Two-Electron State in a Disordered 2D Island: Pairing Caused by the Coulomb Repulsion

M. E. Raikh,[1] L. I. Glazman,[2] and L. E. Zhukov[1]
[1]*Physics Department, University of Utah, Salt Lake City, Utah 84112*
[2]*Theoretical Physics Institute and Department of Physics, University of Minnesota, Minneapolis, Minnesota 55455*
(Received 18 December 1995)

We show the existence of bound two-electron states in an almost depleted two-dimensional island. These two-electron states are carried by special compact configurations of four single-electron levels. The existence of these states does not require phonon mediation, and is facilitated by the disorder-induced potential relief and by the electron-electron *repulsion* only. The density of two-electron states is estimated and their evolution with the magnetic field is discussed. [S0031-9007(96)00822-8]

PACS numbers: 73.20.Dx, 73.40.Gk, 73.40.Sx

In a recent experiment [1,2] the tunneling of electrons from a semimetallic electrode into the localized states (LS's) in a quantum well was studied. By using the electron beam lithography, a dot with a diameter as small as 1 μm was formed. The small size of the dot enabled the authors to detect the individual tunneling acts. These acts manifested themselves as narrow peaks in the differential capacitance measured as a function of the bias applied across the structure. To introduce the LS's in the GaAs quantum well the neighboring AlGaAs region was doped with Si donors.

The authors identified the origin of LS's by studying the evolution of the peak positions with magnetic field, B, perpendicular to the well. They associated the peaks with electronic states of essentially two types: (i) Ground state in the cylindrically symmetric parabolic potential; the B dependence of the corresponding peaks above certain B approached the one for the lowest Landau level. Such a parabolic confinement presumably results from the fluctuations in the concentration of donors in the barrier. (ii) Bound state of an electron at a Si donor which could migrate into the well during the growth. For these states the increase of the energy with B was much slower than for the group (i).

There is a puzzling feature in the data reported in [1]: few of the peaks observed were twice as high as isolated one-electron peaks. This suggests that two electrons tunnel into the well at the same voltage applied. By tracing the B dependences of these peaks the authors have ruled out the possibility that they reflect accidental degeneracies in the energy positions of LS's in some distant minima. All double peaks retained their height within a certain range of B and then split into doublets at some critical value of the magnetic field. Such a behavior indicates that both LS's involved "feel" each other and, thus, are located close in space. On the other hand, it is apparent that two close potential minima cannot accommodate two electrons at the same bias. Even if the energy levels are degenerate, tunneling of one electron would elevate the level for the second electron, so that the subsequent tunneling will occur at the bias larger by the energy of the Coulomb repulsion. The authors mentioned that the physical mechanism which could resolve this paradox is the polaronic effect [3]. Their conjecture was further developed in [4], where the two-electron state in a hybrid hydrogenic-parabolic potential in the presence of electron-phonon interaction was considered. The pair-binding condition used in [4] implicitly assumed that the two electrons share the same lattice deformation, which leads to the enhancement of the polaronic effect. In fact, the polaronic shift per electron in the paired state used in [4] is twice the shift for a single localized electron. Under this assumption bound two-electron states were found even at weak electron-phonon interaction, provided that the distance between the hydrogenlike impurity and the center of the parabolic potential is larger than $8a_0$, where a_0 is the radius of the hydrogenic state. For such distances the enhanced polaronic shift overweighs the Coulomb repulsion. However, we find the underlying assumption hard to justify. Indeed, the spatial scale of the polaronic deformation coincides with the size of a single-electron state [5]; two distant electrons do not share the same deformation, and, therefore, the corresponding enhancement of the polaron shift is suppressed.

In the present paper we demonstrate that the double peaks observed in [1] can be explained without invoking the electron-phonon interactions. Our explanation is based exclusively on electrostatics. We assume that the electrons are strongly localized and neglect the overlap of their wave functions and, correspondingly, the exchange interaction. On the other hand, the modification of the Coulomb interaction between localized electrons due to the presence of an electrode plays an important role in our picture. We show that, in contrast to [4], the objects responsible for two-electron tunneling are the compact groups of LS's and calculate the relative portion of the double peaks.

First of all, let us establish the general criterion for two-electron tunneling. Consider a cluster of N LS's occupied by n electrons. The distribution of electrons over LS's corresponds to the *minimal* possible energy which we denote as E_N^n. The position of the Fermi level in the electrode, E_F^1, at which an additional electron will enter the cluster is determined from the condition

 0031-9007/96/77(7)/1354(4)$10.00

$E_F^1 + E_N^n = E_N^{n+1}$. If two electrons enter the cluster, the corresponding position of the Fermi level, E_F^2, satisfies the relation $2E_F^2 + E_N^n = E_N^{n+2}$. Double peak occurs if $E_F^2 < E_F^1$. This leads us to the following criterion:

$$E_N^{n+2} + E_N^n < 2E_N^{n+1}. \quad (1)$$

Obviously condition (1) cannot be satisfied if $N = 2$. Formally, if we denote the energies of two LS's as ε_1 and ε_2 so that $\varepsilon_1 < \varepsilon_2$, then $E_2^1 = \varepsilon_1$ and $E_2^2 = \varepsilon_1 + \varepsilon_2 + V_{12}$, V_{12} being the interaction energy of two electrons occupying the first and the second LS's. We see that $E_2^2 > 2E_2^1$—the inequality opposite to (1).

Now we will prove that for $N = 3$ the occurrence of a double peak is also forbidden by Eq. (1). By analogy to the consideration above, the case $n = 0$ is obvious for any N since a single electron on a cluster will occupy the LS with the lowest energy level, so that the condition (1) is violated even without the Coulomb repulsion. Thus the only case to be considered is $n = 1$. Let us again order the energies of LS's: $\varepsilon_1 < \varepsilon_2 < \varepsilon_3$. Then $E_3^1 = \varepsilon_1$, and $E_3^3 = \varepsilon_1 + \varepsilon_2 + \varepsilon_3 + V_{12} + V_{13} + V_{23}$. Now there are three candidates for E_3^2 in accordance with three variants of occupation of $N = 3$ cluster by two electrons. It is important that E_3^2 is the *minimal* of these three energies. This means that if the double peak is possible, the condition (1) should be met when we substitute for E_3^2 *each* of these energies. Let us choose two of the three candidates for E_3^2, namely, $\varepsilon_1 + \varepsilon_2 + V_{12}$ and $\varepsilon_1 + \varepsilon_3 + V_{13}$, which correspond to the occupation of the first and second and the first and third LS's, respectively. Substituting them into the right-hand side of Eq. (1), we get the following system of inequalities:

$$\varepsilon_3 - \varepsilon_2 < V_{12} - V_{13} - V_{23}, \quad (2)$$

$$\varepsilon_2 - \varepsilon_3 < V_{13} - V_{12} - V_{23}. \quad (3)$$

We see that since $V_{23} > 0$ the conditions (2) and (3) are inconsistent and, hence, the clusters of three LS's cannot provide double peaks.

Let us turn to the case $N = 4$. Because the number of variants increases dramatically in this case we will restrict our search. Namely, we will assume that the first LS with the lowest energy ε_1 is located in the center of an equilateral triangle while the other three LS's with energies $\varepsilon_2 < \varepsilon_3 < \varepsilon_4$ are located in the vertexes. Then the energy of repulsion takes only two values, V_1 and V_2 (see Fig. 1). The first electron enters the system at $E_F^1 = E_4^1 \equiv \varepsilon_1$. It is easy to see that there are only two energies competing for E_4^2, which are $\varepsilon_1 + \varepsilon_2 + V_1$ and $\varepsilon_2 + \varepsilon_3 + V_2$. All the other two-electron states have higher energies. Similarly, we conclude that there are only two candidates for E_4^3. They are $\varepsilon_1 + \varepsilon_2 + \varepsilon_3 + 2V_1 + V_2$ and $\varepsilon_2 + \varepsilon_3 + \varepsilon_4 + 3V_2$. Let us again assume $n = 1$ in the condition (1). Then, according to the general procedure, one should choose the lowest of two values for E_4^3 and check condition (1) with both candidates for E_4^2. If we pick the first candidate for E_4^3 and $\varepsilon_1 + \varepsilon_2 + V_1$ for E_4^2, this condition reduces to $\varepsilon_3 - \varepsilon_2 < -V_2$, which contradicts our assumption that the energies are ordered. Thus, the only remaining option is that from two candidates for E_4^3 the second one has the lower energy. The corresponding condition for this can be written as

$$\varepsilon_4 - \varepsilon_1 < 2(V_1 - V_2). \quad (4)$$

Now with $E_4^3 = \varepsilon_2 + \varepsilon_3 + \varepsilon_4 + 3V_2$ the system of inequalities, resulting from (1) in a similar way as (2) and (3), takes the form

$$(\varepsilon_4 - \varepsilon_2) + (\varepsilon_3 - \varepsilon_1) < 2V_1 - 3V_2, \quad (5)$$

$$(\varepsilon_4 - \varepsilon_2) - (\varepsilon_3 - \varepsilon_1) < -V_2. \quad (6)$$

Upon summation of these two inequalities we get

$$\varepsilon_4 - \varepsilon_2 < V_1 - 2V_2. \quad (7)$$

On the other hand, we have assumed that $\varepsilon_4 > \varepsilon_2$. Then the principal requirement for a double peak to occur reduces to $V_1 > 2V_2$. If this requirement is met, the inequalities (5) and (6) are consistent. This is illustrated in Fig. 1 where the graphical solution of the system (5) and (6) is shown. In principle, one should also check that the energies of LS's, satisfying the system, satisfy the condition (4) as well. Note, however, that the condition (5) is stronger than (4), so that the latter is automatically

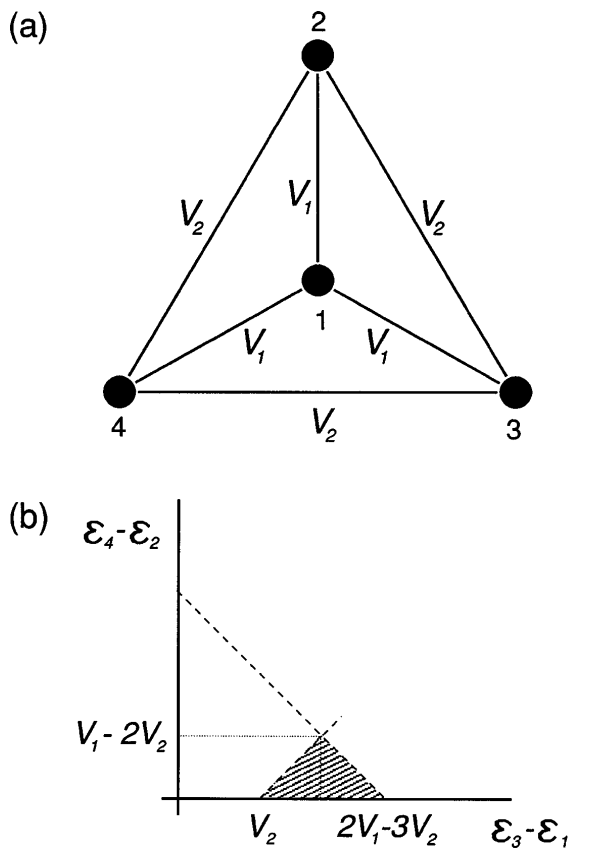

FIG. 1. Cluster of four LS's providing a double peak (a). Graphical solution of the system (5) and (6). Dashed is the region within which this system is satisfied (b).

obeyed, as can readily be seen from the following chain of relations:

$$\varepsilon_4 - \varepsilon_1 < (\varepsilon_4 - \varepsilon_2) + (\varepsilon_3 - \varepsilon_1) < 2V_1 - 3V_2 < 2(V_1 - V_2). \quad (8)$$

Thus we have demonstrated that, within the restricted geometry considered, double peaks can occur provided that $V_1 > 2V_2$. Obviously, the relation between V_1 and V_2 is opposite if the interaction between the localized electrons is simply the Coulomb repulsion. Since the distance between the first and the second LS is $\sqrt{3}$ times smaller than the distance between the second and the third LS (see Fig. 1) we have $V_1 = \sqrt{3}V_2$. The situation changes if a metallic electrode is placed at a distance d from the plane of the localized electrons. Then the Coulomb interaction is modified to

$$V(r) = \frac{e^2}{\kappa}\left(\frac{1}{r} - \frac{1}{\sqrt{r^2 + 4d^2}}\right). \quad (9)$$

The modified interaction falls off as $1/r^3$, and we indeed have $V_1 > 2V_2$ as soon as the distance between the first and the second LS's exceeds $0.33d$.

If the system (5) and (6) is satisfied, the evolution of the occupation of the cluster with increasing the gate voltage (Fermi level position E_F) would be as follows. For $E_F < \varepsilon_1$ all four LS's are empty. At $E_F = \varepsilon_1$ the first LS in the center of the triangle gets occupied. As E_F reaches the value $E_F = (\varepsilon_2 + \varepsilon_3 + \varepsilon_4 + 3V_2 - \varepsilon_1)/2$ an electron from the center moves to one of the vertexes and two electrons arrive from the electrode and occupy two other vertexes. Finally at $E_F = \varepsilon_1 + 3V_1$ the LS in the center gets occupied again.

After realizing that double peaks are possible in principle, we turn to the question: how frequent are they? One could argue that double peaks are allowed only for extremely rare configurations that do not really occur in a finite-size sample. To answer this question we calculate the probability that in a cluster of four LS's the energies and distances between LS's are arranged in such a way that the two-electron tunneling becomes possible after the cluster is singly occupied. We start from the observation that the previous consideration for restricted geometry becomes general if, instead of (9), we assume the model "hard core" interaction between the LS's: $V(r) = U$ for $r < d$ and $V(r) = 0$ for $r > d$. It is important that the interaction takes only two values: U and zero. Then the above analysis for the equilateral triangle applies if the distances, r_{ij}, between the LS's satisfy the following requirements: $r_{12}, r_{13}, r_{14} < d$ and $r_{23}, r_{34}, r_{24} > d$. If these requirements are met, the conditions for the double-peak formation are given by Eqs. (5) and (6) with $V_1 = U$ and $V_2 = 0$. It can be verified directly that for all other configurations of four LS's double peaks are forbidden. Then the calculation of the probability, $\mathcal{P}$, of the occurrence of a double peak can be performed in the following way: we fix the position and the energy of the first LS and find the allowed phase volume for the other three LS's. The advantage of the "hard core" interaction is that the intergrations over coordinates and energies are decoupled from each other. If we denote with g the density of LS's, then the expression for $\mathcal{P}$ can be presented as $\mathcal{P} = g^3 I_1 I_2$, where I_1 and I_2 are the phase volumes in the energy and coordinate spaces, respectively,

$$I_1 = \int_{\varepsilon_1}^{\infty} d\varepsilon_2 \int_{\varepsilon_2}^{\infty} d\varepsilon_3 \int_{\varepsilon_3}^{\infty} d\varepsilon_4 \times \theta(2U + \varepsilon_1 + \varepsilon_2 - \varepsilon_3 - \varepsilon_4) \times \theta(\varepsilon_2 + \varepsilon_3 - \varepsilon_1 - \varepsilon_4), \quad (10)$$

$$I_2 = \int d\mathbf{r}_2 \int d\mathbf{r}_3 \int d\mathbf{r}_4\, \theta(r_{23} - d)\theta(r_{24} - d) \times \theta(r_{34} - d)\theta(d - r_{12})\theta(d - r_{13})\theta(d - r_{14}). \quad (11)$$

The analytical evaluation of the first integral results in $I_1 = U^3/3$. The integral I_2 is obviously proportional to d^6; the numerical factor was found using the Monte Carlo procedure. Finally, we obtain $\mathcal{P} = 0.611(gUd^2)^3$. We see that the portion of double peaks is governed by the dimensionless parameter gUd^2 which is the ratio of the interaction energy and the mean level spacing within the size of the "core."

It is apparent that for the realistic interaction (9) the estimate for $\mathcal{P}$ emerges if one substitutes for U the value $e^2/\kappa d$—the Coulomb interaction at distance d. This gives $\mathcal{P} \sim (ge^2 d/\kappa)^3$. To find the numerical coefficient, the Monte Carlo integration over the nine-dimensional space (6 coordinates and 3 energies) was performed using the program published in [6]. The program generated a random set of dimensionless (in the units of d and $e^2/\kappa d$) coordinates and energies, calculated the values E_4^1, E_4^2, E_4^3, and then checked condition (1). The numerical factor obtained is $(5.1 \pm 0.1) \times 10^{-2}$.

The calculation of g poses a separate problem. One approach to estimate g is to assume that the random potential just smears the edge of the band density of states $g_0 = m/\pi\hbar^2$. Then for energies not very deep in the tail, g is still of the order of g_0. The product $g_0 e^2 d/\kappa$ can be rewritten as $d/\pi a_0$, where $a_0 = \hbar^2\kappa/me^2$ is the effective Bohr radius. It may seem that, if d is large enough, this product could be much larger than 1. However, this is not the case, since with increasing d the interaction of the occupied LS's becomes important. This leads to the suppression of the density of states in the vicinity of the Fermi level (Coulomb gap [7,8]). For the interaction (9) the energy dependence of g was studied both analytically [9] and by computer simulations [10]. It was shown that $g(\varepsilon) = 0.085(d/\kappa e^2) + (2/\pi)(\kappa^2|\varepsilon - E_F|/e^4)$. In our problem the relevant energy scale is $|\varepsilon - E_F| \sim e^2/\kappa d$ so that $ge^2 d/\kappa$ is of the order of 1. In view of the ambiguity in g, our calculation can be considered only as an estimate showing that $\mathcal{P}$ is not small. Indeed, the only small parameter in $\mathcal{P}$ is the numerical factor 0.051. This factor is, in fact, surprisingly large, taking

into account that it emerged as a result of the ninefold integration.

We have addressed only the energy aspects of the double-peak formation. There is also a question about the dynamics of the process. Suppose that the occupation of a single-electron state requires a time τ (τ is inverse proportional to the probability of tunneling). For a two-electron state with a binding energy W it can be shown that this time increases dramatically and becomes of the order of $\tau^2 W/\hbar$. The reason for the enhancement is that two electrons cannot tunnel sequentially because of the energy restrictions. However, if the temperature T is finite, the sequential tunneling becomes possible due to the smearing of the Fermi distribution in the electrode. Assume for simplicity that both single-electron energies in a two-electron state are the same. Then the time required for the sequential occupation of the two-electron state is equal to $\tau \exp(W/2T)$. This time is shorter than $\tau^2 W/\hbar$ if $T > W/2\ln(W\tau/\hbar)$.

Finally, let us discuss the magnetic field dependence of the double peaks. Note that conditions (5) and (6) (formation of a double peak) require the energies of all four LS's in the cluster to be rather close (roughly speaking, they should lie within the interval of the order of $e^2/\kappa d$). With increasing B each LS moves up in energy. It is important that the rate of this motion is different for different LS's. This is obvious if some of the LS's originate from donors, located in the well, while others represent the size quantization levels in the lateral fluctuations of the random potential [1,4]. Since the rate for donors is much slower, condition (1), met at $B = 0$, will get violated at some critical B due to the spread in the level positions. For higher B the double peak will split into two. Even in the case when all four components of the double-peak cluster at $B = 0$ are the ground states in parabolic confinements, their energies will depart from each other with increasing B, thus causing a splitting of the peak. If a confinement is characterized by the position of minimum U_0 and the frequency of the zero-point motion ω_0, the behavior of the energy level with B is given by $\varepsilon(B) = U_0 + \hbar(\omega_0^2 + \omega_c^2/4)^{1/2}$ (ω_c stands for the cyclotron frequency). Suppose that at $B = 0$ two levels, $\varepsilon_1(0)$ and $\varepsilon_2(0)$, are anomalously close in energy (in order to participate in the cluster). This means that the sum $U_0^{(1)} + \hbar\omega_0^{(1)} = \varepsilon_1(0)$ is close to $U_0^{(2)} + \hbar\omega_0^{(2)} = \varepsilon_2(0)$, while separately $U_0^{(1)}$ and $U_0^{(2)}$ can differ, say, by a factor of 2. But in a strong magnetic field we have $\varepsilon_1(B) - \varepsilon_2(B) = U_0^{(1)} - U_0^{(2)}$, so that departure is the typical fate of the initially aligned levels.

Although double peaks at $B = 0$ disappear eventually, new double peaks may emerge with increasing B. Whether this happens or not depends on how the density of LS's within some energy interval changes with B. In the simplest model, the radius of a state grows with its energy. Therefore, the higher in energy is the state, the faster it shifts upwards with B. This leads to the depletion of the "tail" of the density of states. The depletion, in turn, suppresses the number of the double-peak configurations. Experimentally, however, the LS's have different origins, and their rates of energy shifts with B are widely spread. In this situation any certain prediction on the evolution of the number of the double peaks with B is impossible.

In conclusion, we have demonstrated that double peaks in the differential capacitance may result from the interaction-induced correlations in the occupation numbers of LS's within a cluster. A more conventional consequence of these correlations is that adding *one* electron to the cluster might cause a redistribution of neighboring electrons over the LS's in order to reduce the total energy. This process is similar to the formation of a polaron by a lattice surrounding an LS. Note that such a purely electronic "polaron" was studied intensively by Efros and Shklovskii and by Pollak and Ortuño (see, e.g., the reviews [7,8]) in connection with the density of states and inelastic transport in the Coulomb glass. Making a link to these works, our main result can be reformulated as follows: for interactions which fall off steeply enough with distance, the formation of an "electronic *bipolaron*" in certain compact clusters of LS's is energetically favorable.

The authors are grateful to R. C. Ashoori for a valuable comment. L. G. is grateful to E. I. Rashba for illuminating discussion on the deformation fields of a polaron. M. R. acknowledges the discussions with A. L. Efros and F. G. Pikus at the initial stage of the work. The work at the University of Minnesota was supported by NSF Grant No. DMR-9423244.

[1] R. C. Ashoori, H. L. Stormer, J. C. Weiner, L. N. Pfeiffer, S. J. Pearton, K. W. Baldwin, and K. W. West, Phys. Rev. Lett. **68**, 3088 (1992); Physica (Amsterdam) **189B**, 117 (1993).

[2] R. C. Ashoori, H. L. Stormer, J. C. Weiner, L. N. Pfeiffer, K. W. Baldwin, and K. W. West, Phys. Rev. Lett. **71**, 613 (1993).

[3] P. W. Anderson, Phys. Rev. Lett. **34**, 935 (1975).

[4] Y. Wan, G. Ortiz, and P. Phillips, Phys. Rev. Lett. **75**, 2879 (1995).

[5] F. V. Kusmartsev and E. I. Rashba, Fiz. Tekh. Poluprovodn. **18**, 691 (1894) [Sov. Phys. Semicond. **18**, 439 (1984)].

[6] W. H. Press *et al.*, *Numerical Recipes in C* (Cambridge University Press, Cambridge, 1992), 2nd ed.

[7] A. L. Efros and B. I. Shklovskii, in *Electron-Electron Interactions in Disordered Systems*, edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985).

[8] M. Pollak and M. Ortuño, in *Electron-Electron Interactions in Disordered Systems* (Ref. [7]).

[9] A. A. Mogilyanskii and M. E. Raikh (unpublished).

[10] F. G. Pikus and A. L. Efros, Phys. Rev. B **51**, 16 871 (1995).