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Wigner Molecules in Nanostructures.

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Abstract. – The charge density and the pair correlation function of interacting electrons with spin, confined within a quasi–one-dimensional «quantum dot», are calculated by numerical diagonalization. The transition from a dense homogeneous charge distribution to a dilute Wigner-type electron arrangement is investigated. The influence of the long-range part of the Coulomb interaction is explicitly studied. When the interaction is exponentially screened the «crystallized» Wigner molecule is destroyed in favour of an inhomogeneous charge distribution similar to a charge density wave.

Single charges dominate the electronic properties of submicron structures at low temperatures. Due to small capacitances the charging energy associated with the addition of one electron into a given structure can exceed the thermal energy. Two recently discovered important phenomena can be explained by single-electron charging effects: the Coulomb blockade of the d.c.-current through small tunnel junctions [1, 2], and the periodic oscillations of the conductance of quantum dots [3, 4].

In contrast to metallic systems, semiconductor nanostructures allow to reduce the number of electrons in a quantum dot by varying a gate voltage. Optical-absorption experiments were performed on quantum dots which contain only $N = 2...4$ electrons [5]. For very low electron densities additional effects in the transport properties can be expected due to the increasing importance of the Coulomb interaction [6] since the electrons tend to «crystallize» into an inhomogeneous ground state [7]. Besides Coulomb effects, linear [8] and non-linear [9] transport experiments show fine structure in the current voltage characteristics that can be traced black to the granularity of the charge density distribution and to the lowest collective excitations [10], respectively. Crystallized charge density distributions have been assumed recently [11] in the calculation of the transport properties of semiconductor nanostructures. We provide a microscopic justification of this assumption and analyse its range of validity.

In this paper we consider a quasi–one-dimensional (1D) system containing a few interacting electrons with their spin degrees of freedom. We show that the charge density

$\rho(x)$ becomes inhomogeneous for sufficiently large mean electron distance r_s. There are two consecutive regimes of charge localization. First, $\rho(x)$ starts to become inhomogeneous with increasing r_s. When r_s is further increased, $\rho(x)$ vanishes almost completely between well-separated maxima. In analogy with the Wigner lattice, we denote this limiting configuration in a finite system as a «Wigner molecule». A similar effect of the repulsive force between the electrons, but in a strong magnetic field, has been discussed recently by Maksym [12]. We identify two classes of elementary excitations in this limit: vibrations of the charge density and tunnelling between different permutational arrangements of the electrons [6, 13]. The latter are associated with changes in symmetry *and total spin* [13]. They are a consequence of strong electron correlations which are known to dominate the behaviour of low-dimensional electron systems, especially also in high magnetic fields [14]. Exact ground-state correlations in an infinite 1D system for a special type of «screened» interaction, namely $\propto r^{-2}$, but without spin, were considered, for instance, by Sutherland [15]. We show quantitatively that exponential screening of the Coulomb interaction does not destroy the nature of the correlations, and especially the spin-dependent tunnelling excitations. We argue that the correlation-induced granularity of the charge density may be visible in linear transport through quantum dots as a splitting of the conductance peaks [8] when the gate voltage is varied.

We consider N interacting electrons including spin, confined in a 1D square-well potential of finite length L. The height of the potential was assumed to be finite but large. The Hartree-Fock approximation is known [16] to overestimate the ferromagnetic state and cannot reproduce the true ground state which is antiferromagnetic in 1D [17]. We calculate eigenvalues and eigenstates of the Hamiltonian exactly.

The kinetic energy

$$E_H\left(\frac{a_B}{L}\right)^2 \sum_{n,\sigma} \varepsilon_n c^\dagger_{n,\sigma} c_{n,\sigma} \equiv E_H\left(\frac{a_B}{L}\right)^2 H_0 \tag{1}$$

and the Coulomb interaction

$$E_H\left(\frac{a_B}{L}\right) \sum_{\substack{n_1 \dots n_4 \\ \sigma,\sigma'}} V_{n_4 n_3 n_2 n_1} c^\dagger_{n_4\sigma} c^\dagger_{n_3\sigma'} c_{n_2\sigma'} c_{n_1\sigma} \equiv E_H\left(\frac{a_B}{L}\right) H_I \tag{2}$$

scale like $1/L^2$ and $1/L$, respectively. Thus, the latter dominates at large L. The natural energy and length scales are given by the Hartree $E_H = e^2/a_B$, and the Bohr radius, $a_B = \varepsilon\hbar^2/me^2$ (ε relative dielectric constant, m effective electron mass). $c^\dagger_{n,\sigma}$ creates occupation of the n-th one-electron state with energy ε_n ($= (n\pi)^2/2$ for an infinitely high well) and spin σ. $V_{n_4 n_3 n_2 n_1}$ is the matrix element of the interaction potential $V(x, x') = e^2/\varepsilon\sqrt{(x - x')^2 + \lambda^2}$. The cut-off at short distances simulates a small transversal spread $\lambda \ll L$ of the wave functions and leaves the $V_{n_4 n_3 n_2 n_1}$ finite. In the calculations we assumed $\lambda/L = 2 \cdot 10^{-4}$. The eigenvalues of $H = E_H(a_B/L)[(a_B/L)H_0 + H_I]$ depend only weakly on λ when $\lambda \ll L$ [6]. To investigate the effect of the long-range part of the interaction, in some of the calculations an exponential cut-off at large distances was introduced.

The Hamiltonian matrix was diagonalized numerically on the basis of the Slater determinants restricted to M one-electron states. The latter were constructed from the single-electron states $\varphi_n(x)\chi_\sigma$, where φ_n is a spatial function and χ_σ a spinor with $\sigma = \downarrow, \uparrow$.

Considering $1 \leqslant n \leqslant M$ leads to a matrix of the rank $R = \binom{2M}{N}$ ($R < 1.5 \cdot 10^4$, for $M = 10...17$). We used Lanczos procedures when $R > 3 \cdot 10^3$.

The charge density and the pair correlation function were determined from the normalized eigenfunctions $\psi(x_1, \ldots, x_N) = \sum_{\nu=1}^{R} (b_\nu / \sqrt{N!}) \det\{\varphi_{n_i}(x_j)\chi_{\sigma_i}\}$, where ν enumerates the configurations $\{n_1\sigma_1, \ldots, n_N\sigma_N\}$. The coefficients b_ν can be chosen real. In second quantization the charge density is given by

$$\rho(x) = \sum_\sigma \langle \psi_0^{(N)} | \Psi_\sigma^\dagger(x) \Psi_\sigma(x) | \psi_0^{(N)} \rangle, \tag{3}$$

where $|\psi_0^{(N)}\rangle = \sum_{\nu=1}^{R} b_\nu^0 |\nu\rangle$ is the N-electron ground state. $\Psi_\sigma^\dagger(x) = \sum_{n=1}^{M} \varphi_n(x) c_{n,\sigma}^\dagger$ creates an electron at position x with spin σ and $|\nu\rangle = c_{n_1\sigma_1}^\dagger \ldots c_{n_N\sigma_N}^\dagger |0\rangle$ is a non-interacting Slater determinant. Straightforwardly we have $\rho(x) = \sum_{\substack{\nu=1 \\ \nu'=1}}^{R} b_\nu^0 b_{\nu'}^0 \sum_{n,n',\sigma} \varphi_n(x)\varphi_{n'}(x) \langle \nu' | c_{n',\sigma}^\dagger c_{n,\sigma} | \nu \rangle$.

The development of maxima and minima in $\rho(x)$ with increasing mean electron distance $r_s \equiv L/(N-1)$ is shown in fig. 1 for $N = 3, 4$. The finite height of the square-well potential was assumed to be proportional to $1/L^2$, such that the number of non-interacting bound states was independent of L. This implies a non-vanishing but small $\rho(x)$ outside the box.

We distinguish three characteristic regimes of electron densities. For small distances, $r_s \lesssim 0.1 a_B$, the Coulomb interaction perturbs $E_H (a_B/L)^2 H_0$ only weakly, such that $\rho(x)$ is dominated by the lowest occupied single-particle states. This explains the minimum at $x = 0$ (fig. 1). $\rho(x)$ changes qualitatively at $r_s \gtrsim a_B$. A structure consisting of N peaks emerges. The «critical» length for this transition is of the same order as found in [13] for the transformation from an almost non-interacting energy spectrum to the multiplet spectrum dominated by interaction. When r_s increases further, say $r_s \gtrsim 100 a_B$, $\rho(x)$ vanishes almost completely in finite regions between the maxima, indicating a fully established Wigner molecule. In this

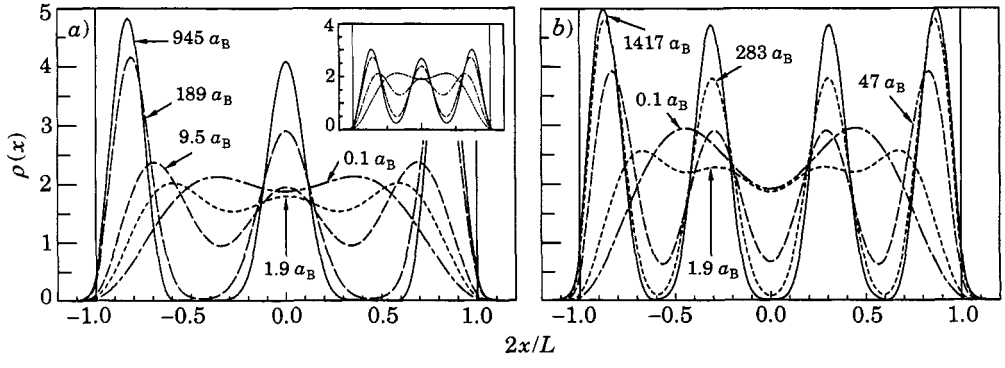

Fig. 1. – Charge density $\rho(x)$ for *a*) $N = 3$ and *b*) $N = 4$ for different L ($0.1 a_B \leqslant L \leqslant 945 a_B$, $M = 13$). The normalization is such that $\int dx\, \rho(x) = N$. When $L \gtrsim 1 a_B$ N peaks begin to emerge. For $L \gtrsim 100 a_B$ the peaks are well separated. Inset: $\rho(x)$ for a pair potential with an exponential cut-off ($\alpha = 10 a_B^{-1}$) for $N = 3$ and $L = 0.1 a_B$ (dotted line); $L = 9.5 a_B$ (dash-dotted line); $L = 472 a_B$ (dashed line); $L = 1417 a_B$ (solid line) ($M = 11$). The minima at large system lengths are less pronounced as for $\alpha = 0$.

limit the ground-state energy can be approximated by that of a chain of elementary charges at equal distances r_s.

In order to investigate the influence of the long-range part of the Coulomb interaction, we introduced an exponential cut-off $V(x, x') \propto \exp[-\alpha|x - x'|]/\sqrt{(x - x')^2 + \lambda^2}$, where $1/\alpha$ is the cut-off distance. The inset of fig. 1 shows $\rho(x)$ for $N = 3$ and $\alpha = 10a_B^{-1}$. Despite the extremely short range of the interaction, pronounced maxima are observed. However, in contrast to $\alpha = 0$ the density between the maxima is much higher even for very large r_s. The inset of fig. 1 rather resembles a charge density wave [18] than a Wigner molecule. We conclude that the long-range part of the Coulomb interaction is essential for the charge condensation and for long-range density-density correlations. The ground-state energy for $\alpha > r_s^{-1}$ does not converge towards the Coulomb energy of N elementary point charges for $L \to \infty$.

For large r_s ($r_s \gg a_B$) the spectrum consists of multiplets (1). Their energetic separation Ω corresponds to vibrational excitations. These excitations are independent of the symmetry of the spatial part of the N-electron wave function and of the total spin. Asymptotically, Ω decreases as $r_s^{-\gamma}$. If one assumes for each peak of the charge density a δ-function or a Gaussian form, then one obtains $\gamma = 3/2$ or 1, respectively, by considering the harmonic part of the Coulomb forces between the charges when $\alpha = 0$ [13]. Power law behaviour is also observed for $\alpha > 0$. However, the exponent changes from $\gamma \lesssim 1.5$ ($\alpha = 0$) to $\gamma \approx 2$ ($\alpha > 0$) (fig. 2). This can be understood by taking into consideration that for $\alpha r_s \gg 1$ the Coulombic forces are so well screened that each electron is practically moving freely within an interval of the length r_s.

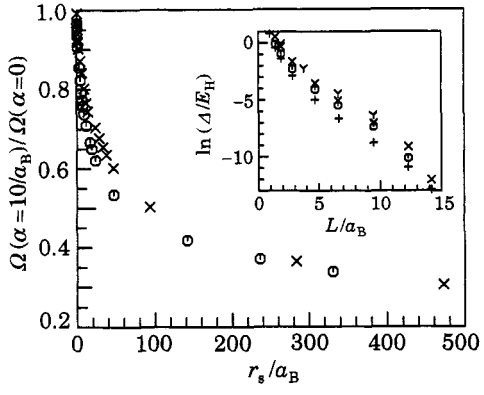

Fig. 2.

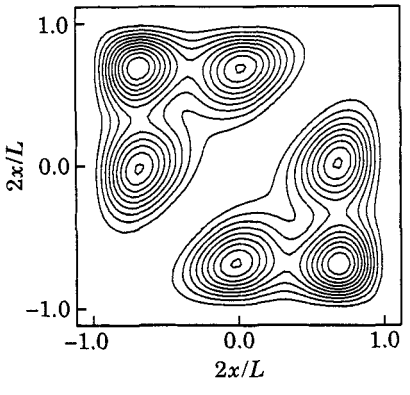

Fig. 3.

Fig. 2. – Influence of a pair potential on the energy difference between the lowest two multiplets Ω (vibrational excitation). The ratio $\Omega(\alpha = 10a_B^{-1})/\Omega(\alpha = 0)$ is plotted *vs.* the particle distance r_s for $N = 2$ (×) and $N = 3$ (○) ($M = 11$). Inset: logarithm of the energy difference Δ between the ground state and the first excited state within the lowest multiplet *vs.* system length L for $N = 2$, $\alpha = 0$ (+) and $\alpha = 10a_B^{-1}$ (○); $N = 3$, $\alpha = 0$ (×) and $\alpha = 10a_B^{-1}$(▽) ($M = 11$).

Fig. 3. – Pair correlation function $\rho_c(x, x')$ (cf. eq. (4)) for the $N = 3$ ground state ($S = 1/2$) at $L = 9.5a_B$ ($M = 13$). The contour lines correspond to equally spaced ρ_c values.

(1) There are 2^N (equal to the dimension of the spin Hilbert space) states per multiplet, differing in total spin and in symmetry with respect to permutations of the spatial space coordinates of the N-electron wave function [13].

On the other hand, the internal splitting Δ is due to tunnelling through the Coulomb barrier between adjacent permutational arrangements of the electrons [13]. Thus, Δ decreases roughly exponentially with increasing L, even for $\alpha > 0$ (inset of fig. 2) since the tunnelling barrier is still present, although its thickness, $o(1/\alpha)$, is considerably reduced as compared to the Coulombic limit. Therefore Δ is larger and the decrease is less rapid than for $\alpha = 0$ when r_s is increased. The enhancement results from the reduced thickness of the screened Coulomb barrier such that the corresponding tunnelling integral ($\propto \Delta$) increases. The modified dependence on r_s is due to the fact that only for $r_s < \alpha^{-1}$ the thickness of the interaction barrier varies with r_s. The tunnelling excitations can be described approximately in terms of correlated, localized basis functions even for $\alpha > 0$. However, in order to obtain similar quantitative agreement with the numerically determined eigenvalues as for $\alpha = 0$, a larger mean electron distance r_s is required to fulfil $\Delta \ll \Omega$. As a consequence, the characteristic length for the transition from almost non-interacting to the interaction-dominated behaviour increases with increasing α.

The pronounced correlations between the electrons in our model are also observed in the pair correlation function of the N-electron state $|\psi^{(N)}\rangle$,

$$\rho_c(x, x') = \sum_{\sigma, \sigma'} \langle \psi^{(N)} | \Psi_\sigma^\dagger(x) \Psi_{\sigma'}^\dagger(x') \Psi_{\sigma'}(x') \Psi_\sigma(x) | \psi^{(N)} \rangle . \tag{4}$$

ρ_c is related to the density-density correlation function ρ_2 via $\rho_2(x, x') = \rho(x)\delta(x - x') + \rho_c(x, x')$. Physically, ρ_c is the probability of finding an electron at position x when another electron sits at x' ($\int dx \int dx' \rho_c(x, x') = N(N-1)$). Figure 3 shows a contour plot of ρ_c for the $N = 3$ ground state ($S = 1/2$) with $L = 9.5 a_B$. Practically the same result is obtained for the spin-polarized excited state ($S = 3/2$), the difference being less than 1% independent of x and x'. In the limit of sufficiently large r_s ($\Delta \ll \Omega$) the differences between ρ_c for different levels of the same multiplet turn out to be exponentially small similar to the corresponding energy differences Δ [2].

It is known [19] that the Hartree-Fock approximation overestimates the tendency of electrons with parallel spins to avoid each other and underestimates the correlations between electrons with antiparallel spins. Therefore large differences between correlation functions for different spin configurations are expected in this approximation. Recently Pfannkuche *et al.* demonstrated this property explicitly for two electrons in a two-dimensional parabolic potential [20]. The weak sensitivity of the pair correlation function on S shows that highly correlated electrons in a «slim» quantum dot cannot be treated within molecular-field approximation.

In conclusion, we have analysed the charge density and the pair correlation function for the lowest eigenstates of $N = 2, 3, 4$ electrons confined in a quasi-1D square well. Three different regimes were identified. At high electron densities the Coulomb interaction is only a weak perturbation. Between $a_B^{-1} \gtrsim r_s^{-1} \gtrsim 10^{-2} a_B^{-1}$ the electron density concentrates around N maxima. Simultaneously the eigenvalue spectrum develops a multiplet structure that is dominated by the interaction. Both the inhomogeneity of the charge density and the multiplet character of the spectrum are not destroyed when the long-range part of the

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[2] In that limit conclusions about ρ and ρ_c of a system of repulsively interacting *bosons* can be drawn. Assuming for simplicity spinless particles, the eigenstates transform totally symmetric under permutations among the boson arrangements. The bosonic state can be regarded as one particular constituent of the vibrational multiplet (with the lowest energy on the scale Δ). Therefore the curves in fig. 1 for $r_s \gtrsim a_B$ and fig. 3 should describe respectively ρ and ρ_c of the ground state for bosons interacting via $V(x, x')$.

interaction is removed. However, there are quantitative changes in the asymptotic behaviour of tunnelling and vibrational excitations at low densities. For the lowest densities (equivalent to $r_s \geqslant 100 a_B$) the formation of a *Wigner molecule* is observed. In this limit where the long-range part of the interaction is crucial, $\rho(x)$ vanishes within finite intervals between the maxima. The structure in the density-density correlation function becomes more pronounced with increasing influence of the interaction but is less sensitive to total electron spin than expected within Hartree-Fock approximation. It is worth mentioning that, to our knowledge, the best numerical estimate for the critical value of r_s for the formation of a Wigner crystal in 3D is also $r_s^{cr} \approx 100 a_B$ [21].

Experiments are frequently performed on 2D quantum dots that are based on GaAs-AlGaAs heterostructures. In many of these the electron density corresponds to the intermediate regime. In typical quantum dots [3, 4] (area of the dot $\approx 10^5\,\mathrm{nm}^2$, number of electrons $\approx 10^2$, effective mass $\approx 0.07 m_e$, dielectric constant ≈ 10) the mean distance between electrons is about $3a_B$. Even for this relatively high electron density in comparison with more recent experiments [5], the charge density distribution cannot be expected to be homogeneous if we assume that the qualitative features of the electronic properties discussed above apply also to the 2D case. In small quantum dots that are weakly coupled to leads and that contain only a finite number of electrons, like the experimentally investigated double-barrier systems [3], a granular charge density should lead to characteristic features in linear-transport properties as a function of the gate voltage V_G. A variation of V_G not only changes the charge density or the electron number inside the dot but probably simultaneously its shape. In [4] the gate electrode is positioned besides the dot region. For this geometry the influence of V_G on the shape of the dot area is obvious. The resonance condition for the appearance of a conductance peak in linear transport, $E_F = E_{N+1} - E_N$, may be fulfilled more than once for given N when V_G is increased. Here E_F is the chemical potential of the leads (where no electron-electron interaction is assumed) and E_N the N-electron *ground-state* energy of the quasi-isolated dot. Such a model could account for the experimentally observed fine structure in the conductance peaks in the *linear*-transport regime reported in [8] which seem to be presently not yet very well understood.

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Additional Remark.

In the meantime a work by H. J. SCHULZ appeared in *Phys. Rev. Lett.*, **71** (1993) 1864, where some of the features discussed here have also been found in the 1D Luttinger liquid when a long-range Coulomb interaction is considered.

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