Polarization transitions in quantum ring arrays

Bahman Roostaei[1] and K. Mullen[1]

1University of Oklahoma, Department of Physics and Astronomy, Norman OK 73019

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We calculate the zero temperature electrostatic properties of charged one and two dimensional arrays of rings, in the classical and quantum limits. Each ring is assumed to be an ideal ring of negligible width, with exactly one electron on the ring that interacts only with nearest neighbor rings. In the classical limit we find that if the electron is treated as a point particle, the 1D array of rings can be mapped to an Ising antiferromagnet, while the 2D array groundstate is a four-fold degenerate “stripe” phase. In contrast, if we treat the electrical charge as a continuous fluid, the distribution will not spontaneously break symmetry, but will develop a charge distribution reflecting the symmetry of the array. In the quantum limit, the competition between the kinetic energy and Coulomb energy allows for a transition between unpolarized and polarized states as a function of the ring parameters. This allows for a new class of polarizable materials whose transitions are based on geometry, rather than a structural transition in a unit cell.

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Advances in technology have made it possible to fabricate structures like dots, and wires on the micro- or nano-scale. Such quantum dots act as artificial atoms, with spectra[1] and shell structure similar to those of real atoms. Recently it has also become possible to assemble structures like dots, and wires on the micro- or nano-scale. We start by considering a 1D array of ideal rings. The radius of each ring is \( R \), and their center-to-center separation is \( D \). On each ring we place one electron with charge \( e \) (fig.1). Because the electrons are confined to move on a ring, their moment is constrained in magnitude but not in direction.

The energy of a 1D array is given by

\[
U_{1D} - U_0 \approx \frac{e^2}{D} \sum_i \left( \frac{3}{2} \cos 2\theta_i + \cos(\theta_i - \theta_{i+1}) \right) - 3 \cos(\theta_i + \theta_{i+1})
\]

\[
= \frac{e^2}{D} \sum_i \left( 2\hat{s}_i \cdot \hat{s}_{i+1} + \frac{3}{2}(\hat{D} \cdot (\hat{s}_i - \hat{s}_{i+1}))^2 \right)
\]

where \( \epsilon \equiv R/D \) and \( U_0 \) is a constant, \( U_0 \equiv \frac{N}{2} \left( 1 + \frac{e^2}{2} \right) \). In the second expression we write the position of the charge as \( \hat{s}_i \), a vector in the 2D plane pointing from the center of the \( i \)-th ring to the charge on that ring. The \( \cos 2\theta \) (or \( \hat{D} \cdot \hat{s} \)) term explicitly breaks the rotational symmetry, driving the system from XY to Ising-like behavior. The last two terms in eq.1 compete: the first driving the system to be ferroelectric (like direction) while the latter, larger term favoring states where neighbors point in opposite directions. Thus the system at zero temperature will order in an AFE pattern in 1D. This is born out by numerical Monte Carlo simulations of the exact Coulomb interaction, which reproduce the AFE pattern on finite size arrays (Fig.1).

If we expand the potential to quadratic order in the classical expressions, we find that the groundstate makes spontaneously break the symmetry, but instead produce charge distributions that reflect the symmetry of the array. In the quantum limit we find that the groundstate makes simultaneous break the symmetry, but instead produce charge distributions that reflect the symmetry of the array. In the quantum limit we find that the groundstate makes simultaneously break the symmetry, but instead produce charge distributions that reflect the symmetry of the array. In the quantum limit we find that the groundstate makes simultaneously break the symmetry, but instead produce charge distributions that reflect the symmetry of the array.

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We present these findings below, and then conclude with a discussion of how they might be tested by experiment.

**Classical Results:** In order to gain intuition for the quantum problem, we will examine the classical case. Here there are two limits: discrete charges and continuous charges. We first examine the limit of ideal point charges on each ring with nearest neighbor interactions only.

We start by considering a 1D array of ideal rings. The radius of each ring is \( R \), and their center-to-center separation is \( D \). On each ring we place one electron with charge \( e \) (fig.1). Because the electrons are confined to move on a ring, their moment is constrained in magnitude but not in direction.

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If we expand the potential to quadratic order in the
displacement and calculate the normal modes we find:

\[ \omega_{\pm}^{(1D)}(k) = 2\omega_0 \sqrt{4 \pm 2 \cos \frac{kD}{2}} \]

where \( \omega_0 \equiv \sqrt{e^2/m^*D^3} \). Both the modes are gapped, since there is an Ising-like term that provides a harmonic restoring force at each site. Note that they are independent of ring radius at this level of approximation.

We may repeat the analysis above for a 2D array of rings. The Coulomb energy for an \( N \times N \) array may be written as

\[ U_{2D} \approx \frac{2N^2}{D} \left( 1 + \frac{5}{4} \epsilon^2 \right) \]

\[ + \frac{\epsilon^2}{D} \sum_{i,j} (2\hat{s}_{x,i,j} \cdot \hat{s}_{x,i+1,j} + \hat{s}_{x,i,j} \cdot \hat{s}_{x,i,j+1}) \]

\[ - \frac{3}{2} (\hat{s}_{x,i,j} \hat{s}_{x,i+1,j} + \hat{s}_{x,i,j} \hat{s}_{x,i,j+1}) \]

where \( i \) and \( j \) denote the \( x \) and \( y \) indices of the ring in the plane. Since each ring in the plane (except the ones on the top and right boundaries) has one neighbor to the right and one above it, this covers the array without double counting. If we neglect edge effects the minimum energy configuration of the system is AFE in one direction (\( x \) or \( y \)) and FE in the other (\( y \) or \( x \)). Since the AFE state is itself doubly degenerate, we have a four-fold degenerate minimum energy state (Fig. 1). This is supported by numerical simulations which enforce (unphysical) periodic boundary conditions on the array. In more realistic 2D finite arrays the long range order is perturbed by a “vortex” like pattern on the boundaries where the charge on each ring resides on the point that puts it on the outermost perimeter of the array.

A normal mode analysis shows that the excitation spectrum is given by

\[ \omega_{\pm}^{(2D)}(k) = \omega_0 \sqrt{6 + 2 \cos k_y D \pm 4 \cos \frac{k_x D}{2}} \]

where we have assumed that the stripes run vertically in the ordered phase, as in Fig. 1. Note that the acoustic mode is now ungapped, at the wavevector \( \vec{k}_0 = (0, \pi/D) \), in contrast to the 1D case. This result holds in the case of only nearest neighbor interactions, where the increase in the energy due to departure from AFE order in the horizontal neighbors is compensated by the motion of vertical neighbors from FE alignment. This mode differs from the magnetoplasmon mode of metal ring arrays.

A second classical limit that contrasts with the point charge model is a continuous charge fluid. That is, each point on the ring has a charge density \( \rho_i(\theta_i) \) such that the integrated charge density per ring is a constant, \( \int \rho_i(\theta_i) d\theta_i = 1 \). We seek a minimal solution to the variational quantity:

\[ I = \frac{1}{2} \int d\theta \int d\theta' \sum_{i,j} \rho_i(\theta) \rho_j(\theta') \]

\[ + \lambda \sum_i \int d\theta \rho_i(\theta) \]

For a 1D ring this is expression is divergent due to the infinite self energy. We can regularize this in several ways. One method is to introduce a short distance cutoff \( \lambda \) to the Coulomb interaction, discretize the integral equation, and then solve the problem numerically. This can also be done analytically, expanding all results to lowest order in the cutoff, \( \epsilon \).

An approximate analytic solution can then be obtained by Fourier expanding the distribution, keeping only the first three modes. We find that amplitude non-trivial Fourier mode as a function of \( \lambda \) and \( \epsilon = R/D \) for \( \rho \) is given by

\[ \bar{\rho}^{(2)}(2) \approx \frac{-3\pi \epsilon^3}{4} \left( \frac{2 - 5 \lambda^2}{-2 + 4 \log(\epsilon) - 4 \log(\lambda)} \right) \]

We compare this analytic result with the numerical approach in Fig. 2. A second method is to obtain analytic results by smearing the ring charge density over a very short cylinder, whose height \( h \) is small compared to its radius. Analytic results are in qualitative agreement with the cutoff expansion.

Quantum mechanical results: The quantum mechanical case at first glance seems similar to the classical fluid charge case. Although there is no self-interaction, the quantum particle has a kinetic energy which opposes localization, hence for low values of interaction strength quantum fluctuations should destroy the polarization. However for large values of interaction it is not a priori obvious whether the polarization pattern beats the symmetry.

Again we assume exactly one charge on each ring and only nearest neighbor Coulomb interaction. The thickness of each ring is much smaller than its radius so that
the only coordinate of the charge is the angular position. We decompose the wavefunction in each ring into a limited number of Fourier modes, \( \psi_i(\theta) = \sum_{n=-\infty}^{\infty} c_n e^{i n \theta} \), and then solve the system numerically in the Hartree approximation. We use a dimensionless Hamiltonian for the \( i \)-th ring:

\[
\hat{h}_i = -\frac{\partial^2}{\partial \theta_i^2} + \delta \sum_j \int_0^{2\pi} \frac{|\psi_j(\theta')|^2}{|\vec{r}_j(\theta') - \vec{r}_i(\theta)|} d\theta' \quad (6)
\]

where the sum is over the nearest neighbors of \( i \)-th ring and \( \delta = (e^2/D) / (\hbar^2/2 m R^2) \) is the interaction strength and energy is measured in units of \( \hbar^2/2 m R^2 \). We impose periodic boundary conditions on the array and by an iterative self-consistent method we find that in a 1D array of rings for a finite value of \( \delta = \delta_c \) there is a change of ground state from totally unpolarized state \( |\vec{P}| = 0 \) to AFE state \( |\vec{P}| = P_0 \) where \( \vec{P} \) is the staggered polarization vector \( \vec{P} = \frac{1}{N} \sum_i (-1)^i \vec{r}_i(\theta) \psi_i(\theta) \) for 1D array. The ground state energy per ring:

\[
E = \frac{1}{N} \sum_i \int_0^{2\pi} \psi_i^*(\theta) \frac{\partial^2 \psi_i(\theta)}{\partial \theta_i^2} d\theta + \frac{\delta}{2N} \sum_{(i,j)} \int_0^{2\pi} \frac{|\psi_i(\theta)|^2 |\psi_j(\theta')|^2}{|\vec{r}_j(\theta') - \vec{r}_i(\theta)|} d\theta' d\theta' \quad (7)
\]

also shows a change in behavior at \( \delta_c \). Results of these calculations are plotted in Fig. 3.

We can obtain analytical insight by assuming that the ground state wave function for all rings in each sublattice is the same, so that we can employ a simple variational wave function for each sublattice \( \psi_A(\theta) = \frac{\sqrt{1-\frac{\pi}{2\pi} \cos(\theta-\phi)}}{\sqrt{2\pi}} + \frac{\sqrt{\frac{\pi}{2\pi} \cos(\theta-\phi)}}{\sqrt{2\pi}} \quad \psi_B(\theta) = \frac{\sqrt{1-\frac{\pi}{2\pi} \cos(\theta-\phi)}}{\sqrt{2\pi}} - \frac{\sqrt{\frac{\pi}{2\pi} \cos(\theta-\phi)}}{\sqrt{2\pi}} \quad \phi = \frac{\pi}{2} \)

alternatively. Inserting these functions into (7) and minimizing the energy using dipole expansion we find an approximate behavior for energy and polarization. In the ground state we find \( \phi = \frac{\pi}{2} \) and

\[
y(\delta, \epsilon) = \begin{cases} \frac{\epsilon}{2\sqrt{1-\frac{\pi}{2\pi} \cos(\theta-\phi)}} & \text{for } \delta \geq \delta_c(\epsilon) = \frac{1}{4} \epsilon^{-2} \\
0 & \text{for } \delta \leq \delta_c \end{cases} \quad (8)
\]

The numerical Hartree results for the energy and polarization are in agreement with the variational calculation when we restrict the number of Fourier modes in the numerical calculation to \( n \in \{ -1, 0, 1 \} \). The results are little changed when more Fourier modes are introduced to the numerical calculation or even when the exact Coulomb interaction is used in our numerical code.

Using the same Hamiltonian as eqn (6) for a 2D array with periodic boundary conditions we are able to find the behavior of the ground state energy and staggered polarization vector as a function of interaction strength. Again by using limited number of Fourier modes for each wave function we find that there is a change of ground state from totally unpolarized state to a striped AFE state in which each column has FE polarization but in opposite direction of its neighbor column. The staggered polarization vector for this vertically oriented case is expressed as \( \vec{P} = \sum_{i,j} (-1)^j \vec{P}_{ij}/N \) where \( j \) counts the columns and \( \vec{P}_{ij} = \int \vec{r}_{ij}(\theta) |\psi_{ij}(\theta)|^2 d\theta \) is polarization vector of each ring. The ground state is four-fold degenerate (the states obtained by flipping every spin and or rotating them all by \( \pi/2 \) yield the same energy) and the U(1) symmetry of unpolarized state has been broken.

Again it is possible to estimate a variational wave function for each ring assuming the wave functions of the rings in each sublattice are the same (infinite system or periodic boundary condition). Using dipole approximation...
where we assume that we can cast it in that form only at very low temperatures. The cos 2θ term breaks the XY-symmetry so that at low temperatures the system is Ising. The kinetic energy term tries to delocalize the spin, similar to the transverse field Ising model. However, the Coulomb interaction is not precisely an AFE Ising interaction; it is a difference of cos (θ − θ′) and cos (θ + θ′) terms that we can cast it in that form only at very low temperatures where we assume that < θ > is sharp on each ring.

In 2D there should be a finite temperature phase transition between the polarized and unpolarized state. While this Letter focuses only on the zero temperature behavior, Monte Carlo simulations of the 1+1D quantum problem not only support the existence of a 1D quantum phase transition, they document the existence of a 2D transition at finite temperature.

Considerations for experiment: The parameter δ determines if the array will spontaneously polarize; in 1D the transition is at δ ≈ 10, while in 2D, it is at δ ≈ 5. It is easy to achieve small values of δ simply by choosing the ring separation to be large. Thus the “quantum” limit where the kinetic energy dominates is simple to obtain. To obtain the antiferroelectrically ordered state we need large δ. We may write this as

\[ \delta = (R^2 / a_0 D) \times (m^* / m) \]

where \( a_0 \) is the Bohr radius and \( m^* \) is the effective mass of the electron. If \( R \) and \( D \) are \( R \) and \( D \) measured in nanometers, and \( m \equiv (m^* / m) \), then \( \delta \approx 18.9 (R^2 / D) \bar{m} \). We require that the rings do not intersect, so that \( \bar{D} \geq 2 \bar{R} \). Thus the ability to achieve large values of δ in semiconductors will depend upon the value of the effective mass. If we set \( \bar{D} = 2 \bar{R} \), then for GaAs (\( \bar{m} = 0.06 \)) 1D arrays of rings with a radius greater than \~10nm will be polarized. For AlAs (\( \bar{m} = 0.4 \)) the crossover radius is about 70nm. Rings with a smaller radius will not spontaneously polarize, but instead be isotropic.

It is well known that in 1D there is no ordered state for \( T > 0 \) for the Ising model. However, this is true in the thermodynamic limit. For small arrays over finite time intervals the system can order. If we wish to observe this behavior we want the characteristic energies of the system to be greater than the temperature. For the Coulomb energy \( kT < e^2 / D \), which we may write as \( DT < 1.8 \times 10^3 \) were \( D \) is measured in nm and \( T \) in Kelvin. For the kinetic energy this means \( kT < \hbar / 2m^* R^2 \); if we measure \( (m / m^*) R^2 T > 40 \) in the same units. For GaAs we can choose \( R \) to be about 14nm at 4K; choosing materials with a smaller effective mass or going to lower temperature allows us to increase the radius.

A AFE polarized ring array will scatter light at a wavelength commensurate with the inter-ring separation, \( D \). In 1D there is a gap \( \sqrt{2} \omega \), which we may write as \( 2 \sqrt{2} \omega (a_0 / D)^{3/2} \). For GaAs rings with a separation \( D = 1000nm \) this gives \( \omega \sim 6.0 \times 10^{10}Hz \). The 2D arrays have a similar sized gap at zone center, but the gap vanishes at one zone edge. The excitation spectrum can be probed optically, but scattering at the edge of the zone is difficult due to the constraints imposed by conservation of energy and momentum. Typically in such cases Raman scattering can be used to investigate the excitations.

Finally these calculations assume that each ring is singly occupied. This might be obtained by fabricating the rings upon a thin insulating layer covering a gate. By tuning the gate voltage we can bias the system so that it is energetically favorable for an electron to tunnel to the rings. The gate will also serve to cutoff long distance interactions between the rings, supporting the assumption of the nearest neighbor interactions used here. Moreover, this letter serves to start investigation into a broad class of problems, such as rings occupied by an optically excited exciton/hole pair or perhaps by a small, varying number of electrons created by a random distribution of dopants.

The topic of quantum dot arrays and their correlations has obvious and useful analogies with solid state models of crystalline arrays of atoms. In this Letter we wish to point out that experimentalists have at their disposal a host of "unnatural atoms" analogs: rings, quantum dot quantum wells, quantum rice, etc. The electrons in these...
nanoscale constituents are confined to orbitals that may not have atomic analogs. Moreover, it may be possible to tune the shape of the constituent to optimize some desired collective property such as frustration in electric or magnetic polarization, high susceptibility or sensitivity to optical polarization of light. Even more rich behavior will develop if we allow electrons to tunnel between these nanoscale periodic structures.

In this Letter we have examined the simple case of arrays of singly charged classical and quantum rings at zero temperature. We have found AFE order in 1D classical systems and a transition to 1D AFE order in the 1D quantum systems. In 2D classical and quantum systems there is a similar striped ordering. The ordered phases have collective excitations that can be measured optically.

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