Charged "Few-Electron–Single Spatially Separated Hole" Complexes in a Double Quantum Well near a Metal Plate

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It is shown that the presence of a metal plate near a double quantum well with spatially separated electron and hole layers may lead to a drastic reconstruction of the system state with the formation of *stable charged* complexes of several electrons bound to a *spatially separated* hole. Complexes of both Fermi and Bose statistics may coexist in the ground state and their relative densities may be changed with the change of the electron and hole densities. The stability of the charged complexes may be increased by an external magnetic field perpendicular to the layers plane. [S0031-9007(96)00943-X]

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Double layers [semiconductor double quantum wells (DQWs)] of spatially separated two-dimensional (2D) electrons (e) and holes (h) are a subject of considerable research interest. The Coulomb e-h interaction leads to interlayer correlations of the carriers and to the possibility of a transition to the superfluid excitonic phase [1,2]. There is a rapidly increasing amount of publications devoted to both theoretical [3–5] and experimental [6,7] investigations of e-h coupling in DQWs; see also references in [4,5,7]. Some experimental evidence has been reported [7] for a stable excitonic ground state in a strong magnetic field which favors the stability of the excitonic phase [8].

In contrast to the locally neutral "excitonic insulator" phase of a bulk sample [9], the excitonic phase of the spatially separated e and h would not possess the *local* electrical neutrality and the motion of strongly coupled spatially separated charges would be accompanied by nonzero electric currents counterflowing on e and h layers [1]. However, these currents are of equal magnitudes, which means that the *total* current along the QWs plane equals zero.

The question addressed in the present Letter concerns an existence of stable *charged* bound complexes of few electrons and holes in the double-layer system with *nonequal* 2D densities n_e and n_h of spatially separated e and h. The two simplest charged (quasi-)Fermi and (quasi-)Bose complexes ("electronic molecules") would be e_2h and e_3h, respectively (for definiteness, we will assume the negative total charge). The charged bosonlike complexes would be of special interest due to the possibility of their Bose-Einstein condensation and superfluidity accompanied with a nonzero *total* current along the DQW plane.

The fermion molecule e_2h known as a negatively charged exciton (X^-) does exist in bulk samples as well as in QWs and quantum dots. A magnetic field favors the existence of the X^- state (see, e.g., [10,11]), and even a series of fermionic "homologies" X_K^- (i.e., $e_{K+1}h_K$) with $K = 2, 3, \ldots$ has been predicted recently [11].

However, no charged *boson* bound states of *few* e and h (not to be mixed with mesoscopic electron-hole droplets where the big numbers of e and h may differ slightly) have been found up to now. Their existence is hindered because of the high increase of the Coulomb energy caused by *extra* electrons. This concerns especially the systems of spatially separated e and h where the Coulomb repulsion of electrons dominates over their attraction to a distant hole.

In this Letter we draw attention to a novel physical situation which takes place in the DQW system of spatially separated e and h layers located near a parallel metal plate (MP). When the MP is close to one of the layers (for definiteness, to the e layer), the e-e Coulomb repulsion is suppressed considerably by the "image charge" polarization of the MP [12]. The influence of a MP on the interaction and collective properties of low-dimensional electron systems is not a new subject; it is known, for instance, that the MP hinders the crystallization of the 2D electron gas ("cold melting" of the 2D electron Wigner crystal [13]). However, to the best of our knowledge, the advantage of using this suppression of the e-e repulsion in systems of spatially separated e and h has not been studied yet.

We demonstrate that the MP close to the e layer may lead to the formation of stable mobile charged complexes e_Nh of several (N) electrons bound to a *spatially separated* hole. We consider N electrons confined to the 2D layer and a single hole located at another parallel layer at a height l over the e layer. The e layer is at a height d over the surface of a metal plate. The potential energy of the e-h system is given by

$$U = -\sum_i [V(\mathbf{r}_i - \mathbf{r}_h; l) - V(\mathbf{r}_i - \mathbf{r}_h; l + 2d)] + \frac{1}{2}\sum_{i\neq j}[V(\mathbf{r}_i - \mathbf{r}_j; 0) - V(\mathbf{r}_i - \mathbf{r}_j; 2d)], \quad (1)$$

where $\mathbf{r}_i$ ($i = 1, ..., N$) and $\mathbf{r}_h$ are 2D radius vectors of the electrons and the hole, respectively; $V(\mathbf{r}; l) \equiv (e^2/\epsilon)[\mathbf{r}^2 + l^2]^{-1/2}$, ϵ is the dielectric constant of a surrounding medium.

Our present consideration is restricted to the case when l is large as compared to the characteristic quantum lengths, which are the effective Bohr radii $a_{e,h}$ of e and h or the magnetic length λ_H (if a strong magnetic field H is applied perpendicular to the DQW plane). In this

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limit, classical configurations play the crucial role and determine the leading contribution to the system energy. This is the feature of systems with spatially separated e and h; in systems with no spatial separation of e and h quantum mechanics is the only remedy against the collapse of the classical charges. Surprisingly, it turns out that even the classical configurations of the DQW-metal system are quite nontrivial. Later we will also discuss effects of quantum fluctuations.

For further comparison, first we describe classical configurations in the double-layer geometry without the MP (i.e., $d = \infty$). We introduce the rectangular coordinate system ($\hat{x}$-$\hat{y}$ axes) on the e layer and set the coordinate origin O exactly under the hole. The only stable classical state ["eh" complex ($N = 1$)] corresponds to the electron located at point O. The energy $U_1 = -e^2/(\epsilon l)$ of this bound state defines the characteristic energy scale of the system. There exist also unstable equilibrium classical configurations $e_N h$ with $2 \leq N \leq 4$ electrons located symmetrically at a ring centered at O. The ring radius ρ is given by

$$\rho = \tan\theta, \qquad \theta = \arcsin\left[\frac{1}{4}\sum_{i=1}^{N-1}\frac{1}{\sin(\pi i/N)}\right]^{1/3}, \quad (2)$$

which provides a saddle-point extremum of the potential energy (1) at $2 \leq N \leq 4$. Energies of these configurations are $U_2 \approx -0.94|U_1|$, $U_3 \approx -0.51|U_1|$, and $U_4 \approx -3.3 \times 10^{-3}|U_1|$, respectively. These configurations are unstable and decay into the stable eh state and free electrons. Though the unstable classical configurations might manifest in nonstationary processes (e.g., optical absorption), they are not important in the equilibrium. According to Eq. (2), at $N > 4$ equilibrium classical configurations $e_N h$ do not exist at all. To summarize, no *stable* classical configurations $e_N h$ with $N > 1$ exist in the DWQ without a neigboring MP.

The presence of a MP at the distance d down from the electron layer may change the situation qualitatively. This is particularly pronounced at $\eta \equiv d/l \ll 1$ and we begin with this range of parameters. The simplest eh state ($N = 1$) corresponds to the electron located at point O. The classical energy of this state is

$$U_1(\eta)/|U_1| = -2\eta/(1 + 2\eta) \approx -2\eta + O(\eta^2), \quad (3)$$

where the second equality refers to the case $\eta \ll 1$. On the contrary, with the increase of η, $U_1(\eta)$ tends to the energy $U_1 = -e^2/\epsilon l$ of the classical eh configuration in the absence of the MP.

At $\eta \ll 1$, the radius ρ of the equilibrium classical configurations e_2h obeys $d \ll \rho \ll l$ and is determined as the extremum of the following approximate expression for the potential energy Eq. (1): $U/|U_1| \approx -4\eta + 6\eta(\rho/l)^2 + \eta^2(l/\rho)^3/4$. We obtain $\rho \approx 0.57\eta^{1/5} l$ which justifies the approximations above. The classical energy of the e_2h configuration is given by

$$U_2(\eta)/|U_1| = -4\eta + 5 \times 2^{-3/5}\eta^{7/5} \approx -4\eta + 3.29\eta^{7/5}. \quad (4)$$

Thus, due to the image charges which decrease the e-e repulsion, the charged complex e_2h has the lower energy (4) than the energy (3) of the neutral complex eh. The e_2h complex is the *stable classical ground state* of the "two-electron–single hole" system. At the same time $2U_1(\eta) < U_2(\eta)$, which guarantees the stability of the eh complexes with respect to the reaction $2eh \rightarrow e_2h + h$. This means that as far as the electron density n_e does not exceed the hole one n_h, all the electrons are bound into the eh complexes. (Here and below both n_e and n_h are assumed to be sufficiently low so that we neglect all the screening effects; the temperature is also assumed to be sufficiently low.) In the range $n_h < n_e < 2n_h$, boson eh and fermion e_2h complexes coexist and their densities equal $2n_h - n_e$ and $n_e - n_h$, respectively. The stability of complexes with respect to adhering to one another is provided by the mutual hole repulsion which is less affected by relatively distant image charges.

For the simplest charged boson complex e_3h we meet a new phenomenon which has not occurred in the absence of the MP—there are *two* possible classical equilibrium configurations e_3h-a and e_3h-b described as follows: (a) three electrons form a regular triangle centered at O; (b) one of three electrons sits at center O (an "inner shell") and the others are located symmetrically with respect to the first one (an "outer shell"). We obtain the radii $\rho \approx 0.72\eta^{1/5} l$ and $\rho \approx 1.01\eta^{1/5} l$ of e_3h-a and e_3h-b configurations, respectively, and the corresponding energies

$$U_{3a}(\eta)/|U_1| = -6\eta + 5 \times 3^{2/5}\eta^{7/5},$$
$$U_{3b}(\eta)/|U_1| = -6\eta + 10(17/16)^{2/5}\eta^{7/5}. \quad (5)$$

As $U_{3a}(\eta) < U_{3b}(\eta)$, the configuration e_3h-b is unstable with respect to the transition into the lower energy "isomer" configuration e_3h-a. Indeed, the stability analysis of the e_3h-b configuration reveals an unstable mode which tends to distort the electron configuration towards the triangle arrangement of the e_3h-a complex. As $U_{3a}(\eta) < U_2(\eta) < U_1(\eta)$ at small η, the charged boson complex e_3h-a realizes the classical ground state of the system of "three electrons and one hole" (note in advance that the roles of e_3h-a and e_3h-b isomers will interchange when η is not small). At the same time, the inequalities $3U_1(\eta) < U_{3a}(\eta)$, $U_1(\eta) + U_2(\eta) < U_{3a}(\eta)$, and $2U_2(\eta) < U_{3a}(\eta) + U_1(\eta)$ forbid the reactions $3eh \rightarrow e_3h\text{-a} + 2h$, $eh + e_2h \rightarrow e_3h\text{-a} + h$, and $2e_2h \rightarrow e_3h\text{-a} + eh$, respectively. This means that the charged boson complexes e_3h-a may appear only at $n_e > 2n_h$. In the range $2n_h < n_e < 3n_h$, the boson e_3h-a and the fermion e_2h complexes coexist; their densities equal $n_e - 2n_h$ and $3n_h - n_e$, respectively.

The problem one meets at higher N is a variety of possible isomer configurations which correspond to different arrangements of electrons over "shells." The energies of type "a" (the "electron ring") and type "b" (with an electron at the center of the electron ring) configurations at $\eta \ll 1$ are given by

$$U_{N\sigma}(\eta)/|U_1| = -2N\eta + 5N\left[k_\sigma + \frac{1}{16}\sum_{i=1}^{N-1-k_\sigma}\frac{1}{\sin^3[\pi i/(N-k_\sigma)]}\right]^{2/5}\eta^{7/5}, \quad (6)$$

where $k_\sigma = 0$ and $k_\sigma = 1$ for the a and b configurations, respectively. At $N > 5$ configuration b becomes a lower energy than configuration a. It is not clear yet whether there are even more favorable configurations. Complete consideration of a "periodic" table of complexes remains a subject for further research. Here we estimate only the maximal allowed value of N (i.e., the number N_c of stable classical complexes in the table) at a given η. A lower bound for this value is determined by the first violation of the condition $U_N(\eta) < U_{N-1}(\eta)$, which reduces to $\partial U_N(\eta)/\partial N = 0$ for large N. If the ground state corresponds to the type b configuration, we obtain the following estimate for N_c:

$$N_c = C/\eta^{1/3} \gg 1, \quad (7)$$

where $C = 2\pi(2/11)^{5/6}$. If other configurations become important, the functional dependence Eq. (7) might still be valid although the numerical factor C might change. According to Eq. (7), at small η the world of stable charged complexes may be rather rich. However, their binding energies at small η are small, which is not favorable for experimental realizations.

When η increases (i.e., the metal plate is removed from the DQW), the binding energies of possible stable configurations increase but the number of these configurations decreases. Our further consideration is restricted to the representative configurations eh, e_2h, e_3h-a, and e_3h-b. Their energies in the intermediate range of η are plotted as functions of η in Fig. 1. In the interval $0 < \eta < 1.32$ the fermionic complexes e_2h are stable $[U_2(\eta) < U_1(\eta)]$ and therefore they would be present at the system ground state at $n_h < n_e \leq 2n_h$. As to the charged boson complexes (at $2n_h < n_e \leq 3n_h$), they are present in the e_3h-a isomeric configuration at $0 < \eta < 0.39$. At $\eta \approx 0.39$ the isomer configuration e_3h-b becomes more favorable. The latter remains stable in the interval $0.39 < \eta < \eta_c = 16$ but it does not exist even as an unstable equilibrium state at $\eta > \eta_c$. The existence of the critical value of η is a consequence of the fact that no equilibrium configuration of type b may exist in the DQW without the MP. As follows from Eq. (1), at $d/l = \eta \to \eta_c - 0$ the radius of the e_3h-b configuration tends to infinity: $\rho/l \approx 194/\sqrt{16-\eta}$; correspondingly, the energy of losing the outer electron shell (i.e., the "ionization" energy) decreases drastically: $U_1(\eta) - U_{3b}(\eta) \propto (16-\eta)^{5/2}|U_1|$.

A succession of the formation of charge complexes with the increase of the electron density differs for different intervals of η. At $0 < \eta < 1.32$, this succession ($eh \to e_2h \to e_3h$) is similar to one described for small η (with the replacement of the a isomer by the b isomer at $\eta \approx 0.39$). However, at $1.32 < \eta$ the fermionic e_2h complexes are not stable, and at $n_h < n_e \leq 3n_h$ *only*

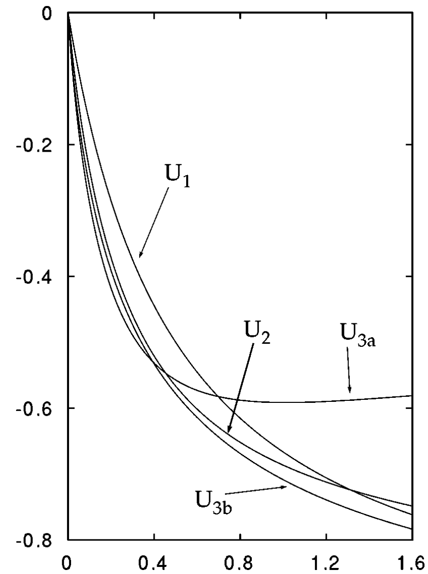

FIG. 1. Energies $\{U\}$ of the classical configurations eh, e_2h, e_3h-a, and e_3h-b in units of $|U_1| = e^2/\epsilon l$ (along the vertical axis) as functions of $\eta = d/l$ (the horizontal axis).

boson complexes eh (with the density $3n_h/2 - n_e/2$) and e_3h-b (with the density $n_e/2 - n_h/2$) may coexist at the ground state of the low density e-h system at $1.32 < \eta < \eta_c = 16$. At $\eta > 16$ only the neutral eh complexes may exist in the ground state and the rest of electrons (at $n_e > n_h$) remain free.

To increase the typical energy scale $|U_1| = e^2/\epsilon l$ of the classical configurations it is desirable to decrease the interlayer distance l. However, this would increase quantum effects and eventually would make the classical description inadequate. The criterion of the classical approach validity is $\xi \ll \rho$, where $\xi \sim \sqrt{\hbar/m\omega_0}$ is an amplitude of "zero" oscillations of the charges around their classical equilibrium positions and ω_0 is a "characteristic" frequency of intracomplex oscillations. For the range of small $\eta = d/l$ and moderate N, the oscillation frequencies differ only by numerical factors (for comparable values of e and h effective masses $m = m_e \sim m_h$) and scale as $\hbar\omega_0 \sim |U_1|da/l^2$ ($a = \hbar^2/me^2$). This gives $\xi \sim l(a/d)^{1/4}$ and determines the range of validity of the classical approach at small η:

$$a(l/a)^{4/9} \ll d \ll l. \quad (8)$$

In the intermediate range of η (i.e., $d \sim l$) the situation is more favorable. Typical intracomplex oscillation frequencies are now estimated as $\omega_0 \sim (e^2/ml^3)^{1/2}$ and the requirement $\xi \ll \rho$ reduces to

$$a \ll d \lesssim l, \quad (9)$$

which is weaker than Eq. (8). These estimations show that in the well pronounced classical regime the typical classical energy scale $|U_1| = e^2/\epsilon l$ is considerably smaller than the exciton Rydberg. However, Eqs. (8) and (9) are only sufficient but not necessary conditions. We may expect that the complexes survive even when the strong left inequalities in Eqs. (8) and (9) are replaced by the usual ones.

Quantum effects may be suppressed by application of a strong magnetic field H perpendicular to the layers plane. The magnetic field has no influence on the structure of classical configurations but it induces a rotation of the electron ring (the Ampère "persistent" current) and reduces the quantum oscillation amplitudes of the charges. At $\lambda_H = \sqrt{\hbar c/eH} < \xi \sim l(a/d)^{1/4}$, the oscillation amplitudes will be of the order of λ_H and at sufficiently strong magnetic fields the condition $\lambda_H \ll \rho \sim l(d/l)^{1/5}$ provides the existence of stable classical charged configurations even at $a \sim d \lesssim l$, i.e., when the classical energies are comparable with the exciton Rydberg. It might also be favorable to use II-VI semiconductor DQWs where a is smaller than in currently used GaAs/AlGaAs and InAs/AlGaSb DQWs [6,7].

To describe the most favorable range of parameters, the simplified quasiclassical consideration should be extended to the quantum one. More elaborate study has to be done to describe the shell structure of this new kind of "artificial atoms," to calculate the spectrum of their vibrational and rotational modes, and to fill the "table of complexes." Collective phenomena in the low-density system of complexes and, particularly, the tempting possibility of the Bose-Einstein condensation of the charged e_3h "bosons" also deserve special research.

The presence of the charged complexes may manifest in the Hall conductivity and cyclotron resonance measurements, drag experiments, and microwave absorption by intracomplex degrees of freedom. Experimental search for the charged boson complexes e_3h (or eh_3, if the MP is closer to the h layer) of the most interest would be, perhaps, more convenient for values d/l which fall into the windows $0.2 < \eta < 0.3$ (for e_3h-a isomer) and $1.2 < \eta < 1.5$ (for e_3h-b isomer): see Fig. 1. Even in these windows, the ionization energy of e_3h complexes amounts to only 2%–4% of $|U_1| = e^2/(\epsilon l)$, which means that experimental investigations of these relatively fragile objects would be more difficult than investigations of the neutral eh excitons. The efforts might be justified by a rich variety of physical phenomena in the novel world of mobile charged electron-hole complexes.

To summarize, the analysis above demonstrates the possibility of the existence of a rich family of stable charged complexes e_Nh (or eh_N) in the double-layer system near a metal plate. These mobile complexes repel each other and may exist in the ground state of the low-density e-h system. Changing densities of the carriers gives rise to a succession of the ground state transformations associated with the change of relative densities of boson and fermion complexes. In the case of the Bose-Einstein condensation of charged boson complexes, the condensate motion would be accompanied by a *nonzero total* electric current along the QW plane. Experimental realization of the suggested system would be of considerable interest.

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