Collective modes in tunneling quantum-dot arrays

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We present a theoretical study of the collective excitations and optical response of two-dimensional arrays of quantum dots. We calculate the dispersion curves and oscillator strength for the various modes when tunneling between nearest-neighbor dots is considered. The self-consistent-field formalism and a Wannier representation, described in the tight-binding approximation, is used to study the behavior of the modes of the system for different lattice parameters. We show that a band of interdot tunneling modes appears due to carrier tunneling between neighbors. The relative oscillator strength of the various interband and intraband excitation modes is calculated from the density-density correlation function. The tunneling modes are found to have a nonvanishing strength, possibly detectable in experiments.

Optical and transport properties of quasi-zero-dimensional structures (called quantum dots), where carriers are confined to a set of discrete energy levels, have been studied experimentally and theoretically with great interest in the past few years.[1] Most experimental studies of optical properties in these structures have been focused on systems where tunneling effects are negligible, since dots have been typically well separated, preventing carrier hopping from dot to dot. However, clever sample designs have allowed the study of a regime in quantum-dot systems, where coupling due to interdot carrier tunneling is very important.[2–4] These studies have shown that tunneling introduces interesting effects, such as anomalous dependence of the plasma oscillation frequency on magnetic field.[2] Tunneling effects are likely to become important also for closely placed quantum dots, systems of great current interest given theoretical predictions for an "antiferroelectric" ground state.[5–7]

Collective excitations in two dimensional (2D) quantum-dot arrays without tunneling have been studied theoretically by several groups.[8,9] Moreover, recent work on tunneling quantum-dot arrays has appeared,[10,11] in which tunneling is considered only via the energy dispersion of different energy states, while the wave function overlap (and corresponding multipole matrix elements) between different quantum dots are neglected in the calculation of the dielectric response function.

In this paper we present a more general theoretical study of tunneling quantum-dot structures where the energy dispersion and the overlap of wave functions coming from different quantum dots are included in the calculation of the polarization matrix. We also allow here for possible intraband and interband mixing due to the Coulomb interactions, an effect not previously included in studies of these systems.[10,11] We show that the general shape of the dispersion curves of collective excitation modes associated with interband transitions is indeed determined mainly by the details of the single-particle level dispersion, producing a strong q dependence.[9,10] On the other hand, we show that inclusion of wave-function overlaps results in a band of collective excitation modes associated with interdot tunneling of electrons in both lateral directions of the array. We present calculations of the oscillator strength for both families of interband and intraband (tunneling) collective excitation modes, obtained from the electron density-density correlation function for this generalized modulated 2D system. These calculations also show that the coupling to external probes exhibits rather complex anisotropies and q dependencies, a somewhat unrecognized result in the literature.

The wave functions of the system are assumed separable and written as

$$| 0, k_x, k_y, i, j\rangle = \xi_0(z)\psi_{ik_x}(x)\psi_{jk_y}(y)\,, \tag{1}$$

where ξ_0 is the wave function of the lowest state in the z direction, $\xi_0(z) = (2/z_0)^{1/2}\sin(\pi z/z_0)$ (the z direction confinement length z_0 is assumed to be much smaller than in the x and y directions). We use a Wannier representation along the x and y directions on the array as

$$\psi_{ik_x}(x) = \sum_l e^{ik_x l d_x}\phi_i(x - l d_x)\,,$$
$$\psi_{jk_y}(y) = \sum_h e^{ik_y h d_y}\phi_j(y - h d_y)\,, \tag{2}$$

where d_x and d_y are the periods of the lattice array, l,h indicate site indices in the x and y directions, respectively, and i,j label the bands.

The density response of the system to a potential perturbation $V(\mathbf{r})$ is then given in the self-consistent-field formalism by[12]

$$n(\mathbf{q}, z) = 2\,|\,\xi_0(z)\,|^2 \sum_{a,a'}\langle a\,|\,V(\mathbf{r})\,|\,a'\rangle\langle a'\,|\,e^{i\mathbf{q}\cdot\mathbf{r}}\,|\,a\rangle\Pi(a, a', \omega)\,, \tag{3}$$

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where $\Pi(a,a',\omega)$ is the polarization term

$$\Pi(a,a',\omega)=\frac{f(a')-f(a)}{E(a')-E(a)+\hbar(\omega+i\eta)}, \tag{4}$$

and the corresponding induced Coulomb interaction is given by

$$V(\mathbf{q},z)=\frac{2\pi e^2}{\epsilon q}\int dz'\, e^{-q|z-z'|}\, n(\mathbf{q},z'), \tag{5}$$

where $\mathbf{q}$ is a 2D vector, $q=\mid \mathbf{q} \mid$, ϵ is the background dielectric constant, $f(a)$ is the Fermi distribution function, and a,a' are composite quantum indices, $a=(k_x,k_y,i,j)$. The energy of the a state is expressed by the usual nearest-neighbor tight-binding energy bands, $E_a=\varepsilon_{ij}-W_x^{ij}\cos k_x d_x - W_y^{ij}\cos k_y d_y$, where W^{ij} are the half bandwidths in each direction. The phenomenological broadening factor η in Eq. (4) is introduced to describe the effects of imperfections in the system, and regularizes the behavior of the polarization function near the poles. Notice that Eq. (3) involves multipolar and Coulomb matrix elements between eigenstates located on different dots, as well as intradot terms. All these are included in our calculations (see $A^l_{ii'}$ and $B^h_{jj'}$ below), while the multipolar terms have been ignored previously in theoretical studies of these systems.[10,11]

Solving Eqs. (5) and (3) self-consistently, and after integrating out the z degree of freedom, allows one to write a secular equation for the normal modes of the system,

$$\sum_{slhHL}\left[\sum_{nm}\frac{4\pi e^2}{\epsilon}\frac{I(q_{nm})}{q_{nm}}A^{*L}_{ii'}(q_x+nG_x)B^{*H}_{jj'}(q_y+mG_y)\Pi A^{l}_{ii'}(q_x+nG_x)B^{h}_{jj'}(q_y+mG_y)-\delta_{ss'}\delta_{LL'}\delta_{HH'}\right]\mathcal{G}^{LH}_s=0, \tag{6}$$

where G_x (G_y) is the smallest reciprocal lattice vector component along the x (y) direction, $\mathbf{q}_{nm}=(q_x+nG_x,\ q_y+mG_y)$, $s=(i,i',j,j')$ is a compound index,

$$I(q_{nm})=\int\int dz\,dz' e^{-q_{nm}|z-z'|}\mid \xi_0(z)\mid^2\mid \xi_0(z')\mid^2, \tag{7}$$

$$A^l_{ii'}(q_x)=\int dx e^{iq_x x}\phi^*_{i'}(x)\phi_i(x-ld_x), \tag{8}$$

$$B^h_{jj'}(q_y)=\int dy e^{iq_y y}\phi^*_{j'}(y)\phi_j(y-hd_y), \tag{9}$$

$$V_{nm}=\int dz\, V(q_{nm},z)|\xi_0(z)|^2, \tag{10}$$

$$\mathcal{G}^{lh}_s\equiv\sum_{nm}V_{nm}A^l_{ii'}(q_x+nG_x)B^h_{jj'}(q_y+mG_y), \tag{11}$$

and $\Pi\equiv\Pi(s,q_x,q_y,L-l,H-h,\omega)$. Notice that Π has been Fourier transformed from the continuum variables k_x and k_y to the discrete *site* indices l and h, to take full advantage of the Wannier function representation.[13] The solutions of Eq. (6) for given q_x and q_y yield the collective excitation frequencies at that wave vector. Notice that the size of the matrix $\mathcal{G}$ (nominally infinite) depends in practice on the total number of levels/bands and neighbors included. The formalism can be used, in general, to study different systems for any value of q_x and q_y or number of subbands.

In the numerical calculations below we assume a square array ($d_x=d_y=d$), that tunneling exists only between nearest-neighbor dots [up to $|l|=1$ in Eq. (8)], and that each dot is well described by three energy levels (the higher two being degenerate) yielding three tunneling bands. We also assume the Wannier functions to be given by those of a local parabolic potential characterized by the harmonic frequency $\omega_0=\varepsilon_{10}/\hbar$. This Gaussian representation simplifies the calculation of the matrix elements considerably and it should give a proper description of the problem.[10,11] It is further assumed that only the lowest band is partially filled, for simplicity. In this case, the dimension of the determinant in Eq. (6) is 20×20.

Figure 1 shows collective excitation dispersion curves in the nontunneling limit along high-symmetry directions in the first Brillouin zone, for typical structure parameters (see caption). Since we focus our attention on the qualitative behavior of the dispersion curves, we have not included the self-consistent correction to the ground state.[14] Absence of this correction *as well as* the interdot coupling in this system causes a small depolarization shift at the Γ point ($q=0$). This shift vanishes for the *single* parabolic dot case, according to the generalized Kohn's theorem.[15] We can see in Fig. 1 drastic changes in the shape of the dispersion curves when the lattice constant decreases. These changes are qualitatively explained by the enhanced Coulomb fields at smaller array

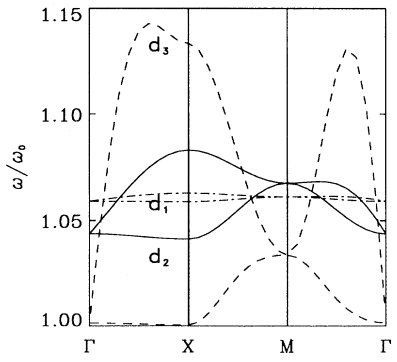

FIG. 1. Dispersion curves for collective modes in the nontunneling limit for a square array of quantum dots. Parameters used are $\varepsilon_{10}=\hbar\omega_0=25$ meV, $\epsilon_b=6.5$, $m^*=0.014m_e$, $z_0=50$ Å, $d_1=1000$ Å, $d_2=500$ Å, $d_3=250$ Å, and $k_{F\max}=\pi/2d$. "Dot size"$=\sqrt{\hbar/m^*\omega}=148$ Å. $\Gamma=(q_x=0,q_y=0)$, $X=(\pi/d,0)$, and $M=(\pi/d,\pi/d)$.

lattice constants and multipolar terms in the potential which are typically negligible for large lattice constants.

In calculating the polarization, we have chosen the resulting Fermi surface for a given density, according to the level dispersion discussed before. This yields a noncircular Fermi surface, having a slight effect on the details of the mode q dependencies. Notice that treating the Fermi vectors k_{xF} and k_{yF} independently for a given density is no longer appropriate,[9,10] since the level dispersion is no longer quadratic in the tunneling systems treated here.

Introducing tunneling in the system, fully accounting for the overlap of wave functions from different dots via the multipole matrix elements $A^{l}_{ii'}$ and $B^{h}_{jj'}$, together with the energy level dispersion of different states, produces a band of extra modes at low energies related to the tunneling degree of freedom. In order to better study the characteristics of this extra band of tunneling collective modes, as well as the expected interband modes, we have performed calculations of the oscillator strength for different wave numbers. Solution of the secular equation (6) only yields mode dispersion curves, while it gives no information on the effective oscillator strength (or coupling to external probes) of the modes. We use the electronic density-density correlation function, which is given by the Dyson's equation

$$\chi(\mathbf{q}_{nm},\mathbf{q}_{n'm'},\omega)=\chi^{0}(\mathbf{q}_{nm},\mathbf{q}_{n'm'},\omega)+\sum_{l,l'}\chi^{0}(\mathbf{q}_{nm},\mathbf{q}_{ll'},\omega)V(\mathbf{q}_{ll'})\chi(\mathbf{q}_{ll'},\mathbf{q}_{n'm'},\omega), \tag{12}$$

where

$$\chi^{0}(\mathbf{q},\mathbf{p},\omega)=\sum_{a,a'}\langle a\mid e^{-i\mathbf{p}\cdot\mathbf{r}}\mid a'\rangle\langle a'\mid e^{i\mathbf{q}\cdot\mathbf{r}}\mid a\rangle\Pi(a,a',\omega)\,. \tag{13}$$

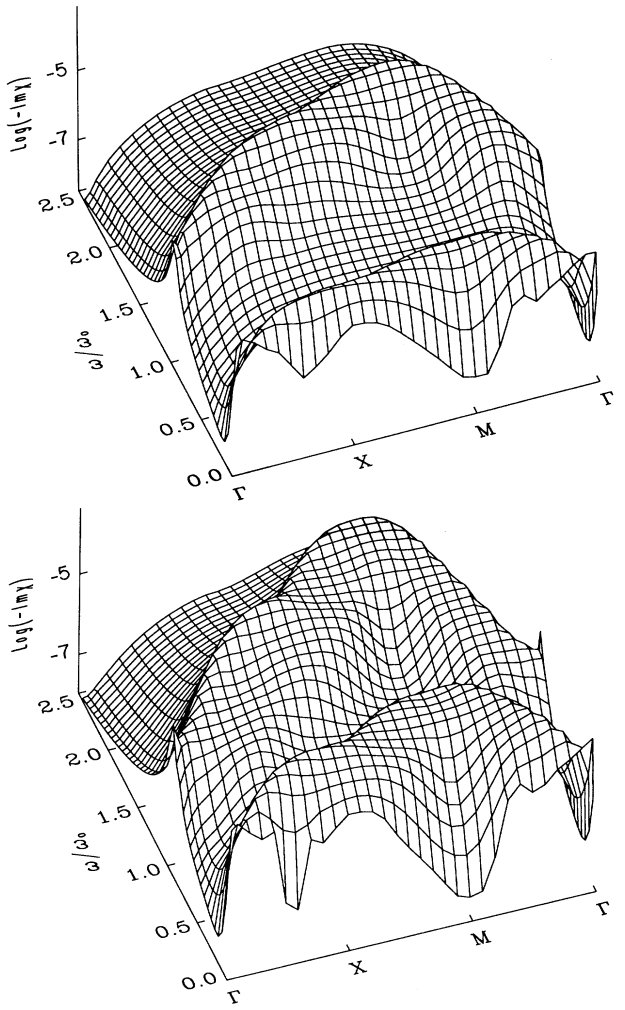

FIG. 2. Plot of $-\mathrm{Im}\chi(q,q;\omega)$ on q-ω plane (notice logarithmic vertical axis, base 10). Notice both intraband and interband set of modes with nonvanishing oscillator strength for low and high energy, respectively. The system parameters used are as in Fig. 1, $d=500$ Å, $\eta=0.02\varepsilon_{10}$, while the bandwidths are (a) top panel, $W_x^{00}=W_y^{00}=0.1\varepsilon_{10}$ and $W_x^{10}=W_2^{10}=0.15\varepsilon_{10}$; (b) bottom panel, $W_{x,y}^{00}=0.2\varepsilon_{10}$ and $W_{x,y}^{10}=0.3\varepsilon_{10}$.

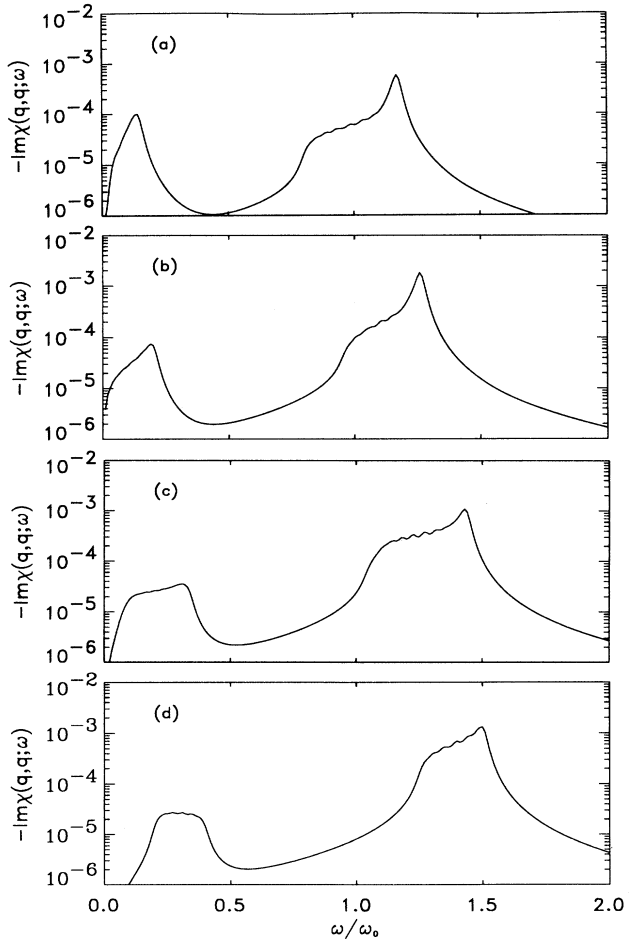

FIG. 3. $-\mathrm{Im}\chi(q,q;\omega)$ versus ω at four different q values: (a) $(\pi/2d,0)$, the half-point on the Γ-X line; (b) X point, $(\pi/d,0)$; (c) the half-point on the X-M line, $(\pi/d,\pi/2d)$; and (d) the M point, $(\pi/d,\pi/d)$. Parameters as in Fig. 2(a). Notice the logarithmic vertical scale.

We can write χ as a matrix in reciprocal space, with compound indices $\chi_{(nm)(n'm')}$, so that $\chi = [I - \chi^0 V]^{-1}\chi^0$.[13] The relative oscillator strength of the collective excitation modes is then evaluated, as it is proportional to the imaginary part of the matrix element $\chi(q_{00}, q_{00}, \omega) = \chi(q, q, \omega)$.[16]

Results of a typical calculation for $-\mathrm{Im}\chi(q, q; \omega)$ are shown in Figs. 2 and 3. These plots show the full response function peaks as the collective modes for this tunneling quantum-dot array system. In changing the bandwidths from zero to $W_x^{00} = W_y^{00} = 0.1\varepsilon_{10}$ and $W_x^{10} = W_x^{10} = W_x^{01} = W_x^{01} = 0.15\varepsilon_{10}$, Fig. 2(a), it is clear that the main interband mode curves acquire strong slopes along the Γ-X and X-M directions. For even larger bandwidths, $W_{x,y}^{00} = 0.2\varepsilon_{10}$ and $W_{x,y}^{10} = W_{x,y}^{01} = 0.3\varepsilon_{10}$, Fig. 2(b), these slopes increase further. This strong q dependence is due to the single-particle level dispersion, as one would expect an effectively weaker Coulomb interaction (and then *smaller* slope) as the carriers are allowed to spread laterally.[10] Notice that in addition to the changes in mode dispersion curves, the interband collective excitations in the nontunneling case (see Fig. 1) become more like a broadband. This "broadening" of the plasma peaks of the system with $\hbar\omega$ around ε_{10} is due to admixtures with the interband single-particle-like transitions made available by the interdot tunneling. A similar background of single-particle-like transitions, shifted due to Coulomb depolarization effects, appears in other systems that allow carrier tunneling, such as superlattices.[13,17] Notice that the main plasma peak rides at frequencies higher than all the possible single-particle transitions and in fact carries most of the oscillator strength (see also Fig. 3).

Also shown prominently in Fig. 2 is the band of low frequency modes in the range $\hbar\omega \leq 0.5\varepsilon_{10}$. The characteristic frequency of these modes increases as a weak function of q, although one would expect that for larger tunneling this function would become closer to a $\sqrt{q}$ dependence, as in a 2D system (see Ref. 11 for a discussion on the one-directional tunneling case). This is qualitatively observed in Fig. 2(b), with a larger tunneling coefficient than in 2(a). We also notice that allowing larger tunneling enhances the oscillator strength of the low-energy tunneling modes. Some of these features are illustrated better in Fig. 3, where $-\mathrm{Im}\chi(q, q; \omega)$ is shown versus the transition frequency ω at four different q values along the Γ-X and X-M lines. At larger q values, we have higher transition frequencies with a somewhat reduced oscillator strength. One should also notice that the single-particle background and tunneling modes have relatively weaker intensities compared with the main interband modes, although their intensities may still be strong enough to be detected by infrared or inelastic light-scattering experiments. Effects of varying electron density will be presented elsewhere, although mode features are weakly dependent in this case of the Fermi level contained in the lowest band.

We have studied the collective excitations and optical response of 2D arrays of quantum dots where tunneling between nearest-neighbor dots is allowed. We show that tunneling strongly affects the shape of the interband mode dispersion curves. We also show that the suitable introduction of the wave-function overlap in the polarization function creates bands of modes near the interband single-particle excitation region and a tunneling plasma mode at low energies. Moreover, we show that the oscillator strength of the various modes shows a rather complex and anisotropic q dependence.

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