Magnetic-field-induced transformations of Wigner molecule symmetry in quantum dots

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A theoretical study has been performed for the ground-state spatial configuration of Wigner molecules with $N=2,\ldots,20$ electrons in quantum dots subjected to an external magnetic field. We have shown that—for $N\geqslant 6$—the Wigner molecule formed in the magnetic field above the maximum-density-droplet instability has a different shape than in the high-field limit. We have found several magnetic-field-induced transitions between molecular phases with different spatial symmetry.

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A quantum dot (QD), a semiconductor nanostructure that confines electrons in three dimensions, can be used as a unique physical laboratory for studying properties of electron systems. In particular, the QD-confined electron systems (artificial atoms) are much more sensitive to the external magnetic field than the natural atoms.[1,2]

The charge distribution in the artificial atom is a result of an interplay between the electron-electron Coulomb interaction and the one-electron effects. If the Coulomb energy is small compared to the one-electron energy-level separation, the occupied spin orbitals are only slightly perturbed by the interaction and the electron distribution reproduces the symmetry of the external potential. If, however, the one-electron energy levels are nearly degenerate, the occupied states are superpositions of a large number of noninteracting electron spin orbitals. In this case, the electrons exhibit a pronounced tendency to be localized at different space sites. Therefore the electron distribution can lose the symmetry of the confining potential and take a form of a set of separate islands. This distribution resembles that of a classical charge-carrier system[3] and is called a Wigner crystal[4] (in the systems with translational symmetry) or a Wigner molecule[5,6] (in the QD's). The Wigner molecules can be formed in large structures,[7–9] for which the quantum size effects disappear. They can also be created in nanostructures by a strong external magnetic field.[10–15]

The application of the external magnetic field leads to relative shifts of the energy levels corresponding to different spin-orbital configurations.[2,16] In consequence, the ground-state configuration changes when the magnetic field increases. At a certain, sufficiently high, magnetic field all the electrons have parallel spins and occupy orbitals with successive magnetic quantum numbers.[13] In this state, called a maximum density droplet[17] (MDD), the electron charge distribution still possesses the symmetry of the confinement potential. If, however, the magnetic field increases further, all the occupied energy levels approach the lowest Landau level and become degenerate. Then, the Coulomb interaction leads to a rapid change of the electron distribution. As a result, the confinement-potential symmetry of the electron density is broken and the Wigner molecule is formed. The breakdown of the MDD (Refs. 18 and 19) has recently been observed[20] in the vertical gated QD.[1]

In the present paper, we consider the shape of the Wigner molecule ground state as a function of the magnetic field above the MDD regime. For this purpose, we apply a theoretical approach designed for a description of Wigner molecules in QD's at high magnetic fields. Let us first consider a single electron in a magnetic field, which is so high that the confinement potential can be neglected in the first approximation. We assume the magnetic field to be perpendicular to the QD region (x-y plane), i.e., $\mathbf{B}=(0,0,B)$, and apply the Landau (nonsymmetric) gauge for the vector potential, i.e., $\mathbf{A}=(-By,0,0)$. The one-electron Hamiltonian has the form

$$h=-\frac{\hbar^2}{2m}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+2i\alpha y\frac{\partial}{\partial x}-\alpha^2y^2\right), \tag{1}$$

where m is the electron effective mass, $\alpha=eB/\hbar=m\omega_c/\hbar$, and $\omega_c=eB/m$ is the cyclotron frequency. The ground-state energy of Eq. (1) is equal to $E_0=\hbar\omega_c/2$, i.e., the energy of the lowest Landau level, which is infinitely-fold degenerate. Due to this degeneracy the ground-state eigenfunction of Eq. (1) can be chosen in many forms. We choose the following one:

$$\Psi_{\mathbf{R}}(x,y)=(\alpha/2\pi)^{1/2}\exp\{-(\alpha/4)[(x-X)^2+(y-Y)^2] -(i\alpha/2)(x-X)(y+Y)\}, \tag{2}$$

which—for arbitrary $\mathbf{R}=(X,Y)$—fulfills the eigenequation of Eq. (1) for eigenvalue E_0. We note that the charge distribution generated by wave function (2) is the Gaussian centered at point $\mathbf{R}$, which can be treated as the center of the Landau orbit. The wave functions of form (2) are convenient for a construction of a multicenter variational basis, which is appropriate for a description of the Wigner molecules. A similar approach, but in the symmetric gauge, has been applied to the Wigner crystals.[4]

Now, we consider the system of N electrons confined in the two-dimensional harmonic-oscillator potential and subjected to the external magnetic field. This system is described by the Hamiltonian

$$H=\sum_{i=1}^{N}\left(h_i+\frac{1}{2}m\omega_0^2r_i^2+\sum_{j<i}^{N}\frac{\kappa e^2}{\varepsilon r_{ij}}\right)-\frac{1}{2}Ng^*\mu_BB, \tag{3}$$

where h_i is Hamiltonian (1) for the ith electron, $\mathbf{r}_i=(x_i,y_i)$, $r_{ij}=|\mathbf{r}_i-\mathbf{r}_j|$, $\kappa=1/4\pi\varepsilon_0$, ε is the dielectric constant, g^* is the effective Lande factor, and μ_B is the Bohr magneton. The last term in Hamiltonian (3) is the Zeeman

TABLE I. Ground-state energy of N-electron Wigner molecule for $B=20$ T. In the second (third) line the results of Ref. 10 (present paper) are listed. Energy is expressed in meV.

N	1	2	3	4	6	10	20
Ref. 10	17.25	40.09	66.44	96.46	166.35	339.93	937.97
Present	17.25	40.15	66.45	96.42	166.27	339.69	936.56

energy of N spin-polarized electrons. We apply the material parameters of GaAs, for which we take on the same values as in Ref. 10, i.e., $m=0.067m_e$, $\varepsilon=12.9$, $g^*=0.54$, and $\hbar\omega_0=3$ meV.

The N-electron problem has been solved by the unrestricted Hartree-Fock method with the one-electron wave functions expanded in multicenter basis (2),

$$\Phi(x,y)=\sum_{i=1}^{N} c_i \Psi_{\mathbf{R}_i}(x,y), \tag{4}$$

where c_i are the linear variational parameters and the basis functions $\Psi_{\mathbf{R}_i}(x,y)$ are taken on in form (2) with parameter α replaced by a nonlinear variational parameter α^*. In the wave function (4), the positions $\mathbf{R}_i=(X_i,Y_i)$ of N centers are found from the equilibrium positions $\mathbf{R}_i^c$ of the classical counterpart of the considered N-electron system as follows: $\mathbf{R}_i=\sigma\mathbf{R}_i^c$, where the scaling factor σ is treated as the second nonlinear variational parameter. It appears that $\alpha^*>\alpha$, since the QD confinement potential enhances the localization of electrons. If the exchange interaction does not vanish, the interelectron repulsion is weakened and the average electron-electron distances are diminished, i.e., $\sigma<1$.

In the parabolic confinement, the classical, equally charged, particles take on the shell-like equilibrium configurations.[3] This feature is qualitatively reproduced in the Wigner molecules. Therefore we label the spatial configurations of the Wigner molecules by the numbers of electrons in the subsequent shells, e.g., N_1-N_2-N_3 denote the state, in which N_1, N_2, and N_3 electrons occupy the inner, middle, and outer shell, respectively.

The present calculations have been performed for $N=1,\ldots,20$ electrons. The results of test calculations with basis (4) are listed in Table I and compared with those of Miller and Koonin,[10] who applied the symmetric gauge and the unrestricted Hartree-Fock method with the basis functions of the definite angular momentum. Table I shows that the present variational estimates are very close to those of Ref. 10 for $N=1,\ldots,3$ and become better for $N\geqslant 4$. We note that the present approach, in which only one basis function is associated with each electron, requires a considerably smaller numerical effort than that of Ref. 10, in which a superposition of a very large number of angular momentum eigenstates is needed to describe the islandlike charge distribution.

Figure 1 shows the ground-state energy of the six-electron system calculated with multicenter basis (4) generated from the scaled classical configurations 0-6 in which the electrons form a hexagon, and 1-5 with one of the electrons localized at the origin and five others forming a pentagon around it.

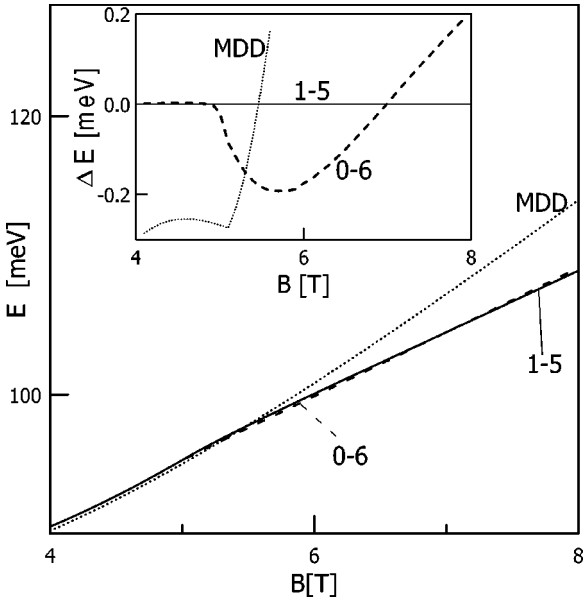

FIG. 1. Ground-state energy E of the six-electron Wigner molecule with shell-structure 1-5 (solid line) and 0-6 (dashed line), and the MDD phase (dotted line) as a function of magnetic field B. Inset shows the energy ΔE of the 0-6 Wigner molecule and MDD determined with respect to the energy of the 1-5 (high-field) phase.

The dotted line shows the results of our additional calculations for the cylindrically symmetric MDD state obtained in the symmetric gauge by the Hartree-Fock method with finite difference approach, free of a possible variational overestimation of the MDD energy. The inset of Fig. 1 shows that the energy calculated with respect to that of the 1-5 configuration exhibits characteristic cusps near $B=5.3$ T. For $B<5.3$ T the MDD state possesses the lowest energy. In the MDD-stability regime, the energy obtained with the multicenter basis for both the 0-6 and 1-5 configurations follow the MDD energy quite well. Figure 2 displays the charge-density distribution for the three configurations of the six-electron system. It is remarkable that the charge density obtained with the 1-5 multicenter basis [cf. Fig. 2(a)] well approximates the MDD charge-density distribution. The slight deviations from the circular symmetry are responsible for the overestimation of the MDD ground-state energy. If the magnetic field increases above 5.3 T, the electron system becomes the Wigner molecule with the 0-6 shell structure [cf. Fig. 2(b)]. If the magnetic field exceeds 7 T, the Wigner

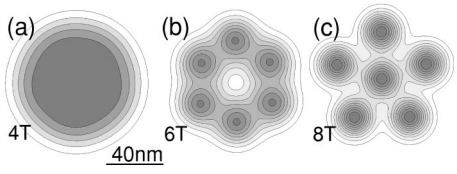

FIG. 2. Electron density distribution for six electrons in (a) approximation of the MDD phase for $B=4$ T calculated with the six-center wave function in 1-5 configuration and the Wigner molecules with shell structures (b) 0-6 for $B=6$ T and (c) 1-5 for $B=8$ T. The darker the shade of grey the larger electron density. The bar shows the length scale.

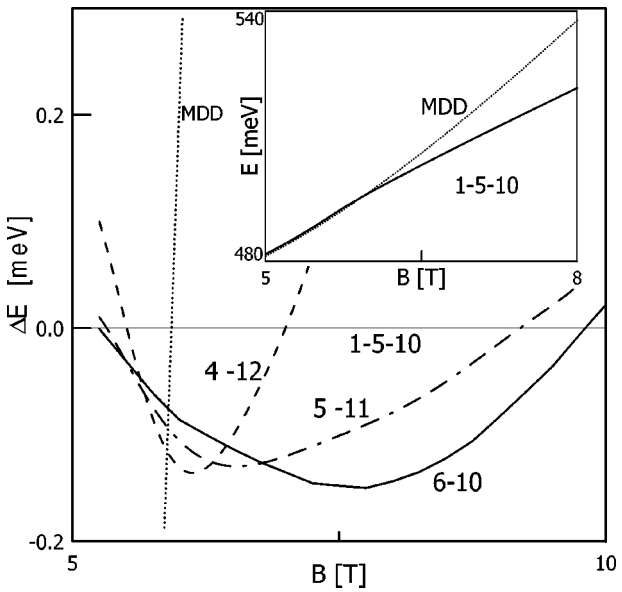

FIG. 3. Energy separation ΔE from the ground-state energy of the 1-5-10 Wigner molecule to those of the 6-10 (solid line), 5-11 (dash-dotted), and 4-12 (dashed) phases. The corresponding results for the 16-electron MDD are plotted by the dotted line. Inset displays the ground-state energy of the 1-5-10 Wigner molecule and MDD.

molecule changes its shape into the 1-5 configuration [cf. Fig. 2(c)], which is the lowest-energy configuration of the system of six classical charge carriers. The present results show that the six-electron Wigner molecule created after the breakdown of the MDD possesses a different shape and symmetry than in the high-field limit. This conclusion is consistent with the result of a recent study of the six-electron system by Maninen *et al.*[14] Kainz *et al.*[21] considered Wigner clusters in parabolic QD's using a multicenter basis in the symmetric gauge. Their vatiational wave function[21] does not allow for formation of the MDD phase, but should be quite equivalent to Eq. (4) in the high-field limit. However, the authors[21] do not discuss the transformations of the Wigner-molecule symmetry.

For the 16-electron system the results depicted in Fig. 3 show that the magnetic field induces several phase transitions. The Wigner molecule created after the breakdown of the MDD possesses the 4-12 shell structure. With the increasing magnetic field the spatial configuration changes first into 5-11 and next into 6-10 structure. Finally, in the high-field regime, the 1-5-10 shell structure is created. Figure 4 shows the charge-density distribution for the four phases of the 16-electron Wigner molecule.

Figure 5 shows the phase diagram calculated for the Wigner molecules with $N=2,\ldots,20$ electrons. We have found that for $N\geqslant 6$ the Wigner molecules undergo several phase transitions until they reach the semiclassical limit[3] of the point-charge structure. This property is related to the fact that—for $N\geqslant 6$—the classical counterpart of the system considered possesses several configurations with nearly the same energy, but different symmetry. It is remarkable that the transitions between different phases of the Wigner molecule show a distinct regularity. At lower magnetic fields, the electrons prefer to occupy the outer shells of the Wigner molecule. If the magnetic field increases, the electron charge-density islands shrink (cf. Figs. 2 and 4) and the more strongly localized electrons can fit into the inner shells of the molecule.

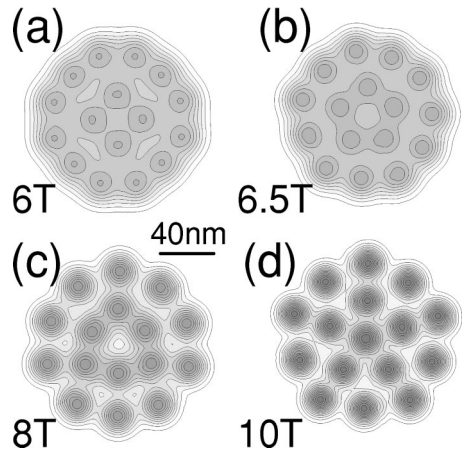

FIG. 4. Electron density distribution for the 16-electron Wigner molecule with shell structure (a) 4-12 for $B=6$ T, (b) 5-11 for $B=6.5$ T, (c) 6-10 for $B=8$ T, and (d) 1-5-10 for $B=10$ T.

In the limit of the infinite magnetic field, the charge distribution associated with Landau orbitals (2) tends towards that of the point charges. The exchange interaction vanishes along with the overlap between the different orbitals. Therefore, in this limit, the shape of the Wigner molecule becomes identical with the shape of its classical counterpart.[3] At lower magnetic field the orbitals are spread out and the overlaps between them do not vanish. In this magnetic-field regime, the shape of the ground-state electron distribution changes due to the finite extension of Landau orbitals (2) and the exchange interaction between the electrons.

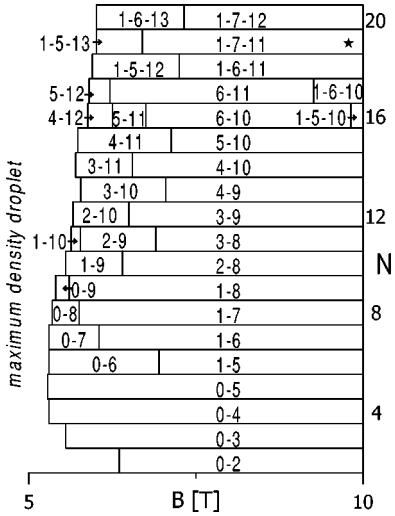

FIG. 5. Phase diagram for N-electron Wigner molecules. The lowest-energy shell structures are shown as a function of magnetic field B. The boundary of the MDD-stability region is depicted on the left part of the plot. (★) For $N=19$ the 1-6-12 phase appears at $B=15.8$ T.

The instability of the MDD phase under influence of the high magnetic field has been observed experimentally[20] in cylindrical QD's as cusps on the borders of transport windows,[16] which occur when the confined-charge distribution undergoes a reorganization from the MDD into the Wigner molecule. In the present calculations, we have assumed a rotational symmetry of the confinement potential, for which the ground state of the Wigner molecule is degenerate, since the rotation of the system by an arbitrary angle results in energetically equivalent charge distributions. Therefore we can construct the ground state with the rotationally invariant charge-density distribution by taking a superposition of the rotated Wigner-molecule states. The spatial configurations depicted in Figs. 2 and 4, without the rotation symmetry, should be understood in terms of the relative electron-electron distances. However, these lower-symmetry phases can be realized if the rotational symmetry is perturbed, for instance, by a small anisotropy of the confinement potential. In the vertical QD's,[20] the anisotropy is inevitable because of the presence of ionized impurities in the neighborhood of the QD. This anisotropy should stabilize the Wigner-molecule configuration with a fixed space orientation.

In summary, we have presented a systematic study of phase transitions in Wigner molecules induced by an external magnetic field. We have shown that for $N \geqslant 6$ the symmetry of the Wigner-molecule phase, that emerges from the MDD phase, is different than that obtained in the semiclassical, high-field limit. We predict the existence of several phases of the Wigner molecule with different symmetries, that should be observed in the magnetic-field regime above the MDD-breakdown transition. In spite of a qualitative nature of the present results, we suggest the interpretation of the additional cusps on the single-electron-transport plots, observed[20] beyond the MDD stability regime, in terms of the transformations of the Wigner molecules. It is interesting that—contrary to the two-dimensional Wigner crystal, which always posses a fixed (triangular) symmetry—the Wigner molecule can appear in several phases with a different symmetry.

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