Lapse of transmission phase and electron molecules in quantum dots

S.A. Gurvitz

Department of Particle Physics, Weizmann Institute of Science, Rehovot 76100, Israel

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The puzzling behavior of the transition phase through a quantum dot can be understood in a natural way via formation of the electron molecule in the quantum dot. In this case, the resonance tunneling takes place through the quasistationary (doorway) state, which emerges when the number of electrons occupying the dot reaches a certain “critical” value, \( N_{cr} \). Our estimation of this quantity agrees with the experimental data. The dependence of \( N_{cr} \) on the dot’s size is predicted as well.

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One of the challenging problems in mesoscopic physics is the puzzling behavior of the transmission phase through a quantum dot, embedded in an Aharonov-Bohm ring. It was found in series of experiments performed by the Weizmann group\(^\text{1,2,3}\) that all transmission amplitudes through different resonant levels of a quantum dot are in phase. This necessarily implies an unexpected role of the Pauli principle that prevents different conductance resonances to be in phase. In addition, it was found in recent measurements\(^\text{4,5}\) that this phenomenon takes place even when the number of electrons inside the dot reaches a certain “critical” value \( N_{cr} \). In spite of many publications addressed to these experiments, a full satisfactory understanding has been found yet\(^\text{6-10}\).

In this Rapid Communication we demonstrate that the observed phase-lapse behavior of the transmission amplitude can be naturally explained by imposing the formation of electron (Wigner) molecules inside quantum dots, proposed in recent publications\(^\text{6,7,8,9}\). Moreover, this framework allows us to estimate \( N_{cr} \) and then to determine how it is varied with a size of the dot. In order to explain our model in a proper way, we first elaborate the physical nature of the transmission phase in the case of noninteracting and interacting electrons. In particular, we concentrate on the role of the Pauli principle that prevents different conductance resonances to be in phase.

The puzzling behavior of the transition phase through a quantum dot can be understood in a natural way via formation of the electron molecule in the quantum dot. In this case, the resonance tunneling takes place through the quasistationary (doorway) state, which emerges when the number of electrons occupying the dot reaches a certain “critical” value, \( N_{cr} \). Our estimation of this quantity agrees with the experimental data. The dependence of \( N_{cr} \) on the dot’s size is predicted as well.

Let us consider the resonant tunneling through a quantum dot, represented by a potential \( U_D(x) \), Fig. 1. The bottom of this potential can be moved by the plunger electrode, so that one observes the current sweeping through different resonant states \( (E_k) \) of the dot. We would treat this problem in the framework of a tunnel Hamiltonian approach. This approach is more transparent for evaluation of the transmission phase than the standard scattering theory, in particular, when the Pauli principle and the electron-electron interaction are taken into account. We introduce therefore the following tunneling Hamiltonian: \( H = H_L + H_R + H_D + H_T \), where

\[
H_L(R) = \sum_{l(r)} E_{l(r)} a_{l(r)}^{\dagger} a_{l(r)}, \quad H_D = \sum_k E_k d_k^{\dagger} d_k + H_C, \quad H_T = \left( \sum_{l,k} \Omega_l^{(k)} d_k^{\dagger} a_l + l \leftrightarrow r \right) + H.c. \quad (1)
\]

Here, \( a_{l(r)}^{\dagger} a_{l(r)} \) is the creation (annihilation) operator of an electron in the reservoirs and \( d_k^{\dagger} (d_k) \) is the same operator for an electron inside the dot. For simplicity, we consider electrons as spin-less fermions. The term \( H_C \) denotes the Coulomb interaction between electrons in the dot and \( \Omega_l^{(k)} [\Omega_r^{(k)}] \) is the coupling between the states \( E_l(E_r) \) and \( E_k \) of the reservoir and the dot, respectively. In the absence of magnetic field, all couplings \( \Omega_l^{(k)} \) are real.

All parameters of the tunneling Hamiltonian \( H \) are related to the initial microscopic description of the system in the configuration space. For instance, the coupling \( \Omega_l^{(k)} \) is given by the Bardeen formula\(^\text{10}\):

\[
\Omega_l^{(k)} = -\frac{\hbar^2}{2m} \int_{x \in \Sigma_l(r)} \phi_k(x) \nabla_n \chi_l(r)(x) d\sigma \quad (2)
\]

where \( \phi_k(x) \) and \( \chi_l(r)(x) \) are the electron wave functions inside the dot and the reservoir, respectively, and \( \Sigma_l(r) \) is a surface inside the left (right) barrier that separates the dot from the corresponding reservoir. It is important to point out that \( \phi_k(x) \) in Eq. (2) is a bound state wave function for the “inner” potential. The latter coincides with the original potential inside the surface \( \Sigma \).
and a constant outside this region. On the other hand, \( \chi_{(r)}(x) \) is a non-resonant scattering wave function in the “outer” potential, which coincides with the original potential outside the surface \( \Sigma \) and a constant inside this region.\(^\text{12}\)

In one-dimensional case (Fig. 1), the separation surface \( \Sigma \) becomes the separation point, \( \bar{x} \), inside the barrier, Fig. 1. Then Eq. (2) can be rewritten as\(^\text{12}\)

\[
\Omega_{l(r)}^{(k)} = - (\kappa_k/m) \phi_k(\bar{x}_{l(r)}) \chi_{l(r)}(\bar{x}_{l(r)}),
\]

(3)

where \( \kappa_k = \sqrt{2m(U_D(\bar{x}_{l(r)}) - E_k)} \) and \( \phi_k(x) \) is the bound state wave function in the potential \( \bar{U}_D(x) \) (Fig. 1). The separation points \( \bar{x}_{l,r} \) are to be taken inside the left (right) barrier as indicated in Fig. 1 and far away from the classical turning points.\(^\text{12}\)

We start with non-interacting electrons, \( H_C = 0 \) in Eq. (1). Then the electron transport through the level \( E_\lambda \) can be described by the time-dependent Schrodinger equation \( i\hbar \partial_t |\Psi(t)\rangle = \hat{H} |\Psi(t)\rangle \) for a single electron. Taking the stationary limit we obtain the Landauer formula for the total current, with the transmission amplitude given by the Bright-Wigner formula\(^\text{12}\)

\[
t_\lambda(E) = N \frac{\Omega_L^{(\lambda)} \Omega_R^{(\lambda)}}{E - E_\lambda + i(\Gamma_L^{(\lambda)} + \Gamma_R^{(\lambda)})/2},
\]

(4)

where \( N = -2\pi |\Omega_L^{(\lambda)} \Omega_R^{(\lambda)}|^2 \) and \( \Gamma_{L(R)}^{(\lambda)} = 2\pi |\Omega_{L(R)}^{(\lambda)}|^2 \) are the partial widths, and \( \Omega_{L(R)}^{(\lambda)} \) is the density of states in the left (right) reservoir. We assumed that \( \Omega_L^{(\lambda)} \equiv \Omega_R^{(\lambda)} \) are weakly dependent on \( E_{l(r)} \).

The corresponding evolution of the resonance transmission phase for different states \( |\lambda\rangle \) is determined by the sign of the product of \( \Omega_L^{(\lambda)} \Omega_R^{(\lambda)} \). Since the reservoir states \( \chi_{l,r} \) are not affected by the plunger voltage, one finds from Eq. (3) that the evolution of the sign \( \Omega_L^{(\lambda)} \Omega_R^{(\lambda)} \) is given by the sign of the product \( \phi_L(x) \phi_R(x) \). Hence, it is clear that the non-interacting electron model cannot explain the same sign for all resonances, observed in Ref.\(^\text{2}\) (see also Refs.\(^\text{12,14}\)).

Consider \( N \) interacting electrons trapped inside the dot. Despite the electron-electron interaction, the coupling amplitudes \( \Omega_{L,R} \) can still be evaluated by using the same multi-dimensional overlapping formula\(^\text{2}\), as in the non-interacting case. Indeed, the many-body tunneling can be considered as one-body tunneling, but in the many-dimensional space. In this case, the wave-function \( \chi_{(r)}(x) \) is replaced by \( \chi_{(r)}(x_{N+1}) \Phi_N^{(0)}(x_1, \ldots, x_N) \), where \( \chi_{(r)} \) is the wave function of tunneling electron in the left (right) reservoir and \( \Phi_N^{(0)} \) is the ground state wave function of \( N \) electrons inside the dot. The wave-function \( \phi_{0}(x) \) corresponds to \( \Phi_{N+1}^{(0)}(x_1, \ldots, x_{N+1}) \), which is the lowest energy state (ground state) of \( N + 1 \) electrons in the inner potential of the dot \( \bar{U}_D \) in Fig. 1.

Taking \( n \) along a coordinate of the tunneling electron, \( x_{N+1} \), we can integrate over \( x_1, \ldots, x_N \) in Eq. (2), thus reducing this equation to Eq. (3) with \( \phi_n \) being replaced by the overlap function\(^\text{2}\)

\[
\varphi_N(x_{N+1}) = \langle x_{N+1}, \Phi_{N}^{(0)} | \Phi_{N+1}^{(0)} \rangle.
\]

(5)

Therefore, the sign of \( \Omega_L^{(\lambda)} \Omega_R^{(\lambda)} \) is determined by the sign of \( \varphi_N(x_{N+1}) \).

By applying the mean-field approximation, we can write \( |\varphi_N^{(0)} \rangle \) and \( |\Phi_N^{(0)} \rangle \) as a product of one-electron states (orbitals) in the effective single-particle potential, \( \bar{U}_D + U_C \), where \( \bar{U}_D \) is the inner part of quantum-dot potential (Fig. 1) and \( U_C(x) \) is the mean-field describing the electron-electron interaction. As a result, the overlap function \( \varphi_N(x) \) is a bound state wave function in the potential \( \bar{U}_D(x) + U_C(x) \), corresponding to one of the orbitals. Since the lowest energy state is always nodeless, one might assume that \( \varphi(x) \) is also a nodeless one, so that the sign of \( \varphi_N(x_{N+1}) \) would be the same sign for all resonances. This, however, is not correct because of the Pauli principle. Indeed, due to the antisymmetrization, any two orbitals in the product of the wave functions representing \( |\Phi_N^{(0)} \rangle \) cannot be the same. Since the lowest state is already occupied, the wave function \( \varphi_N(x_{N+1}) \) must correspond to a higher non-occupied orbital, and therefore it cannot be nodeless. Hence, the Pauli principle would create serious problems in any attempt to explain the same sign for all resonances in a framework of the mean-field description of the electron-electron interaction.

Note that this problem cannot be resolved even by assuming large coupling with reservoirs, so that the resonances are overlap. Indeed, the problem is related only to the inner component of the resonant state, Eqs. (2) and (3). The latter is eventually brought by the plunger below the Fermi level, \( \mu_R \), blocking an appearance of the resonance above the Fermi level with a similar inner component.

The same situation holds in a more general case, when the interaction term \( U_C \) varies with each new electron trapped inside the dot, \( U_C \rightarrow U_C^{(N)} \) (Koopman’s theorem is violated). One finds that due to the central symmetry of the self-consistent potential such a variation of \( U_C \) with \( N \) would not affect the number of nodes in the overlap function \( \varphi_N(x_{N+1}) \). As a result, the sign of the transmission amplitude would fluctuate between \( \pm 1 \) for different resonances.

We illustrate this point by evaluating the overlap function \( \varphi_N(x_{N+1}) \), Eq. (3), for \( N = 0 \) and \( N = 1 \). In the first case, \( \varphi_0(x_1) \) coincides with the wave function of the lowest energy state, \( \Phi_1^{(0)}(x_1) \equiv \hat{\phi}_0(x_1) \), in the inner potential \( \bar{U}_D \), Fig. 1. This wave function is nodeless. The second overlap function is \( \varphi_1(x_2) = \langle x_2, \Phi_1^{(0)} | \Phi_2^{(0)} \rangle \), where \( \Phi_2^{(0)}(x_1, x_2) = |\phi_0(x_1) \phi_1(x_2) - \phi_0(x_2) \phi_1(x_1)|/\sqrt{2} \) is the lowest energy state of two electrons in the potential \( \bar{U}_D + U_C^{(2)} \). Here \( \phi_{0,1} \) represent the two first orbitals in
This potential. One easily finds that
\[
\phi_1(x_2) = \int \Phi_1^0(x_1)\Phi_1^0(x_1, x_2)dx_1 = c_0\phi_1(x_2), \quad (6)
\]
where \(c_0 = \int \phi_0(x_1)\phi_0(x_1)dx_1/\sqrt{2}.\) (The second term is zero, since \(\phi_0\) and \(\phi_1\) are orthogonal due to the opposite parities). Therefore, \(\phi_1\) contains one node, so that the corresponding transition amplitude changes its sign.

The same behavior of the overlap function would persist for any \(N\). For instance, one easily obtains for \(N = 2\) that \(\phi_2(x_1) = c_{10}\phi_1(x_3) - c_{13}\phi_1(x_3),\) where the coefficients \(c_{10} = \langle \phi_1 | \phi_0 \rangle, c_{13} = \langle \phi_0 | \phi_2 \rangle\) and \(\phi, \phi\) are the orbitals in the potentials, \(\tilde{U}_D(x) + U^{(1)}(x)\) and \(\tilde{U}_D(x) + U^{(2)}(x)\), respectively. Since \(c_{13} \ll c_{10}\), the overlap function \(\phi_2\) would contain an additional node in a comparison to \(\phi_1\). Thus, by assuming the \(N\) dependence of the mean-field effective potential, we are still not able to explain the puzzling behavior of the transmission phase.

The above consideration was based on symmetry arguments applied to electrons moving in a spherically symmetric mean-field central potential. In fact, the central mean-field picture for two-dimensional quantum dots was challenged in recent publications.\(^{6,7,8}\) It was suggested that due to the strong inter-electron repulsion inside the dot, spontaneous symmetry breaking takes place leading to the formation of electron molecules. As a result, the electrons appear on the ring (around) the dot’s center. This idea was substantiated by unrestricted Hartree-Fock calculations or by using other computational techniques.\(^{6,7,8}\)

In principle, if the symmetry is broken, the overlap function \(\phi_N(x_{N+1}),\) Eq. (6), could be very different from the corresponding orbital \(\phi_0(x_N + 1)\) in the spherical symmetric potential. Therefore, it is desirable to investigate the evolution of the transmission phase in this case. Consider again the overlap function \(\phi_1(x_2) = \langle x_2, \Phi_1^0 | \Phi_1^0 \rangle,\) but now without the mean-field approximation, as in Eq. (6). In fact, by taking the parabolic confining potential, the two-electron wave function \(\Phi_2^0\) can be exactly calculated,\(^{6,7,8}\) since relative and center-of-mass coordinates of two electrons are decoupled in the total Hamiltonian. As a result, \(\Phi_2^0(x_1, x_2) = \phi_{cm}(x_1 + x_2)\phi_r(x_2 - x_1),\) where \(\phi_r(-x) = -\phi_r(x)\) due to the Pauli principle. Such a wave function peaks for \(x_1 = -x_2\) and therefore it would bear the features of a two-electron molecule. One finds from Eq. (5),
\[
\phi_1(x_2) = \int \phi_0(x_1)\phi_{cm}(x_1 + x_2)\phi_r(x_2 - x_1)dx_1. \quad (7)
\]
Taking into account that the values of \(x_1\) which mainly contribute to the integral (7) are localized inside the dot and that the wave function \(\phi_r(x)\) is the odd one, we find that the overlap function changes its sign when the argument varies from \(\bar{x}_1\) to \(\bar{x}_2\). Fig. 1. Hence, \(\phi_1\) displays one node, as in the spherically symmetric mean-field potential, Eq. (6).

One can continue with the same arguments for the three and more electron molecules, where the electrons are placed on the ring. The corresponding ground state wave functions \(|\Phi_N^0\rangle\) would represent a fully anti-symmetrized product of the original (site) nodeless orbitals.\(^{6,7,8}\) Yet, the overlap function Eq. (6) cannot be nodeless. As a result, the sign of \(\phi_N(x_r)\) would fluctuate with \(N\). One can demonstrate it rather easily for \(N = 3, 4\). Although it would be hard to extend such a demonstration for large \(N\), there is no reason to expect that the sign of \(\phi_N(x)\) ceases to fluctuate when \(N\) increases.

It seems from the above arguments that the rotational symmetry breaking (the electron-molecule formation) cannot explain the evolution of the transmission phase observed in the experiments.\(^{6,7,8}\) Nevertheless, there is an additional feature of the electron molecule, which has not been yet utilized. That is due to the electrons located on the ring (rings) would develop an additional (inner) electrostatic trap inside the dot when their number \((N)\) is large enough. As an example, we display in Fig. 2 such a potential, \(V_C(x) = \sum N e^2/r|x - x_j|,\) produced by 14 electrons equally distributed on the ring, where \(e = 13.6\) is the dielectric constant of the medium. The radius of the ring \((R = 50\text{ nm})\) is taken close to the dot’s size in Ref. 13. The radial profile of this potential along the angle \(\theta = \pi/N\), where the potential height on the ring is minimal, is shown in Fig. 2b for two values of \(N\). It appears that the trap is not well developed for \(N = 6\), but it is already pronounced for \(N = 14\).

A minimum number of electrons in the dot sufficient to develop the trap with one bound state inside it can be estimated from the condition that the barrier height, \(h_N\) in the Fig. 2b, reaches the ground state energy \(\varepsilon_0\). We estimate the latter as \(\pi^2\hbar^2/m^*R^2\), where \(m^*\) is the effective electron mass \((m^*/m_0 = 0.067)\). For instance, one finds \(c_0 = 4.5\text{ meV for } R = 50\text{ nm}\). Then the condition \(h_N = \varepsilon_0\) corresponds to \(N \simeq 10\), which is a minimal ("critical") number of electrons, \(N_{cr}\), enabled to hold a resonance state. This value is an approximate agreement with that found in.\(^{3}\) In fact, a more elaborate, semiclassical estimations of \(N_{cr}\) approximately produce the same number \([h_N \simeq 10\text{ meV for } N = 14, \text{Fig. 2b}]\)\(^{12}\).

The state \(|\varepsilon_0\rangle\) in the inner part of the trap, \(V_C, \text{Fig. 2b}\), is not stable due to the symmetry breaking, leading to formation of the electron molecule. Nevertheless, this state is important in formation of the \((N + 1)\)-electron molecule by adding an additional electron to the \(N\)-electron system. Indeed, one expects that the overlap function (6) for the electron states on the ring is suppressed in comparison to the same overlap for the central mean-field potential. The reason is that all electrons are shifted from their positions whenever an additional electron is placed on the ring. This is in contrast to the mean-field description, where the \(N\)-electron core is not modified. On the other hand, if the electron is placed in the center of the ring, it distorts the remaining \(N\) electrons in a minimal way. We expect therefore that the...
corresponding overlap function is large, as in the case of the central mean-field potential. Hence, such an unstable state $|\varepsilon_0\rangle$ in the middle of the dot would play a role of a “doorway” state in formation of the $(N + 1)$-electron molecule.

It follows from the same arguments that the electron transport would proceed through such an unstable state when the quantum dot coupled with the reservoirs. Since this doorway state is of the lowest energy in the inner trap, $V_C$ (Fig. 2), it is nodeless. The crucial point here is that this state is eventually not occupied, when it is brought by the plunger below the Fermi levels of the reservoirs. Indeed, it is not turned to a stable state below the Fermi levels due to the symmetry breaking, but it always decays to the ring states. Therefore, this state is never blocked by the Pauli principle to carry the resonant transport through it, when it is above the Fermi level $\mu_R$, Fig. 1. As a result, all transmission amplitudes for any $N > N_{cr}$ would be in phase.

In fact, by taking into account the electron spin, one finds that two electrons with the same spatial (nodeless) wave functions are allowed to occupy the lowest energy states. Therefore, even if the state $|\varepsilon_0\rangle$ in the center of the dot becomes a stable one for some values of $N$, the resonant transport would proceed through an unstable state of the two electrons (with opposite spin) inside the dot. The corresponding overlap function would be again nodeless.

Note that although the doorway-state energy is the lowest one for the inner trap, $V_C$, it exceeds the energy of the ring states. Therefore, the ring states would appear inside the bias voltage before the doorway state. We can assume, however, that the ring states are not well separated in the energy from the doorway state, which dominates the resonant current. It was also taken into account that in the presence of the Coulomb interaction, the shift of the resonance energy due to tunneling is different for different levels. In particular, the broad resonance is shifted down more than the narrow one. As a result the doorway state could have a lower energy than the ring states.

One of the consequences of our model is an existence of the critical number of electrons in the dot, which is necessary for formation of the resonant state inside the dot $(N_{cr})$. This number would vary with the dot’s size. Such a dependence of $N_{cr}$ on the radius of the dot $(R)$, obtained from our estimation, $h_N = \varepsilon_0$, is shown in Fig. 3. One finds from this figure that this dependence is rather weak. The critical number slightly decreases with an increase of the dot’s size.

In summary, we demonstrated that the unusual behavior of the resonant phase, observed in interference experiments, can be considered as a strong evidence for formation of electron molecules in quantum dots. This structure would produce an electrostatic trap, containing an unstable (doorway) state localized in the center of the dot, whenever the number of electrons occupying the dot is large enough, $N > N_{cr}$. Then such an unstable state would carry the electron transport through the dot irrespective of the value of $N$. This would appear as if the different transmission amplitudes are in phase. Our prediction for the dependence of $N_{cr}$ on the dot’s radius can be experimentally verified.
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17. In fact, a two-ring structure is expected for $N = 14$ electrons. Although this creates a more complicated trap, our simple estimations of $N_{cr}$, based on the effective one-dimentional potential, Fig. 2(b), remain the same. For such estimations it is sufficient to consider all electrons on one ring of the average radius of two rings. Note that $N_{cr}$ is weakly dependent on the ring's radius, Fig. 3.
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