Magneto-optics in a square-well quantum dot

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We calculate the optical transition coefficient of two electrons confined in a square-well quantum dot under a magnetic field. We determined that there are many different types of absorption, some induced when interacting electrons are under a magnetic field. As the magnetic field becomes strong, a spin-singlet–spin-triplet transition of the ground state occurs, resulting in a drastic change in optical transition spectra. In a strong magnetic field, an optical transition with double the cyclotron frequency is induced by electron-electron interaction and the confining potential.

A square-well quantum dot, in which electrons are confined by a heterostructure of compound semiconductors,[1] is a simple but typical zero-dimensional nanostructure, as is a quantum dot with parabolic confinement.[2–4] This square-well quantum dot has many-body effects of electrons on optical transitions,[5] which are never observed in parabolically confined systems.[6,7] This is because spatial coordinates of electrons in a square-well quantum dot cannot be divided into center-of-mass coordinates and relative coordinates, in contrast with electrons in a parabolically confined quantum dot. In a very small square-well quantum dot, electron correlation of a few electrons is weak, so the optical transition spectra are similar to those of a single electron. Let us denote Γ_1 as the least energy absorption, and Γ_4 as the second-least energy absorption. When a square-well quantum dot containing two electrons becomes larger than 30 nm, electron-electron interaction affects optical transitions—the intensity of Γ_1 becomes weaker, and two kinds of absorption, Γ_2 and Γ_3, appear between Γ_1 and Γ_4. When the size of this quantum dot is much larger than 50 nm, Γ_2 becomes very strong compared to Γ_3 and Γ_4. This increase is due to the strong correlation of electrons, which if the quantum dot is as large as 1 mμm, can result in a Wigner lattice.

If two electrons are confined in a square-well quantum dot with a side L, the density of the electron is $2/L^2$. When L is about 20 nm, this electron density is comparable to the density of electrons in a double heterostructure used for a typical quantized Hall sample, i.e., about 5×10^{15} (m^{-2}).[8] When a magnetic field is applied to this quantum dot, magnetic length $l_B=\sqrt{\hbar/eB}$ is expected to be 26 nm when $B=1$ T. Thus we can expect correlated electrons affected by an interplay between the magnetic length and the size of quantum dots. In this paper, the effects of a magnetic field on the optical transition of interacting electrons in a square-well quantum dot are investigated based on numerical diagonalizations of a Hamiltonian using the effective-mass approximation.

We consider a quantum dot in which electrons are confined by a heterostructure of compound semiconductors. As this quantum dot is assumed to be a well with finite depth, we introduce a Hamiltonian for two electrons in a two-dimensional space with an effective-mass approximation:

$$\hat{H}=\frac{1}{2m}\sum_{i=1,2}[(\mathbf{p}_i+e\mathbf{A}_i)^2+V(\mathbf{r}_i)]+\frac{e^2}{4\pi\epsilon_r\epsilon_0}\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}+g^*\mu_B B\sum_{i=1,2}s_{i,z}, \tag{1}$$

$$V(x,y)=\begin{cases}-V_0 & \text{if } |x|<L/2 \text{ and } |y|<L/2\\ 0 & \text{otherwise.}\end{cases} \tag{2}$$

We assume that an electron in a semiconductor material can be described by effective mass $m=0.067m_0$ for GaAs, where m_0 is the free-electron mass. The shape of a quantum dot is determined by the one-body potential $V(x,y)$, where a side of the square quantum dot is denoted by L. The heterostructure is assumed to be $Al_{0.45}Ga_{0.55}As/GaAs$, so V_0 is taken to be 0.36 eV and ϵ_r is 10.9 for GaAs. The last term in the Hamiltonian is the Zeeman energy where the effective Landé factor g^* is taken to be -0.44, and μ_B is the Bohr magneton. We take a vector potential

$$(A_{i,x},A_{i,y})=B[-y_i\alpha,x_i(1-\alpha)] \tag{3}$$

in order to introduce a magnetic field B, where α is introduced for a gauge transformation. $\alpha=\frac{1}{2}$ provides a circular gauge.

When $|n\rangle$ is the nth eigenstate of the Hamiltonian H, and E_n is its eigenenergy, an optical transition matrix element from the pth state $|p\rangle$ to the qth state $|q\rangle$ can be derived from

$$\langle q|\hat{H}_{\mathrm{em}}|p\rangle=i\frac{e}{\hbar}(E_q-E_p)\mathbf{A}_{\mathrm{ex}}(0)\cdot\langle q|\left(\sum_{j=1,2}\mathbf{r}_j\right)|p\rangle \tag{4}$$

within the dipole approximation, where $\mathbf{A}_{\mathrm{ex}}(0)$ is the vector potential of external radiation fields. The absorption coefficient

$$\Gamma_{p,q}=\left|\langle q|\sum_j x_j|p\rangle\right|^2+\left|\langle q|\sum_j y_j|p\rangle\right|^2 \tag{5}$$

is proportional to the intensity of absorption from $|p\rangle$ to $|q\rangle$, thus the optical transition is allowed as a two-electron phenomenon if $\Gamma_{p,q}\neq0$. To sum up, we determine $|n\rangle$, then

calculate Γ.[5] By these dipole matrix elements, we calculate the quantity $\Theta(E)$ proportional to the absorption of photon with energy E defined as

$$\Theta(E)=\eta\sum_{p,q}\Gamma_{p,q}\frac{\rho_p-\rho_q}{[E-(E_q-E_p)]^2+\eta^2}, \tag{6}$$

where $\hbar/\eta$ is a characteristic relaxation time.[9] In our calculations η is taken to be 0.2 meV, resulting in $\hbar/\eta\sim 3\times 10^{-12}$ s. This relaxation time is smaller than that expected in quantum dots,[10,11] so absorption spectra in actual quantum dots may be more sharp than the absorption spectra shown here. ρ_p is the Boltzmann factor $e^{-\beta E_p}/\mathcal{Z}$ of thermal distribution. We assume that a very low temperature $\beta^{-1}=0.1$ meV enables us to ignore the optical transition between excited states with the same spin. Therefore, the partition function $\mathcal{Z}$ is calculated by considering the least-energy state among spin-singlet states and the least-energy state among spin-triplet states.

Let us briefly discuss our method of calculating $|n\rangle$. Before we construct Hamiltonian matrix elements with the eigenfunctions of a harmonic oscillator, we define the scale parameter ξ as $(x_j,y_j)=(\xi X_j,\xi Y_j)$ so as to make X_j and Y_j dimensionless variables. We use functions $\psi_n(X)\psi_m(Y)$ as the basis of a single electron: $\psi_n(X)=A_nH_n(X)\exp(-X^2/2)$, where the $H_n(X)$'s are Hermite polynomials and the A_n's are the normalization constants. Matrix elements of the single-electron term in the Hamiltonian can be easily obtained using the error function $\mathrm{erf}(z)=\int_0^z dt\exp(-t^2)$. The size of the quantum dot L appears in the matrix elements of the potential term, so we introduce the ratio $\zeta=L/\xi$, which will be treated as a variational parameter. Matrix elements of the Coulomb interaction term, i.e., $\langle i_1,i_2;i_3,i_4|e^2/(4\pi\epsilon_r\epsilon_0 r)|j_1,j_2,j_3,j_4\rangle$, can be obtained analytically.

Though each matrix element depends on ζ, exact energy eigenvalues of the Hamiltonian must be independent of ζ because of ζ is not a physical variable. However, if we numerically diagonalize a Hamiltonian matrix made of a finite set of base functions, the obtained eigenvalues will depend on ζ. Therefore, we treat ζ as a variational parameter in our numerical calculations in order to minimize the ground-state energy. We used six bases for each direction and particle.[12] Another check on the sufficiency of the number of base functions can be made by a gauge transformation described by α. When a perturbation expansion on the order of the coupling constant of electron-electron interaction or a $1/N$ expansion on the order of the number of components of material fields is used for analysis, the gauge invariance of each order is guaranteed by the Ward-Takahashi identity.[13] However, as in our case, a Hamiltonian matrix constructed by a finite set of base functions is usually not invariant under the gauge transformation, though we know that the exact solution must be invariant under any gauge transformation. Thus, if our solutions of low-lying states are not affected by gauge transformation, these solutions seem to be accurate. We have checked our numerical results using a gauge transformation described by α. The results were calculated using $\alpha=0.5$, and these results are invariant under $\alpha=0.5\pm 0.2$.

The least-energy absorption of two electrons, denoted by Γ_1, is due to a transition from the ground state to an excited state containing the single-electron first excited states. One of these first excited states has odd parity in the x direction, and the other has odd parity in the y direction. When a magnetic field is applied to the quantum dot, the circular property of the magnetic field causes an energy difference between these two states, resulting in a split of the absorption energy of Γ_1. Let us denote these optical transitions by Γ_{1-} and Γ_{1+}.[14] When noninteracting electrons are under a strong magnetic field, the absorption energy of Γ_{1-} goes to 0, and that of Γ_{1+} goes to $\hbar\omega_c$, where $\omega_c=eB/m$ is the cyclotron frequency. Figure 1 shows the intensity of absorptions when L is taken to be 40 nm. The absorption induced by electron-electron interaction, as denoted by Γ_2, is also divided into two kinds by an applied magnetic field. The kind with lower absorption energy, which is the third-least energy absorption under the magnetic field seen in Fig. 1, is enhanced by the magnetic field, and becomes comparable to $\Gamma_1 s$ when $B\sim 6$ T. On the other hand, the other kind originating from Γ_2 becomes weaker as the magnetic field becomes strong, so this kind cannot be seen in Fig. 1. Therefore, an applied magnetic field encourages some kinds of absorption induced by electron-electron interaction, and discourages other kinds. This can be understood by considering the angular momentum of these states.

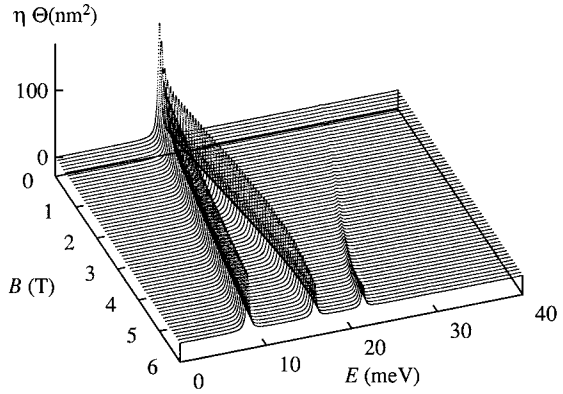

FIG. 1. Intensity of absorption as a function of magnetic field B and absorption energy E when a side of the quantum dot L is 40 nm. The effective Landé factor g^* is -0.44.

As the quantum dot becomes larger in a weak magnetic field, electron-electron interaction causes a transition of the ground state from the spin-singlet state to the spin-triplet state. In Fig. 2, the energy spectra of two interacting electrons in a square-well quantum dot are shown as a function of the magnetic field when L is 60 nm. Note that g^* is taken to be 0 in Fig. 2. When B is smaller than about 3 T, the ground state is spin singlet, denoted by the broken line. On the other hand, the ground state becomes a triplet state when B is larger than about 3 T. This single-triplet transition has the same origin as in a quantum dot with parabolic confinement.[15,16] Let us turn to the optical transition coefficient. In Fig. 3, the intensity of the absorption is shown when L is 60 nm. When $B=0$, Γ_1 has an absorption energy of about 6 meV. Γ_2, which is induced by electron-electron interaction, is also seen at $E\sim 15$ meV. When the strength of the magnetic field is less than about 2 T, Γ_1 behaves similarly to Γ_1 in Fig. 1. In the region of $B\sim 2$ T, a drastic change in absorption spectra is brought about by a change of

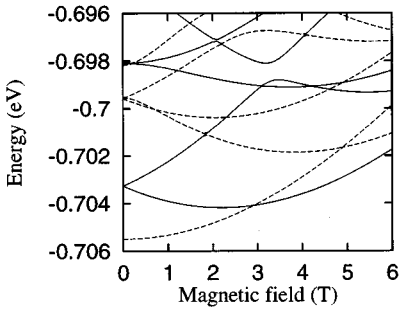

FIG. 2. Energy spectra of two electrons in a quantum dot with $L=60$ nm and $g^*=0$ as functions of B. The spin-singlet–spin-triplet transition is seen at $B\sim 3$ T. Broken lines denote spin-singlet states, and solid lines denote spin-triplet states.

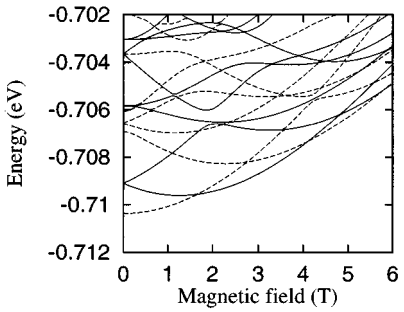

FIG. 4. Energy spectra of two electrons in a quantum dot with $L=80$ nm and $g^*=0$ as functions of B. Broken lines denote spin-singlet states and solid lies denote spin-triplet states. Multiple singlet-triplet transitions are seen when B is changed from 0 to 6 T.

the magnetic-field strength, because of the spin-singlet–spin-triplet transition of the ground state discussed above. The least and the next-least energy absorptions in this region behave similarly to absorptions between singlet states observed when the applied magnetic field is weak; however, the energy split induced by the magnetic field is smaller than that of singlet states. This is because triplet states are apt to have a higher angular momentum because of Pauli's principle, even without a magnetic field. Let us turn to optical transitions with higher energy, which have remarkable properties. With absorption energies of about 10 meV or more, very strong optical transitions are seen, as in Fig. 3. These optical transitions are due to electron-electron interaction, because they do not appear when there is no electron-electron interaction. Triplet states are influenced more than singlet states, because the strong repulsion between electrons causes a drastic change in orbital wave functions having a higher angular momentum.

When a quantum dot becomes larger, spin-singlet–spin-triplet oscillations can be observed even in a small range of magnetic fields. In Fig. 4, energy spectra of interacting electrons are shown when $L=80$ nm and $g^*=0$, where spin-singlet–spin-triplet transitions are seen at $B\sim 1.7$ T, $B\sim 4.1$ T, and $B\sim 5.6$ T. In Fig. 5, the intensity of the absorption is shown when L is 80 nm. In this quantum dot, the energy difference between the least-energy state among singlet states and that among triplet states is so small that a mixture of optical transitions of singlet and triplet states is seen at a considerable temperature, i.e., ~ 1 K. In a strong magnetic field much greater than 1 T, higher-energy absorption is very complex, and consists of many different types of optical transitions. An optical transition with a cyclotron frequency—its absorption energy is approximately 1.7 B (meV)—is composed of several types. Note that the absorption lines in Fig. 5 exhibit an anticrossing behavior as the magnetic field changes. This behavior is also caused by electron-electron interaction, as is seen in a Coulomb-coupled pair of parabolic quantum dots[17] and in a square-well quantum dot under an external electric field.[18]

It is remarkable that optical transitions at double the cyclotron frequency are very strong in a strong magnetic field. These transitions, which also exhibit anticrossing behavior, are induced when interacting electrons are confined in a square-well quantum dot under a magnetic field. When electrons are under a strong magnetic field, each electron has an effective size determined by the magnetic length. Therefore, an electron confined by the magnetic field behaves as an atom with an excitation energy of $\hbar\omega_c$. When the atom is in its ground state, the bare electron is in the first Landau level. If the atom is excited to the first excited state, the bare electron is in the second Landau level. In a free space, these atoms can move and interact with each other by the Coulomb

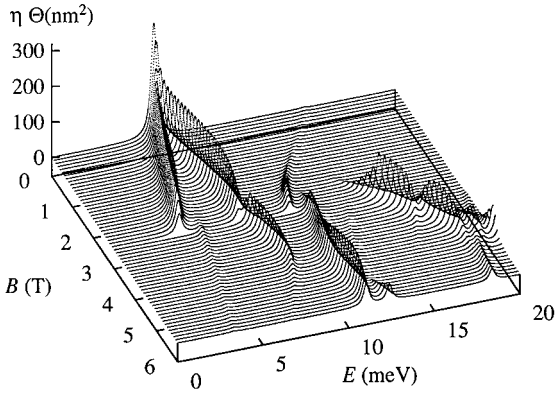

FIG. 3. Intensity of absorption as a function of magnetic field B and absorption energy E when two interacting electrons are confined in a square-well quantum dot with $L=60$ nm. The effective Landé factor g^* is -0.44.

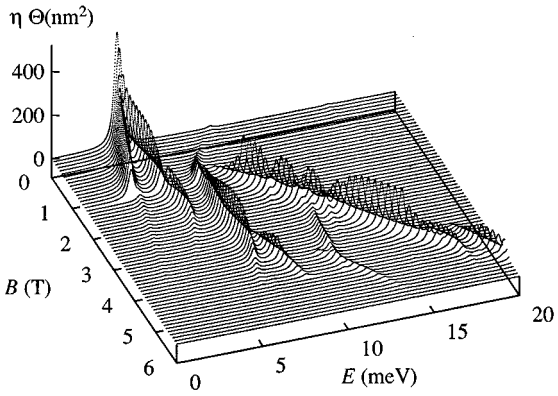

FIG. 5. Intensity of absorption as a function of magnetic field B and absorption energy E when $L=80$ nm. The effective Landé factor g^* is -0.44.

interaction of bare electrons because each Landau level has a large degree of degeneracy. When two atoms are confined in a square-well quantum dot, interaction between atoms can affect optical transition spectra of each atom. Optical transitions at double the cyclotron frequency may be due to this kind of effect.

We have calculated the absorption coefficient of two electrons confined in a square-well quantum dot under a magnetic field. There are several types of absorption induced by the Coulomb interaction in a magnetic field. In a stronger magnetic field, the spin-singlet–spin-triplet transition of the ground state has a considerable effect on absorption spectra. This effect occurs in a weaker magnetic field if electrons are confined in a larger quantum dot. Absorption at double the cyclotron frequency is due to interaction between two atoms which consist of an electron in a magnetic field. Therefore, we can expect a structure of interacting electrons confined by a heterostructure of compound semiconductors under a magnetic field in far-infrared absorption spectroscopy that is well worth studying.

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