Three-electron bunches in occupation of a Coulomb cluster with $N = 5$ sites

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Attraction of like charges in a localized system implies that, upon increasing the Fermi energy, the occupation of the system changes as $n \to (n+2)$, while the occupation, $(n+1)$, is skipped. In this way, the attraction translates into the bunching of electrons. For a localized system of $N = 4$ sites, attraction of electrons manifests itself in skipping of $n = 2$ occupation. The origin of the attraction is the rearrangement of the occupations of the surrounding sites, which plays the role of a polaronic effect. We consider a Coulomb cluster with $N = 5$ sites, and we demonstrate that, with screened Coulomb repulsion, *three-electron* bunching becomes possible, i.e., the change of occupation $n = 1 \to n = 4$ with $n = 2$ and 3 occupations skipped.

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I. INTRODUCTION

The question of whether two electrons can attract each other without lattice dynamics being involved has been posed in many papers. Another formulation of this question is whether the formation of negative-U centers [1], which are believed to be due to phonons, is possible as a result of electron-electron repulsion. One of the appeals of having purely electronic negative-U centers is that such centers can be viewed as precursors of a purely electronic superconducting state.

On the theory side, electron attraction in repulsive systems was invoked to (i) explain the valence-skipping phenomenon (see, e.g., Refs. [2–6]), i.e., when the valence of certain elements does not occur in the compounds that they form; (ii) derive pairing within certain limits of the Hubbard model [7], which essentially amounts to replacing phonons by magnons [8]. Also, as demonstrated in Ref. [9], the formation of charge-2e excitations, i.e., "trimers," is favored in doped transition-metal dichalcogenide bilayers near half-filling. The origin of pairing in Ref. [9] with two types of sites having different energy is the reduction of *electrostatic* energy, which dominates over kinetic energy. A minimal model of a trimer requires four sites. Note that such a four-site model of pairing was previously studied in Ref. [10] in a different relation.

On the experimental side, early experiments brought attention to the issue of the possible attraction of localized electrons [11–13]. These experiments addressed one-by-one magnetotunneling events of electrons from an electrode into a big semiconductor island. A certain portion of events revealed bunching of electrons into pairs. There is a problem with attributing this bunching to the attraction of electrons within a four-site model because incoherent tunneling of two electrons takes too much time. Later experiments [14,15] suggested that two-electron events take place at the edge and are related to the formation of the edge states in magnetic field.

Demonstration of pairing due to repulsion on a truly microscopic level was reported in Ref. [16]. The experimental setup in Ref. [16] was very similar to the four-site cluster (two-site polarizer and double-well quantum dot). In a certain domain of gate voltages, one dot of a double-well system was either empty or accommodated a pair of electrons as a result of reoccupation of the dots constituting the polarizer.

Due to the flexibility of their nanotube-based technique, the authors of Ref. [16] suggested several variants for scaling-up their setup. In particular, their approach can tackle the question of whether more complex many-particle processes can be realized experimentally.

In the present paper, we demonstrate that adding one extra site to the four-site model opens the possibility of *three-electron* bunches. To establish a criterion for such three-electron bunches, we introduce E_N^n, which is a minimal energy of n electrons in a cluster of N sites. The next electron enters the cluster when the Fermi level position in the surrounding system is equal to

$$\mu_1 = E_N^{n+1} - E_N^n. \tag{1}$$

Two electrons enter the cluster at

$$\mu_2 = \frac{E_N^{n+2} - E_N^n}{2}. \tag{2}$$

The formation of a 2e pair takes place when $\mu_2 < \mu_1$, i.e., when the usual condition $(E_N^{n+2} + E_N^n) < 2E_N^{n+1}$, illustrated in Fig. 1, is met. Continuing this reasoning, three electrons enter the cluster at

$$\mu_3 = \frac{E_N^{n+3} - E_N^n}{3}. \tag{3}$$

Then a 3e-bunch is favored over a single electron, and a 2e-bunch is favored under the conditions $\mu_3 < \mu_1$ and $\mu_3 < \mu_2$.

II. TWO-ELECTRON BUNCHING IN A FOUR-SITE CLUSTER

For didactic reasons, in this section we review the steps unveiling the two-electron bunching in the four-site model. An extension to the five-site model involves all the same steps.

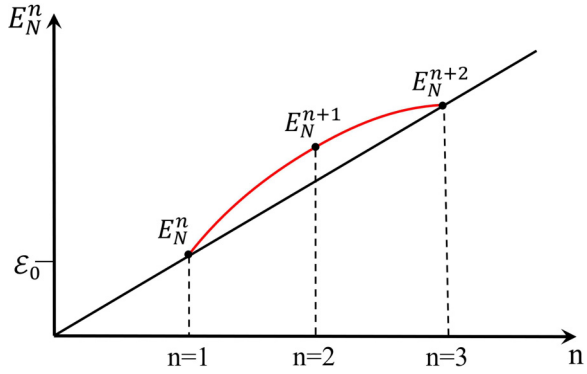

FIG. 1. A criterion, $E_N^n + E_N^{n+2} < 2E_N^{n+1}$, of the attraction of two electrons implies a concavity of the curve E_N^n. As a result, the population evolves as $n \to (n+2)$.

On the other hand, the flow of logics offers an insight about possible extensions.

Step I. A four-site cluster is illustrated in Fig. 2(a). We examine the evolution of the minimal energy of configurations with varying n. The four-site cluster is completely filled when $n = 4$, i.e., only one configuration is possible. It is also obvious that, for $n = 1$, the minimal energy corresponds to filling the central site with energy $\varepsilon_0 < \varepsilon_1$. Thus $E_4^1 = \varepsilon_0$. When $n = 2$, the electron in the center can either stay there or move to the vertex in order to reduce the potential energy. Thus, the candidates for E_4^2 are

$$\tilde{E}_4^2 = \varepsilon_0 + \varepsilon_1 + W \text{ or } \tilde{\tilde{E}}_4^2 = 2\varepsilon_1 + V. \tag{4}$$

Equally, there are two candidates to minimize the energy when the population of the cluster is $n = 3$, namely

$$\tilde{E}_4^3 = \varepsilon_0 + 2\varepsilon_1 + 2W + V \text{ or } \tilde{\tilde{E}}_4^3 = 3\varepsilon_1 + 3V. \tag{5}$$

Step II. At this step, we make two crucial assumptions: $\tilde{E}_4^2 < \tilde{\tilde{E}}_4^2$ and $\tilde{E}_4^3 > \tilde{\tilde{E}}_4^3$, which translate into the following inequalities:

$$\begin{aligned} 2\varepsilon_1 + V &> \varepsilon_0 + \varepsilon_1 + W, \\ 3\varepsilon_1 + 3V &< \varepsilon_0 + 2\varepsilon_1 + V + 2W. \end{aligned} \tag{6}$$

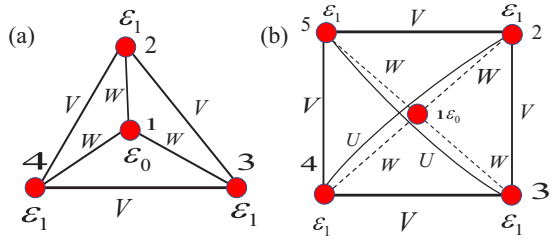

FIG. 2. (a) Illustration of the four-site model. The energy of the central site, ε_0, is smaller than the energies of the corner sites, ε_1. For a purely Coulomb system, the repulsions W and V are related as $3^{1/2}$. For a gate-screened interaction, they are related as $3^{3/2}$. (b) Illustration of the five-site model. The repulsions W, U, and V are related as $2 : 2^{1/2} : 1$ for a purely Coulomb interaction, and as $2^3 : 2^{3/2} : 1$ for the gate-screened interaction.

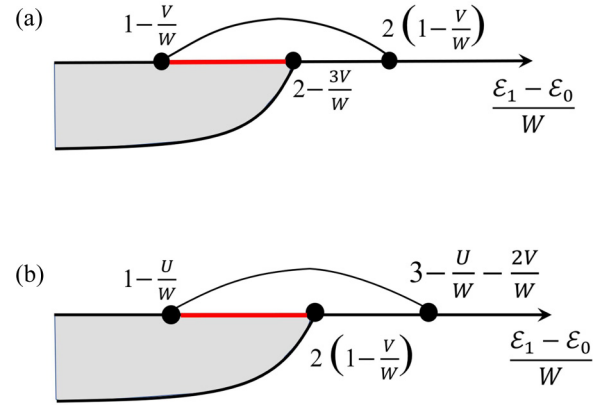

FIG. 3. Different domains of the dimensionless imbalance, $\frac{\varepsilon_1-\varepsilon_0}{W}$, of the site energies: (a) Four-site model. In the domain of imbalances, $\{1-\frac{V}{W}, 2(1-\frac{V}{W})\}$, the energy of the two-electron configuration is minimal when one electron occupies the central site, while for the three-electron configuration the energy is minimal when all three electrons reside in the vertices. In the gray region, the condition of concavity, illustrated in Fig. 1, is satisfied, so that 2e-pairing occurs in the domain marked with red. (b) Five-site model. In the red domain, the occupation of the cluster with chemical potential evolves as $n = 1 \to n = 4$.

These inequalities imply that it is energetically unfavorable for two electrons to vacate the center, but it is favorable for three electrons to vacate the center. Note that Eq. (6) can be presented in a concise form,

$$1 - \frac{V}{W} < \frac{\varepsilon_1 - \varepsilon_0}{W} < 2\left(1 - \frac{V}{W}\right). \tag{7}$$

When the conditions Eq. (6) are met, we have

$$E_4^2 = \tilde{E}_4^2, \; E_4^3 = \tilde{\tilde{E}}_4^3. \tag{8}$$

Step III. At this step, we require that the dependence E_4^n is "concave," as illustrated in Fig. 1. Using Eq. (8), the pairing condition $E_4^1 + E_4^3 < 2E_4^2$ takes the form

$$\varepsilon_0 + 3(\varepsilon_1 + V) < 2(\varepsilon_0 + \varepsilon_1 + W). \tag{9}$$

Note that the above condition restricts the energy difference between the center and the vertex sites: $\frac{\varepsilon_1-\varepsilon_0}{W} < 2 - \frac{3V}{W}$.

Step IV. We now go back to the assumptions made above, Eq. (7), and test whether they are consistent with concavity. This test is illustrated in Fig. 3. We see that the second inequality is satisfied automatically. To satisfy the first inequality, one needs $V < \frac{W}{2}$. As seen from Fig. 2(a), for purely Coulomb interaction, the relation between V and W is $V = \frac{W}{3^{1/2}}$. Thus, the first condition is not satisfied. It can be satisfied, however, if the gate is present at a distance, d, above the plane of the cluster. Then the Coulomb interaction, $V(\rho) = \frac{e^2}{\rho}$, gets modified to $V(\rho) = e^2[\frac{1}{\rho} - \frac{1}{(\rho^2+4d^2)^{1/2}}]$. Then, for $\rho \gg d$, we have $V = \frac{W}{3^{3/2}} < \frac{W}{2}$, so that in the domain

$$W - V < \varepsilon_1 - \varepsilon_0 < 2W - 3V, \tag{10}$$

shown in Fig. 3(a) with red, 2e-pairing is possible.

III. THREE-ELECTRON BUNCHING IN A FIVE-SITE CLUSTER

We now generalize the reasoning from the previous section to the five-site model illustrated in Fig. 2(b). The central site is surrounded by four vertexes. Three repulsion energies, shown in the figure, are related as $U < V < W$ in accordance with distances between the corresponding sites. Similarly to the four-site model, we assume that the energy, ε_0, of the central site is lower than ε_1, the energy of the vertex sites. Obviously, $E_5^1 = \varepsilon_0$. Our strategy in a search for three-electron bunching is based on the argument that, by placing electrons in the vertexes, the configuration loses its on-site energy, but gains due to the reduction in potential energy.

Step I. There are three candidates for the double occupation of the cluster. They are

$$\tilde{E}_5^2 = \varepsilon_0 + \varepsilon_1 + W \text{ or } \tilde{\tilde{E}}_5^2 = 2\varepsilon_1 + V \text{ or } (\tilde{\tilde{E}})_5^2 = 2\varepsilon_1 + U. \tag{11}$$

Since $V > U$, the energy $\tilde{E}_5^2$ is smaller than $\tilde{\tilde{E}}_5^2$, leaving us with only two candidates. One can also see that there are three candidates for the triple occupation, namely

$$\tilde{E}_5^3 = 3\varepsilon_1 + 2V + U \text{ or } \tilde{\tilde{E}}_5^3 = 2\varepsilon_1 + \varepsilon_0 + 2W + U, \text{ or}$$
$$(\tilde{\tilde{E}})_5^3 = 2\varepsilon_1 + \varepsilon_0 + 2W + V. \tag{12}$$

We can now compare $\tilde{\tilde{E}}_5^3$ to $(\tilde{\tilde{E}})_5^3$ and realize that the latter has higher energy since $V > U$. This again leaves us with two candidates for $\tilde{E}_5^3$.

Finally, the occupation of the cluster with four electrons is possible in two configurations having different energies. These energies are

$$\tilde{E}_5^4 = \varepsilon_0 + 3\varepsilon_1 + 3W + 2V + U \text{ or } \tilde{\tilde{E}}_5^4 = 4\varepsilon_1 + 4V + 2U. \tag{13}$$

Obviously, the state with energy $\tilde{E}_5^4$ is fourfold-degenerate depending on which vertex is empty.

Step II. At this point, we make three crucial assumptions:

(1) $\tilde{\tilde{E}}_5^2 > \tilde{E}_5^2$,

(2) $\tilde{\tilde{E}}_5^3 > \tilde{E}_5^3$,

(3) $\tilde{\tilde{E}}_5^4 < \tilde{E}_5^4$.

The assumptions are made in order to ensure that the ground state with $n = 2$ includes the center site, while the ground states with $n = 3$ and 4 include the sites in the vertexes. Using Eqs. (11), (12), and (13), the above assumptions can be rewritten as

$$\varepsilon_0 + \varepsilon_1 + W < 2\varepsilon_1 + U \Rightarrow 1 - \frac{U}{W} < \frac{\varepsilon_1 - \varepsilon_0}{W}, \tag{14}$$

$$\varepsilon_0 + 2\varepsilon_1 + 2W + U > 3\varepsilon_1 + 2V + U$$
$$\Rightarrow 2\left(1 - \frac{V}{W}\right) > \frac{\varepsilon_1 - \varepsilon_0}{W}, \tag{15}$$

$$\varepsilon_0 + 3\varepsilon_1 + 3W + 2V + U > 4\varepsilon_1 + 4V + 2U$$
$$\Rightarrow 3 - \frac{2V}{W} - \frac{U}{W} > \frac{\varepsilon_1 - \varepsilon_0}{W}. \tag{16}$$

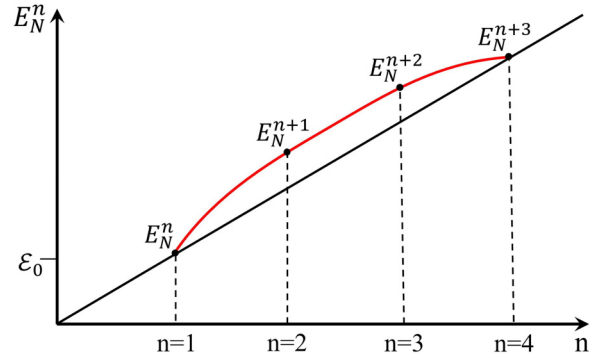

FIG. 4. Illustration of three-electron bunching in the cluster of N sites. The bunch is possible under the conditions $\frac{1}{3}(E_N^{n+3} - E_N^n) < \frac{1}{2}(E_N^{n+2} - E_N^n) < E_N^{n+1} - E_N^n$.

Now it is convenient to restructure the above inequalities into two separate ranges as

$$1 - \frac{U}{W} < \frac{\varepsilon_1 - \varepsilon_0}{W} < 2\left(1 - \frac{V}{W}\right),$$
$$1 - \frac{U}{W} < \frac{\varepsilon_1 - \varepsilon_0}{W} < 3 - 2\frac{V}{W} - \frac{U}{W}. \tag{17}$$

Whether the three-electron bunches are allowed or not depends on whether or not the domains Eq. (17) overlap, as illustrated in Fig. 3(b). After making the above assumptions, we can specify the ground-state configuration for each n, namely

$$E_5^2 = \tilde{E}_5^2,\ E_5^3 = \tilde{E}_5^3,\ E_5^4 = \tilde{\tilde{E}}_5^4. \tag{18}$$

Step III. We now require that the ground-state energies E_5^n are arranged as shown in Fig. 4, or, in other words, we require that these energies satisfy the conditions

$$\tfrac{1}{3}\left(E_5^4 - E_5^1\right) < \tfrac{1}{2}\left(E_5^3 - E_5^1\right) < E_5^2 - E_5^1. \tag{19}$$

The first and the second conditions can be cast into a traditional form

$$\frac{\varepsilon_1 - \varepsilon_0}{W} > 2\frac{V}{W} + \frac{U}{W},$$
$$\frac{\varepsilon_1 - \varepsilon_0}{W} < 2 - 2\frac{V}{W} - \frac{U}{W}. \tag{20}$$

We see that, in the same way as Eq. (17), the necessary requirements for 3e bunches restrict the asymmetry in single-electron energies both from below and above.

Step IV. Now a nontrivial question arises: is there a domain in which the conditions Eqs. (17) and (20) are consistent with each other? It is apparent that for purely Coulomb repulsion, the requirements Eq. (20) cannot be met. Indeed, with purely Coulomb interaction, one has $\frac{U}{W} = \frac{1}{2}$, while $\frac{V}{W} = \frac{1}{2^{1/2}}$. Then the right-hand side in the second inequality Eq. (20) requires that the asymmetry, $\frac{\varepsilon_1 - \varepsilon_0}{W}$, exceeds 1.54, while the second inequality requires that this asymmetry is smaller than 0.46. Turning to screened Coulomb repulsion, we have $\frac{U}{W} = \frac{1}{8}$, while $\frac{V}{W} = \frac{1}{2^{3/2}}$. Then Eq. (20) restricts the asymmetry to the interval (0.83, 1.17). At the same time, Eq. (17) restricts this interval to (0.875, 1.29). We see that two restricting intervals overlap proving that 3e-bunches are allowed.

Turning to the general form of the electron-electron, we compare the right-hand sides of Eqs. (17) and (20). This comparison suggests that the most stringent upper limitation on the asymmetry of the on-site energies reads $\frac{\varepsilon_1-\varepsilon_0}{W} < 2-\frac{2V}{W}-\frac{U}{W}$. Assuming that the interaction of localized electrons falls off with distance as $r^{-\alpha}$, we find that the repulsions W, U, and V are related as $2^{\alpha}:2^{\frac{\alpha}{2}}:1$. In other words, the relation $\frac{U}{W}=(\frac{V}{W})^2$ holds for any α. We can now address the following question: how fast should the repulsion fall off with distance to allow for the 3e bunches? At critical α the upper bound on asymmetry coincides with the lower bound, $\frac{\varepsilon_1-\varepsilon_0}{W} > 2\frac{V}{W}+\frac{U}{W}$. Equating the two bounds, we find $\alpha=\alpha_c=\frac{\ln(3+2^{3/2})}{\ln 2}\approx 2.55$. In this regard, it is instructive to compare the conditions for 2e-bunching and 3e-bunching in the five-site model. The analysis of the criterion $(E_5^1+E_5^3)<2E_5^2$ for 2e-bunching leads to the restriction

$$2\frac{V}{W}-\frac{U}{W}<\frac{\varepsilon_1-\varepsilon_0}{W}<2-2\frac{V}{W}-\frac{U}{W}, \tag{21}$$

which yields the criterion $\alpha > 2$. Naturally, this criterion is "softer" than the criterion for 3e-bunching.

IV. CONCLUSION

The conditions for the formation of the 3e-bound state in the repulsive system are more restrictive than the conditions for the formation of the 2e-bound state. We do not know whether bunches containing more than three electrons are possible, but our finding motivates us to search for them. It is straightforward to expect that such a bunching manifests itself in the counting statistics of electron transfer; see, e.g., Refs. [17–19]. As a final remark, we note that, even in the absence of the gate, the modification of the Coulomb repulsion can take place as a result of polarization charges induced in a neighboring plane [20,21]. If the plane is separated from the localized sites by a distance, r_0, the Coulomb repulsion at distances $r \gg r_0$ is modified from $1/r$ to $1/r^3$. This rapid fall-off of the repulsion allows for 3e bunching.

In our study we have completely neglected the hopping between the sites. This neglect is applicable when the tunneling amplitude, t, is much smaller than the Coulomb repulsions U, V, and W. As t increases, the effect of bunching disappears. A general argument in favor of this disappearance is that the bunching relies on the discreteness of electrons. On the other hand, the allowance of tunneling between the sites tends to "homogenize" the electron density, and consequently suppresses the discreteness. The above argument is similar to the criterion for the formation of the Wigner crystal, i.e., the kinetic energy should be smaller than the potential energy. Finite t plays the role of kinetic energy.

Demonstration of bunching in a five-cite model suggests that, similar to the honeycomb lattice of transition-metal dichalcogenide [9], a phase-centered square lattice undergoes, at a certain filling, a transition into an inhomogeneous state.

As was mentioned in the Introduction, in experiment the bunching of electrons manifests itself in two setups. In the first setup, discrete jumps in the capacitance of a big semiconductor island [11–13] are observed upon gradual increase of the bias between the island and the gate. In the second setup, transport through a system of two quantum dots defined electrostatically in a carbon nanotube [16] is measured. Normally, this transport is characteristic of a Coulomb blockade. However, in the presence of a polarizer, represented by another nanotube crossing the bare nanotube and containing two additional dots between which an electron can hop, the picture of transport changes. Then the measured stability diagram indicates a crossover from interdot repulsion to interdot attraction. This suggests that occupation of the combined system of a nanotube plus a polarizer crosses from one to three. The most natural realization of the four-site model is the system [16] to which one dot is added.

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