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Theory of collective excitations in a two-dimensional array of quantum dots

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We present a quantum theory of collective excitations in a two-dimensional array of quantum dots. The collective excitations are induced by Coulomb coupling of the quantum dots, and can have energies significantly higher than the energy-level splittings in individual quantum dots. These collective excitations should be experimentally detectable.

Recently, it has become experimentally possible to fabricate zero-dimensional quantum structures—quantum dots.[1-7] In such structures, electrons are confined in all three spatial dimensions and electron states are discrete. In light of the remarkable richness of physical phenomena which has been uncovered since electron confinement to two dimensions was made possible, it is expected that electron confinement to even lower dimensions will create a new exciting field both for basic physics research and device applications.

Discrete energy levels in quantum dots have been observed in a recent experiment by Reed *et al.*[1] A typical quantum dot in their experiment is a disklike structure embedded in a column of semiconductor materials. Electrons are confined in a thin layer (about 50 Å) of $In_xGa_{1-x}As$ in between two $Al_xGa_{1-x}As$ barrier layers. The diameters of the columns are in the range of 1000–2500 Å. Thus, the confinement in the direction along the columns is much stronger than in the lateral directions. Energy-level splittings in such quantum dots are of the order of 25 meV. In order to obtain enough signal strength, it is common in experiments to fabricate arrays of quantum dots. Although the quantum dots are electrically insulated from each other in the sense that electrons cannot transfer from one quantum dot to another, the long-range Coulomb force couples the quantum dots and this Coulomb coupling can lead to collective excitations in the system. This is analogous to the situations in multiwire superlattices and multilayer superlattices. In multiwire superlattices, the Coulomb coupling of excitations between one-dimensional (1D) subbands of the wires leads to collective intersubband plasmons.[8,9] In multilayer superlattices similar intersubband plasmons occur[10,11] and have been observed experimentally.[12] In this Rapid Communication, we propose a quantum theory of collective excitations in a system of quantum dots which form a rectangular lattice.

First we consider a square lattice of quantum dots. The results will be extended to the rectangular lattice later. We choose the array of quantum dots to be in the xy plane, and the z axis to be along the columns. Since electron confinement in the z direction is much stronger, it is only necessary to consider the lowest-energy level in the z direction. In the x and y directions, quantum dots are periodically spaced and wave functions of electron states for different quantum dots do not overlap. Thus, in x and y directions, tight-binding wave functions are perfectly suited for describing this system. The fabricated quantum dots have a disklike shape; thus, for a single quantum dot, the x and y directions are equivalent. This implies that the excited states in a quantum dot will be degenerate (not counting spin degeneracy).

In light of the above discussion we can write the wave functions of the system as

$$|k_x,k_y,i,j\rangle = \xi_z \psi_{ik_x}(x)\psi_{jk_y}(y)\,, \tag{1}$$

where ξ_z is the wave function of the lowest state in the z direction and

$$\psi_{ik_x}(x) = \sum_n e^{ik_x nd}\phi_i(x-nd)\,, \tag{2}$$

$$\psi_{jk_y}(y) = \sum_n e^{ik_y nd}\phi_j(y-nd)\,. \tag{3}$$

In the above equations d is the period of the square lattice and i and j label the states in the x and y directions. The energy of the Lth level $E(L)$ depends on both i and j. If the confining potential of a quantum dot is parabolic, then we can write $L=i+j$. The first excited energy level has $L=1$ and is doubly degenerate, corresponding to states with $i=0$, $j=1$ and $j=0$, $i=1$. This degeneracy exists as long as the quantum dot has xy symmetry, irrespective of the detailed form of the confining potential.

To study collective excitations of the system we use the self-consistent-field formalism of Ehrenreich and Cohen.[13] We start from the integral form of the Poisson equation

$$V(\bar{\mathrm{r}}) = \int \frac{d\bar{\mathrm{r}}' e^2 n(\bar{\mathrm{r}}')}{\epsilon|\bar{\mathrm{r}}-\bar{\mathrm{r}}'|}\,,$$

where $\epsilon = 4\pi\epsilon_0\epsilon_b$, and ϵ_b is the background dielectric constant. Taking Fourier transforms in the xy directions we get

$$V(\bar{\mathrm{q}},z) = \frac{2\pi e^2}{\epsilon q}\int dx' e^{-q|z-z'|} n(\bar{\mathrm{q}},z')\,, \tag{4}$$

where $\bar{\mathrm{q}}$ is a 2D vector (q_x,q_y) and $q=|\bar{\mathrm{q}}|$. The density response is given by

$$n(\bar{q},z)=2|\xi_z|^2\sum_{a,a'}\langle a|V(\bar{r})|a'\rangle\langle a'|e^{i\bar{q}\cdot\bar{r}}|a\rangle\frac{f(L')-f(L)}{E(L')-E(L)+\hbar\omega}. \tag{5}$$

In (5) a,a' are composite quantum indices, with $a=(k_x,k_y,i,j)$, and f is the Fermi function. We combine (4) and (5) and integrate out the z degree of freedom, and get

$$V(\bar{q})=\frac{4\pi e^2}{\epsilon q}I(q)\sum_{a,a'}\langle a|V(\bar{r})|a'\rangle\langle a'|e^{i\bar{q}\cdot\bar{r}}|a\rangle\frac{f(L')-f(L)}{E(L')-E(L)+\hbar\omega}, \tag{6}$$

where

$$V(\bar{q})=\int dz\,V(\bar{q},z)|\xi_z|^2,$$

and

$$I(q)=\int\int dz\,dz'e^{-q|z-z'|}|\xi_z|^2|\xi_{z'}|^2. \tag{7}$$

Using the tight-binding wave functions (2) and (3) and assuming that the Wannier wave functions for different quantum dots do not overlap, the matrix element $\langle a'|e^{i\bar{q}\cdot\bar{r}}|a\rangle$ is particularly easy to calculate. This matrix element can be decomposed into x and y components and

$$\langle k_x',i'|e^{iq_xx}|k_x,i\rangle=\delta_{k_x'-k_x-q_x,nG}A_{ii'}(q_x), \tag{8}$$

$$\langle k_y',j'|e^{iq_yy}|k_y,j\rangle=\delta_{k_y'-k_y-q_y,nG}B_{jj'}(q_y). \tag{9}$$

G is the reciprocal wave vector $2\pi/d$ and

$$A_{ii'}(q_x)=\int dx\,e^{iq_xx}\phi_{i'}^*(x)\phi_i(x), \tag{10}$$

$$B_{jj'}(q_y)=\int dy\,e^{iq_yy}\phi_{j'}^*(y)\phi_j(y). \tag{11}$$

The other matrix element $\langle a|V(\bar{r})|a'\rangle$ can be written as

$$\langle a|V(\bar{r})|a'\rangle=\sum_{\bar{q}'}V(\bar{q}')\langle a'|e^{-i\bar{q}'\cdot\bar{r}}|a\rangle. \tag{12}$$

Combining (8)–(12) and (6) we get

$$V(\bar{q})=\frac{4\pi e^2}{\epsilon q}I(q)\sum_{nm}V(q_x+nG,q_y+mG)\sum_{\substack{i,i'\\j,j'}}A_{ii'}^*(q_x+nG)B_{jj'}^*(q_y+mG)A_{ii'}(q_x)B_{jj'}(q_y)\Pi_{LL'}(\omega), \tag{13}$$

where the polarization

$$\Pi_{LL'}(\omega)=\sum_{k_x,k_y}\frac{f(L')-f(L)}{E(L')-E(L)+\hbar\omega}. \tag{14}$$

Let us denote V_{nm} to be $V(q_x+nG,q_y+mG)$, q_{nm} to be the absolute value of the wave vector (q_x+nG,q_y+mG), and define

$$U_{nm}=I(q_{nm})/q_{nm}. \tag{15}$$

Then, from (13) we have

$$V_{nm}=\frac{4\pi e^2}{\epsilon}U_{nm}\sum_{\substack{ii'\\jj'}}\Pi_{LL'}(\omega)A_{ii'}(q_x+nG)B_{jj'}(q_y+mG)\sum_{n'm'}V_{n'm'}A_{ii'}^*(q_x+n'G)B_{jj'}^*(q_y+m'G). \tag{16}$$

Equation (16) serves as the basic equation for deriving all collective excitations of the system. For a rectangular lattice, the G associated with q_x should be replaced by $2\pi/d_x$, and the G associated with q_y should be replaced by $2\pi/d_y$. If individual quantum dots do not have xy symmetry then the labels L, L' should be replaced by ij and $i'j'$, respectively.

Solving (16) is equivalent to solving a multidimensional matrix equation and in general can be quite involved numerically. In the following, we solve (16) for a simple case for which analytical solutions are obtainable. This is the case where each quantum dot has only two energy levels. Although real systems should be more complicated, solving this simple case demonstrates how Eq. (16) can be solved and provides a picture of the basic physics involved. Generalizations to the more complicated cases will then be straightforward.

Since the system is dispersionless the polarization $\Pi_{LL'}(\omega)$ is nonzero only when $L\neq L'$. In our notation, $L=0$ corresponds to the ground energy level and $L=1$ corresponds to the first excited energy level. If there are only two energy levels in each quantum dot, (16) becomes

$$V_{nm}=(4\pi e^2/\epsilon)U_{nm}\Pi(\omega)[A_{00}(q_x+nG)B_{01}(q_y+mG)g_1(q_x,q_y)+A_{01}(q_x+nG)B_{00}(q_y+mG)g_2(q_x,q_y)], \tag{17}$$

where

$$g_1(q_x,q_y)=\sum_{n,m}V_{nm}A_{00}^*(q_x+nG)B_{01}^*(q_y+mG), \tag{18}$$

$$g_2(q_x,q_y)=\sum_{n,m}V_{nm}A_{01}^*(q_x+nG)B_{00}^*(q_y+mG). \tag{19}$$

The $\Pi(\omega)$ in (17) is given by

$$\Pi(\omega)=\Pi_{01}(\omega)+\Pi_{10}(\omega)$$
$$=\sum_{k_x,k_y}\frac{[f(0)-f(1)]2E_{10}}{(\hbar\omega)^2-E_{10}^2}=\frac{(n_{s0}-n_{s1}/2)E_{10}}{(\hbar\omega)^2-E_{10}^2}, \tag{20}$$

where E_{10} is the energy difference between the two energy levels and n_{s0}, n_{s1} are average areal electron densities of the two energy levels. We multiply (17) by either $A_{00}^*(q_x+nG)B_{01}^*(q_y+mG)$ or $A_{01}^*(q_x+nG)B_{00}^*(q_y+mG)$ and then sum over n and m, and get the following two equations:

$$g_1(q_x,q_y)=\Pi(\omega)c_{11}g_1(q_x,q_y)+\Pi(\omega)c_{12}g_2(q_x,q_y), \tag{21}$$

$$g_2(q_x,q_y)=\Pi(\omega)c_{21}g_1(q_x,q_y)+\Pi(\omega)c_{22}g_2(q_x,q_y), \tag{22}$$

with

$$c_{11}=(4\pi e^2/\epsilon)\sum_{n,m}U_{nm}|A_{00}(q_x+nG)B_{01}(q_y+mG)|^2,$$
$$c_{22}=(4\pi e^2/\epsilon)\sum_{n,m}U_{nm}|A_{01}(q_x+nG)B_{00}(q_y+mG)|^2,$$
$$c_{12}=(4\pi e^2/\epsilon)\sum_{n,m}U_{nm}A_{00}^*(q_x+nG)B_{01}^*(q_y+mG)$$
$$\times A_{01}(q_x+nG)B_{00}(q_y+mG),$$

and $c_{21}=c_{12}^*$. From (21) and (22) we have

$$\begin{vmatrix}\Pi(\omega)c_{11}-1 & \Pi(\omega)c_{12}\\ \Pi(\omega)c_{21} & \Pi(\omega)c_{22}-1\end{vmatrix}=0. \tag{23}$$

Equation (23) is quadratic in $\Pi(\omega)$ and gives two collective excitation modes. Their frequencies are given by

$$(\hbar\omega)^2=E_{10}^2+\frac{E_{10}(2n_{s0}-n_{s1})(c_{11}c_{22}-|c_{12}|^2)}{c_{11}+c_{22}\pm[(c_{11}-c_{22})^2+4|c_{12}|^2]^{1/2}}. \tag{24}$$

The two collective modes given by (24) are nondegenerate except for some special cases. At first it seems surprising that although the first excited single-particle energy level is twofold degenerate, the collective excitations associated with electron excitations from the ground level to the first excited level have two nondegenerate modes. The lifting of the degeneracy is due to the fact that the system can support both longitudinal and transverse collective modes and excitation energies for these two modes are different except for some special wave vectors.

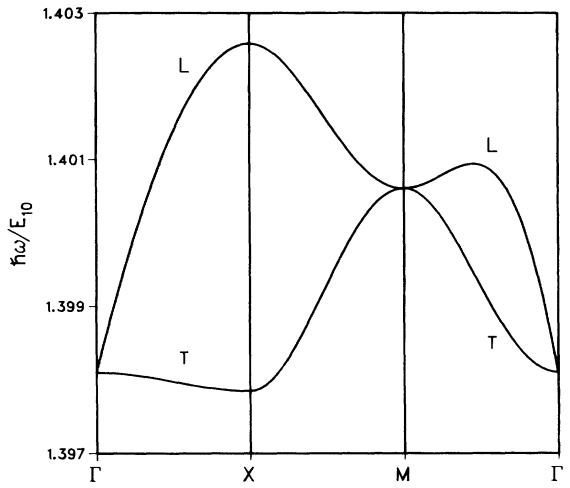

FIG. 1. Dispersions of collective modes for a square lattice of quantum dots. Each quantum dot is assumed to have two energy levels. Parameters used are (Ref. 14) $E_{10}=25$ meV, $\epsilon_b=6.5$, $m^*=0.041m_e$, $d=1000$ Å, $z_0=50$ Å, and $n_{s0}-n_{s1}/2=2\times10^{10}$ cm^{-2}. L and T represent longitudinal and transverse modes, respectively.

Figure 1 shows the dispersion of the two collective modes as given by (24). In producing Fig. 1 we assume that the confining potential of the quantum dots takes the parabolic form $(m^*/2)(E_{10}/\hbar)^2(x^2+y^2)$, and that the z-component wave function is the "particle in a box" wave function $\xi_z=(2/z_0)^{1/2}\sin(\pi z/z_0)$. These assumptions are appropriate for systems studied by Reed *et al.*[1] Γ, X, and M are points in the wave-vector space that correspond to (0,0), $(\pi/d,0)$, and $(\pi/d,\pi/d)$, respectively. Along the lines of ΓX and $M\Gamma$, we can explicitly identify the longitudinal and transverse modes. Deviations of the frequencies of the collective modes from the energy-level spacing of quantum dots can be significant. Raman-scattering experiments should be able to detect the collective modes. Indeed, since optical experiments are much easier to perform on quantum dot systems than transport experiments, the prospects for the experimental observation of the collective modes in quantum dot systems should be good. However, the dispersions of the longitudinal and transverse modes and their splittings are very small, making the experimental resolution of the two modes difficult.

In conclusion, we have developed a quantum theory of collective excitations in a 2D array of quantum dots. The collective modes should be experimentally detectable and can have energies significantly higher than the energy-level spacings in quantum dots.

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