Optical properties of quantum dots in a magnetic field

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(Submitted 24 January 1991)
Zh. Eksp. Teor. Fiz. **99**, 1853–1870 (June 1991)

We discuss the theory of optical phenomena in quasi-zero-dimensional systems (quantum dots, or QD) in the presence of a magnetic field. Intraband processes (IR absorption, resonant Faraday effect, and inelastic light scattering far from interband resonances) are investigated within the framework of a parabolic model for the QD lateral potential. This model is in good agreement with the available experiments, and also allows us to obtain certain exact results for the QD as a many-body interacting system, specifically a generalization of the Kohn theorem that allows us to obtain the intensity of the IR absorption lines. For interband processes we point out the possibility of a unique "phase transition" in the absorption spectrum and in Raman scattering with respect to magnetic field, and clarify the structure of the exciton resonance for various quantum-dot models.

1. INTRODUCTION

In recent years there has been considerable interest in the study of electronic processes in quasi-zero-dimensional systems, the so-called "quantum dots" (QD). Current submicron technology allows structures to be fabricated in which the motion of two-dimensional electrons is localized in the plane of the boundary in both directions. Typical geometric sizes of such QD are a few thousand angstroms; however, the characteristic diameter of the region occupied by electrons can be considerably smaller due to the action of the lateral potential. In this situation, motion in the plane of the boundary can be quantized to a considerable degree for semiconductors with small effective masses (GaAs, InSb), so that in fact the QD constitutes an artificial atom in which the number of electrons can be controllably varied from one to several hundred.[1-3]

The magnetocapacitance measurements of Hansen *et al.* made on GaAs/AlGaAs heterostructures proved convincingly that the structure of the QD energy spectrum is completely discrete (due to "superquantization" with respect to all three spatial directions). It is natural that the next problems of interest should involve optical phenomena in QD, especially in view of their possible applications to optoelectronics. The first experiments on infrared absorption in QD have already been carried out,[1-3] and questions have arisen in the context of these experiments regarding the relation between the frequencies of single-particle excitations and collective modes (i.e., involving the depolarization shift). In this paper we will explain one result of these experiments that was unexpected at first glance. We also will discuss the theory of certain other optical effects in QD, e.g., Raman scattering, Faraday rotation, interband and exciton absorption, that have not yet been observed experimentally; these effects can be very useful for obtaining information regarding such new and interesting systems in solid state physics.

2. ENERGY SPECTRUM OF QUANTUM DOTS AND GENERALIZATION OF KOHN'S THEOREM

The conditions for obtaining a QD must be such that the lateral potential of the "empty atom" (i.e., the initial form of the potential well before the QD is occupied by mobile carriers) can be approximated as a parabola

$$U(\rho)=\frac{m\Omega^2\rho^2}{2},$$

where m is the effective mass, Ω is the characteristic frequency of the parabola, and $\rho=(x^2+y^2)^{1/2}$ is the in-plane distance from the center of the QD. Motion along the z axis can be treated in the ultra-quantum limit, so that the atom may be considered to be planar. This approximation has been validated by the numerical calculations of Kumar, Laux, and Stern,[5] who solved the system of Schroedinger and Poisson equations self-consistently. It turns out that this simple model of a parabolic QD allows us to explain a very interesting result obtained in these infrared absorption experiments—the fact that the resonant frequency is independent of the number of electrons in the QD (i.e., independent of its "atomic number"). Also interesting is the fact that this system constitutes a rare example of an experimentally interesting many-body system whose characteristics can be found exactly for arbitrary interactions between particles.

A well-known theorem of Kohn[6] proves that the resonance absorption of a uniform high-frequency electric field by a system of interacting electrons in a magnetic field B takes place at the cyclotron frequency $eB/mc=\omega_c$ independent of the form of the interaction if the latter depends only on the difference between particle coordinates. This result was originally proved for a spatially homogeneous system. Brey, Johnson, and Halperin[7] recently showed that this "exclusion" of the interaction occurs also for particles moving in a one-dimensional parabolic potential. Kohn's method of proof,[6] which was used in Ref. 7 as well as in subsequent generalizations for two- and three-dimensional anisotropic cases,[8,9] was to work with the commutation relations for the operators of creation and annihilation in order to establish that the distances between exact energy levels of the many-particle system, which are connected by dipole optical transitions, equal the corresponding single-particle quantities.

Here we will use another method, which allows us to find not only the exact values of the resonant frequencies of the system of N particles in the potential $U(\rho)$ and in the presence of a magnetic field, but also the intensity of the corresponding lines, a result not previously derived in Refs. 6–9. Furthermore, we have discovered that it is possible to solve the problem of a QD in a magnetic and uniform electric field exactly.

 0038-5646/91/061037-10$03.00

Let us write the Hamiltonian of the system under discussion in the symmetric vector potential gauge $\mathbf{A} = 1/2[\mathbf{B}\rho]$ (here we set $\hbar = 1$):

$$\hat{H} = -\frac{1}{2m}\left\{\sum_{k=1}^{N}\left[\frac{\partial^2}{\partial\rho_k^2} + \frac{ieB}{c}\left(x_k\frac{\partial}{\partial y_k} - y_k\frac{\partial}{\partial x_k}\right) - \left(\frac{eB}{2c}\right)^2\rho_k^2\right] + \frac{m\Omega^2}{2}\sum_k\rho_k^2 + \sum_{j<k}u(\rho_j-\rho_k) + \hat{H}_{spin}, \tag{1}$$

where $u(\rho_j - \rho_k)$ is the pairwise interaction potential, and H_{spin} is the spin part of the system energy in the magnetic field.

We introduce the variables $\mathbf{R}$ and $\mathbf{X}_1, \mathbf{X}_2, \ldots, \mathbf{X}_{N-1}$ in place of ρ_k ($k = 1, 2, \ldots$), allowing us to extract the motion of the center of mass; these quantities are normalized in a special way (see Ref. 10):

$$\mathbf{R} = \sum_{k=1}^{N}\frac{\rho_k}{N^{1/2}}, \quad \mathbf{X}_1 = \frac{\rho_1-\rho_2}{(1\cdot 2)^{1/2}}, \quad \mathbf{X}_2 = \frac{\rho_1+\rho_2-2\rho_3}{(2\cdot 3)^{1/2}},$$

$$\mathbf{X}_{N-1} = \frac{\rho_1+\rho_2+\ldots-(N-1)\rho_N}{((N-1)N)^{1/2}}. \tag{2}$$

The transformation (2) preserves that part of the Hamiltonian $\hat{H}$ which does not contain the interaction $\Sigma\bar{u}(\rho_j - \rho_k)$; furthermore, after the transformation to these new variables this part of the Hamiltonian does not contain any dependence on R. Then the new Hamiltonian has the form

$$H(\mathbf{R}, \mathbf{X}_1, \ldots, \mathbf{X}_{N-1}) = H_0(\mathbf{R}) + H'(\mathbf{X}_1, \ldots, \mathbf{X}_{N-1}),$$

$$\hat{H}_0 = -\frac{1}{2m}\left[\frac{\partial^2}{\partial\mathbf{R}^2} + \frac{ieB}{c}\left(X\frac{\partial}{\partial Y} - Y\frac{\partial}{\partial X}\right)\right] + \frac{m\tilde{\omega}^2}{2}R^2, \tag{3}$$

where the combined frequency is $\tilde{\omega} = (\Omega^2 + \omega_c^2/4)^{1/2}$, and X, Y are the components of the vector $\mathbf{R}$.

Thus, the coordinate part of the wave function of the system has the form

$$\Psi = \psi_{nM}(\mathbf{R})\,\Phi(\mathbf{X}_1, \ldots, \mathbf{X}_{N-1}),$$

where ψ_{nM} is a solution of the Schroedinger equation for a two-dimensional isotropic oscillator in a magnetic field normal to the plane of oscillation; n and M are respectively the radial and azimuthal quantum numbers, while the energy level is given by the expression (see Ref. 11)

$$E_{nM} = (2n+|M|+1)\tilde{\omega} + \omega_c M/2, \quad n = 0, 1, 2, \ldots, \quad M = 0, \pm 1, \pm 2, \ldots \tag{4}$$

We remark that $\psi_{nM}(\mathbf{R})$ is always symmetric with respect to all the particles; therefore, the Pauli principle must be satisfied through the function Φ and the spin factor. From Eqs. (3) and (4) it follows that for arbitrary electron-electron interactions the intervals between levels of the energy spectrum of the system are the same as in the analogous single-particle problem (of course, many other intervals are also present, corresponding to the excitation of the internal degrees of freedom $\mathbf{X}_1, \ldots, \mathbf{X}_{N-1}$).

In the dipole approximation the interaction with the electromagnetic field is described by the Hamiltonian

$$\hat{H}_{int} = eF(t)\sum\rho_k = eFN^{1/2}\mathbf{R},$$

which does not contain the variables $\mathbf{X}_1, \mathbf{X}_2, \ldots, \mathbf{X}_{N-1}$ and therefore is diagonal with respect to the quantum numbers of the function Φ. Consequently, the optical absorption of this system of N particles will be the same as the absorption of a two-dimensional isotropic oscillator in the field of a wave with amplitude $N^{1/2}F$. This question will be discussed in Sec. 3; for now let us find the polarizability of the QD in an arbitrary constant electric field $\mathbf{F}\perp\mathbf{B}$. For example, let the field be directed along the y axis. Then the wave function can be written in terms of $\psi(\mathbf{R})$ and $\Phi(\mathbf{X}_1, \ldots, \mathbf{X}_{N-1})$ in the following way:

$$\Psi = \exp(ip_0X)\,\psi_{nM}(X, Y+Y_c)\,\Phi(\mathbf{X}_1, \ldots, \mathbf{X}_{N-1}), \tag{5}$$

where

$$p_0 = \frac{-eFN^{1/2}\omega_c}{2\Omega^2}, \quad Y_c = \frac{eFN^{1/2}}{m\Omega^2}.$$

The change in the system energy corresponds to an overall shift of all the levels by the quantity

$$\Delta E = -e^2F^2N/2m\Omega^2.$$

Thus, the polarizability of the QD (in its plane) does not depend on magnetic field, interelectron interactions, or the state of the "atom" (i.e., the electronic configuration of the QD), and for any F it equals the N-fold polarizability of the harmonic oscillator $e^2/m\Omega^2$.

3. INFRARED ABSORPTION

By solving the problem of the interaction of the QD with an electromagnetic field within perturbation theory, we obtain the values of the resonant frequencies in the dipole approximation:

$$\omega_+ = \tilde{\omega} + \frac{\omega_c}{2}, \quad \omega_- = \tilde{\omega} - \frac{\omega_c}{2},$$

for transitions with $\Delta n = 0$ and $\Delta M = \pm 1$, respectively (waves with left- and right-handed circular polarizations are absorbed at different frequencies). These selection rules exhaust the possible dipole transitions from the ground state of the QD, for which we must have $n = M = 0$; this follows from the positivity of the energy E_{nM} in (4). Thus, the IR absorption spectrum consists of two lines at ω_+ and ω_-, whose position does not depend on the number of electrons in the QD; this fact was also noted in Ref. 1. The intensity of these lines is determined by the oscillator strengths

$$f(\omega_+) = N\frac{\omega_+}{\tilde{\omega}}, \quad f(\omega_-) = N\frac{\omega_-}{\tilde{\omega}}. \tag{6}$$

In the region of strong magnetic fields $\omega_c \gg \Omega$ we have the asymptotic forms

$$f(\omega_+) \to \text{const}, \quad f(\omega_-) \propto B^{-2},$$

both intensities increase linearly with N, i.e., the number of electrons in the dot.

Within the framework of the parabolic model we can proceed further and find the intensity of IR absorption in a field $F(t)$ of arbitrary intensity (although uniform as before).

Let us begin with the case $B = 0$. Then the variables separate in Cartesian coordinates and the problem reduces to a uniform oscillator whose point of suspension moves according to a specified law

$$\mathbf{R}_0(t)=\frac{-e\mathbf{F}(t)N^{1/2}}{m\Omega^2}.$$

In this case the solution to the time-dependent Schroedinger equation has the form (see, e.g., Ref. 12)

$$\Psi(t)=\exp\left[i\mathbf{p}(t)(\mathbf{R}-\mathbf{r}(t))\right]\psi_0(\mathbf{R}-\mathbf{r}(t))\Phi(\mathbf{X}_1,\ldots,\mathbf{X}_{N-1}),$$

$$\mathbf{p}(t)=-eN^{1/2}\int_{t_0}^{t}\mathbf{F}(t')\cos(\Omega(t-t'))\,dt',$$

$$\mathbf{r}(t)=-\frac{eN^{1/2}}{m\Omega}\int_{t_0}^{t}\mathbf{F}(t')\sin(\Omega(t-t'))\,dt', \qquad (7)$$

where ψ_0 is the wave function at the time t_0 when the field is first switched on. If we now calculate the current density in the state $\Psi(t)$, we obtain

$$\mathbf{j}(\mathbf{R},t)=-\frac{e\mathbf{p}(t)}{m}|\psi_0(\mathbf{R}-\mathbf{r}(t))|^2, \qquad (8)$$

from which it is easy to find the work done by the field $F(t)$ on the system. For adiabatic switching-on of a monochromatic field

$$F(t)=F_0\exp(i\omega+\delta)t$$

the total energy absorbed by the system within the time T is given by the expression

$$Q(T)=\frac{Ne^2F_0^2\exp(2\delta T)}{4m}\frac{\Omega^2+\omega^2+\delta^2}{(\omega^2-\Omega^2-\delta^2)^2+4\omega^2\delta^2}. \qquad (9)$$

We can proceed analogously for the case where a magnetic field is present. The equations of motion for the Heisenberg operators corresponding to the coordinates x, y are classical in form, and as a consequence of their linearity are easy to solve. From their solution we find the current operator by averaging it with respect to the initial state and calculating $Q(T)$. The result depends on the sign of the circular polarization of the wave. Thus, for the field

$$\mathbf{F}(t)=F_1e^{\delta t}(\mathbf{i}\cos\omega t+\mathbf{j}\sin\omega t),$$

where $\mathbf{i}, \mathbf{j}$ are unit vectors for the x and y axes respectively, we obtain

$$Q_-(T)=\frac{Ne^2F_1^2\exp(2\delta T)}{2m}\frac{\Omega^2+\omega^2+\delta^2}{(\omega-\omega_-)^2(\omega+\omega_+)^2+\delta^2(2\omega+\omega_c)^2}, \qquad (10)$$

where F_1 is the amplitude of the circular wave (corresponding to $F_0/2^{1/2}$ in Eq. (9) for linear polarization). The resonance in Eq. (10) is reached for $\omega=\omega_-$. For the opposite polarization we must substitute $-\omega$ for ω in Eq. (10); then the resonance will occur at ω_+. In both cases

$$2\omega_{\mp}\pm\omega_c=2\tilde{\omega},$$

i.e., the widths of the resonance equal δ.

The energy absorbed per unit time (dQ/dT for $\delta\to 0$) naturally gives the same value for the oscillator strengths as Eq. (6). For example, at the resonance $\omega=\omega_+$, we have from (10)

$$\frac{dQ_-}{dT}=\frac{\pi}{2}\frac{e^2F_1^2}{m}N\frac{\omega_-}{\tilde{\omega}}\delta(\omega-\omega_-).$$

Thus, the exact quantum mechanical expression for the power absorption is linear in the intensity of the wave and contains only the one-photon resonance $\omega_\pm$, which coincides with the first-order perturbation theory result. In this case, however, the occupation of all the levels is different from zero at any instant of time; the distinctive feature of the parabolic model is the fact that the power absorbed averaged over time corresponds only to transitions with frequency $\omega_\pm$.

4. DEPOLARIZATION SHIFT

The depolarization shift of the IR absorption resonance arises because of dynamic screening: the effective field acting on an "individual" electron is different from the field of the incident wave due to electron-electron interactions. It is sufficient to include these within the framework of the self-consistent field approximation. Thus, the mechanism for the effect is the same as that which establishes the dielectric permittivity; however, it was shown in Ref. 14 for the example of a quantum film that the resonance is shifted in an inhomogeneous system, so that the frequency of IR absorption is larger than the distance between the single-particle levels of the system (e.g., the film subbands). These level spacings can be found from measurements that do not involve high-frequency electric fields, e.g., from the Shubnikov–de Haas effect. Consequently, the depolarization shift is the difference between the collective and single-particle excitation frequencies. For intersubband transitions its magnitude is proportional to the surface density of electrons for the case of a quantum film or an inversion channel, and to the line density for a quantum wire. The results of Secs. 2 and 3 imply that for a single QD with a parabolic lateral potential this depolarization shift is exactly equal to zero.

However, the system probed by the experiments of Refs. 1–3 was a square lattice of QD's. Of course, this system as a whole is not described by the parabolic model; consequently, it should exhibit some depolarization shift. In these experiments the period of the QD lattice was sufficiently large (0.25–1 μm) that tunneling could be neglected; hence, the only coupling between QD that requires modeling is electrostatic. Thus, the problem reduces to finding the Lorenz-Lorentz correction for the two-dimensional system consisting of dipoles induced in each QD by an incident wave and by the fields of the other QD. Because the wavelength of the excitation IR wave is much larger than the period of the QD lattice, we will neglect retardation effects.

Assume that a uniform effective field $\mathbf{F}_{\text{eff}}\exp(-i\omega t)$ acts on the QD whose position is given by the lattice vector $\mathbf{a}_n$. The wave function of the N-body system including the field $\mathbf{F}_{\text{eff}}$ is given by Eq. (7), and can be written in the form

$$\Psi(t)=\exp\{i\mathbf{p}(t)[\mathbf{R}-\mathbf{r}(t)]\}\psi_0$$

$$\times\left(\frac{\boldsymbol{\rho}_1+\ldots+\boldsymbol{\rho}_N}{N^{1/2}}-N^{1/2}\boldsymbol{\Delta}(t)\right)\Phi(\mathbf{X}_1,\ldots,\mathbf{X}_{N-1}),$$

where

$$\boldsymbol{\Delta}(t)=\frac{-e\mathbf{F}_{eff}\exp(-i\omega t)}{m(\Omega^2-\omega^2)}.$$

Returning to the old variables $\rho_1,\ldots,\rho_N$, we verify that the function Ψ depends only on the differences

$\rho_1 - \Delta(t), \ldots, \rho_N - \Delta(t)$. Therefore, the charge density in the QD excited by the field $\mathbf{F}_{\text{eff}}$ can be written in terms of the unperturbed density $n_0(\rho)$:

$$n(\boldsymbol{\rho}, t) = n_0(\boldsymbol{\rho} - \boldsymbol{\Delta}(t)).$$

This result is valid only for a field that is uniform within the QD, so that we must assume that the electronic radius of the QD is much smaller than the period of the lattice. In the experimental systems fabricated to date this condition is fulfilled, although not by a large margin (roughly one order of magnitude). In what follows, we will neglect effects that are nonlinear in the field; if we expand the difference $n_0(\rho - \Delta) - n_0(\rho)$, we find that the value of the induced dipole moment of the QD is determined, as it should be, by the dynamic polarizability of the latter:

$$\alpha(\omega) = \frac{Ne^2}{m(\Omega^2 - \omega^2)}.$$

The equation used to determine the effective field self-consistently has the form

$$\mathbf{F}_{eff}(\mathbf{a}_m) = \mathbf{F}_0 \exp(i\mathbf{k}\mathbf{a}_m) - \frac{\partial}{\partial \mathbf{a}_m} \sum_{l \neq m} \frac{\alpha(\omega)(\mathbf{a}_m - \mathbf{a}_l)}{|\mathbf{a}_m - \mathbf{a}_l|^3} \mathbf{F}_{eff}(\mathbf{a}_l). \tag{11}$$

Here $\mathbf{k}$ and $\mathbf{F}_0$ are respectively the wave vector and amplitude of the incident wave. In the long-wavelength approximation we find from (11) that

$$\mathbf{F}_{eff} = \mathbf{F}_0 \left[1 - \frac{C\alpha(\omega)}{L^3} \right]^{-1}, \tag{12}$$

and for a planar square lattice

$$C = {}^1/_2 \sum_{n,m}{}' (n^2 + m^2)^{-3/2} \approx 4{,}52,$$

L is the lattice period, and the dash implies that the sum does not include the point $n = m = 0$. The position of the IR absorption resonance can be determined using the expressions derived in Sec. 3 by making the substitution $F_0 \to F_{\text{eff}}$. Then for the position of the absorption resonance we have

$$\omega_{res}^2 = \Omega^2 - \delta_p^2, \quad \delta_p^2 = \frac{CNe^2}{mL^3}. \tag{13}$$

Thus, for a system of "parabolic" QD's we expect a negative depolarization shift (which contradicts all of the experimental examples known up to now). The reason for this is easy to understand: there is no internal depolarization field in the parabolic QD model, since all the particles shift as a whole, while it is well-known that polarization effects from outside a dielectric body enhance the action of an external field (hence we have $F_{\text{eff}} > F_0$ in Eq. (12) for $\alpha(\omega) > 0$, i.e., for $\omega < \Omega$). We also note that in this case the shift δ_p^2 is proportional to N/L^3, and not to the average surface particle density N/L^2.

In the experiments involving QD on InSb (Ref. 1), for which we have $L = 0.25\,\mu$m, $\Omega = 7$ meV, and $N_{\max} = 20$, we obtain the estimate $\delta_p^2/\Omega^2 < 4\%$. For the system based on GaAs investigated in Ref. 2, we have $L = 1\,\mu$m, $\Omega = 2$ meV, $N_{\max} = 210$, which implies $\delta_p^2/\Omega^2 < 2\%$. The depolarization shift calculated here is finite in zero-dimensional (QD) and one-dimensional (quantum wire) systems; however, it equals zero for a multilayer lattice of quantum films (quantum wells) with a parabolic potential. In fact, a shift of the electronic distribution in the direction normal to the film which preserves the uniformity in the two other directions does not change the electric field outside the film, so that the layers of the superlattice remain electrically "uncoupled." Therefore, if we do not include tunnelling, absorption by intraband transitions in a superlattice has a resonance at the single-particle frequency Ω.

Including the effect of a magnetic field reduces to calculating the dynamic polarizability of the QD for $B \neq 0$. In this case, by analogy with what was given above, we find an effective field F_{eff}. Using the same Heisenberg operators introduced in Section 3 for the coordinates, we find the dipole moment of the QD induced by a circularly-polarized wave; then for the polarizabilities $\alpha_{\pm}(\omega)$ we obtain

$$\alpha_{\pm}(\omega) = \frac{Ne^2/m}{(\omega_+ \mp \omega)(\omega_- \pm \omega)}, \tag{14}$$

for the cases of right- and left-handed circular polarization respectively. The equation for the resonance frequencies differs from the analogous equation for the case of a single QD only by the renormalization $\Omega^2 \to \Omega^2 - \delta_p^2$. Thus, the resonances are spaced according to

$$(\omega_{res})_{\pm} = \left(\Omega^2 - \delta_p^2 + \frac{\omega_c^2}{4} \right)^{1/2} \pm \frac{\omega_c}{4}. \tag{15}$$

In this sense the magnetic field does not affect the depolarization shift.

For the case of a linearly-polarized incident wave, the relation between the effective field and the "bare" field is tensorial. In this case the value of the depolarization shift is determined by mixing of the other polarization into the effective field. Thus, for $\mathbf{F}_0$ (where $F_{0x} = \cos \omega t, 0$) we obtain

$$(F_{eff})_y = \frac{\omega \omega_c \delta_p^2 F_{0x} \sin \omega t}{(\Omega^2 - \delta_p^2 - \omega^2)^2 - \omega^2 \omega_c^2}; \tag{16}$$

of course the positions where resonances occur $(\omega_{res})_{\pm}$ are again given by Eq. (15).

5. ADDITIONAL MODES AND DEVIATIONS FROM THE IDEAL MODEL

Let us note here that our generalized Kohn theorem is valid only for (1) a parabolic potential and bounded motion of the particles, and (2) a uniform high-frequency exciting field. Violation of either of these conditions leads to the appearance of additional resonances in the IR absorption and (or) to a dependence of the positions of the resonances on the number of particles in the QD. It is also interesting to consider the possibility of crossing or pseudocrossing of terms of the QD as the magnetic field changes; such crossings were observed experimentally in Ref. 4 in the magnetocapacitance dependence and in Ref. 2 in the IR absorption. In this case, perturbations in the absorption spectrum of the QD are analogous to well-known corrections in molecular spectroscopy, e.g., the Fermi resonance. However, there is an obvious advantage to studying the QD system, namely the possibility of controllable variation of all the important parameters of these effects.

A splitting of the IR absorption resonance associated

with pseudocrossing of terms is possible only when nonparabolicity of the lateral potential is included. If the conditions under which the QD lattice is fabricated result in QD with square symmetry, the first nonparabolic correction to the potential $U(\rho)$ has the form

$$U_1=a(x^4+y^4)+2bx^2y^2,$$

where a and b are constants. For $a\neq b$ the perturbation U_1 is associated with states with $\Delta M=\pm 4$. For example, a Fermi resonance between the terms $n=0$, $M=1$, and $n=0$, $M=-3$ occurs for $\omega_c=2\Omega/3^{1/2}$, so that we have $\omega_{res}=3^{1/2}\Omega$. This additional resonance is excited by a uniform field from the state $|0,0\rangle$ because of a small (for $U_1\ll U$!) admixture of the function ψ_{01} into the upper state, which is a superposition of the states $|0,1\rangle$ and $|0,-3\rangle$. The intensity of this additional resonance is smaller than the intensity of the ground-state transition $|0,0\rangle\cdots\rightarrow|0,1\rangle$, where the corresponding parameter of smallness is $(a-b)^2/m^4\Omega^6$.

The same additional resonance, now corresponding to the transition $|0,0\rangle\cdots\rightarrow|0,-3\rangle$, can be observed if the field that excites it is nonuniform in the system plane (in this case the nonparabolicity is inessential). Such nonuniformity can arise as a consequence of polarization effects discussed in Sec. 4.

Nonuniformity of the field F_{eff} is a small effect if the electron radius of the QD ρ_0 is much smaller than the lattice period L. For a QD lattice having a center of symmetry, the next term after the uniform field in the expansion of F_{eff} is of order $\alpha(\omega)\rho_0^2F_0/L^5$. The corresponding correction to the interaction Hamiltonian for electrons with the IR wave has the following form for the case of a square lattice:

$$H_{int}=\lambda\sum_{k=1}^{N}\frac{e\rho_k^3\alpha(\omega)}{8L^5}[(F_{eff})_x+i(F_{eff})_y]\exp(3i\varphi_k), \qquad (17)$$

where ρ_k, φ_k are cylindrical coordinates of particles in the QD.

$$\lambda=-\frac{105}{2}\sum_{n,m}{}'(n^2+m^2)^{-5/2}+420\sum_{n,m}{}'\frac{m^2n^2}{(n^2+m^2)^{9/2}}\approx -177$$

(here we have calculated the octupole contribution to H_{int} from each QD, assuming that the latter is acted on by the fields of the remaining QD which to the required degree of accuracy can be treated as dipoles). An estimate of the ratio of matrix elements for these transitions gives

$$\frac{M(0,0\rightarrow 0,-3)}{M(0,0\rightarrow 0,1)}\sim\frac{0,2}{(m\tilde{\omega}L^2)}$$

in the resonance region, where $\alpha(\omega)\sim L^3$ [see Eq. (12)]. In this case we assume that the resonance γ is rather narrow, $\gamma\Omega\ll\delta_p^2$, so that the depolarization shift must be included. In the opposite limiting case the ratio of matrix elements has an additional small factor $\delta_p^2/\Omega\gamma$. In both cases the intensity of the additional peak falls off as B^{-2} with increasing magnetic field in the region $\omega_c\gg\Omega$.

Both of the mechanisms leading to the appearance of the additional resonances discussed in this section are associated with perturbations that depend not only on the coordinates of the center of mass $\mathbf{R}$, but also on $\mathbf{X}_1,\ldots,\mathbf{X}_{N-1}$. Therefore, along with higher modes of the collective oscillations discussed here ($\omega=2\omega_{\pm}$, $3\omega_{\pm}$, etc.), in principle we should also observe absorption lines associated with individual electrons due to excitation of the internal degrees of freedom $\mathbf{X}_k$. Observation of these lines would give information on the electronic structure of the QD analogous to that given by spectroscopy of ordinary atoms.

6. FARADAY EFFECT

In the presence of a magnetic field the dynamic polarizability of the QD is a tensor even without including the depolarization effects. Therefore an initially linearly-polarized wave becomes elliptical after passing through a plane containing a QD lattice.[1)] In the most real experimental situations there exist two small parameters that determine the effect, $(\omega_c\tau)^{-1}$ and $N_se^2\tau/mc=\sigma_0/c$, where τ is a phenomenological relaxation time for the electrons and N_s is the average surface density of mobile carriers: $N_s=N/L^2$. The electrodynamic problem to be solved is that of the passage of a wave through a plane in which surface currents are distributed that give rise to the same wave. This problem is completely analogous to the passage of a particle through a one-dimensional delta-function barrier. For a normally-incident wave, the angle of rotation of the large axis of the polarization ellipse θ and the degree of ellipticity ε are given by the following expression far from resonance ($|\omega-\omega_{\pm}|\tau\gg 1$):

$$\theta=\frac{2\pi eN_s}{B}\frac{\omega^2\omega_c^2}{(\omega^2-\omega_+^2)(\omega^2-\omega_-^2)},$$
$$\varepsilon=\left(\frac{2\pi eN_s}{B}\right)^2\frac{\omega^3\omega_c^3(\Omega^2-\omega^2)}{(\omega^2-\omega_+^2)^2(\omega^2-\omega_-^2)^2}. \qquad (18)$$

The change in the sign of ε for $\omega=\Omega$ implies a change in the phase of the ratio F_y/F_x by a factor of π. The angle of rotation of the ellipse changes sign twice as ω varies from $\omega\ll\omega_-$ to $\omega\gg\omega_+$. In the region $\omega_-\ll\omega\ll\omega_+$, which is nonempty when the condition $\omega_c>\Omega$ holds, θ depends only weakly on ω:

$$\theta\approx\frac{-2\pi eN_s}{B}\sim\frac{\sigma_0}{c(\omega_c\tau)}.$$

The magnitude of θ reaches its maximum value near the resonances $\omega_{\pm}$ (but not at the points $\omega=\omega_{\pm}$ themselves!):

$$\max|\theta_{\pm}|=\pi\sigma_0/2c.$$

The quantity ε has two maxima

$$|\varepsilon(\omega_+)|=|\varepsilon(\omega_-)|=\frac{\pi\sigma_0}{c}$$

at the points $\omega_{\pm}$, and is very small outside the neighborhood of these resonances [of order $\sigma_0/(c\omega_c\tau)^2$]. Based on these estimates, the peak values of θ and ε (both of order σ_0/c) should be comparatively easy to measure for the QD structures available at this time. Thus, in the GaAs system investigated in Ref. 2, we have $N_s=2\cdot 10^{10}$ cm^{-2}, so that for a relaxation time τ corresponding to the rather moderate mobility $1\cdot 10^5$ cm^2/V·sec we obtain

$$|\theta_{\max}|\sim|\varepsilon_{\max}|\sim 10^{-2}.$$

7. RAMAN SCATTERING

It is well known that a linear harmonic oscillator cannot cause inelastic light scattering, at least the dipole approxi-

mation, which corresponds to a second-order treatment of the term $\mathbf{A}\hat{\mathbf{v}}$ in the Hamiltonian for interaction of light with electrons ($\mathbf{A}$ is the vector potential of the optical field, and $\hat{\mathbf{v}}$ is the velocity operator of an electron). The same assertion applies to the two-dimensional magnetic oscillator (4), which corresponds to a collective mode of the QD. As we showed above, this is the only degree of freedom excited by a uniform field; taking into account selection rules for dipole transitions it is easy to verify that to second order in $(\mathbf{A}\hat{\mathbf{v}})$ only elastic processes are possible: $|0,0\rangle \cdots \to |0, \pm 1\rangle \cdots \to |0,0\rangle$.

However, inelastic scattering becomes possible when we take into account the finite value of the photon momentum, in which case the primary contribution to the effect comes from the term A^2 in the interaction Hamiltonian, which should be expanded with respect to $\mathbf{k}\rho_i$, where $\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$ is the momentum transfer during scattering. Thus,

$$\hat{H}_{int} = a_{k_2}^{+} a_{k_1} N^{h}(\mathbf{kR}), \tag{19}$$

where a_{k2}^{+}, a_{k1} are photon creation and annihilation operators. Raman scattering is obtained in first-order perturbation theory with respect to the interaction (19); it is subject to the same selection rule as IR absorption, i.e., $\Delta M = \pm 1$, and corresponds to a frequency transfer $\Delta\omega = \omega_1 - \omega_2 = \omega_{\pm}$. The scattering cross-section is

$$\sigma_{\pm} = \left(\frac{e^2}{2mc^2}\right)^2 N \frac{\omega_2}{\omega_1} \frac{k^2}{m\tilde{\omega}} |\mathbf{e}_1 \mathbf{e}_2^{*}|^2, \tag{20}$$

where $\mathbf{e}_1$, $\mathbf{e}_2$ are polarization vectors for the incident and scattered waves, respectively. In a spatially uniform system, the A^2 scattering described here is caused by fluctuations in the charge density, i.e., plasma waves are excited in the electron gas. As we will see, in a QD the analogous collective modes ω_+ and ω_- are excited.

All the statements in this section apply to nonresonant Raman scattering, when we have $\omega_1, \omega_2 < E_g$, the width of the bandgap. The usual enhancement of the effect for $\omega_1 \to E_g$ that is used in experiment takes us out of the framework of a purely parabolic model, and will be discussed in the next section.

8. INTERBAND PROCESSES

In this section we will discuss interband magnetoabsorption, resonant inelastic light scattering, and exciton effects in a QD. Since states of the valence band now enter into the process, the electron-electron interactions cannot be "eliminated" by any transformation of coordinates, and the parabolicity of the QD potential no longer plays a significant role. Nevertheless, we will use this model in the usual single-electron formulation of the problem in order to reduce the calculations to their final (rather simple) expressions. It is hoped that the results we obtain will be adequate to describe the experiments, since interband processes do not depend critically on the form of the lateral potential of the QD. The QD magnetoabsorption is calculated in the usual way (see, e.g., Ref. 15), obviously including the spatial nonuniformity of the QD electric field. In the absence of a magnetic field, the envelopes of electron and hole wave functions overlap weakly, since the QD is a potential well for one type of carrier and a barrier for the other. A cursory investigation of the form of the "effective potential energy" [a "magnetic" parabola $m\omega_c^2\rho^2$ plus an electrostatic parabola $U(\rho)$] shows that when the magnetic field exceeds a certain critical value B_0, the motion of the holes becomes finite, and is localized in the same region of the QD as the electrons. This critical magnetic field is determined by the relation

$$\omega_v^2 = \frac{4m_c\Omega^2}{m_v},$$

where m_c, m_v are effective masses of the electrons and holes, respectively, and ω_v is the cyclotron frequency for holes. For $B > B_0$ the interband absorption is caused by discrete-to-discrete transitions, and its spectrum consists of a set of lines labeled by the three quantum numbers n_c, n_v, and M (here we are talking about the quantum numbers of an individual electron). The selection rule in this case is simply $\Delta M = 0$; $\Delta n = n_c - n_v$ is arbitrary since the envelope wave functions of the c- and v-bands are eigenfunctions of different Hamiltonians if $m_c \neq m_v$. The frequencies of the lines are given by the expression

$$\omega(n_c, n_v, M) = E_g + 2(n_c\tilde{\omega}_c + n_v\tilde{\omega}_v) + (|M|+1)(\tilde{\omega}_c + \tilde{\omega}_v) + \frac{M}{2}(\tilde{\omega}_c + \tilde{\omega}_v). \tag{21}$$

Here $n_c, n_v = 0,1,2,\ldots$, $M = 0, \pm 1, \pm 2, \ldots$, and

$$\tilde{\omega}_c^2 = \frac{\omega_c^2}{4} + \Omega^2, \quad \tilde{\omega}_v^2 = \frac{\omega_v^2}{4} - \Omega^2 \frac{m_c}{m_v}.$$

Thus, we should observe a unique phase transition with respect to magnetic field in the interband magnetoabsorption spectrum, consisting of "ignition" of the straight lines with frequencies (21) in the region $B > B_0$. Near threshold the intensity I of these lines depends on the magnetic field according to the law

$$I(M) \propto (B^2 - B_0^2)^{(|M|+1)/2}, \quad B > B_0. \tag{22}$$

This result follows from the normalization of the envelope function of the holes, which become localized for $B > B_0$.

Raman scattering corresponds to the process illustrated in Fig. 1; it can only occur when the QD is occupied by electrons (in contrast to absorption, where the transitions are possible even for an "empty" QD). The matrix element for the process is quadratic in the overlap integral of the envelopes; accordingly, the intensities of the Raman lines behave like $(B^2 \pm B_0^2)^{|M|+1}$ with respect to magnetic field near threshold and contain the resonant enhancement factor $(E_g^2 - \omega_1^2)^{-2}$. The energy transfer in this case equals $2\tilde{\omega}\Delta n_c$, where $\Delta n_c = 1,2,\ldots$; as before, the condition $\Delta M = 0$ should hold. Thus, inelastic light scattering constitutes a direct method for measuring the spacing between those energy levels of the QD that are not associated with allowed IR transitions.

9. EXCITONS IN A QD

Turning now to a discussion of exciton effects in a QD, we must first address the question of whether or not an exciton can "survive" in the very strong field caused by the lateral potential, i.e., it is necessary to discuss the probability of tunneling ionization of the exciton. In view of the large number of characteristic parameters, we cannot treat this prob-

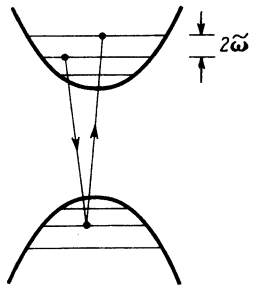

FIG. 1. Electron transitions for resonant light scattering in a QD.

lem in its most general formulation, which would require too much space; we therefore limit ourselves to the case of excitons based on heavy holes, whose subband is closest to the conduction band since it is least affected by size quantization along the z axis. We will discuss the two systems that were investigated experimentally in Refs. 1 and 2. In both of these systems the following conditions are fulfilled: $a^* \ll L$, $Ry^* \ll U_0$, where U_0 is the characteristic depth of the potential contour of the QD lattice, and a^* and Ry^* are respectively the effective Bohr radius and Rydberg energy in the conduction band. These conditions allow us to assume that the field that destroys the excitons is uniform at sufficiently large distances from the center of the QD. The central parabolic region requires a separate discussion.

Let us begin with the case of a uniform field, while still assuming that $B_0 = 0$. The problem can be solved in the quasiclassical approximation when the external electric field is much smaller than $F^*_{at} = \tilde{e}/a_c^{*2}$, i.e., the effective Coulomb unit of field intensity; $\tilde{e}$ is the effective charge, which includes the effective dielectric permittivity of the medium. In the two-dimensional case it is convenient to introduce planar parabolic coordinates:

$$\xi,\ \eta = (x^2+y^2)^{1/2} \pm x,$$

for which the Schroedinger equation for an exciton in a uniform electric field admits separation of variables. The calculations are completely analogous to the case of field ionization of a hydrogen atom (see Ref. 16). We obtain the following expression (in effective Coulomb units) for the decay probability of the ground state of a planar exciton:

$$w_0 = \frac{2^7}{(2\pi F)^{1/2}} \exp\left(-\frac{16}{3F}\right). \tag{23}$$

The exponent in this exponential is eight times as large as its three-dimensional analogue, because the logarithm of the transmission coefficient of a triangular barrier is proportional to $E_0^{3/2}/F$, where E_0 is the energy distance from the top of the barrier to the level from which the ionization takes place; in two dimensions E_0 is four times as large as in three dimensions.

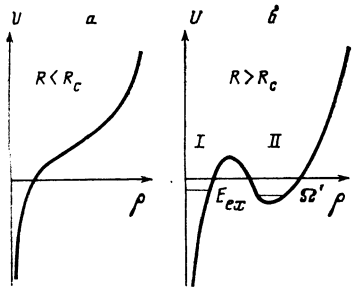

FIG. 2. Potential energy of an exciton in the parabolic model.

We next investigate the parabolic region. The QD systems based on InSb of Ref. 1 and GaAs of Ref. 2 require different approaches, since $\Omega \gg Ry^*$ holds in the first case, while in the second case we have $Ry^* \gg \Omega$. For the case $\Omega \gg Ry^*$ (InSb) the electron is a fast subsystem and generates the adiabatic potential V_{ad} for heavy holes (see Ref. 17). Calculating this potential

$$V_{ad} = -\tilde{e}^2 \int \frac{|\psi_{00}(\rho_e)|^2}{|\rho_e - \rho_h|} d^2\rho_e$$

using the function $\psi_{00}(\rho_e)$ for the ground state of a two-dimensional oscillator of frequency Ω, we find

$$V_{ad}(\rho_h (m_c\Omega)^{1/2} \ll 1) \approx \pi^{1/2}\tilde{e}^2 (m_c\Omega)^{3/2}\rho_h^2;$$

$$V_{ad}(|\rho_h| \to \infty) \sim -\frac{\tilde{e}^2}{|\rho_h|},$$

where ρ_h is the radius vector of the hole. Since the repulsion of the hole potential of the QD is

$$V_h(\rho) = \frac{-m_c\Omega\rho_h^2}{2},$$

it is clear that in the case $\Omega \gg Ry^*$ under discussion an exciton cannot form in the central part of the QD. For the system based on InSb this assertion is also valid for regions at the periphery of the QD, where Eq. (23) is applicable: estimates show that the exponent in the exponential of (23) equals approximately 0.4, i.e., $W_0 \gg 1$.

For the case of GaAs ($\Omega \ll Ry^*$) it is convenient to transform to coordinates $\rho = \rho_e - \rho_h$ and $\mathbf{R}$, i.e., the exciton center of mass. The fast system now corresponds to the relative motion of the electron and hole. The total potential energy has the form

$$U(\mathbf{R}, \boldsymbol{\rho}) = -\frac{\tilde{e}^2}{|\rho|} + \gamma\frac{m_c\Omega^2}{2}\rho^2 + m_c\Omega^2(\boldsymbol{\rho}\mathbf{R}), \tag{24}$$

where

$$\gamma = \frac{m_v - m_c}{m_v + m_c}.$$

In the adiabatic approximation, $\mathbf{R}$ in (24) is treated as a parameter, and depending on its absolute value the potential $U(\rho,\mathbf{R})$ can have the forms shown in Fig. 2a or Fig. 2b. Breakup of the exciton is possible only when

$$R > R_c = \frac{2\gamma^{1/4}a^*Ry^*}{\Omega},$$

when there is a certain direction ρ (such that $\rho \downarrow\uparrow \mathbf{R}$) for which the minimum of $U(\rho,\mathbf{R})$, which equals $-m_c\Omega^2R^2/2\gamma$, is sufficiently low, and when the level of zero-point oscillations in region II, which equals

$$\Omega' = \Omega\left(1 - \frac{m_c}{m_v}\right),$$

becomes lower than the ground state level of the exciton

$E_{ex} = -2Ry^*$. Ionization of the exciton in this case consists of a transition from region I to region II. The exponent of the tunnelling exponential that describes this process, which equals

$$\frac{R}{\gamma a^*}\left[1-\left(1-\frac{R_c^2}{R^2}\right)^{1/2}\right]+\ln\frac{2R}{a^*}\left[1-\left(1-\frac{R_c^2}{R^2}\right)^{1/2}\right]$$

becomes of order unity only for

$$R \approx \frac{R_c^2}{2\gamma a^*} = 2a^*\left(\frac{Ry}{\Omega}\right)^2,$$

which corresponds to approximately 8500 Å under conditions of the experiment in Ref. 2. The QD in Ref. 2 are formed by cylindrical elevations on the surface of the GaAs whose radii are smaller than this value, so that the excitons have a rather long lifetime over the entire region of the QD.

Let us turn now to the motion of the exciton center of mass. When the condition $\Omega \ll Ry^*$ is fulfilled, the last two terms in (24) can be treated by perturbation theory. The term $m_c\Omega^2\rho\mathbf{R}$ corresponds to the following uniform electric field acting on the exciton:

$$\mathbf{F} = \frac{m_c\Omega^2\mathbf{R}}{e}.$$

In the general case of a QD system with an arbitrary potential contour, in view of the condition $a^* \ll L$ we can obviously introduce a locally uniform field $\mathbf{F}(\mathbf{R})$; then to second order in perturbation theory we obtain the potential energy of the center of mass in the form

$$U_c = \frac{-\alpha\mathbf{F}^2(\mathbf{R})}{2},$$

where α is the polarizability of the exciton. For the ground state of a two-dimensional exciton we have $\alpha = 21/128\ a^{*3}$.

The probability of forming an exciton is determined by the factor

$$P = \left|\int \Phi(\mathbf{R})\, d^2\mathbf{R}\right|^2,$$

where Φ is the wave function of the center of mass, which describes motion in the potential $U_c(\mathbf{R})$. If we assume that the parabolic approximation to the QD potential under the conditions of Ref. 2 is valid up to the lateral surface of the cylinder, then the motion of the exciton as a whole takes place in an annular region bounded by the rectangular barrier of the work function for $R = d$ (the radius of the cylinder) and by the parabola $-\alpha(m_c\Omega^2R)^2/2e^2$ from the small-R side (see Fig. 3). Thus, excitons should "condense"

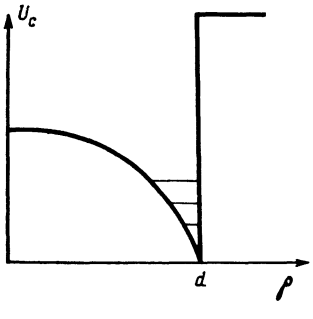

FIG. 3. Effective potential well for an exciton in a QD.

near the surface of the cylinders that make up the QD lattice. This naturally imposes rigorous technological requirements in order to avoid capture at surface defects and accelerated recombination that may hinder observation of the exciton lines.

It is easy to estimate the number of levels for a particle of mass $m_c + m_v$ in a potential well like the one shown in Fig. 3 for $d \sim (3\text{–}4)\cdot 10^3$ Å (see Ref. 2). The number turns out to be rather large, so that the quasiclassical considerations are applicable; then we may approximate U_c near $R = d$ by a linear function in the Schroedinger equation for $\Phi(\mathbf{R})$. Separating out the angular variable

$$\Phi = e^{il\varphi}\psi(\xi), \quad \xi = d - R$$

and assuming (where possible) that $R = d$, we obtain for $\psi(\xi)$

$$\psi'' - \frac{\psi'}{d} + \left[2M_{ex}E - \frac{l^2}{d^2} + \alpha\frac{m_c^2\Omega^4}{e^2}M_{ex}(d^2 - 2d\xi)\right]\psi = 0. \quad (25)$$

Here $M_{ex} = m_c + m_v$.

The quasiclassical spectrum and the wave functions of Eq. (25) are easily found. Obviously we must set $l = 0$, since if this is not true then $P = 0$, i.e., only those exciton states can form for which the wave function of the center of mass is axially symmetric. Calculating P using the quasiclassical solution to Eq. (25), we find the following intensity distribution I_n of components of the exciton transition ($n \gg 1$):

$$I_n \sim \frac{d}{(M_{ex}W_n)^{1/2}},$$

where

$$W_n = \frac{1}{2}\left(\frac{9\pi^2}{M_{ex}}\alpha^2\frac{m_c^4\Omega^8}{e^4}d^2\right)^{1/3}\left(n+\frac{3}{4}\right)^{2/3}. \quad (26)$$

The position of the nth component is determined by the spectrum of the energy of radial motion

$$E_n = -\alpha\frac{m_c^2\Omega^4d^2}{2e^2} + W_n.$$

If we now make a lattice of QD from GaAs, while bounding the motion of the electrons with a smooth ($L \gg a^*$) potential contour (analogous to the situation in Ref. 1), the points where $\mathbf{F}^2(\mathbf{R})$ is a maximum also form a lattice, at whose vertices excitons should "condense."[2)] For a square lattice, $U_c(\mathbf{R})$ corresponds to the potential energy of an isotropic two-dimensional harmonic oscillator near these lattice sites, with a certain frequency ω_0. Then we can show that the "fine" structure of the exciton transition is made up of equidistant lines of the same intensity, i.e., P does not depend on the radial quantum number (the azimuthal quantum number, as before, should be equal to zero). The integrals between components of the structure equal $2\omega_0$.

Finally, let us investigate the effect of a magnetic field. We will assume that B is large enough that the magnetic length is much smaller than a^*, and that d is even larger. In the Appendix we show that in this case the motion of the center of gravity of the exciton is described by the effective Hamiltonian (A10), which for the quantum-dot potential takes the form

$$H_{eff}=\frac{\hat{P}^2}{2M_0}-\alpha_0\frac{m_c^2\Omega^4}{2e^2}R^2-\frac{c}{B^2}([\mathbf{BF}]\hat{\mathbf{P}})+\frac{c}{B}\gamma\frac{m_c\Omega^2}{e}+\varepsilon_0.$$

The quantities α_0, M_0, and ε_0 are determined in the Appendix. This equation is written for an exciton that is in its ground state.

Because the operator $([\mathbf{BF}]\hat{\mathbf{P}})$ is proportional to the derivative with respect to angle, the solution may once more be sought in the form $\exp(il\varphi)\psi(\xi)$. The equation for $\psi(\xi)$ is analogous to (25) with renormalized values of the polarizability and exciton mass ($\alpha\to\alpha_0$, $M\to M_0$). Then we will have the following energy spectrum ($l=0$):

$$E_n=\varepsilon_0+\frac{c}{B}\gamma\frac{m_c\Omega^2}{e}-\alpha_0\frac{m_c^2\Omega^4}{2e^2}d^2+W_n.$$

The intensity of the absorption is given by the expression

$$I_n\sim\frac{d}{(M_0W_n)^{1/2}}.$$

The quantity W_n is determined by Eq. (6) with the replacements $\alpha\to\alpha_0$, $M\to M_0$. The spacing between discrete quantization levels of the exciton in the QD potential changes with magnetic field according to the law $B^{-3/2}$, while the intensity satisfies $I_n\propto B^{3/2}$.

APPENDIX

Let us consider the problem of the motion of a magnetoexciton in a slowly-varying weak external field with potential $\varphi(\mathbf{R})$. In this approximation the exciton may be considered as a particle moving in an effective potential. The Hamiltonian of the system electron-plus-hole is written in the form

$$\hat{H}=\frac{1}{2m_c}\left(\hat{\mathbf{P}}_e+\frac{e}{c}\mathbf{A}_e\right)^2+\frac{1}{2m_v}\left(\hat{\mathbf{P}}_h-\frac{e}{c}\mathbf{A}_h\right)^2-\frac{e^2}{\varepsilon|\mathbf{r}_e-\mathbf{r}_h|}+e(\varphi(\mathbf{r}_h)-\varphi(\mathbf{r}_e)). \tag{A1}$$

Here $\mathbf{r}_{e,h}$ are the electron and hole coordinates, respectively. In Eq. (A1) we first transfer to the new coordinates

$$\mathbf{R}=\frac{m_c\mathbf{r}_e+m_v\mathbf{r}_h}{m_c+m_v},\quad \boldsymbol{\rho}=\mathbf{r}_e-\mathbf{r}_h.$$

Following Refs. (19) and (20), we will seek a solution to Eq. (A1) in the form

$$\Psi(\mathbf{R},\boldsymbol{\rho})=\exp\left\{i\frac{e}{2c}([\mathbf{B}\boldsymbol{\rho}]\mathbf{R})\right\}\psi(\mathbf{R},\boldsymbol{\rho}). \tag{A2}$$

Then we arrive at the following equation for the function $\psi(\mathbf{R},\rho)$:

$$\left[\hat{H}_0(\boldsymbol{\rho})+\frac{e}{M_{ex}c}([\mathbf{B}\boldsymbol{\rho}]\hat{\mathbf{P}})+\frac{\hat{P}^2}{2M_{ex}}+e\varphi\left(\mathbf{R}-\frac{m_c}{M_{ex}}\boldsymbol{\rho}\right)-e\varphi\left(\mathbf{R}+\frac{m_v}{M_{ex}}\boldsymbol{\rho}\right)\right]\psi=E\psi. \tag{A3}$$

Here $\hat{\mathbf{P}}=-i\nabla_{\mathbf{R}}$; the operator H_0 describes the internal motion of the exciton:

$$\hat{H}_0=-\frac{\nabla_\rho^2}{2\mu}+\frac{ie}{2\mu c}\gamma(\mathbf{B}[\boldsymbol{\rho}\nabla_\rho])+\frac{e^2}{8\mu c^2}B^2\rho^2-\frac{e^2}{\varepsilon\rho}, \tag{A4}$$

$$\mu=\frac{m_cm_v}{M_{ex}}.$$

We introduce an orthogonal system of functions:

$$\varphi_{nk}=\varphi_n(\boldsymbol{\rho})e^{i(\mathbf{kR})},\quad \hat{H}_0\varphi_n=\varepsilon_n\varphi_n \tag{A5}$$

and expand the ψ-function in terms of them:

$$\psi=\sum_{k,n}C_{nk}\varphi_{nk}(\mathbf{R},\boldsymbol{\rho}). \tag{A6}$$

Substituting (A6) into Eq. (A3), multiplying by $\varphi_{n'k'}$, and integrating with respect to $\mathbf{R}$ and ρ, we arrive at the following system of equations for C_{nk}:

$$\sum_{nk}\left(\frac{k^2}{2M_{ex}}\delta_{n'k',nk}+\frac{e}{M_{ex}c}([\mathbf{k},\mathbf{B}]\boldsymbol{\rho}_{n'n})\delta_{kk'}+eV_{n'k',nk}\right)C_{nk}=(E-\varepsilon_{n'})C_{n'k'}. \tag{A7}$$

In this expression $\rho_{n'n}$ and $V_{n'k',nk}$ are matrix elements of the operators ρ and

$$V=\varphi\left(\mathbf{R}-\frac{m_c}{M_{ex}}\boldsymbol{\rho}\right)-\varphi\left(\mathbf{R}+\frac{m_v}{M_{ex}}\boldsymbol{\rho}\right)$$

with respect to the system of functions φ_{nk}. If we assume that the size d of the QD is much larger than the size of the exciton, we can expand the operator $\hat{V}$ in the series

$$V=(\mathbf{F}(\mathbf{R})\boldsymbol{\rho})+\frac{\gamma}{2}\rho_i^2\frac{\partial F_i}{\partial R_i}+\dots.$$

Here $\mathbf{F}=-\nabla\varphi(\mathbf{R})$. The size of the exciton in the magnetic field is

$$l_{ex}\sim\min(a^*,a_B),\quad a_B=(c/eB)^{1/2}.$$

In perturbation theory Eq. (A7) can be diagonalized by transforming to a new system of envelope functions $C_{nk}\to\overline{C}_{nk}$ (Ref. 21). Transforming to R-space, we obtain the following equation for the envelope function:

$$f_n(R)=\sum_k\overline{C}_{nk}e^{i(\mathbf{kR})}.$$

Assuming that the state is n-fold degenerate, we have

$$\left[\frac{\hat{P}^2}{2M_{ex}}-\frac{\alpha_{ij}}{2}\left(F_i+\frac{[\hat{\mathbf{P}}\mathbf{B}]_i}{M_{ex}c}\right)\left(F_j+\frac{[\hat{\mathbf{P}}\mathbf{B}]_j}{M_{ex}c}\right)+\frac{e\gamma}{2}\frac{\partial F_i}{\partial R_i}(\rho_i^2)_{nn}+\varepsilon_n\right]f_n(R)=Ef_n(R). \tag{A8}$$

This equation contains only terms of leading order in the quantities $el_{ex}F/(\varepsilon_n-\varepsilon_m)$ and l_{ex}/d. The polarizability is determined by the expression

$$\alpha_{ij}=2e^2\sum_{m\neq n}\frac{\langle n|\rho_i|m\rangle\langle m|\rho_j|n\rangle}{\varepsilon_m-\varepsilon_n}.$$

Equation (A8) describes an exciton in an arbitrary magnetic field. For the case of large magnetic fields ($a_B\ll a^*$) we can neglect the Coulomb energy in Eq. (A4); then the wave functions φ_n are harmonic oscillator functions whose energies are

$$\varepsilon_n=\omega_c(s+{}^1/_2(|s'|-\gamma s'+1)),\quad n=(s,s'),$$

$$s=0,1,2,\dots,\quad s'=0,\pm1,\pm2,\dots.$$

The polarizability in the ground state has the form ($s = s' = 0$)

$$\alpha_{xx}=\alpha_{yy}=\alpha_0, \quad \alpha_{xy}=-\alpha_{yx}=-i\gamma\alpha_0,$$

where $\alpha_0 = M_{ex}c^2/B^2$. Neglect of the Coulomb energy in calculating α_{ij} leads to the disappearance of the kinetic energy in Eq. (A8). In order that the effective mass of the exciton be finite, it is necessary to include the Coulomb energy via perturbation theory.[20] Calculating φ_n, ε_n using perturbation theory, we find the following correction to α_{xx} for $s = s' = 0$:

$$\alpha_{xx}=\alpha_0\left(1-\frac{M_{ex}}{M_0}\right), \quad M_0=\frac{2\varepsilon}{e^2}\left(\frac{2eB}{\pi c}\right)^{1/2}. \tag{A9}$$

Then Eq. (A8) leads to the expression

$$\left[\frac{\hat{P}^2}{2M_0}-\frac{c}{B^2}([\mathbf{BF}]\hat{\mathbf{P}})-\frac{M_{ex}c^2}{2B^2}F^2+\frac{c}{B}\gamma(\nabla\mathbf{F})+\varepsilon_0\right]f=Ef. \tag{A10}$$

Here

$$\varepsilon_0=\omega_c\left(\frac{1}{2}-\left(\frac{\pi}{2}\right)^{1/2}\frac{a_B}{a^*}\right)$$

is the energy of the exciton ground state in the absence of the lateral potential. The quantity

$$\frac{M}{M_0}\sim\frac{a_B}{a^*}$$

is retained in the equation only for the terms $\hat{P}^2$, ε_0; for the operators F^2 and $F\hat{P}$ this parameter can be neglected. Eq. (A10) is valid with respect to the parameters

$$a_B/d, \quad ea_BF/\omega_c, \quad a_B/a^*.$$

[1)] Strictly speaking, the transmitted wave is broken up into individual beams, because the QD system acts like a diffraction grating. It is clear, however, that because of the large wavelength of the IR radiation ($kL \ll 1$) the primary contribution is given by the zero-order spatial Fourier harmonic of the transmitted radiation, which we now are discussing.

[2)] This was in fact the situation realized in Ref. 18; however, the authors of this paper investigated the IR spectrum but not the exciton transitions.

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Translated by Frank J. Crowne