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Interacting electrons in a one-dimensional quantum dot

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The spectral properties of up to *four* interacting electrons confined within a quasi-one-dimensional system of finite length are determined by numerical diagonalization including the spin degree of freedom. The ground-state energy is investigated as a function of the electron number and of the system length. The limitations of a description in terms of a capacitance are demonstrated. The energetically lowest-lying excitations are physically explained as vibrational and tunneling modes. The limits of a dilute, Wigner-type arrangement of the electrons, and a dense, more homogeneous charge distribution are discussed.

INTRODUCTION

Interaction effects play a crucial role in the understanding of the electrical transport properties of very small condensed-matter systems at low temperatures.[1] Examples are (1) the Coulomb blockade,[2,3] where charging energies of single electrons suppress the current through a dissipatively shunted tunnel junction, (2) single-electron tunneling oscillations[4] of the voltage across such a junction at constant current, and (3) resonancelike oscillations of the conductance of quantum dots, being periodic in multiples of the elementary charge inside the dot.[5,6]

An important feature in all conductance measurements on tunnel junctions and quantum dots is the relative isolation of the sample region from the "external world." In the blockade experiments this is achieved by a shunt impedance representing the (phase randomizing) influence of a coupling to the electromagnetic environment. For quantum dots, weak coupling is achieved by highly resistive tunnel junctions. Here the time scale for a change of the electron number is large compared to the other inverse energies involved, namely the Fermi energy of the external wires, the charging energy, and the characteristic energy of the dissipative heat bath. Therefore, the electron number on the time scale of all relaxation processes approximately becomes a good quantum number. Due to its relative isolation, the quantum properties of the disconnected dot can be considered as one of the dominating factors in the single-electron phenomena. The Coulomb interaction should be taken into account, because the charging energy is the most relevant energy scale of the problem. The commonly used overall description by means of a capacitance C (Refs. 7 and 8) is not completely obvious[9,10] and needs to be justified. This is of particular relevance if the number of charge carriers is reduced to only about ten in semiconductor samples.[6,11] Then both Coulomb interaction and kinetic (confinement) energy start to play a role. Clarifying the interplay between these two energy contributions[12] has been one of the main motivations for our quantitative study of a model of $N \leq 4$ interacting electrons in a one-dimensional (1D) square-well potential. We shall see that the noncommutativity of the two corresponding energy operators has an important influence on the ground-state energy and on the excitation spectrum. The ground-state energy in general cannot be obtained by the charging formula $Q^2/2C$ where Q is the total charge. The excitation energies have no similarity to the discrete level structure pertaining to the confinement. We shall present a qualitative picture, which allows one to understand physically the spectrum in the correlated case.

In the following section we describe our model and the calculational method. Section III addresses the question of a capacitance definition in such correlated systems. One possibility for investigating this question is to compare the quantum-mechanical ground-state energy of N interacting, confined electrons with the charging energy of a corresponding capacitor. In contrast to previous work,[13-15] we have been able to treat up to $N=4$ electrons with high accuracy.

The excitation spectrum is discussed in Sec. IV for various electron densities. An inhomogeneous distribution is established for low charge density with essentially regularly spaced elementary charges as a consequence of the dominating Coulomb repulsion in that limit. We designate this arrangement of electrons as a *Wigner molecule,* in analogy to the Wigner electron lattice in dilute infinite systems with Coulomb interaction. Two kinds of elementary excitations are identified, *vibrational* and *tunneling* modes, that are characteristic for dilute and intermediate densities, respectively.

According to experimental situations, we conclude that for the system parameters that characterize inversion-layer-based quantum dots, the quantum mechanics of the electron-electron interaction cannot be completely discarded. In particular, it should be possible to observe the features related to the existence of the low-lying correlated excitations by suitable sample fabrication.

INTERACTING ELECTRONS IN A SQUARE-WELL POTENTIAL

For the electron-electron interaction, we use the form

$$V(x,x') \propto \frac{1}{\sqrt{(x-x')^2+\lambda^2}} , \tag{1}$$

which behaves Coulombically at large distances. λ is a measure for the width of the electron wave functions in the transverse direction. In most of our calculations we take $\lambda/L = 2\times10^{-4} \ll 1$, where L is the system length. Then the eigenvalues of the Hamiltonian

$$H = E_H \frac{a_B}{L}\left[\frac{a_B}{L}H_0 + H_I\right] \tag{2}$$

depend only weakly on λ. $E_H = e^2/a_B$ is the Hartree energy, $a_B = \varepsilon\hbar^2/me^2$ the Bohr radius, ε the relative dielectric constant, and m the electron mass. In most cases we present eigenvalues of the operator enclosed by round brackets. The relative importance of the kinetic energy in the 1D square-well potential

$$H_0 = \sum_{n,\sigma} \epsilon_n c^{\dagger}_{n,\sigma} c_{n,\sigma} \tag{3}$$

($\epsilon_n \propto n^2, n$ integer) decreases as compared to the Coulomb energy

$$H_I = \sum_{n_1,\ldots,n_4,\sigma_1,\sigma_2} V_{n_4n_3n_2n_1} c^{\dagger}_{n_4\sigma_1} c^{\dagger}_{n_3\sigma_2} c_{n_2\sigma_2} c_{n_1\sigma_1} \tag{4}$$

with increasing system length L. The matrix elements $V_{n_4n_3n_2n_1}$ are real and do not depend on the electron spin σ. The total spin S is therefore conserved and all eigenvalues are $(2S+1)$-fold degenerate. The symmetry relations are $V_{n_4n_3n_2n_1} = V_{n_4n_2n_3n_1} = V_{n_1n_3n_2n_4} = V_{n_3n_4n_1n_2}$ and $V_{n_4n_3n_2n_1} = 0$ if $\sum_i n_i$ is odd. For $\lambda/L \ll 1$,

$$V_{n_4n_3n_2n_1} \approx -8\pi \int_0^{\infty} dq[\ln(q\lambda/L) + C]\hat{f}_{14}(q)\hat{f}_{23}(q) ,$$

where $\hat{f}_{ij}$ is the Fourier transform of the product $\varphi^*_{n_i}(x)\varphi_{n_j}(x)$ of the one-electron eigenfunctions of H_0, and $C = 0.577$, the Euler constant.

For the numerical diagonalization, single-particle states $c^{\dagger}_{n\sigma}|0\rangle$ with $1 \le n \le M$ were chosen (usually $M = 9, \ldots, 17$, depending on the calculation). The properly antisymmetrized, noninteracting N-particle basis $\psi^{(N)}_{\nu}$, including the spin degree of freedom, is of dimensionality $R = \binom{2M}{N}$, $1 \le \nu \le R$. In our calculations, R was restricted to 1.5×10^4, even using Lanczos procedures. To avoid loops over all R^2 matrix elements of the Hamiltonian, we used the following economic procedure to occupy the matrix. Starting from the $(N-2)$-particle basis, the application of two creation operators onto a certain $\psi^{(N-2)}_{\mu}$ generates, say, the N-particle state $\psi^{(N)}_{\nu}$ with proper sign. $\psi^{(N)}_{\nu}$ corresponds to a certain row ν of the Hamiltonian matrix. Creating from the same $\psi^{(N-2)}_{\mu}$ a (different or the same) $\psi^{(N)}_{\nu'}$ identifies a certain column ν'. The *independent* summation over all possible two-particle excitations and subsequent summation over all $(N-2)$-particle states generates eventually all nonvanishing entries [including sums from $n_4 = n_1$ and/or $n_3 = n_2$, cf. (4)] of the Hamiltonian matrix.

Typical examples of N-electron energy spectra are shown in Fig 1. In the presence of interaction, $N > 1$, the density of states is much more inhomogeneous as a function of the energy. The lowest eigenvalues form *multiplets* of extremely small width when $L \gg Na_B$. The total number of states within each of these multiplets, including degeneracies, is 2^N.

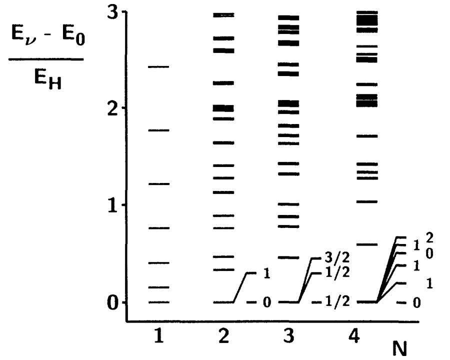

FIG. 1. Typical spectra of model (2) for various N and $L = 9.45a_B$. For $N \ge 2$ the low-lying eigenvalues form groups of (fine-structure) multiplets, the total number of states per multiplet being equal to the dimensionality of the spin Hilbert space 2^N. For clarity, the lowest multiplets are magnified indicating the total spin of each level. The ground-state energy is subtracted, respectively.

GROUND-STATE ENERGIES

Figure 2 shows the dependence of the ground-state energy per particle E_0/N on the particle number N for different L. The data are multiplied by L in order to eliminate the trivial L dependence. The charging model would yield a straight line for the ground-state energy as a function of the particle number $E_0(N)$ when plotted in the same way. In very small systems E_0/N deviates from a linear $(N-1)$ dependence due to the discreteness of the spectrum of H_0. On the other hand, for systems with large L the formation of an inhomogeneous charge density (Wigner molecule, see below) prevents the ground state of few electrons energy to obey $E_0/N \propto (N-1)$. A better approximation is obtained by considering the Coulomb energy of N point charges at equal distances

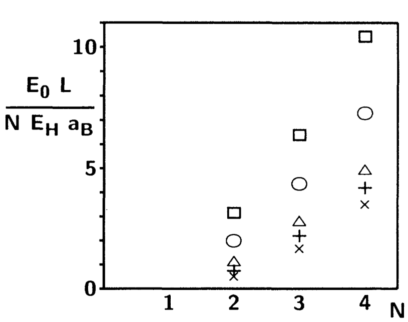

FIG. 2. Ground-state energies per particle E_0/N multiplied by L/a_B vs the particle number N for $L = 6.61a_B$(□), $L = 16.1a_B$(○), $L = 94.5a_B$ (△), $L = 944.8a_B$ (+). (×) denotes the energy of N fixed-point charges equally spaced at distances $L/(N-1)$. The quantum-mechanical ground-state energies (slowly) approach these values as $L \to \infty$.

$r_s = L/(N-1)$.

The importance of this charge "crystallization," which is a consequence of the charge quantization, for the capacitance per unit length C/L, can be visualized for equidistant point charges e in 1D with a charge-density distribution

$$\rho(x) = \sum_{j=1}^{N} \delta(x - x_j) \ , \quad x_j = \frac{j-1}{N-1} L \ .$$

For a total charge Ne the capacity of such an arrangement is defined as

$$C(N) := (Ne)^2 / 2U \ ,$$

where

$$U = e^2 \sum_{\substack{i,j \\ i \neq j}} \frac{1}{|x_i - x_j|}$$

is the charging energy. For $N \geq 3$,

$$U = \frac{e^2}{L}(N-1) \sum_{j=1}^{N-1} \frac{j}{N-j} = \frac{e^2}{L} N(N-1) \sum_{j=2}^{N} \frac{1}{j} \ ,$$

and therefore

$$C(N)/L = \frac{N}{2(N-1)} \left[\sum_{j=2}^{N} \frac{1}{j} \right]^{-1} \ .$$

In contrast to the classical capacitance of a homogeneously charged and long cylinder, this capacitance per unit length is independent of L but explicitly dependent on the charge. In particular, for small N the classical concept of a capacitance obviously in inapplicable. Also in higher dimensionalities we expect considerable fluctuations of the capacitance as a function of the charge due to an inhomogeneous charge density.

On the other hand for short systems with a more homogeneous charge distribution, quantum-mechanical corrections to the ground-state energy also do not allow the use of the capacitance formula. In Fig. 3, E_0/N is shown as a function of r_s. The deviations from the capacitancelike $1/L$ behavior occurring below $r_s \lesssim 50 a_B$ cannot be attributed to a simple additive influence of the kinetic part of the Hamiltonian. The ground-state energy, of the noninteracting system has already been subtracted in Fig. 3.

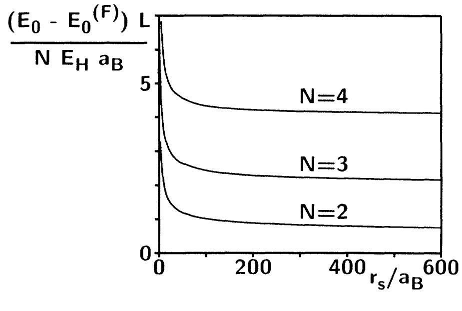

FIG. 3. Ground-state energy per particle multiplied by L/a_B as a function of the electron distance $r_s := L/(N-1)$; the corresponding noninteracting ground-state energy is subtracted. Pronounced deviations from the Coulombic $1/L$ behavior occur below $r_s \lesssim 50 a_B$. These deviations cannot be attributed to a simple additive influence of the kinetic part of the Hamiltonian.

LOW-LYING EXCITATIONS

For $L \gg N a_B$ the spectrum of the low-lying excitations can be understood using the picture of a Wigner molecule. The one-particle density shows N approximately equidistant peaks.[16] One type of excitation in such an arrangement is of a phonon kind due to the Coulomb forces between the charges. Similar to the one-particle density these excitations are insensitive to the total spin and the symmetry properties of the wave function.

To estimate the asymptotic behavior of typical phonon frequencies Ω as a function of the electron distance r_s, we assume δ functions or Gaussians for in the charge density of each peak. The Gaussians can be considered to emerge from the harmonic-oscillator ground-state wave functions due to the linearized electrostatic potential for one certain electron, leaving the other electrons fixed. The result of this crude estimate is an $r_s^{-\gamma}$ decrease of Ω with $\gamma = \frac{3}{2}$ (δ-function density) or $\gamma = 1$ (Gaussian density). Figure 4 shows $\Omega L / a_B$ as a function of r_s for different N, Ω is the distance between the lowest two multiplets in our spectra. The behavior at large r_s is consistent with $\gamma \gtrsim 1$. This indicates that the charge-density distribution is more localized than a Gaussian. For $r_s \lesssim 100 a_B$ the strong deviations from the asymptotic behavior signal the breakdown of the Wigner molecule.

The multiplets consist of a series of energy levels which, compared to Ω, are extremely close to each other. Each level is $(2S+1)$-fold degenerate since all eigenstates of H are simultaneously eigenstates to $\hat{S}^2$. For $N \geq 3$ the states are, in general, not products of a spatial part and a spin part.[17] The Pauli principle only requires their transformation according to the totally antisymmetric irreducible representation of the symmetric group S_N.

In the following a qualitative understanding for the fine-structure spectrum within one multiplet is developed, starting from the notion of the Wigner molecule. Quantitative results will be published elsewhere.[18] The total potential, including the interaction (1), has $N!$ equivalent

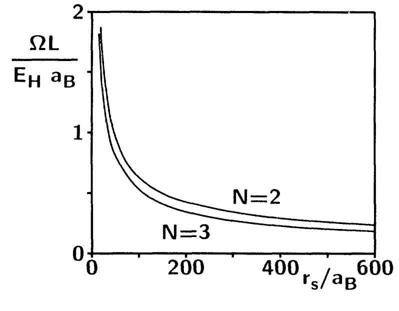

FIG. 4. Energy difference Ω between the two lowest multiplets, multiplied by L/a_B vs the mean particle distance r_s for $N=2$ and $N=3$. For $r_s \gtrsim 100 a_B$ the asymptotic power-law behavior is recovered.

minima in the configuration space which is an N-dimensional hypercube L^N. There are no further equivalent minima, because S_N is the *only* symmetry group of the problem. For sufficiently low electron densities (the corresponding r_s will be quantified by numerical results below), the probability amplitude of the eigenfunctions is well localized around the vicinity of the potential minima. The barrier of the Coulomb interaction in the 1D case separates adjacent minima.

Each of the probability amplitude peaks may be taken as one of the basis functions of a finite dimensional "pocket-state basis"[19] $\{|j\rangle\}$, with $1 \le j \le N!$. Matrix elements of the Hamiltonian in this basis $\langle j|H|j'\rangle \equiv H_{jj'}$ mutually connect different permutations of N-electron states and behave like tunneling integrals. The smallness of the $H_{jj'}$ is crucially required to approximate the true interacting eigenfunctions of H by eigenvectors of the Hamiltonian in this truncated basis. They are given by certain linear combinations of the $\{|j\rangle\}$. Correspondingly, the true energy eigenvalues are approximated by eigenvalues of the pocket-state Hamiltonian matrix. Since the amplitudes of the $|j\rangle$ decrease roughly exponentially with the distance from its center, the $H_{jj'}$ also have this property and thus the pocket-state approach improves with increasing r_s. It is important to note that this description is not based on any single-particle basis set as a starting point. It becomes increasingly reliable with increasing influence of the interaction. However, the diagonalization of the Hamiltonian matrix in this basis cannot be achieved by Fourier transformation, since S_N is not abelian.

As a further approximation, we neglect all $H_{jj'}$ except those that correspond to nearest neighbors in the configuration space L^N. For sufficiently large r_s this is justified by the exponentially fast decay of the pocket state far away from its center. Due to the symmetry of the problem, all of the remaining tunneling integrals $H_{jj'} = t$ must be equal and are the only nontrivial entries in the Hamiltonian matrix. Therefore, the differences between the eigenvalues scale with the common factor t.

In Fig. 5 the energy difference Δ between the two lowest numerically obtained eigenvalues is plotted versus the system length L. The assumption of an asymptotically exponential decay of the pocket states suggests that $\Delta \propto \exp(-L/L_\Delta)$ with $L_\Delta \approx 1.5a_B$ from Fig. 5. This establishes a length scale beyond which the spectrum of noninteracting electrons is changed into one showing the multiplet structure that is characteristic for the influence of the Coulomb interaction. The tunneling energies Δ depend on λ, because the height of the barriers between the potential minima are proportional to λ^{-1} [cf. (1)].

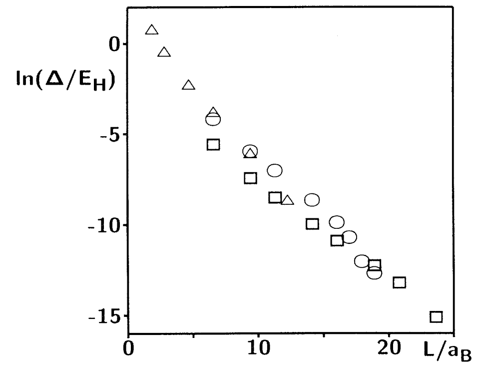

FIG. 5. Logarithm of the energy difference Δ between the ground state and the first excited state within the lowest multiplet vs the system length for $N=2$, $M=11$ (□), $N=3$, $M=13$ (○), and $N=4$, $M=10$ (△). From the slope we estimate $L_\Delta \approx 1.5a_B$.

CONCLUSION

We have calculated numerically the energy spectra of up to $N=4$ electrons confined in a quasi-one-dimensional square-well potential of finite length. The discussion in terms of the pocket-state basis suggests that our classification of the energy eigenvalues should remain valid also in 2D or 3D systems, if the width of the system does not exceed the width of one pocket-state wave function. For $N=2$ we can reproduce the fine structure in the lowest multiplet calculated by Bryant[13] with its sequence $S=0,1,0$ of total spins and with equal level distances for sufficiently large size of a rectangular two-dimensional quantum dot, which is ten times longer than it is wide.

We have demonstrated that the ground-state energies $E_0(N)$ deviate from the N^2 behavior assumed in the charging model because of the formation of a Wigner-molecule-like structure at sufficiently low electron densities (quantization of the charge) and the quantum-mechanical influence of the kinetic energy (noncommutativity of H_0 and H_I). Only in sufficiently large systems *and* at sufficiently high electron densities can a capacitancelike behavior be obtained.

We have obtained three different regimes for the electron densities to characterize the excitation spectra. The Wigner molecule is found to be fully established for densities $1/r_s$ below $10^{-2}a_B^{-1}$ (Fig. 4; see also Ref. 16). Nevertheless, the description of the interacting spectrum in terms of the pocket-state picture does already hold at much larger electron densities. Only for $L \lesssim L_\Delta \approx 1.5a_B$, the confinement energy starts to dominate the Coulomb energy and the spectrum approaches the noninteracting limit (Fig. 5). Neither the ground-state energy nor the level spectrum is given by a sum of kinetic and potential energy eigenvalues separately.

Experiments are frequently performed on $Al_xGa_{1-x}As$/GaAs-based heterostructures which rather correspond to a 2D situation. It is not obvious how far our 1D classification for the length and energy scales of few Coulombically interacting electrons can be applied to that case. If we assume that at least the qualitative aspects of our classification into different regimes for the electron density can be used, the intermediate regime should apply in most circumstances. Given the geometry and the electron numbers in typical quantum dots[5] (area of the dot $\approx 10^5$ nm^2, number of electrons $\approx 10^2$ effective mass $\approx 0.07m$, dielectric constant ≈ 10), a mean distance of $r_s \approx 3a_B$ can be estimated. For this relatively high electron density and number, the *ground-state energy,* which is in first approximation the relevant quantity that enters a dc-transport experiment, can roughly be estimated by using the charging model. However, the *excitation energies* are qualitatively different from the ones expected within the noninteracting picture. They are importantly characterized by the fine-structure-level spectrum and the total spin. In experimental situations as they have been realized recently by Meurer, Heitmann, and Ploog[11] with only a few electrons per quantum dot, the charging model cannot even be expected to yield correct results for the ground-state energy.

It should be possible to observe the excitation spectrum obtained in this paper in optical measurements[20,11] (if the potential is not strictly harmonic[15]) and in nonlinear transport measurements[12] at low temperatures. At least for quasi-1D geometries, where the spatial width of the dot region is of comparable size or smaller than the width of the pocket-state wave function, we predict a close relationship between the energies of low-lying excitations and the number of electrons in the dot. Only a finite number of tunneling-type excitations should exist for fixed electron numbers. The energy scale for a 100-nm-long 1D structure on $Al_xGa_{1-x}As/GaAs$ basis can be estimated from Fig. 1 to be $\Delta \approx 0.1-0.5$ meV for $N=2,\ldots,4$. However, this value still depends on the effective width of the structure.

For the nonlinear transport measurements two remarks should be made: (i) the excitation spectra of N and $(N+1)$ electron states are completely different—even the typical splittings may differ by almost an order of magnitude. They are not equidistant and independent of N; (ii) the total spin of the electrons inside the dot region can be changed only by $\pm\frac{1}{2}$ when an electron is added or removed at the finite conductance situation. This reduces the number of transport channels available. For example, the conductance channel connected with the energy difference $E_{S=3/2}^{(N=3)} - E_{S=0}^{(N=2)}$ [$\mu_d(N=2)$ in the nomenclature of Ref. 12, S is the total spin] should not appear.

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