A statistical model of quantum dot arrays with Coulomb coupling

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We present a study of a statistical model of arrays of quantum dots, in which electrons are confined by semiconductor heterojunctions in all three dimensions, with Coulomb coupling. Our model describes repulsively interacting localized electrons whose number can vary with changes in chemical potential. By means of a Monte Carlo simulation, it is shown that, at low temperatures, some stable electron densities hardly changing in certain ranges of chemical potential appear, which are associated with suppressed fluctuation and characteristic spatial electron distributions. Spin fluctuation, which gives magnetic susceptibility, exhibits essentially different behavior depending on temperature and Coulomb energies. © *1995 American Institute of Physics.*

I. INTRODUCTION

Various phenomena taking place in artificial low-dimensional quantum structures such as quantum wires and quantum dots, which are realized by virtue of new technologies, have been the subject of extensive studies and the possibility of applying them to devices has been suggested.[1] Since coupled quantum dot arrays created by a grid potential imposed on a two-dimensional electron gas, also called lateral surface superlattices, are in some aspects different from ordinary superlattices, they have been of interest and their novel properties have been demonstrated.[2,3] On the other hand, by means of a semiconductor heterojunction, we can construct quantum dots in which electrons are confined by heterojunctions in all three dimensions. Since the confinement of electrons in these dots is quite strong and the size of them can be reduced, it will be possible to fabricate quantum dot arrays in which the dots are very close together and Coulomb coupling between the dots plays an important role. In this article, we discuss some aspects of the quantum dot arrays with Coulomb coupling. In particular, considering that the number of electrons in the quantum dot array can be modulated via an applied voltage by means of a tunnel capacitor structure,[4–6] we investigate a statistical model of the quantum dot array in which the number of electrons can vary by changing the chemical potential.

II. MODELING

We assume a tunnel capacitor to consist of an electron supply layer, a tunnel barrier, a two-dimensional quantum dot array layer, a thick barrier layer, and an electrode (see Fig. 1). This structure allows us to change the number of electrons in the quantum dot array layer; an applied voltage between the electron supply layer and the electrode changes the energy difference between the confined energy levels of the quantum dots and the Fermi energy of the electron supply layer, i.e., it changes the chemical potential of the quantum dot array layer, and electrons can enter and exit the array layer through the tunnel barrier. For a single quantum dot, modulation of the electron number in the dot is observed by capacitance spectroscopy and energy levels in the dot have been determined.[5,6] In the quantum dot array, it is assumed that both the size of dots and the spacing between adjacent dots are $\lesssim 10$ nm. When such a system is fabricated, deviation in the size of the dots is inevitable. Assuming 10 nm cubed GaAs dots embedded in AlGaAs, the confined ground-state energy levels measured from the edge of the conduction band for bulk GaAs are approximately 10^2 meV. If the size of the dots has a deviation of several % (deviation of several Å), these energies have a deviation of several meV. Since the transfer energy between two dots is about 10^{-2} meV assuming the spacing is 5 nm and about 10^{-5} meV assuming the spacing is 10 nm, the transfer energies are much smaller than the deviation of the energy levels estimated above and the wave functions of electrons tend to be localized at each dot due to the effects of randomness. On the other hand, neglecting the screening effect, the intradot Coulomb energy and the interdot Coulomb energy for adjacent dots are approximately 10 meV and several meV, respectively. Thus, we can expect that the Coulomb coupling will play an important role in the system. Moreover, modulating the screening effect by controlling the position of the positive charge layer, the Coulomb coupling may be tuned.

In order to investigate the above situation, we employ a finite temperature Monte Carlo simulation of a model of repulsively interacting electrons represented by the following Hamiltonian:

$$\mathcal{H}=\sum_i \epsilon_i(n_i^+ + n_i^-)+\sum_i U_i n_i^+ n_i^- + \frac{1}{2}\sum_{i\neq j} V_{ij}(n_i^+ + n_i^-)(n_j^+ + n_j^-), \tag{1}$$

where the dynamical variable $n_i^+(n_i^-)$ is assumed to be 0 or 1, which denotes the number of up (down) spin electrons in the ith dot. The parameters ϵ_i and $U_i \geq 0$ denote the quantized energy level and the intradot Coulomb energy of the ith dot, respectively. The interdot Coulomb energy between the ith dot and jth dot is represented by $V_{ij}=V_{ji}\geq 0$. In this model, electrons are assumed to be localized at each dot and we have taken into account only one bound state localized at each dot, which has twofold degeneracy with respect to the freedom of spin. This is justified when the dot size is sufficiently small, because of a large separation between the

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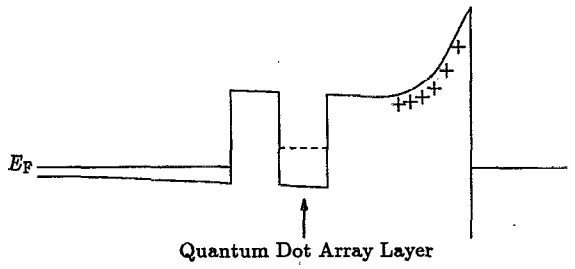

FIG. 1. Conduction band profile of the tunnel capacitor structure. E_F is the Fermi level in the electron supply layer.

ground-state energy and an excited-state energy or the absence of excited states. The total number of electrons $N=\Sigma_i(n_i^+ + n_i^-)$ can vary with change in the chemical potential and the statistical properties of the model are determined by the grand partition function $Z_G = \Sigma_{n_i^\mp=0,1}\exp[-\beta(\mathcal{H}-\mu N)]$, where β and μ are the inverse temperature and the chemical potential, respectively.

In this model, we are interested in the behavior of the statistical average of the electron density, i.e., the electron number per dot, $\rho=\langle N\rangle/N_{\rm dot}=\langle\Sigma_i(n_i^+ + n_i^-)\rangle/N_{\rm dot}$, where $\langle\cdot\rangle$ denotes the statistical average and $N_{\rm dot}$ is the total number of dots. Fluctuation of the electron density is given by

$$\sqrt{\langle(N/N_{\rm dot}-\langle N\rangle/N_{\rm dot})^2\rangle}=\sqrt{\partial\rho/\partial(\beta\mu)}/\sqrt{N_{\rm dot}},$$

in which the factor $1/\sqrt{N_{\rm dot}}$ arises from the law of large numbers. We define the electron density fluctuation $\delta\rho=\sqrt{\partial\rho/\partial(\beta\mu)}=\sqrt{\langle(N-\langle N\rangle)^2\rangle}/N_{\rm dot}$, which is related to the capacitance of the system and from which the factor from the law of large number is divided. Since our model has no spin interaction, the statistical average of the magnetization of the system $M=\Sigma_i(n_i^+ - n_i^-)$ always vanishes. However, the fluctuation of the magnetization, which gives the magnetic susceptibility, exhibits a characteristic behavior. Thus we define the spin fluctuation $\delta m = \sqrt{\langle(M-\langle M\rangle)^2\rangle}/N_{\rm dot}$, from which the factor from the law of large numbers is also divided.

III. SIMULATION

Hereafter, we restrict our attention to the case that electron–electron interaction is short range; only the intradot Coulomb energy with the same value U and the interdot Coulomb energy for nearest neighbor dots with the same value V are taken into account. Similar situations were considered in order to investigate some materials with a fixed number of electrons.[7] However, we are interested in a system in which the number of electrons can be changed by the chemical potential; this situation can be realized in the quantum dot arrays. Figure 2 shows the behavior of the electron density and its fluctuation as the chemical potential changes for square-lattice arrays of quantum dots. The energy levels of the dots are assumed to randomly distribute obeying Gaussian distribution whose average is zero (without loss of generality) and standard deviation is denoted by $\delta\epsilon$. We use ten random samples of 36×36 square lattices with the periodic boundary condition and thermal averages is again averaged for the samples. The parameter is taken to be $\beta U=20.0$, 10.0, 5.0, and $V/U=0.2$, 0.3, and $\delta\epsilon/U=0.1$. If the intradot Coulomb energy is assumed to be 10 meV, $\beta U=20.0$, 10.0, and 5.0 correspond to temperatures of about 6,

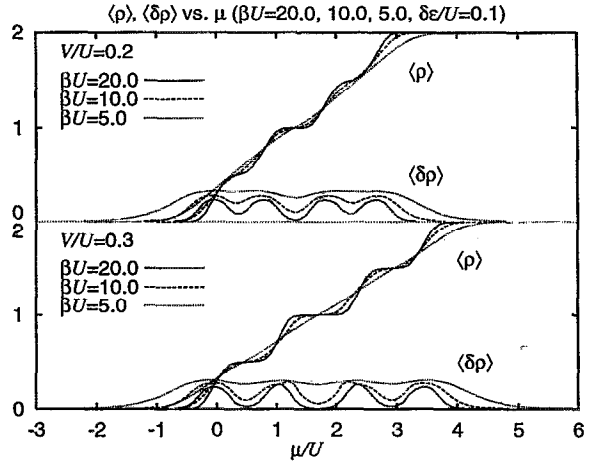

FIG. 2. The behavior of the electron density and its fluctuation as functions of the chemical potential. At low temperatures, there exist some plateaus in the electron density with suppressed fluctuation. For $V/U=0.2$ and $V/U=0.3$, a very similar behavior is seen.

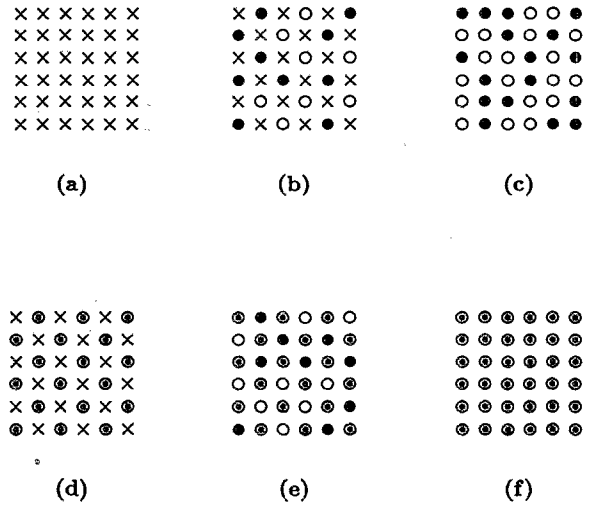

FIG. 3. Various characteristic spatial electron distributions. Open and closed circles denote quantum dots occupied by up- and down-spin electrons, respectively. A quantum dot occupied by two electrons (both up and down spin) is represented by an open circle with a closed circle inside. An unoccupied quantum dot is represented by a cross. (a) Every dot is unoccupied by electrons. Both the electron density and the spin fluctuation vanish. (b) A dot occupied by one electron and an unoccupied dot alternate. The electron density equals a half and the spin fluctuation equals $1/\sqrt{2}$. (c) Every dot is occupied by one electron. The electron density equals unity and the spin fluctuation takes the maximum value of unity. (d) A dot occupied by two electrons and an unoccupied dot alternate. The electron density equals unity and the spin fluctuation vanishes. (e) A dot occupied by one electron and a dot occupied by two electrons alternate. The electron density equals three halves and the spin fluctuation equals $1/\sqrt{2}$. (f) Every dot is occupied by two electrons. The electron density equals two and the spin fluctuation vanishes.

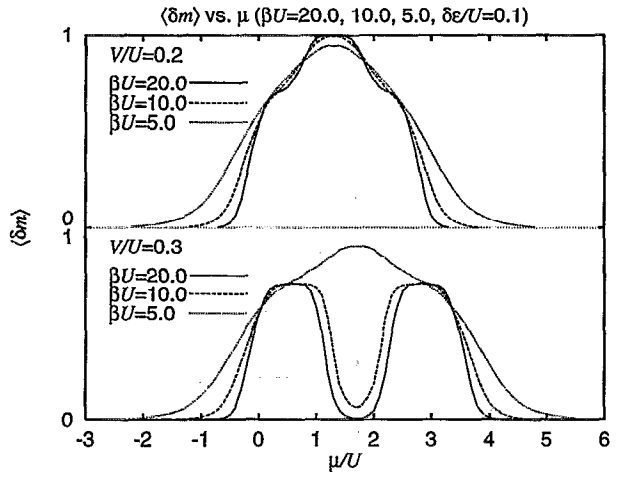

FIG. 4. The behavior of the spin fluctuation as a function of the chemical potential. For $V/U=0.2$ and $V/U=0.3$, a remarkably different behavior is seen at low temperatures. At high temperatures this difference vanishes.

12, and 24 K, respectively, and $\delta\epsilon/U=0.1$ corresponds to a deviation of the quantized energies of 1 meV. In the regions of very low and very high chemical potential, the electron density vanishes corresponding to Fig. 3(a), and equals two corresponding to Fig. 3(f), respectively. In the intermediate region, electron density plateaus exist at low temperatures ($\beta U=20.0$, 10.0), namely one half, unity, and three halves, which is especially clear in the case of lower temperatures ($\beta U=20.0$). The behavior of the electron density in the case of $V/U=0.2$ and 0.3 is very similar. There is, however, a distinction between them, which is elucidated from the behavior of the spin fluctuation, which is shown in Fig. 4. In the plateaus of the electron density of one half and three halves, the spin fluctuation is approximately $1/\sqrt{2}$. In fact, in the one-half and three-halves plateau, the probability of the appearance of the distributions shown in Figs. 3(b) and 3(e) is very large, respectively. The spin fluctuation of both these distributions is $1/\sqrt{2}$. In contrast, in the plateau of the electron density of unity, the spin fluctuation is unity for the case of $V/U=0.2$, and suppressed for $V/U=0.3$. For the case of $V/U=0.2$, the intradot Coulomb interaction dominates. As a result, in the unity plateau, the probability of the appearance of the distribution shown in Fig. 3(c), in which the spin fluctuation takes the maximum value unity, is very large. On the other hand, for $V/U=0.3$, the interdot interaction is dominant. Thus, in the unity plateau, the probability of the appearance of the distribution shown in Fig. 3(d), in which the spin fluctuation vanishes, is very large. In the result of our simulation, although the spin fluctuation does not completely vanish, suppression of the spin fluctuation at low temperatures is seen. This effect can appear for bipartite lattices with the nearest neighbor Coulomb interaction. At very low temperatures, whether the intra- or interdot interaction is dominant is determined by the relation between V/U and the number of nearest neighbors z ($z=4$ for the square lattice); if $V/U<1/z$, intradot interaction dominates, and if $V/U>1/z$, interdot interaction dominates. Our simulation demonstrates that, at high temperatures ($\beta U=5.0$), this difference disappears. Notice that the distributions shown in Figs. 3(c) and 3(d) have very different entropies; the one shown in Fig. 3(c) has entropy $k_B \log 2$ per dot due to the freedom of spin, and the other shown in Fig. 3(d) has no entropy. Therefore, the latter is rather weak in thermal fluctuation and, at high temperatures, even if $V/U>1/z$, the probability of the appearance of the distribution shown in Fig. 3(c) can be rather large due to its entropy.

IV. CONCLUSION

As discussed above, we can expect, for a quantum dot array with Coulomb coupling at low temperatures, some stable electron densities associated with suppressed fluctuation and a large probability of the appearance of characteristic spatial electron distributions, even in the presence of randomness. This can be viewed as many-valued stability taking place in the system. In order to suppress the fluctuation, conventional electron devices make use of the law of large numbers. However, the suppressed fluctuation in the system is due to a kind of charging effect. If we make use of this many-valued stability, we can suppress the fluctuation without the law of large numbers, i.e., even if $N_{\rm dot}$ is not so large. This many-valued stability will be useful for new electron devices. Similar effects of suppressed fluctuation due to a charging effect were experimentally investigated in the early days[4] and also more recently.[5,6,8] However, there is a distinction between them and our case. In our cases not only the intradot Coulomb interaction but also the interdot one, which induces characteristics spatial distributions, is important.

The effect of spin fluctuation discussed above is similar to the Kubo effect,[9] which takes place in independent fine metallic particles due to the charging effect. Arranging Coulomb coupled semiconductor quantum dots using heterojunctions, the new effects related to chemical potential, Coulomb energies, and temperature should be observed.

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