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Spontaneous polarization in quantum-dot systems

K. Kempa, D. A. Broido, and P. Bakshi
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02167-3811
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We show that the dipole-dipole interactions can lead to a spontaneous polarization of quantum dots. For a square lattice of dots we find that the phase transition into the polarized state is of the second order, and leads to an antiferroelectric arrangement.

Recent advances in semiconductor technology have lead to the fabrication of quantum-dot structures,[1-7] in which carriers are confined in all three spatial dimensions. Quantum dots can be viewed as artificially structured atoms. Most common arrangements of dots are two-dimensional (2D) periodic arrays, which can be construed as *planar crystals*. Therefore, some phenomena that occur in crystals might also be expected to occur in such dot systems. In this Rapid Communication we explore one such phenomenon: the spontaneous phase transition into a ferroelectric or antiferroelectric state. Such a phenomenon is of intrinsic scientific interest and, in addition, could lead to important device applications, such as nanometer-size switching elements for novel computer architectures.[1]

Thus far, most of the investigations of quantum-dot systems have focused primarily on the far-infrared response of individual dots,[4-7] and have neglected the interdot interactions. In a previous paper[8] we have shown that the strength of the interdot interaction depends on the ratio N/a^3, where N is the number of electrons per dot and a is the interdot spacing. For most current semiconductor dot systems, the interdot effects are negligible. However, for closely spaced dots that contain a sufficiently large number of electrons, this interaction can be large. In this paper we show that under such conditions a phase transition may occur in which the dot system will polarize spontaneously. In particular, we show that a second-order phase transition can occur that leads to both ferroelectric and antiferroelectric arrangements, with the latter being the most stable phase.

We consider a system of quantum dots arranged in a 2D lattice that is taken to lie in the x-y plane. We model the dots as interacting point dipoles.[8] This model assumes that the electronic charge density in each dot remains well separated from that of its neighbors. The internal properties of each dot are reflected through a dipole polarizability $\alpha(\omega)$, which is determined from the intradot physics.

Previously,[8] we studied the electromagnetic response of such a system of dots within the linear response theory. We found that the dispersion relation of the transverse plasma mode in this system is[9]

$$1=4\pi\alpha(\omega)\xi(k)/a^3 \tag{1}$$

where, for a square lattice of dots with lattice constant a,

$$\xi(k)=v(0)+2\sum_{l=1}^{\infty} v(l)\cos(kla) \tag{2}$$

and

$$v(l)=-\frac{1}{4\pi}\sum_{n=-\infty}^{\infty}\frac{1}{(l^2+n^2)^{3/2}}\left(1-\frac{3n^2}{l^2+n^2}\right), \quad n\neq 0 \text{ for } l=0. \tag{3}$$

The first few values of $v(l)$ are $v(0)=0.383$, $v(1)=-1.16\times10^{-2}$, $v(2)=-1.54\times10^{-5}$, $v(3)=-6.4\times10^{-8}$.

Since the confining potential inside a dot is roughly parabolic,[4,8,10,11] the N interacting electrons in each dot can be viewed[8,10] as a single quasiparticle with charge Ne and mass Nm oscillating in a parabolic well whose frequency is ω_0. Therefore, $\alpha(\omega)$ can be assumed to have the form $\alpha(\omega)=Ne^2/\epsilon m(\omega_0^2-\omega^2)$, where ϵ is the dielectric constant of the dot material and m is the electron effective mass. The dispersion relation of the transverse plasma mode is then given by

$$\omega^2(k)=\omega_0^2-\xi(k)\omega_p^2, \quad \omega_p^2=\frac{4\pi Ne^2}{\epsilon ma^3}. \tag{4}$$

Note that the downward shift from ω_0 corresponds to a softening of the mode, and that a significant shift in ω can be achieved most efficiently by reducing the interdot separation a. When the interdot interaction is strong enough that $\omega(k)=0$, the plasma mode becomes "frozen out" and describes an arrangement of dipole moments in the dot system arising from static deformations of the electronic charge densities within each dot. This point corresponds to the onset of spontaneous polarization in the dot system. For a given lattice spacing a, the critical number of electrons required for this to happen, $N_c(k)$, is given from Eq. (4) with $\omega(k)=0$, i.e., $\omega_0=[\xi(k)]^{1/2}\omega_p$:

$$N_c(k)=\frac{\epsilon m\omega_0^2}{4\pi e^2\xi(k)}a^3. \tag{5}$$

It can be seen, from Eqs. (2)–(4), that the dispersion is negative. As a result, the intersection with the $\omega=0$ axis occurs first at the boundary of the first Brillouin zone: $k=\pi/a$. If N exceeds $N_c(\pi/a)$, the $\omega=0$ crossing of the dispersion occurs for $k_1<\pi/a$, predicting a stable static arrangement with wavelength $2\pi/k_1$. The higher-k modes ($k_1<k\leq\pi/a$), however, now become unstable [$\mathrm{Im}(\omega)\neq0$] and spontaneously begin to grow exponentially on a time scale $\tau(k)=[\mathrm{Im}(\omega)]^{-1}=\omega_0^{-1}[N/N_c(k)-1]^{1/2}$. Thus the dominant instability occurs for $k=\pi/a$. Nonlinear effects should set in over a time period a few times τ.

We now analyze this phenomenon through an electrostatic analysis, including nonlinear effects. For a static ($\omega=0$) external electric field, $\mathbf{E}=E(x)\hat{\mathbf{y}}$, the dipole moments of the dots become polarized in lines that are parallel to $\mathbf{E}$. The dipole moment $P(l)$ of a dot lying on the lth line can be written in terms of a local field expansion of the form

$$P(l)=\alpha_1 E_{\rm loc}(l)+\alpha_3 E_{\rm loc}^3(l)+\cdots,$$
$$E_{\rm loc}(l)=E(la)+\frac{4\pi}{a^3}\sum_{l'} v(l')P(l'), \tag{6}$$

where the local field at the line l, $E_{\rm loc}(l)$, is composed of the external field and that produced by the other dipole lines. The linear polarizability $\alpha_1=\alpha(0)=Ne^2/\epsilon m\omega_0^2$, while the third-order polarizability α_3 and all higher orders are determined from the anharmonicity of the potential seen by the quasielectron in the dot.[12] This potential, including the first anharmonic term, can be expressed as

$$V(R)=\tfrac{1}{2}Nm\omega_0^2R^2+\tfrac{1}{4}\gamma R^4+\cdots, \tag{7}$$

where R is the displacement of the quasielectron from the center of the dot and γ is a positive constant. It is then straightforward to show that[12]

$$\alpha_3=-\frac{N\gamma}{\epsilon^2}\left(\frac{e}{m\omega_0^2}\right)^4. \tag{8}$$

For a plane-wave variation of the field, $E(x)=Ee^{ikx}$, the solution of Eqs. (6) is obtained by writing $P(l)=Pe^{ikla}$. Then, we obtain

$$P=\chi_1 E+\chi_3 E^3+\cdots, \tag{9}$$

where the linear and third-order susceptibilities χ_1 and χ_3 are given by

$$\chi_1=\frac{\alpha_1}{1-(4\pi/a^3)\alpha_1\xi(k)}=\frac{Na^3}{4\pi\xi(k)[N_c(k)-N]}, \tag{10a}$$

$$\chi_3=\frac{\alpha_3}{[1-(4\pi/a^3)\alpha_1\xi(k)]^4}=\alpha_3\left(\frac{N_c(k)}{N_c(k)-N}\right)^4. \tag{10b}$$

Spontaneous polarization occurs for a given k when the denominator in Eqs. (10) vanishes. Note that this condition is identical with that arising from Eqs. (1) and (4) for $\omega=0$.

The stable phases associated with the transition into this new state can be determined from a thermodynamic analysis.[13] The Landau free-energy density F can be expanded in powers of P as

$$F=\tfrac{1}{2}\sum_l aP^2(l)+\tfrac{1}{4}\sum_l bP^4(l)+\cdots-\sum_l E(la)P(l)$$
$$=\tfrac{1}{2}AP^2+\tfrac{1}{4}BP^4+\cdots-CEP, \tag{11}$$

where A, B, and C are constants. It has been assumed that temperature $T=0$ K. Minimizing F with respect to P yields

$$CE=AP+BP^3+\cdots, \tag{12}$$

and combining Eqs. (9) and (12) yield

$$\frac{A}{C}=\frac{1}{\chi_1},\quad \frac{B}{C}=-\frac{\chi_3}{\chi_1^4}=-\frac{\alpha_3}{\alpha_1^4}. \tag{13}$$

For vanishing external electric field $E=0$, this yields a nonvanishing moment:

$$P_s=\left(\frac{-A}{B}\right)^{1/2}=D\left(\frac{N_c(k)-N}{\alpha_3}\right)^{1/2},$$
$$D=\frac{1}{N_c^2}\left(\frac{Na^3}{4\pi\xi}\right)^{3/2}. \tag{14}$$

When the phase transition occurs, $N>N_c$, and the numerator becomes negative. Therefore, since from Eq. (8), $\alpha_3<0$, the transition is second order. The minimum of F is given by

$$F(P_s)=-N\Delta[N-N_c(k)]^2,\quad \Delta=\frac{1}{\gamma}\left(\frac{4\pi e^2\xi(k)}{a^3\epsilon}\right)^2. \tag{15}$$

From Eqs. (15) and (5) it is clear that $F(P_s)$ decreases with increasing $\xi(k)$ which, in turn, increases with k [see Eqs. (2) and (3)]. Therefore, the most stable configuration [lowest $F(P_s)$] occurs at the boundary of the first Brillouin zone: $k=\pi/a$. The corresponding dipole moments along line l are $P(l)=P(-1)^l$. Adjacent lines then have oppositely directed moments, and the arrangement of lines is antiferroelectric. For $k<\pi/a$, other less stable antiferroelectric arrangements are generated with longer wavelengths, $\lambda=2\pi/k$. A ferroelectric arrangement occurs for $k=0$ when $P(l)=P$. This is the least-stable arrangement since it leads to the smallest value of ξ [largest $F(P_s)$]. We note that the longitudinal mode that arises in the mode analysis discussed above leads to an arrangement of dipoles that is even less energetically favorable except for $k=0$ where it becomes degenerate with the transverse branch. We therefore will not discuss this arrangement here.

That the antiferroelectric arrangement is the most stable can be seen from the form of the dipole-dipole interaction. For two neighboring dipoles lying in the same line, the minimum energy arrangement occurs when the dipoles are parallel to each other. In contrast, for two adjacent lines the minimum energy arrangement occurs when the neighboring dipoles are antiparallel to each other. Thus, the minimum-energy arrangement of the system of dipoles corresponds to antiferroelectric lines.

It is interesting to note that in the three-dimensional case, the ferroelectric phase is more stable than the antiferroelectric phase for the body-centered-cubic and face-centered-cubic arrangements.[14,15] The corresponding "body-centered" square (bcs) and "face-centered" square lattices in two-dimensions are identical and correspond to a simple square (ss) lattice rotated by 45°. However, the minimum energy phase for dipoles in the bcs lattice still corresponds to the antiferroelectric lines. Furthermore, this phase is less energetically favorable than that of the original ss lattice. It is, however, possible that some other 2D geometrical arrangements of the dot system would

favor the ferroelectric phase over the antiferroelectric phase.

The above analysis is strictly valid only for a system of point dipoles. However, it is reasonable to expect that the resulting conclusions are also approximately correct even for systems where the dot sizes are comparable to the interdot distances, provided that the electronic charge densities in neighboring dots remain well separated. In current systems of semiconductor quantum dots the interdot spacings are generally a few thousand angstroms. This leads to a corresponding critical electron number, N_c, far exceeding the capacity of an individual dot, and therefore, no spontaneous polarization occurs. For example, taking the parameters from Ref. 4 (InSb dots, $a=2500$ Å, $\omega_0=7.5$ meV, $\epsilon=17.9$, $m=0.014m_0$), the most stable (antiferroelectric) arrangement, for which

$$\xi(\pi/a)=v(0)+2\sum_{l=1}^{\infty} v(l)(-1)^l \approx 0.406 ,$$

occurs for $N_c \approx 400$ [see Eq. (5)]. This is much larger than the actual electron numbers, $N=2$–20 in the experiment.[4] However, if an equivalent system with $a=1500$ Å could be constructed, the corresponding critical electron number would be a much more reasonable $N_c \approx 84$ (provided that $\omega_0 \approx 7.5$ meV). Such an arrangement may be within the range of what is currently technologically achievable.[16] Reducing a further, if feasible, will require even smaller N_c, since the scaling is $N_c \sim a^3$. For example, for $a \approx 1000$ Å, the critical electron number would be $N_c \approx 25$, almost the actual electron number used in the experiment.

In mesa-etched dots the resonance frequency increases with N. This is in contrast to the gated-dot systems,[4] where ω_0 remains independent of N. Thus the latter may be the best candidates to observe the phase-transition phenomena. One should also explore more complex geometrical arrangements such as hexagonal lattices, interpenetrating lattices of dots of different sizes, etc. . . .

In conclusion, we have investigated the conditions under which spontaneous polarization will occur in a 2D square array of dots modeled as point dipoles. We have shown that if dots are brought sufficiently close and contain enough electrons, such spontaneous polarization should occur, provided that the resonance frequency of a dot does not increase significantly with the number of electrons in the dot. The phase transition is of second order, and, in the absence of external electric fields, the antiferroelectric phase is the most stable. We would like to encourage experimental effort to verify existence of this phenomenon.

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