at the heterojunction interface resulting in a zero derivative signal. At higher gate voltages electrons occupy the discrete energy states of the quantum dots resulting in capacitance maxima whenever the Fermi energy crosses a discrete energy level.[16] The gate voltage derivative is recorded in order to enhance the signal against the background capacitance. The period of the oscillations changes systematically with the dot size of the samples: largest in samples with 200-nm dots and smallest in samples with 400-nm dots. Also, the oscillation amplitude of samples with 400- and 300-nm squares increases significantly between $T=4.2$ K and $T=0.7$ K, whereas the oscillation amplitude of 200-nm squares is insensitive to temperature changes in this range. The amplitude of the oscillations varies with the gate voltage in the $B=0$ T trace of Fig. 1. Such modulation of the oscillation amplitude is reproducible and clearly observable in 300- and 400-nm squares. The number of recorded oscillations in samples of 200-nm dot size is too low to observe a corresponding behavior, since the applied gate voltage is limited by the onset of leakage current. The positions of the levels at low gate voltages ($V_G < -0.1$ V) change only slightly at low magnetic fields ($B<1$ T), whereas the positions of levels at higher gate voltages exhibit a complex behavior at very low magnetic fields ($B<0.2$ T). At a magnetic field of about $B=1$ T a splitting of the oscillation maxima is clearly observed in samples with 300-nm squares. The field value at which this splitting occurs depends on the dot size of the samples. In samples with 400-nm squares the splitting occurs at a magnetic field of $B=0.6$ T; in 200-nm samples at $B=1.5$ T.

The gate voltage positions of the oscillation maxima in dC/dV_G are plotted versus the magnetic field in Fig. 2. Weak oscillations are plotted as open circles as shown in Fig. 1. The magnetic field dependence of the maxima positions at $B=1$ T, where the splitting occurs, suggests a level anticrossing. Furthermore, we observe that those peaks dominating at low magnetic fields weaken as soon as satellite peaks occur at slightly higher gate voltage. With increasing magnetic field the former vanish and the latter become dominant.

Figure 3 elucidates the behavior described above. The magnetic field of adjacent traces increases by 0.01 T and the traces are slightly offset for clarity. Besides illustrat-

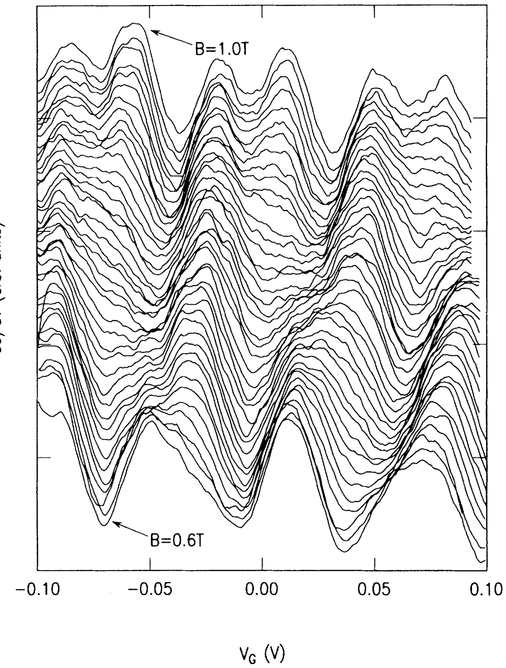

FIG. 3. Gate voltage derivative of the sample capacitance in a small magnetic field range (cf. Fig. 2). Traces were recorded in 0.01-T steps.

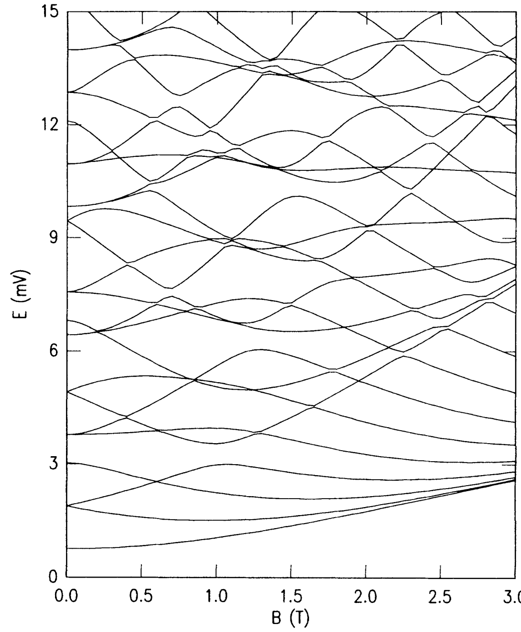

FIG. 4. Calculated energy spectra of an electron system confined in a square of width 120 nm.

ing in the upper half ($B>0.8$ T) the onset of the strong level splitting at about $B=1$ T this figure also shows that similar but less well-resolved level crossing occurs at lower magnetic fields. For instance, the lowest trace ($B=0.6$ T) exhibits a pronounced asymmetry in the shape of the maximum at $V_G=-0.04$ V with the maximum position shifted to lower gate voltage and a shoulder evolving at the high-gate-voltage slope. The shoulder becomes a separate peak at 0.04 T higher field. At a magnetic field of about 0.68 T the asymmetry is inverted with the shoulder on the low-gate-voltage side of the maximum. The same behavior occurs at slightly higher fields in the branch starting at $B=0.6$ T with an oscillation maximum at $V_G=0.01$ V.

We have calculated the magnetic field dependence of the electron energies in a quantum dot using the decoupled approximation. Furthermore, since the electron system is in the quantum limit with respect to motion in the z direction, we can neglect the energy dispersion for motion in this direction. Because the quantum oscillations are measured in terms of gate voltage not energy, we do not anticipate perfect agreement between calculated spectra and experimental data. However, the main features of the measured data are present in the calculated spectra. An analytical calculation of the single-electron energy spectrum was first done assuming a harmonic confinement in the x-y plane with rotational symmetry with respect to the z axis[1] $V(x,y)=m^*\times\Omega^2(x^2+y^2)/2$. The electron energies are then described by two quantum numbers (n,l):

$$W_{n,l}=\tfrac{1}{2}\hbar\omega_c l+(2n+|l|+1)(\tfrac{1}{4}\hbar^2\omega_c^2+\hbar^2\Omega^2)^{1/2}, \quad (1)$$

with ω_c the cyclotron frequency and $\hbar l$ the angular momentum of the electron. Model calculations previously performed for quantum wires[16] in the absence of magnetic field and extended to 0D structures in the classical limit[17,18] show that the self-consistent potential changes with gate voltage. It is parabolic at the threshold gate voltage but becomes square-well-like with increasing gate voltage. The results presented in Fig. 4 are calculated for electrons confined in a rectangular square-well potential with finite ($V_0=600$ meV) walls and width $W=120$ nm. These values approximate the self-consistently calculated potential at a gate voltage about 200 mV above the threshold voltage. The energy spectrum is obtained by diagonalizing the Hamiltonian in magnetic field calculated with a finite basis of solutions at zero magnetic field. The spectrum at zero magnetic field represents the well-known energy levels of a particle in a box with degeneracies of different levels corresponding to the fourfold symmetry of a square. This degeneracy is lifted by the magnetic field. Each branch corresponds to a twofold-degenerate energy level since electron spin is neglected. Spin splitting of the levels is observed at far higher magnetic fields than those discussed here ($B>4$ T). The evolution of each branch depends in detail on the assumptions made for the shape and symmetry of the confinement potential.[2,3] At low magnetic field ($l_B\approx W$) and high quantum numbers, a complicated level crossing commences. This agrees invariant to a change of the scale in real space, the magnetic fields where different states cross each other scale with the dot size. As discussed above, we observe the level crossings at higher fields with decreasing square size of the samples.

At high magnetic fields different branches coalesce at the Landau energies $(\nu+\frac{1}{2})\hbar\omega_c$, $\nu=0,1,2,\ldots$. In Fig. 4 this behavior is seen for the two lowest Landau energies at magnetic fields larger than $B=2$ T. In this field regime the magnetic length is smaller than the potential width and it becomes possible to distinguish between surface states with energies between the Landau levels and bulklike states close to the Landau energies. Experimentally, we observe an increase in the oscillation amplitudes at high fields indicating that more and more energy levels condense into bulk Landau states. However, every state is not resolved in our measurements, and the strengths of the observed oscillations vary significantly (cf. Fig. 3). In contrast, our model predicts equally strong maxima, since each branch corresponds to a twofold-degenerate energy state. Only certain maxima are expected to grow strong at high magnetic fields when many levels merge into Landau levels or, occasionally, at low fields, when several levels happen to cross one another. Our calculation of the energy spectra does not include broadening of the energy levels by potential fluctuations originated, e.g., by impurities or surface roughness of the electrostatic confinement. The impact of fluctuations in the confinement potential on the broadening of a level depends on the probability distribution of the wave function in the quantum dot. A level with large probability amplitude in the center of the dot will be less affected by surface roughness than a state with large probability for finding the electron near the surface. We may, therefore, expect that states in which the electron wave function is predominantly in the center of the dot exhibit larger maxima in the measurement. In the harmonic-oscillator model with rotational symmetry [cf. Eq. (1)] such states have quantum numbers $n=0$ and $l\geq 0$. They increase monotonically with magnetic field and are the first states to approach the Landau energies $(\nu+\frac{1}{2})\hbar\omega_c$, $\nu=0,1,2,\ldots$. In fact the dominant peaks in the measurements behave similarly except where level anticrossing occurs.

A theory that explains the observations quantitatively must take into account that not only the Fermi energy, but also the confinement potential, changes with the gate voltage. Furthermore, it is predicted that electron correlation plays an important role.[19] The exact form of the potential and the shape of the dot will determine the detailed behavior of the surface states. Our results also indicate that inclusion of surface roughness is important to explain the signal strength. A systematic study of the perimeter contributions to the Landau quantization in quantum dots may reveal a variety of detailed information about the artificially imposed confinement that has not been accessible so far.

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[(a)]Permanent address: Universidade de Campinas, Department de Fisica Do Estado Solido e Ciencia dos Materiais, 13081, Campinas, São Paulo, Brazil.

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