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Electromagnetic response of quantum dots

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We show that the many-body interactions do not produce any significant shifts in the far-infrared absorption frequency for a system of well-separated quantum dots with parabolic confining potentials. This is in contrast to a result based on the random-phase approximation but in agreement with recent experimental results.

In recent years it has become possible to fabricate semiconductor systems in which quantum confinement of electrons in all three dimensions can be observed.[1-5] These "quantum dots" are of increasing interest because they can be viewed as artificial atoms whose properties can be well controlled through the material parameters and geometry, and which can be arranged in various periodic or aperiodic structures. Furthermore, it has been suggested[6] that future computer architectures might be based on such structures.

The study of electrons in such systems is usually done by various spectroscopies, most of which utilize electromagnetic radiation. Recent measurements[1] of the electromagnetic response of a system of quantum dots show that the far-infrared (FIR) absorption frequency is independent of the number of electrons, N, occupying each dot. This is in striking contrast to a calculation[7] based on the random-phase approximation (RPA), which predicts significant N-dependent shifts.

In this paper we provide a possible explanation of the lack of variation with N of the observed resonance frequency noted in Ref. 1. In particular, we show that for a system of well-separated dots the interdot electrodynamic interaction produces a shift in the FIR absorption frequency that depends on an effective plasma frequency for the system. For many samples investigated so far,[1-5] this effective plasma frequency is very small, and therefore, in practice, the interdot interactions produce a negligible shift. We obtain this result employing two approaches: In the first, the dots are modeled as point dipoles allowing interdot correlations to be treated exactly; in the second, the confining potential for each dot is assumed to be parabolic and a Hamiltonian approach is used to treat both intra- and interdot interactions. This latter approach also explains that there is no shift due to intradot interactions, irrespective of N. This result, basically a variation of the Kohn's theorem,[8] has been recently obtained in the context of wide parabolic quantum wells by Brey *et al.*[9]

A quantum dot is characterized by quantization of electronic motion in all the three spatial dimensions. We consider an array of quantum dots arranged in a two-dimensional lattice that is taken to lie in the x-y plane. The confinement in the z direction is assumed to be much stronger than that in the x-y plane, so that the dots can be viewed as two-dimensional disks.[5] In current experiments the interdot distances are much larger than the effective dot size. Then the charge densities in the individual dots are well separated (no wave-function overlaps), and for the purpose of studying the interdot effects we begin by modeling the dots as interacting point dipoles.[10] The individual-dot dipole polarizability, $\alpha(\omega)$, which reflects the internal properties of the dot, is to be determined from the intradot physics. The Maxwell's equations of the system take the general form

$$\mathbf{P}(\mathbf{R}_j)=\alpha(\omega)\left[\mathbf{E}(\mathbf{R}_j)-\sum_{j'}\phi(\mathbf{R}_j-\mathbf{R}_{j'})\cdot\mathbf{P}(\mathbf{R}_{j'})\right] \tag{1a}$$

$$-\nabla[\nabla\cdot\mathbf{E}(\mathbf{r})]+\nabla^2\mathbf{E}(\mathbf{r})+\frac{\omega^2}{c^2}\mathbf{E}(\mathbf{r}) =-\frac{4\pi\omega^2}{c^2}\sum_j\hat{\mathbf{e}}\hat{\mathbf{e}}\cdot\mathbf{P}(\mathbf{R}_j)\delta(\mathbf{r}-\mathbf{R}_j)\ , \tag{1b}$$

where $\hat{\mathbf{e}}$ is the unit vector in the direction of polarization, and the dipole-dipole interaction is

$$\phi=\frac{1}{R^3}\left[1-3\frac{\mathbf{R}\mathbf{R}}{R^2}\right] .$$

Specifically, we consider a square lattice of dots in the x-y plane with lattice constant a. The fields $\mathbf{E}$ and $\mathbf{P}$ are taken parallel to the y direction. In the nonretarded limit ($c\rightarrow\infty$), and using standard plane-wave expansions[11,10] of $\mathbf{E}$ and $\mathbf{P}$, we obtain the following dispersion relation for the transverse mode (propagating along x with wave number k and frequency ω):

$$1=-4\pi\alpha(\omega)\beta(k)/a^3\ , \tag{2}$$

where

$$\beta(k)=v(0)+2\sum_{l=1}^{\infty}v(l)\cos(kl\,a)\ , \tag{3a}$$

$$v(l)=\frac{1}{4\pi}\sum_{n=-\infty}^{\infty}\frac{1}{(l^2+n^2)^{3/2}}\left[1-\frac{3n^2}{l^2+n^2}\right],\quad l\neq0\ . \tag{3b}$$

The first few values of $v(l)$ are $v(0)=-\zeta(3)/\pi=-0.382\,624$, $v(1)=1.1579\times10^{-2}$, $v(2)=1.538\times10^{-5}$, and $v(3)=6.4\times10^{-8}$. Note that $\beta(0)=v(0)+2\sum v(l)=-0.359\,43\ldots$, which is equal to the value $v(0)$ of Ref. 10 for the interaction between a square lattice of point dipoles and an in-plane dipole.

For large interdot separations, the plasma modes [solutions of Eq. (2)] occur near the poles of $\alpha(\omega)$. Since the polarizability per dot increases with the number of electrons per dot, N, one may rewrite $\alpha(\omega)=N\alpha'(\omega)$, where $\alpha'(\omega)$, the polarizability per electron in the dot, is (by virtue of the oscillator sum rule) relatively insensitive to N. Then the effective scale size for the interdot shift from the poles of $\alpha'(\omega)$ is given by an effective "plasma frequency" $\omega_p^2=4\pi Ne^2/\epsilon ma^3$, where ϵ is the dielectric constant of the medium and m is the effective mass of an electron. The dispersion (k dependence) of the mode, which is governed by the $v(l)\ll v(0)$, is an order of magnitude smaller than the shift. The dispersion relation for the longitudinal mode ($\mathbf{E}\|\mathbf{k}$) can also be derived similarly, and leads to the same shift for $k=0$, and its dispersion, though different, is also very small.

For the InSb sample of Ref. 1 (parameters: $a=2500$ Å, $\epsilon=17.9$, $m=0.014m_e$, resonance frequency $\omega_0=7.5$ meV, and $N=3$–20), the interdot shift $\Delta\omega$, which is proportional to ω_p^2, is negligible ($\Delta\omega/\omega_0=-0.36$ to -2.4%) and we conclude that any shift of the FIR absorption frequency (or lack thereof) has to be ascribed to the intradot physics.[12] Furthermore, since $v(l)\ll v(0)$ the dispersion (k dependence) of the mode is extremely small. In fact, these statements hold true for other relevant dot systems investigated to this point.[2–4] In order to produce any significant shift in the absorption frequency due to interdot effects, one has to substantially increase ω_p^2. Note that this can be achieved most efficiently by reducing the interdot separation, a, since $\omega_p^2\sim a^{-3}$.

Now we consider the electromagnetic response of a single isolated dot. If the dot is occupied by a single electron, there are no interelectron effects to consider, and the (FIR) electromagnetic absorption should occur at the bare level spacings. Applying the RPA to this system, and thus making the electron interact with its own mean field, is conceptually incorrect, and is in clear violation of elementary quantum mechanics.[13]

We next ask under what conditions can we expect the intradot, many-electron physics to produce *no shift* of the[1] FIR absorption frequency with N, as has been observed. In general, the electron-electron interaction produces N-dependent shifts. However, it will not, if the N-electron Hamiltonian can be separated into the form

$$H=H_{\text{c.m.}}(\mathbf{R},\mathbf{P})+H'\,, \tag{4}$$

and the center of mass (c.m.) variables $\mathbf{R}=(1/N)\sum_i\mathbf{r}_i$ and $\mathbf{P}=\sum_i\mathbf{p}_i$, commute with H'. The radiation dipole operator $\sum_i e\mathbf{r}_i=eN\mathbf{R}$, being a pure c.m. variable, then does not couple to H', which contains all the electron-electron interactions. Thus the dipole operator, for a uniform field, induces transitions only between the states of $H_{\text{c.m.}}$, and absorption can occur only at the corresponding interlevel separations, irrespective of the details of the electron-electron interactions. If, in addition, these level separations are independent of N, the system will have the desired property.

Now we turn to the question of what *specific* confining potentials $V_c(\mathbf{r}_i)$ can lead to such separability. The Hamiltonian for the N-electron system is given by

$$H=\sum_{i=1}^{N}\left[\frac{\mathbf{p}_i^2}{2m}+V_c(\mathbf{r}_i)\right]+\sum_{i>j}u(r_{ij})\,,$$

$$\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j,\quad r_{ij}=|\mathbf{r}_{ij}|\,. \tag{5}$$

We note that the identity for quadratic forms,

$$\sum_i\mathbf{p}_i^2=\frac{1}{N}\mathbf{P}^2+\frac{1}{N}\sum_{i>j}\mathbf{p}_{ij}^2,\quad \mathbf{p}_{ij}=\mathbf{p}_i-\mathbf{p}_j \tag{6}$$

renders the kinetic-energy term separable in the sense of Eq. (4), since $[\mathbf{R},\mathbf{p}_{ij}]=0$. The electron-electron interaction term, $u(r_{ij})$, obviously depends only on the relative coordinates $\mathbf{r}_{ij}$ which commute with $\mathbf{P}$. Thus the separability of H essentially rests on whether we can rewrite the confining potential term, $\sum_i V_c(\mathbf{r}_i)$, in a separable form. The first term in the Taylor expansion,

$$V_c(\mathbf{r}_i)\equiv V_c(\mathbf{r}_i'+\mathbf{R})=V_c(\mathbf{r}_i')+\mathbf{R}\cdot\nabla V_c(\mathbf{r}_i')+\cdots\,, \tag{7}$$

clearly commutes with $H_{\text{c.m.}}$ since $\mathbf{r}_i'=\mathbf{r}_i-\mathbf{R}$ commutes with $\mathbf{P}$. Each of the higher-order terms, upon summation over i, has to be separable, or must vanish. This is a severe constraint on the form of $V_c(\mathbf{r}_i)$, and, in fact, only the constant, linear, and quadratic potentials give rise to separability. The first two forms do not produce confinement. For the quadratic potential, $V_c(\mathbf{r}_i)=\frac{1}{2}m\omega_0^2\mathbf{r}_i^2$, we have explicitly

$$H_{\text{c.m.}}=\frac{\mathbf{P}^2}{2Nm}+\tfrac{1}{2}Nm\omega_0^2\mathbf{R}^2\,, \tag{8}$$

$$H'=\frac{1}{N}\sum_{i>j}\left[\frac{\mathbf{p}_{ij}^2}{2m}+\tfrac{1}{2}m\omega_0^2\mathbf{r}_{ij}^2+Nu(\mathbf{r}_{ij})\right]. \tag{9}$$

As mentioned earlier, the dipole operator only couples to $H_{\text{c.m.}}$ and, due to the harmonic-oscillator structure of $H_{\text{c.m.}}$ in Eq. (8), only adjacent levels produce nonvanishing matrix elements. Therefore we conclude that the electromagnetic FIR absorption spectrum of the many-electron system with the parabolic confinement will have only one, N-independent absorption peak, corresponding to the bare interlevel energy separation of the confining potential, ω_0. The same result can also be obtained by a variation of the Kohn's theorem,[8] as has been shown for wide parabolic quantum wells in Ref. 9. Clearly, no other forms of $V_c(\mathbf{r}_i)$ lead to separability, and, as a consequence thereof, an N-independent absorption frequency. This strongly suggests that the confining potentials in Ref. 1, and possibly other experiments, are parabolic.[14]

We wish to emphasize that $H_{\text{c.m.}}$ is the only relevant part of the Hamiltonian for the purpose of determining the FIR electromagnetic response (at $k=0$) of the quantum dot. In fact, one can replace the interacting system of N electrons by a single quasiparticle, with charge $e^*=Ne$ and mass $M=Nm$, oscillating in a parabolic well with frequency ω_0. The corresponding polarizability is then given by $\alpha(\omega)=e^{*^2}/M(\omega_0^2-\omega^2)=Ne^2/m(\omega_0^2-\omega^2)$, where the latter expression can also be viewed as the sum of the polarizabilities of N noninteracting electrons.

Effects of a magnetic field, $\mathbf{B}$, can be easily included in the Hamiltonian approach. When $\mathbf{B}$ is perpendicular to the plane of the lattice, it can be shown that FIR absorption occurs, in spite of the electron-electron interactions, at two frequencies:

$$\omega_\pm=(\omega_0^2+\tfrac{1}{4}\omega_c^2)^{1/2}\pm\tfrac{1}{2}\omega_c, \quad \omega_c=eB/mc \ . \tag{10}$$

The proof of separability is straightforward. Now,

$$H_{\text{c.m.}}=\frac{\mathbf{P}^2}{2Nm}+\tfrac{1}{2}Nm(\omega_0^2+\tfrac{1}{4}\omega_c^2)\mathbf{R}^2+\frac{\omega_c}{2}L_z \ , \qquad L_z=XP_Y-YP_X \ . \tag{11}$$

While Eq. (10) has been employed before to explain the experimental results,[1,3,4] its derivation was based on a single-electron picture (neglecting all electron-electron interactions). We have shown here that for a system of N *interacting* electrons the same formula holds. This proof provides an explanation of why the simple theory used in Refs. 1, 3, and 4 yielded good agreement with experiments.

In contrast to the above results showing no dependence of the FIR absorption peak on N, a calculation[7] based on the RPA, in which a parabolic potential is employed with two electrons per dot, gives a large positive shift (~40 %) of the absorption peak from ω_0. This discrepancy raises the question of the applicability of the RPA for dot systems. The RPA, in principle, cannot be used for a dot with few electrons. Furthermore, it can produce accurate results *only* when it is used in conjunction with a self-consistent Hartree calculation of the ground state. There seems to be considerable confusion in the literature regarding this point. The proper procedure is to solve the coupled Schrödinger-Poisson equations, in which the total potential consists of the initial confining potential (parabolic in this case) *plus* the Hartree potential, which is determined self-consistently with the charge density being obtained from the square of the electron wave functions. We have performed a Hartree-RPA calculation for parabolic quantum dots[15] and we find that this two-step method reproduces the exact result (i.e., absorption occurring at ω_0) to a high degree of accuracy. A similar result has been obtained in Ref. 9 for wide parabolic quantum wells. Basically, a significant downward shift in the ground-state intersubband separations exactly compensates the upward shift due to the RPA. In contrast, Ref. 7 omits the ground-state (Hartree) calculations and therefore the large upward RPA shift remains uncompensated.

We must stress, however, that the special case of the parabolic potential may not necessarily be considered a test of the applicability of the Hartree-RPA approach. As shown above, the dipole interaction operator probes in this case only the part of the Hamiltonian that is insensitive to electron-electron interactions, and therefore a Hartree-RPA calculation, which treats incompletely these interactions, can be expected to produce a correct result.

The Hamiltonian separability approach continues to be useful for nearly parabolic systems (through standard perturbation theory, treating departures from the parabolic form as perturbations). It also allows a unified treatment of the interdot and intradot FIR response. We can show that the total Hamiltonian for a system of ν quantum dots, with a parabolic confining potential for each dot, is essentially separable. All the electron-electron interactions, interdot as well as intradot, go into H', and play no role in determining the FIR absorption frequency. All the quadratic momentum and coordinate terms separate in the same way as before, except that $\mathbf{P}$ and $\mathbf{R}$ now refer to the total momentum and the center-of-mass variable of the whole system. The electron-ion[16] interactions consist of intradot and interdot parts. The intradot electron-ion interaction for each dot is already taken into account through the parabolic confining potential. The interdot term, on the other hand, is not separable, but approximate separability can be achieved to a high degree of accuracy through its Taylor expansion in the electron coordinate,

$$-\frac{e^2}{|\mathbf{d}-\mathbf{r}|}=-\frac{e^2}{d}+e^2\mathbf{r}\cdot\frac{\mathbf{d}}{d^3}+\frac{e^2}{2d^5}[x^2(d^2-3d_x^2)+y^2(d^2-3d_y^2)-6xyd_xd_y]+\cdots \ , \tag{12}$$

where $\mathbf{d}=\mathbf{b}+\boldsymbol{\rho}$, $\mathbf{r}$ is an electron coordinate in a given dot, and $\boldsymbol{\rho}$ is an ion coordinate in any of the other dots, $\mathbf{b}$ being the lattice vector connecting the two dots under consideration.

The first term on the right-hand side of Eq. (12) only alters the reference energy and is irrelevant for the interlevel separations. The second term, summed over all the ions in all the other dots, represents the effect of the net interdot ionic field at the center of the dot under consideration; for a lattice of dots with translational symmetry, this term will vanish. The third term, summed over the ions, leads to an additional force on the electron at $\mathbf{r}$ due to all the ions in the other dots; it is related to the quadrupole moment of the ion distribution around the dot under consideration. By symmetry, the effect for each dot is the same. Essentially, this quadratic term in $\mathbf{r}$ alters the restoring force on the electron and represents the main effect of the interdot electron-ion interactions. The higher-order terms (beyond the quadratic) can be ignored when the interdot separation is much larger than the wave-function spread for an electron, and then separability is achieved. The full Hamiltonian now only consists of terms quadratic in momenta and coordinates, apart from the electron-electron interaction term, and is thus separable by the same arguments as in the case of a single dot: $H = H_{\text{c.m.}}(\mathbf{R},\mathbf{P}) + H'$. Specifically,

$$H_{\text{c.m.}} = \frac{P^2}{2N\nu m} + \tfrac{1}{2}N\nu m\,\Omega^2 R^2 , \tag{13}$$

where Ω represents the effective oscillation frequency of the center of mass of all the electrons in the system. It is also the resonance frequency of the system.

The specific value of Ω depends on the ion-charge distribution. The additional "potential" due to the ions in the other dots, represented by Eq. (12), shifts the restoring force constant $m\omega_0^2$ in a given dot by

$$\sum \frac{e^2}{d^5}(d^2 - 3d_x^2) = \sum \frac{e^2}{d^5}(d^2 - 3d_y^2) = -\sum \frac{e^2}{2d^3} ,$$

where the summation extends over all the ions in all other dots. We have assumed that the x and y distributions are the same. The resulting FIR absorption frequency for a system of dots is then given by

$$\Omega^2 = \omega_0^2 - \sum \frac{e^2}{2md^3} . \tag{14}$$

This expression has a simple physical meaning: the interdot electron-ion interactions soften the restoring force on the electrons in a given dot; thus the effect of forming a lattice of dots is to decrease the absorption frequency from that of a single dot. Therefore the shift from ω_0 is negative, in contrast to the normally positive shift due to electron-electron interactions. The interdot electron effects are absent here, as those interactions are in H', which does not couple to the total radiation dipole operator, $\mathbf{D} = eN\nu\mathbf{R}$.

For the ion distribution corresponding to the point dipole ($\rho \equiv 0$) model, with N ions at each site, $\mathbf{b} = la\hat{\mathbf{x}} + na\hat{\mathbf{y}}$, Eq. (14) reduces to

$$\Omega^2 = \omega_0^2 - \gamma\omega_p^2 , \tag{15}$$

where ω_p has been defined earlier, and the sum $\gamma \equiv (1/8\pi)\sum'(l^2+n^2)^{-3/2}$ (l,n do no vanish simultaneously) can be shown to be equal to $-\beta(0)$ of Eqs. (3). Equation (15) is consistent with the result obtained from the dipole-dipole approximation [Eq. (2), for $k=0$] if $\alpha(\omega)$ is taken to be $Ne^2/m(\omega_0^2-\omega^2)$, as is appropriate for a harmonic oscillator.

In summary, we have shown that a system of well-separated quantum dots with parabolic confining potentials will have an (FIR) absorption frequency that will be insensitive to N, the number of electrons per dot. This might explain the experimentally observed[1] insenstivity to N. This result will also hold for other systems with parabolic initial confining potentials (e.g., quantum wires, quantum wells, etc.). A dispersion relation is obtained for the interdot effects in the dipole-dipole approximation, and the magnitude of the interdot shift and dispersion are shown to be negligible for relevant experiments. We have also developed a Hamiltonian approach, based on the concept of separability of center-of-mass and relative (or difference) variables. This method provides a proof that only parabolic confining potentials lead to exact separability and N independence. Approximate separability may prevail under more general conditions; a systematic development of this approach for a broader class of systems will be given elsewhere. We have already shown here how the Hamiltonian separability approach allows a unified treatment of the interdot and intradot physics of a system of quantum dots. We have emphasized here a first-principles quantum-mechanical approach, without recourse to any many-body approximations.

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[12] In this context the interpretation by the authors of Ref. 1 that the metallic electrode completely screens the interdot interactions is irrelevant, since these interactions are negligible anyway.

[13] $|\Psi|^2$ is the probability density and not the charge density. In the Hartree (or RPA) approximation, $|\Psi|^2$ is assumed to be a charge density, which is approximately valid only for a statistical ensemble of many electrons; therefore it *cannot* be used in general for few-electron systems. An interesting comment on the historical context of the confusion regarding the intepretation of $|\Psi|^2$ is given in *Feynman Lectures in Physics* (Addison-Wesley, Reading, MA, 1989); Vol. III, pp. 21–26.

[14] The details of how the parabolic potential is produced by the neutralizing positive point charges may vary from one experiment to another.

[15] D. A. Broido, K. Kempa, and P. Bakshi (to be published).

[16] Here, "ions" refer to the neutralizing positive point charges, e.g., ions of dopants for the quantum dots etched from modulation-doped heterostuctures.