Effect of the Coulomb interaction on far-infrared absorption in a square-well quantum dot

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We calculate far-infrared-absorption spectra and the absorption coefficient of two electrons confined in a square-well quantum dot as the strength of the Coulomb interaction between the electrons is changed. We determine that there are many different types of absorption, some induced by the Coulomb interaction. This is because electrons are confined by a heterostructure of compound semiconductors, so a square-well potential allows the electron-electron interaction to affect the far-infrared-absorption spectra. When a side of the quantum dot becomes larger than 50 nm, one or more types of absorption become much more intense than the others. This suggests that a strong correlation between electrons enhances certain types of absorption.

I. INTRODUCTION

Many-body effects of electrons confined in a zero-dimensional region, i.e., a quantum dot,[1] are of much interest. When electrons are confined by a depletion region of semiconductors, whose confinement can be modeled by a parabolic potential, many-body effects on heat capacity[2] and magnetization[3,4] have been reported.

However, we cannot expect many-body effects on far-infrared absorption in a quantum dot with this parabolic confinement.[5] This is because the size of the quantum dot, typically 100 nm, is much less than the wavelength of photons to be absorbed, typically 100 μm, so the dipole approximation holds to a high degree of accuracy. The dipole approximation has no many-body effects on far-infrared-absorption spectra.[2] It is not possible to observe how many electrons are in a quantum dot with parabolic confinement, although we know that the absorption intensity is proportional to the number of electrons.[6] However, drastic changes in absorption spectra induced by the Coulomb interaction are expected in a confining structure whose shape is slightly different for a parabolic structure.[7]

The next stage of research on zero-dimensional confinement is studying a system in which electrons are confined by a compound semiconductor heterostructure, for example, GaAs/$Al_xGa_{1-x}As$. Such a confinement can be modeled by a well-like potential,[8,9] so we can expect many-body effects on far-infrared-absorption spectroscopy. In this paper, we calculate far-infrared-absorption spectra and the absorption coefficient of two electrons confined in a square-well quantum dot as the strength of the Coulomb interaction between electrons is changed. We obtained several types of absorption induced by the Coulomb interaction. We will emphasize the effect of the Coulomb interaction on the absorption coefficient.

II. METHOD OF CALCULATION

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

$$\hat{H}=\sum_{j=1,2}\left[(\mathbf{p}_j+e\,\mathbf{A})\cdot\frac{1}{2m(\mathbf{r}_j)}(\mathbf{p}_j+e\,\mathbf{A})+V(\mathbf{r}_j)\right]+\alpha\frac{e^2}{4\pi\epsilon_r\epsilon_0}\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}\ , \tag{1}$$

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

$$m(x,y)=\begin{cases}m_{\text{in}} & \text{if } |x|<L/2 \text{ and } |y|<L/2\\ m_{\text{out}} & \text{otherwise}\ .\end{cases} \tag{3}$$

We assume that an electron in a semiconductor material can be described by an effective mass $m(\mathbf{r})$ which depends on the kind of semiconductor material. α is introduced to describe the strength of the Coulomb interaction. The shape of a quantum dot is determined by the one-body potential $V(x,y)$. 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 m_{in} is taken to be $0.067m_0$ and m_{out} is taken to be $0.104m_0$, where m_0 is the free-electron mass. V_0 is taken to be 0.36 eV and ϵ_r is 10.9 for GaAs.

In our system, $\mathbf{p}_j$ and $1/m(\mathbf{r}_j)$ do not commute because the effective mass depends on where the electrons are. Therefore, the electromagnetic interaction term in the Hamiltonian (1) within the dipole approximation is

$$\hat{H}_{\text{em}}=e\,\mathbf{A}(0)\cdot\sum_{j=1,2}\left[\frac{1}{2m(\mathbf{r}_j)}\mathbf{p}_j+\mathbf{p}_j\frac{1}{2m(\mathbf{r}_j)}\right]. \tag{4}$$

This expression is reduced into

$$\hat{H}_{\text{em}}=-i\frac{e}{\hbar}\,\mathbf{A}(0)\cdot\sum_{j=1,2}[\mathbf{r}_j,\hat{H}_0]\ , \tag{5}$$

where $\hat{H}_0$ is $\hat{H}$ without $\mathbf{A}(\mathbf{r})$. When $|n\rangle$ is the nth eigenstate of the Hamiltonian H_0, and E_n is its eigenenergy, an

optical transition matrix element from the ground state $|0\rangle$ to an excited state $|n\rangle$ can be derived from

$$\langle n|\hat{H}_{\rm em}|0\rangle = i\frac{e}{\hbar}(E_n - E_0)\,\mathbf{A}(0)\cdot\langle n|\left[\sum_{j=1,2}\mathbf{r}_j\right]|0\rangle\ , \quad (6)$$

so this noncommutativity does not influence the expression of the matrix elements of $\hat{H}_{\rm em}$ within the dipole approximation, in which we ignore the spatial dependence of $\mathbf{A}(\mathbf{r})$. If a photon to be absorbed is polarized in the x direction, the absorption coefficient

$$\Gamma = \left|\left\langle n\left|\sum_j x_j\right|0\right\rangle\right|^2 \quad (7)$$

is proportional to the intensity of absorption from the ground state to the excited state $|n\rangle$, if an electron can transit rapidly from the excited state $|n\rangle$ to the ground-state emitting photons. A coefficient for absorption of a photon polarized in the y direction is the same as in the x direction because our system has symmetries of x and y. If $\Gamma\neq 0$, the optical transition from the ground state $|0\rangle$ to the excited state $|n\rangle$ is allowed as a two-electron phenomenon. To sum up, we determine $|n\rangle$, then calculate Γ.[10]

Let us discuss our method for 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 $\langle X|n\rangle=\psi_n(X)$ as a basis:

$$\psi_n(X) = A_n H_n(X)\exp\left[-\frac{X^2}{2}\right]\ , \quad (8)$$

where the $H_n(X)$'s are the Hermite polynomials and the A_n's are the normalization constants. We need quadruple tensorial products of the base functions

$$\langle X_1,Y_1;X_2,Y_2|j_1,j_2;j_3,j_4\rangle = \psi_{j_1}(X_1)\psi_{j_2}(Y_1)\psi_{j_3}(X_2)\psi_{j_4}(Y_2) \quad (9)$$

for the orthogonal basis set because our system includes two electrons in the two-dimensional space . Matrix elements of the kinetic term and the one-body potential term in the Hamiltonian H_0 can be obtained easily using the error function $\mathrm{erf}(x)=\int_0^x dz\,\exp(-z^2)$. The size of the quantum dot L appears in the matrix elements of the potential term, so we introduce ratio $\zeta=L/\xi$, which will be treated as a variational parameter. Matrix elements of the Coulomb interaction term in H_0,

$$\langle i_1,i_2;i_3,i_4|\frac{e^2}{4\pi\epsilon_r\epsilon_0 r}|j_1,j_2;j_3,j_4\rangle\ ,$$

can be written as

$$\frac{e^2}{4\pi\epsilon_r\epsilon_0\xi}\prod_{m=1}^{4}\left[\sum_{k_m=0}^{i_m}\sum_{l_m=0}^{j_m}\eta_{i_m k_m}\eta_{j_m l_m}\right] \times S(k_1+l_1,k_2+l_2,k_3+l_3,k_4+l_4) \quad (10)$$

if we define η_{ik} by $\phi_i(z)=e^{-z^2}/2\sum_{k=0}^{i}\eta_{ik}z^k$ and

$$S(a,b,c,d)=\int_{-\infty}^{\infty}dX_1dY_1dX_2dY_2 \times\frac{X_1^aY_1^bX_2^cY_2^d}{\sqrt{(X_1-X_2)^2+(Y_1-Y_2)^2}} \times e^{-(X_1^2+Y_1^2+X_2^2+Y_2^2)}\ , \quad (11)$$

which can be obtained analytically.

Though each matrix element depends on ζ, exact energy eigenvalues of the Hamiltonian H_0 must be independent of ζ because 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 because systems treated in this paper are simpler than coupled quantum dots, whose energy spectra were calculated using the same numerical method with eight bases.[11]

The orbital wave function $\phi_n(\mathbf{r}_1,\mathbf{r}_2)$ of the states $|n\rangle$ must be even or odd when particles are exchanged. When $\phi_n(\mathbf{r}_1,\mathbf{r}_2)$ is even, the two-electron eigenstate $|\Phi_{n0}\rangle$ has spin 0, as in

$$|\Phi_{n0}\rangle=\frac{1}{\sqrt{2}}\phi_n(\mathbf{r}_1,\mathbf{r}_2)(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)\ . \quad (12)$$

When $\phi_m(\mathbf{r}_1,\mathbf{r}_2)$ is odd, the two-electron eigenstate $|\Phi_{mi}\rangle$ has spin 1, as in

$$|\Phi_{m1}\rangle=\frac{1}{\sqrt{2}}\phi_m(\mathbf{r}_1,\mathbf{r}_2)(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)\ , \quad (13)$$

$$|\Phi_{m2}\rangle=\phi_m(\mathbf{r}_1,\mathbf{r}_2)|\uparrow\uparrow\rangle\ , \quad (14)$$

$$|\Phi_{m3}\rangle=\phi_m(\mathbf{r}_1,\mathbf{r}_2)|\downarrow\downarrow\rangle\ . \quad (15)$$

In their dipole approximation, a spin-flip transition is impossible. The ground state should have spin 0, so photon absorptions in the dipole approximation are due to transitions between states having spin 0. The absorption coefficient Γ, as defined above, can be written as

$$\Gamma=\left|\int d\mathbf{r}_1d\mathbf{r}_2\phi_n(\mathbf{r}_1,\mathbf{r}_2)\left[\sum_j x_j\right]\phi_0(\mathbf{r}_1,\mathbf{r}_2)\right|^2\ . \quad (16)$$

Note that if $\phi_n(\mathbf{r}_1,\mathbf{r}_2)$ is an odd state when particles are exchanged, Γ in the above expression is automatically zero because we have the relation

$$\begin{aligned}&\int d\mathbf{r}_1d\mathbf{r}_2\phi_n(\mathbf{r}_1,\mathbf{r}_2)x_1\phi_0(\mathbf{r}_1,\mathbf{r}_2)\\ &=\int d\mathbf{r}_2d\mathbf{r}_1\phi_n(\mathbf{r}_2,\mathbf{r}_1)x_2\phi_0(\mathbf{r}_2,\mathbf{r}_1)\\ &=\int d\mathbf{r}_2d\mathbf{r}_1[-\phi_n(\mathbf{r}_1,\mathbf{r}_2)]x_2\phi_0(\mathbf{r}_1,\mathbf{r}_2)\ .\end{aligned} \quad (17)$$

III. RESULTS

In a square quantum dot whose side is less than 30 nm, quantum states of two electrons are very weakly correlated, and energy levels can be obtained approximately by

states occupied by two electrons with Pauli's principle.[12] Therefore, interpretation of two-electron states by single-particle states is helpful in such a small quantum dot, and far-infrared-absorption spectra are obtained from selection rules which govern optical transitions. These selection rules are determined by the symmetries of wave functions—absorption is allowed if $\Gamma \neq 0$. Let us denote single-particle energy eigenstates in a two-dimensional square-well quantum dot by $|\varphi_{pq}\rangle$, so that

$$\hat{P}_x|\varphi_{pq}\rangle=(-1)^p|\varphi_{pq}\rangle\ , \tag{18}$$

$$\hat{P}_y|\varphi_{pq}\rangle=(-1)^2|\varphi_{pq}\rangle\ , \tag{19}$$

where $\hat{P}_x$ and $\hat{P}_y$ are parity operators in the x and y directions, respectively, and p and q are non-negative integers. When a single electron is contained in a square quantum dot, optical transitions from the ground state to $|\varphi_{10}\rangle$ and $|\varphi_{01}\rangle$ are allowed, and the optical transitions yield the first least-energy absorptions. The second least-energy absorptions are due to transitions to $|\varphi_{30}\rangle$ and $|\varphi_{03}\rangle$. When two electrons are contained in a very small quantum dot, the ground state $|\Phi_0\rangle$ of two electrons can be written approximately as

$$|\Phi_0\rangle\sim\frac{1}{\sqrt{2}}\varphi_{00}(\mathbf{r}_1)\varphi_{00}(\mathbf{r}_2)(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)\ . \tag{20}$$

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 state, for example

$$|\Phi_1\rangle\sim\tfrac{1}{2}[\varphi_{10}(\mathbf{r}_1)\varphi_{00}(\mathbf{r}_2)+\varphi_{00}(\mathbf{r}_1)\varphi_{10}(\mathbf{r}_2)] \times(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)\ . \tag{21}$$

Another absorption of two electrons, denoted by Γ_4, is due to a transition from the ground state to an excited state, for example

$$|\Phi_2\rangle\sim\tfrac{1}{2}[\varphi_{30}(\mathbf{r}_1)\varphi_{00}(\mathbf{r}_2)+\varphi_{00}(\mathbf{r}_1)\varphi_{30}(\mathbf{r}_2)] \times(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)\ . \tag{22}$$

If we assume a square quantum dot with an infinite well, single-particle wave functions can be described as

$$\varphi_{pq}(x,y)=\frac{2}{L}\sin\left[\frac{\pi(p+1)(x-L/2)}{L}\right] \times\sin\left[\frac{\pi(q+1)(y-L/2)}{L}\right], \tag{23}$$

where L is a side of the square. The absorption coefficients of the two noninteracting electrons $\Gamma_1^{(0)}$ and $\Gamma_4^{(0)}$ can easily be obtained by

$$\Gamma_1^{(0)}=2\left|\frac{2L}{\pi^2}\int_0^\pi dt\ t\sin(t)\sin(2t)\right|^2 \tag{24}$$

$$=2\left[\frac{4}{3\pi}\right]^4 L^2\sim 6.49\times10^{-2}L^2 \tag{25}$$

and

$$\Gamma_4^{(0)}=2\left|\frac{2L}{\pi^2}\int_0^\pi dt\ t\sin(t)\sin(4t)\right|^2 \tag{26}$$

$$=\frac{1}{2}\left[\frac{8}{15\pi}\right]^4 L^2\sim 4.15\times10^{-4}L^2\ . \tag{27}$$

These optical transitions can be described as a single-electron transition between single-particle states. As the size of a quantum dot becomes large, however, optical transitions must be described as two-electron phenomena because electron correlations become important. Therefore, far-infrared-absorption spectra are determined by the condition $\Gamma \gg 0$.

In Fig. 1, Γ_1 is calculated as L is changed from 20 to 30, 40 , . . . , 100 nm. Without the electron-electron interaction, i.e,. $\alpha=0$, Γ_1 increases linearly with an offset as L^2 increases. This offset is due to the spreading of wave functions outside the quantum dot because the square of the dipole matrix element is proportional to L^2 if an electron is confined by an infinite well. Even if α is changed from zero, i.e., a Coulomb interaction is introduced, the predominance of Γ_1 remains but the value of Γ_1 becomes smaller. This tendency becomes more obvious with larger L. This is because the Coulomb interaction causes electrons to avoid each other, so an electron must move in order to absorb a photon. Γ_1 vs the absorption energy is shown in Fig. 2. We can observe that not only the absorption coefficient but the absorption energy is influenced by the Coulomb interaction. However, if we change the strength of the Coulomb interaction, Γ_1 vs the absorption energy will be almost on the same curve, as observed in Fig. 2. (This may be because the Coulomb repulsion reduces the effective size of electrons.) Therefore, it is difficult to extract many-body effects from characteristics of Γ_1 vs the absorption energy.

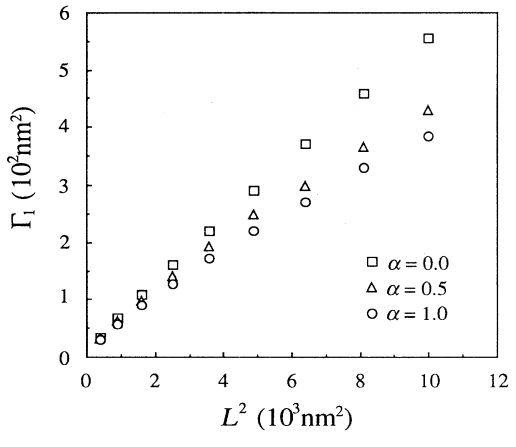

FIG. 1. Γ_1 which is proportional to the intensity of the strongest absorption of two electrons in a square-well quantum dot. A side of the quantum dot L is changed from 20 to 30, 40, . . . , 100 nm, and the horizontal axis indicates the square of L. Open squares are Γ_1 without electron-electron interaction, and open circles are Γ_1 when the Coulomb interaction is included. Open triangles are when a half of the Coulomb interaction is introduced.

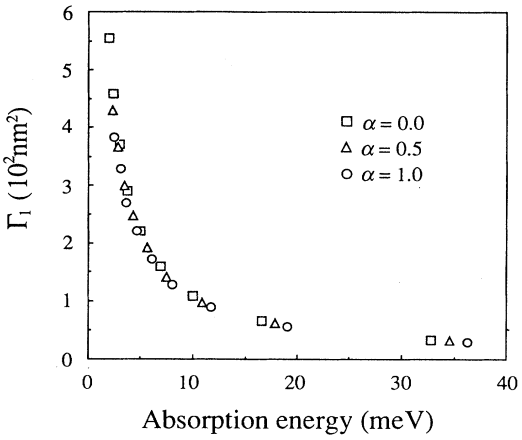

FIG. 2. Γ_1 vs absorption energy is plotted as L is changed from 20 to 30, 40, . . . , 100 nm. The absorption energy is affected by the Coulomb interaction as well as the absorption coefficient.

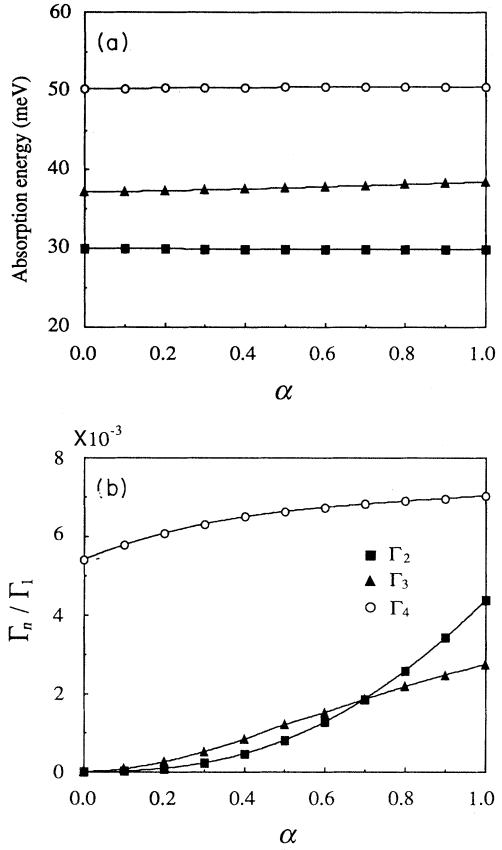

FIG. 3. Absorption energy and Γ_2, Γ_3, and Γ_4 relative to Γ_1 for optical transitions between the ground state and higher-energy states in a square-well quantum dot whose side L is 40 nm. (a) Absorption energy as a function of α. (b) Absorption coefficient relative to Γ_1 as a function of α. In both figures, Γ_2 is denoted by solid squares, Γ_3 is denoted by solid triangles, and Γ_4 is denoted by open circles. Γ_2 and Γ_3 will be zero if electron-electron interaction is ignored, i.e., $\alpha=0$. These two types of absorption are induced by the Coulomb interaction between electrons.

Let us turn to absorption between the ground state and higher-energy states. When $L=40$ nm, the absorption energy of Γ_1 is about 10 meV, as shown in Fig. 2. We can also observe a weaker absorption of about 50 meV, denoted by Γ_4. This absorption exists even if there is no electron-electron interaction. If we introduce the Coulomb interaction, we can observe different types of absorption with energies between Γ_1 and Γ_4. The type of absorption with relatively small energy is denoted by Γ_2, and the type of relatively large energy is denoted by Γ_3, as shown in Fig. 3(a). In Fig. 3(b), the absorption coefficients Γ_2, Γ_3, and Γ_4 relative to Γ_1 are plotted as functions of α, where L is fixed to be 40 nm. As α is increased from zero, different absorption types Γ_2 and Γ_3, whose energies are about 30 and 37 meV, appear. These types of absorption are due to the Coulomb interaction. As α increases, these types of absorption become stronger, as does Γ_4. The absorption energy of Γ_2 and Γ_4 hardly changes as the strength of the Coulomb interaction changes, but that of Γ_3 changes slightly, as seen in Fig. 3(a). These changes in absorption energy are small because electrons are confined in such a small quantum dot. However, different types of absorption are brought on by the disruption of the symmetry of wave functions caused by electron-electron interaction.

When a quantum dot become large, the correlation between electrons becomes stronger, and two electrons with opposite spin would sit on opposite ends of the same diagonal (as in a Wigner lattice).[12] We will analyze absorption induced by the Coulomb interaction in a larger quantum dot. As L is changed from 20 to 30, 40, . . . , 100 nm, Γ_2, Γ_3, and Γ_4 relative to Γ_1, vs the absorption energy is shown in Fig. 4, where α is taken to be 1. When L becomes larger, the behavior of Γ_2/Γ_1 and that of Γ_3/Γ_1 and Γ_4/Γ_1 are very different. The relative coefficients of Γ_3 and Γ_4 are hardly changed by increasing L. On the other hand, the relative coefficient of Γ_2 increases rapidly when L is greater than 50 nm. As shown in Fig. 2, Γ_1 increases as L becomes larger—Γ_n except Γ_2 behave simi-

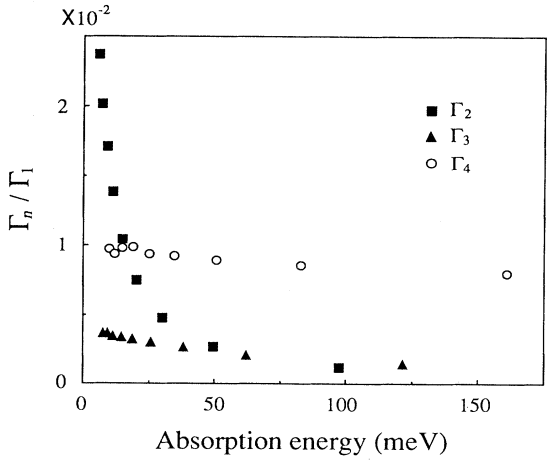

FIG. 4. Γ_2, Γ_3, and Γ_4 relative to Γ_1, vs absorption energy as L is changed from 20 to 30, 40, . . . , 100 nm. As L increases, the absorption energy decreases. When L increases beyond 50 nm, Γ_2/Γ_1 increases rapidly, in contrast to Γ_3/Γ_1 and Γ_4/Γ_1.

larly. Because we can expect a strong correlation between electrons when L is greater than 50 nm, this peculiar behavior of Γ_2 suggests that a strong correlation between electrons resulting in Wigner lattice states enhances a certain type of photon absorption, i.e., Γ_2.

IV. DISCUSSION AND CONCLUSION

Electrons interact with surrounding electromagnetic fields through a single-electron interaction term in the Hamiltonian. Thus the mean properties of electrons are important for electromagnetic interaction in a free space if the effective size of electrons is smaller than the wavelength of the photon to be absorbed. In this system, variables in the Hamiltonian decompose into center-of-mass coordinates and relative coordinates. A system of electrons confined by a parabolic potential inherits this kind of decomposition, so we cannot expect many-body effects on far-infrared-absorption spectra. However, electrons in general confinement no longer have such properties. Therefore, by examining far-infrared absorption, we can observe many-body effects of electrons confined into manmade structures.

If we ignore electron-electron interaction, the Schrödinger equation of two electrons in a two-dimensional space is identical to that of a single electron in a four-dimensional space whose eigenfunctions have a high degree of symmetry. An electron-electron interaction breaks some symmetry of wave functions of noninteracting electrons, so additional types of absorption will appear. If we apply an external electric field to a quantum dot, the symmetry will be broken in a way different from that caused by electron-electron interaction. This is well known as the Stark effect in quantum structures.[13] The concurrence of these two types of symmetry breakers is of much interest, and far-infrared absorption of interacting electrons in a quantum dot under an external electric field and its application will be discussed elsewhere.[14]

We have calculated the absorption coefficient of two electrons confined in a square-well quantum dot. The absorption coefficient and the absorption energy are affected by the Coulomb interaction, and additional types of absorption are induced. These types of absorption are due to the breakdown of symmetry of wave functions by the Coulomb interaction. In a larger quantum dot, we also observe the enhancement of an intensity of a certain type of absorption, whose enhancement may be induced by a strong correlation between electrons. Therefore, we can expect a rich structure in a far-infrared-absorption spectroscopy of interacting electrons confined by a heterostructure of compound semiconductors, and this rich structure will illuminate many-body effects of electrons in manmade structures.

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