RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 45, NUMBER 4 15 JANUARY 1992-II

Spin-singlet–spin-triplet oscillations in quantum dots

M. Wagner
Hitachi Cambridge Laboratory, Hitachi Europe Limited, Cambridge CB3 0HE, United Kingdom

U. Merkt and A. V. Chaplik*
Institut für Angewandte Physik, Universität Hamburg, Jungiusstrasse 11, 2000 Hamburg 36, Germany
(Received 12 August 1991)

Two interacting electrons confined to a disk on a semiconductor surface are considered in a perpendicular magnetic field. As it is appropriate for experimental realizations, we use a two-dimensional harmonic-oscillator well to confine the electrons in the plane of the disk. We predict oscillations between spin-singlet and spin-triplet ground states as a function of the magnetic field strength. Phase diagrams describing this peculiar manifestation of the electron-electron interaction in a quantum dot are calculated for GaAs and experiments to verify them are proposed.

Nanostructure technologies allow the lateral confinement of two-dimensional electron gases in heterojunctions or metal-oxide-semiconductor structures to widths comparable to the effective Bohr radius a^* of the host semiconductor.[1] In this case we have electron systems with discrete energy spectra that are commonly called zero-dimensional systems or quantum dots.[2–9] Since their widths in the x-y plane are much larger than their extent in the z direction, which is the growth direction of the underlying semiconductor structure, quantum dots may be regarded as artificial atoms with disklike shapes. Electron numbers as low as one or two per dot have already been realized.[6,9]

So far, quantum dots have been investigated experimentally by capacitance-voltage spectroscopy[3] and transport measurements,[4,5] as well as by far-infrared spectroscopy.[6–9] Capacitance-voltage and transport measurements are not favorable for the study of isolated dots since they require coupling to external contacts. Particularly for small dots their interpretation is additionally hampered by Coulomb blockade. Despite these difficulties, much information on the *single-electron* energy spectra could be deduced from transport data.[5] In many cases the value of far-infrared spectroscopy[6–9] is limited as a consequence of the approximately harmonic shapes of the confining potentials and the associated validity of the generalized Kohn theorem.[10,11] This theorem states that, for strictly harmonic potentials, dipole radiation can only probe the center-of-mass motion of all electrons but is inadequate to see any effect due to the electron-electron interaction.

Here, we predict spin oscillations of the ground state of two electrons in a harmonic quantum dot as a function of the magnetic field strength, which are a peculiar consequence of the electron-electron interaction and the Pauli exclusion principle. Hence, they are a direct manifestation of the *two-electron* states in the quantum dot. The oscillations should be accessible to a different type of experiment, namely spin susceptibility[12] and magnetization measurements[13] that previously have been successfully applied to study electronic properties of two-dimensional electron gases in $GaAs/Ga_{1-x}Al_xAs$ and related heterostructures.

In experimentally realized dots, the motion in the z direction is always frozen out into the lowest electric subband $E_i=0$. Since the corresponding extent of the wave function is much less than the one in the x-y plane, we can treat the dots in the two-dimensional limit of thin disks. For most dots, a harmonic oscillator is a very good approximation to describe the lateral confinement of the electrons.[2–6,14] Hence we consider two electrons of effective masses m^* in the $z=0$ plane in the harmonic potential $\frac{1}{2}m^*\omega_0^2(x^2+y^2)$ of characteristic frequency ω_0 or oscillator length $l_0=(\hbar/m^*\omega_0)^{1/2}$. The perpendicular magnetic field ($\mathbf{B}\|z$) is in the symmetric gauge described by the vector potential $\mathbf{A}=\frac{1}{2}(-y,x,0)\mathbf{B}$. A dielectric constant ϵ accounts for the host semiconductor. Ignoring the Zeeman spin splitting for the present, the Hamiltonian can be separated into center-of-mass and relative-motion terms as

$$H=\frac{[\mathbf{P}+Q\mathbf{A}(\mathbf{R})]^2}{2M}+\frac{1}{2}M\omega_0^2R^2+\frac{[\mathbf{p}+q\mathbf{A}(\mathbf{r})]^2}{2\mu}+\frac{1}{2}\mu\omega_0^2r^2+\frac{e^2}{4\pi\epsilon\epsilon_0}\frac{1}{r} \quad (1)$$

by introducing the center-of-mass coordinates $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$, $\mathbf{P}=\mathbf{p}_1+\mathbf{p}_2$, the total mass $M=2m^*$, and charge $Q=2e>0$, as well as the relative coordinates $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$, $\mathbf{p}=(\mathbf{p}_1-\mathbf{p}_2)/2$, the reduced mass $\mu=m^*/2$, and charge $q=e/2$.

This separability and the cylindrical symmetry of the problem allow us to write the two-particle wave function in plane polar coordinates $\mathbf{r}=(r,\phi)$ in the form $\Psi(\mathbf{R})\xi(r)\exp(im\phi)$. The spatial part of the total wave function is symmetric or antisymmetric with respect to particle permutation ($\phi\rightarrow\phi+\pi$) for even, respectively odd, azimuthal quantum numbers m. Since the Pauli exclusion principle requires the total wave function to be antisymmetric, we therefore have spin singlet ($S=0$) and triplet ($S=1$) states for even and odd m.[15] The energy eigenvalues of the Hamiltonian in Eq. (1) are the sum of the center-of-mass energy and the energy of the relative

motion. The former is given by[10]

$$E_{N,M}=\hbar(2N+|M|+1)\left[\omega_0^2+\left(\frac{\omega_c}{2}\right)^2\right]^{1/2}+\hbar\frac{\omega_c}{2}M \tag{2}$$

with cyclotron frequency $\omega_c=eB/m^*$, radial ($N=0,1,2,\ldots$), and azimuthal ($M=0,\pm1,\pm2,\ldots$) quantum numbers. The energy of the relative motion has corresponding quantum numbers n and m and includes the electron-electron interaction. The spin of the two electrons leads to an additional Zeeman energy

$$E_S=g^*\mu_B BS_z=g^*\frac{m^*}{m_e}\frac{\hbar\omega_c}{2}S_z \tag{3}$$

described by an effective Landé factor g^*. Thus triplet states split into three distinct levels, while singlet states remain unchanged. The exact eigenvalues $E_{n,m}$ of the relative motion including the Zeeman energy are calculated numerically.[15]

We now investigate the ground state of the two-electron system as a function of dot size and magnetic field strength. Since the center-of-mass quantum numbers N,M and the quantum number m are conserved by the Coulomb interaction, the ground state has the quantum numbers $N=0$, $M=0$, $n=0$, $m\leq 0$, and only m is to be determined. The important feature of the ground state to be discussed here is that its angular momentum $\hbar m$ does depend on the Coulomb interaction. In Fig. 1 we have plotted the energy of the states $N=0$, $M=0$, $n=0$, $m\leq 0$ for vanishing Landé factor $g^*=0$ and dot size $l_0/a^*=3$ as a function of the ratio ω_c/ω_0 of cyclotron and oscillator frequency. In Fig. 1(a) we neglect the Coulomb interaction and the $m=0$ state is always the ground state. If, however, we include the Coulomb interaction in Fig. 1(b), the state $m=0$ remains as a ground state only for low magnetic fields. As the magnetic field increases, this state rises in energy while the states $m=-1,-2,-3,\ldots$ drop, thus leading to a sequence of different ground states $m=0,-1,-2,-3,\ldots$ as the magnetic field is swept. Since the total spin of the two electrons is $S=[1-(-1)^m]/2$, this entails an alternating sequence of singlet and triplet states.

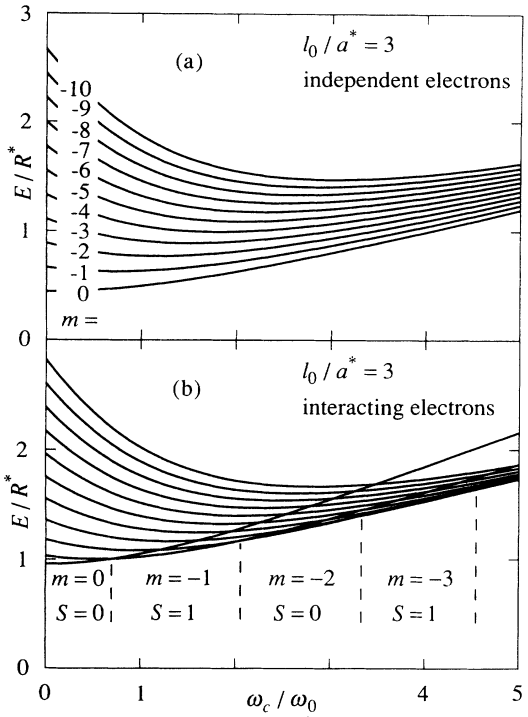

FIG. 1. Eigenenergies in units of the effective Rydberg constant R^* vs the ratio ω_c/ω_0 for a dot size $l_0/a^*=3$ and Landé factor $g^*=0$. The family of states $N=0$, $M=0$, $n=0$, $m\leq 0$ is shown (a) without and (b) including Coulomb interaction between the two electrons. As the ratio ω_c/ω_0 increases, the Coulomb interaction leads to a sequence of different ground states $m=0,-1,-2,\ldots$, and concomitant changes of the total spin $S=0,1,0,\ldots$.

The reason for these changes of symmetry of the ground state is found in the competition of the various energies contributing to the energy of the relative motion. On the one hand, a higher angular momentum $\hbar m$ means higher rotational energy, but on the other hand, the average distance between the two electrons is then increased and hence the Coulomb energy gets smaller. With the relative strength of the Coulomb interaction varying as $[1+(\omega_c/2\omega_0)^2]^{-1/4}l_0/a^*$, the optimum number m thus depends on the dot size and on the magnetic field.

By properly designing the dot size l_0/a^* and adjusting the relative strength of the magnetic field ω_c/ω_0, it should therefore be possible to investigate different ground states. A suitable tool to visualize this is a phase diagram in the $l_0/a^*-\omega_c/\omega_0$ plane. The transition $m\rightarrow m-1$ between a singlet and a triplet ground state is given by the condition $E_{0,m}=E_{0,m-1}$ ($m\leq 0$). These are the only possible transitions as long as the Zeeman spin splitting is ignored. For a negative Landé factor $g^*<0$, the spin-splitting energy in magnetic fields will lower the energy of the spin $S_z=+1$ component of the triplet states while leaving the singlet states unchanged. Thus the phases of the triplet states will increase at the cost of the singlet phases, and eventually, for high magnetic fields, the singlet ground states are totally suppressed. Singlet-triplet transitions only exist for $E_{0,m}<E_{0,m+1}$ (m even), and for $E_{0,m-1}<E_{0,m-2}$ (m odd). In particular, the relation $E_{0,m}=E_{0,m-1}=E_{0,m-2}$ (m odd) defines triple points where singlet phases cease to exist. Beyond this point we are left with phase transitions between triplet states described by the condition $E_{0,m}=E_{0,m-2}$ (m odd).

In Fig. 2 we show phase diagrams based on an exact numerical diagonalization of the Coulomb interaction. In Fig. 2(a) the Landé factor is set to zero. Then we have only singlet-triplet phase transitions $m\rightarrow m-1,m-2,\ldots$ starting with the $m=0$ singlet phase at zero magnetic field. In Fig. 2(b) we assume the Landé factor $g^*=-0.44$ and effective mass $m^*=0.067m_e$ of bulk GaAs conduction-band electrons. For higher magnetic fields the singlet ground states now completely vanish and only triplet-triplet phase transitions m (odd)$\rightarrow m-2,m-4,\ldots$ are left. For a typical experimental dot size of $l_0/a^*=3$ the last singlet phases present are at $\omega_c/\omega_0<0.69$ and $2.2<\omega_c/\omega_0<2.9$.

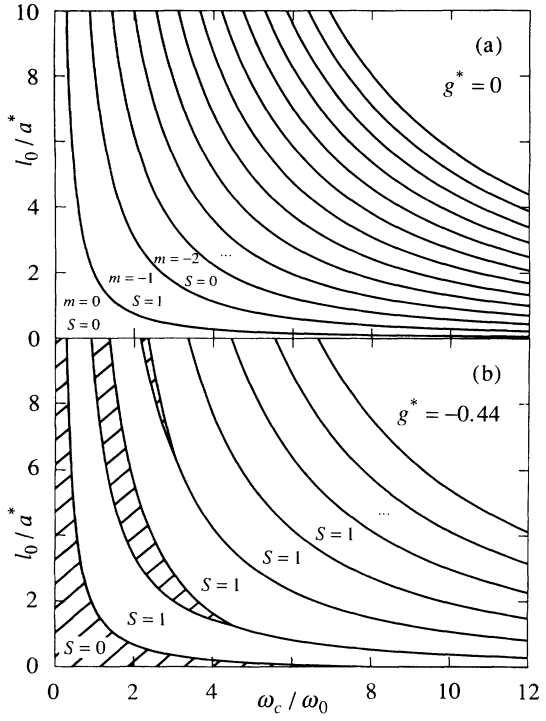

FIG. 2. Phase diagram for singlet and triplet ground states calculated (a) in the absence and (b) in the presence of Zeeman spin splitting with a Landé factor $g^* = -0.44$ appropriate for GaAs. In the absence of spin splitting the singlet phase $m = 0$ ($S = 0$) in zero and low magnetic fields is followed by $m = -1$ ($S = 1$), $m = -2$ ($S = 0$), ... phases as the magnetic field strength is increased. In the presence of spin splitting the singlet phases are strongly suppressed, and only the hatched $m = 0, -2, -4$ singlet phases are left.

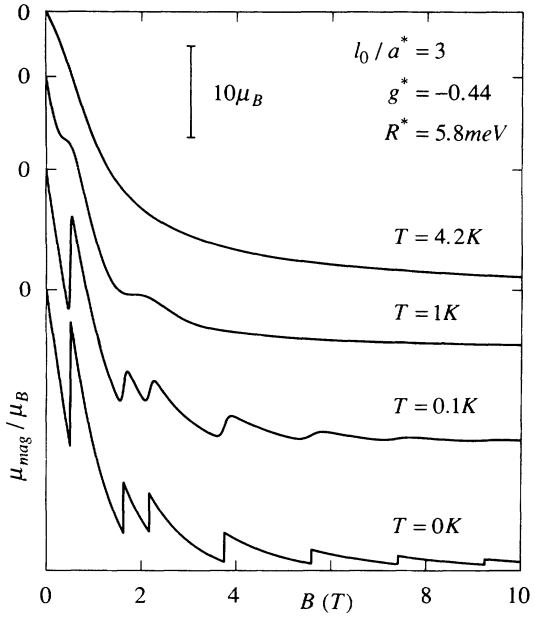

FIG. 3. Magnetic moment μ_{mag} in units of the Bohr magneton μ_B of a single quantum dot with two electrons vs magnetic field strength for GaAs parameters. The curves for various temperatures have been successively displaced for clarity. Oscillations caused by changes of the azimuthal angular momentum and the total spin become visible for temperatures less than about 1 K.

Essential features of these phase diagrams can already be understood, if we calculate the Coulomb energy E_{Coulomb} in first-order perturbation theory, which is valid for $[1+(\omega_c/2\omega_0)^2]^{-1/4} l_0/a^* \ll 1$. In this case we find ($n = 0$)

$$E_{\text{Coulomb}} = \frac{l_0}{a^*} \hbar\omega_0 \left[1 + \left(\frac{\omega_c}{2\omega_0}\right)^2\right]^{1/4} \frac{(2|m|-1)!!}{(2|m|)!!} \left(\frac{\pi}{2}\right)^{1/2}. \tag{4}$$

For dot sizes $l_0/a^* \gtrsim 1$, strong magnetic fields ($\omega_c \gg \omega_0$), and $|g^*|m^*/m_e \ll 1$ we then obtain for singlet-triplet phase transitions $m \rightarrow m-1$

$$\frac{l_0}{a^*} = \frac{1}{\sqrt{2\pi}} \frac{(2|m|+2)!!}{(2|m|-1)!!} \times \left[\left(\frac{2\omega_0}{\omega_c}\right)^{3/2} + 2\left(\frac{\omega_c}{2\omega_0}\right)^{1/2} (-1)^m g^* \frac{m^*}{m_e}\right] \tag{5}$$

and for triplet-triplet phase transitions $m \rightarrow m-2$ (m odd)

$$\frac{l_0}{a^*} = \frac{1}{\sqrt{2\pi}} \frac{2(2|m|+4)!!}{(2|m|-1)!!(4|m|+5)} \left(\frac{2\omega_0}{\omega_c}\right)^{3/2}. \tag{6}$$

By using Eqs. (5) and (6), the exact triple point at $\omega_c/\omega_0 = 4.48$, $l_0/a^* = 1.27$ in Fig. 2(b) is approximated to be $\omega_c/\omega_0 = 4.76$, $l_0/a^* = 1.16$. Hence, the perturbational results can be used advantageously to estimate phase diagrams for other effective masses m^* or Landé factors g^*.

Based on the exact eigenenergies, the magnetic moment $\mu_{\text{mag}}(B)$ of a single dot with two electrons is plotted in Fig. 3 in units of the Bohr magneton $\mu_B = e\hbar/2m_e$ for various temperatures. At $T = 4.2$ K we find a significant but smooth diamagnetic behavior while for temperatures less than 1 K strong oscillations become visible. These contributions with amplitudes of more than $10\mu_B$ exceed the paramagnetic spin contribution $-g^*\mu_B$ of two independent electrons by more than an order of magnitude and show most obviously the two-particle nature of the ground state at intermediate magnetic field strengths. The fact that these oscillations are Coulomb induced is easily understood for zero temperature where we have $\mu_{\text{mag}}(B, T=0) = -\partial E_{\text{ground}}/\partial B$. By comparing Figs. 1(a) and 1(b) we see that discontinuities in this derivative occur at the phase boundaries and are therefore a clear-cut consequence of the Coulomb interaction. For instance, the sharp drop of the magnetic moment at $B = 0.51$ T is caused by the first singlet-triplet transition $m = 0 \rightarrow m = -1$. In high magnetic fields ($\omega_c \gg \omega_0$) the oscillations vanish and we are left with the saturation moment $\mu_\infty/\mu_B = -2m_e/m^* - g^* = -29.4$, valid for two independent electrons in GaAs.

To conclude, we predict spin singlet-triplet and triplet-triplet transitions of the ground state of two *interacting* electrons in quantum dots in a perpendicular magnetic field. In principle, our prediction can be verified by spin

susceptibility or magnetization measurements at low temperatures ($T < 1$ K). We are aware of the intensity problems resulting from the low number of electrons, even if arrays of 10^9 dots/cm^2 are used. Perhaps it is particularly challenging that the analogous singlet-triplet transition from para- ($S=0$) to ortho- ($S=1$) helium, predicted at about $B=4\times10^5$ T in the vicinity of white dwarfs and pulsars,[16] also remains to be observed.

We thank J. Huser for the supply of numerical routines and acknowledge financial support from the Deutsche Forschungsgemeinschaft.

---

*Permanent address: Institute of Semiconductor Physics, U.S.S.R. Academy of Science, Novosibirsk, U.S.S.R.

[1]G. W. Bryant, Phys. Rev. Lett. **59**, 1140 (1987).

[2]M. A. Reed, J. N. Randall, R. J. Aggarwal, R. J. Matyi, T. M. Moore, and A. E. Wetsel, Phys. Rev. Lett. **60**, 535 (1988).

[3]W. Hansen, T. P. Smith III, K. Y. Lee, J. A. Brum, C. M. Knoedler, J. M. Hong, and D. P. Kern, Phys. Rev. Lett. **62**, 2168 (1989); **64**, 1991 (1990); W. Hansen, T. P. Smith III, K. Y. Lee, J. M. Hong, and C. M. Knoedler, Appl. Phys. Lett. **56**, 168 (1990).

[4]L. P. Kouwenhoven, F. W. J. Hekking, B. J. van Wees, C. J. P. M. Harmans, C. E. Timmering, and C. T. Foxon, Phys. Rev. Lett. **65**, 361 (1990).

[5]P. L. McEuen, E. B. Foxman, U. Meirav, M. A. Kastner, Y. Meir, N. S. Wingreen, and S. J. Wind, Phys. Rev. Lett. **66**, 1926 (1991).

[6]Ch. Sikorski and U. Merkt, Phys. Rev. Lett. **62**, 2164 (1989); **64**, 3100 (1990); Surf. Sci. **229**, 282 (1990).

[7]A. Lorke, J. P. Kotthaus, and K. Ploog, Phys. Rev. Lett. **64**, 2559 (1990).

[8]T. Demel, D. Heitmann, P. Grambow, and K. Ploog, Phys. Rev. Lett. **64**, 788 (1990).

[9]B. Meurer, D. Heitmann, and K. Ploog (unpublished).

[10]P. A. Maksym and T. Chakraborty, Phys. Rev. Lett. **65**, 108 (1990).

[11]P. Bakshi, D. A. Broido, and K. Kempa, Phys. Rev. B **42**, 7416 (1990).

[12]M. Dobers, Surf. Sci. **229**, 126 (1990).

[13]J. P. Eisenstein, H. L. Stormer, V. Narayanamurti, A. Y. Cho, A. C. Gossard, and C. W. Tu, Phys. Rev. Lett. **55**, 875 (1985); J. P. Eisenstein, Appl. Phys. Lett. **46**, 695 (1985).

[14]A. Kumar, S.E. Laux, and F. Stern, Phys. Rev. B **42**, 5166 (1990).

[15]U. Merkt, J. Huser, and M. Wagner, Phys. Rev. B **43**, 7320 (1991).

[16]G. Thurner, H. Herold, H. Ruder, G. Schlicht, and G. Wunner, Phys. Lett. **89A**, 133 (1982).