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Abstract.— Equilibrium geometries of quasi two-dimensional jellium systems are calculated in the local density approximation, closely following the "Ultimate Jellium Model" of Ref. [1]. The background charge is assumed to be fully deformable in a layer between two parallel planes, whereas the wave functions in the direction perpendicular to such a "jellium slice" are confined to their ground state. Like for jellium clusters in three dimensions [1], we find that for various system sizes, a trend towards a breaking of axial and inversion symmetries is observable.

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1 Introduction

The concept of shell structure is a very general one, and today many systems on the mesoscopic size scale are known which exhibit pronounced shell effects. Perhaps one of the most beautiful examples of electronic quantum confinement are metallic clusters in their close analogy to the nuclear shell model [2]. Using the Jellium model in the framework of density functional theory, the enhanced stabilities for clusters with a "magic" size [3] were easily explained [4, 5] by spherical shell closures: For a filled shell, the density of states at the Fermi surface has a minimum and thus leads to particularly stable configurations. For non-closed shells, however, a high level density near the Fermi surface is energetically unfavourable. Without external constraints, the system tends to stabilize itself by spontaneously breaking its symmetry. This is most easily achieved by the mechanism of Jahn-Teller deformations [6]. It opens up an

energy gap at the Fermi surface and thus results in a lower total energy. While this behaviour is well investigated for the case of three-dimensional Fermi systems like nuclei [2] and metal clusters [7], it seems interesting to us to study Jahn-Teller deformations for systems with reduced space dimensionality.

Keldysh suggested in 1967 that excitons can condense into droplets of an electron-hole Fermi liquid [8]. It was already pointed out in [1] that the ultimate jellium model is valid for the description of such electron-hole drops, if the effective masses of the electrons and holes are the same. Having in mind two-dimensional excitonic states, which are found to be considerably lower in energy than the three-dimensional exciton in the same semiconductor material [9], it is of interest to apply the 3D calculations of [1] to the quasi-2D case. Clearly, a direct comparison between systems like electron-hole liquids in semiconductors and the model suggested here is not possible. We believe, however, that the results obtained, as particularly the potential energy surfaces and the various minimum shapes, are of general nature. For the three-dimensional case, it was already shown [10] that even the shapes of the lightest nuclei show a striking similarity to those of jellium clusters [1]. We expect that a similar analogy holds for the quasi-2D Fermion systems.

In the present work, we calculate free shape relaxations of a jellium which is confined between two parallel planes. We hereby restrict ourselves to the discussion of systems with even particle numbers, using the methods of density functional theory in the local density approximation (see [11] for a review) and apply the ultimate jellium model of Ref. [1].

Recently, similar calculations for planar Jellium clusters, including a discussion of spin effects, have been performed by Kohl *et al.* [12]. In the present work, we avoid any symmetry restrictions for the electronic densities by using a plane wave basis [1].

Our calculations reveal that the shapes of the slices show a trend towards breaking of axial and inversion symmetries. For small systems with up to 34 valence electrons, only few of the magic configurations are found to be of circular shape. Strong octupole-like deformations occur frequently and are often very likely corresponding to the ground-state geometries.

2 The quasi two-dimensional model

Our model for the quasi two-dimensional electron gas clusters is directly based on the so-called Ultimate Jellium Model of Ref. [1] for the three-dimensional case. There, it was shown that if the background charge is completely deformable, it everywhere equals the density of the valence electrons. Thus,

there are no Coulomb contributions to the total energy, and the system is bound by the exchange-correlation energy only.

Consider now an “ultimate jellium slice” which is confined between two planar, parallel surfaces of distance z_0 . In order to make the actual calculation to be strictly two-dimensional, we fix the Kohn-Sham single particle wave functions in z -direction and require zero wave function at the planar surfaces. Thus, we set

$$\phi(z) = \sqrt{\frac{2}{z_0}} \cos\left(\frac{\pi z}{z_0}\right). \quad (1)$$

The total single-particle wave functions are then given by

$$\phi_i(\mathbf{r}) = \phi_i(x, y) \phi(z). \quad (2)$$

Here, we consider only even electron numbers N . Thus, all states $i = 1, 2, \dots, N/2$ are doubly occupied. The electron density is then given by

$$\rho(\mathbf{r}) = 2 \sum_{i=1}^{N/2} |\phi_i(\mathbf{r})|^2. \quad (3)$$

The total energy of the slice can be written as a functional of the electron density ρ or, more precisely, as a functional of ϕ_i :

$$E[\phi_i] = T[\phi_i] + E_{xc}[\rho]. \quad (4)$$

The total kinetic energy of the electrons $T[\phi_i]$ equals the sum of the kinetic energy contributions of all occupied single-particle states. The exchange-correlation functional $E_{xc}[\rho]$ is given by

$$E_{xc}[\rho] = \int d\mathbf{r} \rho(\mathbf{r}) e_{xc}(\rho(\mathbf{r})), \quad (5)$$

where the Perdew-Zunger parametrization [13] of the Ceperley-Alder [14] correlation energy e_{xc} is used.

Minimizing the total energy functional Eq. (4) with respect to the “free” wave functions $\phi_i(x, y)$ then leads to the Kohn-Sham [15] equations for the quasi-2D case,

$$\left[-\frac{\hbar^2}{2m_e} \nabla_{xy}^2 + V(x, y) \right] \phi_i(x, y) = (\varepsilon_i - t_z) \phi_i(x, y), \quad (6)$$

where

$$V(x, y) = \int_{-z_0/2}^{z_0/2} dz |\phi(z)|^2 V_{xc}(\rho(\mathbf{r})) \quad (7)$$

corresponds to the z -averaged exchange-correlation potential V_{xc} and $t_z = \hbar^2 \pi^2 / (2m_e z_0^2)$ is the kinetic energy of the electrons in the z -direction.

3 Method of calculation

For the iterative solution of the two-dimensional Kohn-Sham-equations Eq. (6), we used the plain wave basis

$$\phi_{\mathbf{k}} = L^{-1} e^{i\mathbf{k} \cdot (x, y)} \quad \text{with} \quad \mathbf{k} = \frac{2\pi}{L} (n_1, n_2), \quad (8)$$

where $n_i = k, k-1, \dots, -k$. The integer k determines the number of plane waves, and L is the side length of the mesh. In coordinate space, we use $4k+1$ points per axis with a spacing of $L/(4k+1)$.

The iteration is started by making an initial guess for the two-dimensional potential $V(x, y)$ at the lattice points. Then, the Fast Fourier Transform (FFT) is used to expand the potential in the plane-wave basis, and the Hamiltonian matrix is computed. After diagonalization, the FFT is again used for getting the single-electron eigenfunctions in the space-lattice points. The electron density is now easily obtained from Eq. (3). The new potential $V(x, y)$ is obtained from Eq. (7) by performing the z -integration numerically. The whole procedure is then repeated until full convergence is obtained, and no symmetry restrictions are imposed.

The numerical calculations are made with $z_0 = 3.9a_0$, i.e. the Wigner seitz radius of sodium. For electron numbers up to $N = 34$, practically exact solutions were obtained by using $L = 57a_0$ and $k = 14$. The resulting Hamiltonian matrix was then of dimension $29 \times 29 = 841$.

The initial potential was chosen to be of square-well type. Using polar coordinates, its radial dependence was defined by the expansion

$$R(\varphi) = R_0 [1 + \sum_n \epsilon_n \cos(n\varphi)]. \quad (9)$$

For each system, the iteration was then started from six different geometries of the initial potential, selected to represent basic two-dimensional shapes. They correspond to quadrupole ($n = 2$), octupole ($n = 3$) and hexadecapole ($n = 4$)-like deformations of a circle ($\epsilon_n = 0$). The latter was chosen as the simplest first guess of the initial potential. For the set of quadrupole-deformed initial shapes with $n = 2$, we chose $\epsilon_2 = 0.2$, which corresponds to a stadium-like shape. A weak octupole deformation yielding a triangular-like shape is obtained for $\epsilon_3 = 0.1$. A square with rounded corners corresponds to $\epsilon_4 = 0.1$. A pear shape is obtained by a combination of quadrupole and octupole deformations, setting the parameters to $\epsilon_2 = 0.2$ and $\epsilon_3 = 0.3$, and a cross-like shape is finally obtained with $\epsilon_4 = 0.6$.

To make sure that during the selfconsistent solution of the Kohn-Sham equations the system could find a deformation which differs from the initial shape, a weak random perturbation was added to each initial potential.

4 Results

The calculated ground state energies as a function of particle number N are shown in Fig. 1 below. A contour plot of the self-consistent densities $\rho(x, y) = 2 \sum_{i=1}^{N/2} |\phi_i(x, y)|^2$ resembling the shapes is shown in Fig. 2 (see next page).

Figure 1: Ground-state energies as a function of electron number N . Weak shell closures, resulting in only a small dip, are found at the magic numbers of both a disk and harmonic oscillator in two dimensions.

For the smallest slices with up to ten electrons, only one possible equilibrium geometry was found: The calculations started with the six different initial potentials defined above all converged into the same ground-state. Shell closures occur at $N = 2, 6$ and 12 , corresponding to the first three magic numbers of both a two-dimensional harmonic oscillator and a circular square well [12, 16].

Figure 2: Contours of the self-consistent densities $\rho(x, y)$, resembling the possible ground-state geometries of the quasi-2D jellium slices. The lowest contour is drawn at a density $\rho = 0.005a_0^{-2}$, and the highest at $\rho = 0.028a_0^{-2}$, where a_0 is the Bohr radius. The diameter of the perfect disk with $N = 34$ is $d \approx 33a_0$, giving the length unit in this figure.

The system with $N = 6$ breaks inversion symmetry and is of octupole-like shape.

Going from inversion-symmetric shapes at $N = 8$ and $N = 10$, for the next shell closure at $N = 12$ again an almost triangular shape is energetically preferable. We now find a large number of isomers having only a small energy difference to the ground-state.

Similar to the 3D model of Ref. [1], we find that those slices that lack inversion symmetries for their ground state are very soft against deformations of odd multipolarity. In these cases, a fairly high number of iterations was needed to obtain full self-consistency.

Table 1 shows for which of the initial potentials the lowest energies (labelled "gs") were obtained. It lists the number of isomers together with their energy differences to the ground state for systems with $N \geq 10$. From this table it can be seen that in some cases (particularly for $N = 26$ and $N = 28$), only one of the six different initial potentials could lead to the lowest energy and therefore does not necessarily correspond to the true ground-state.

$N = 14$ shows a combination of octupole- and hexadecapole-like deformations, whereas $N = 16$ is again found to be inversion-symmetric. The change in geometry from $N = 18$ to $N = 20$, corresponding to a shell closure in the two-dimensional oscillator, is remarkable: For $N = 18$, one finds a triangular geometry with a cusp at one of its corners, which for $N = 20$ is filled up: Here, a triangular ground-state shape similar to the case $N = 12$ occurs.

For $N = 24$, one finally obtains a perfect circular shape, corresponding to a shell closure in a circular, hard-walled potential [12, 16]. A particular similarity between the shapes for $N = 18$ and $N = 28$, as well as $N = 20$ and $N = 30$ is observed, with $N = 30$ again corresponding to a harmonic oscillator shell closure and having triangular geometry.

In agreement to the results of Kohl *et al.* [12], the next "disk" shell closing is found for $N = 34$, again corresponding to a perfectly circular ground-state shape.

For very strong deformations of the initial potential, as for example two circles which are almost separated, we found that one is easily trapped in a local minimum of the potential energy surface. Indeed, with such starting geometries the calculations quickly converge to isomeric configurations having much higher energy than those isomers that are energetically close to the ground state. This happens in particular for those slices with $N \geq 16$. In none of the cases, however, very strongly deformed initial potentials lead to lower binding energies than those which were already found by the weakly deformed starting geometries which are given in the table below.

N	circle	stadium	triangle	square	pear	cross
10	gs	gs	gs	gs	gs	gs
12 (HO,D)	gs	i1 (11.8)	gs	gs	gs	i1
14	i1 (1.02)	gs	i1	gs	gs	gs
16	gs	gs	gs	i1 (6.51)	gs	i1
18	gs	gs	gs	gs	gs	i1 (4.73)
20 (HO)	gs	i1 (6.07)	gs	gs	i2 (6.57)	i3 (6.97)
22	gs	i1 (0.70)	gs	gs	i2 (1.41)	i3 (1.87)
24 (D)	gs	i1 (5.95)	gs	gs	gs	i2 (6.94)
26	i2 (1.45)	gs	i1 (0.12)	i2	i1	i2
28	i1 (1.54)	i2 (3.12)	gs	i1	i2	i3 (3.31)
30 (HO)	i2 (2.61)	i1 (2.05)	gs	gs	i3 (4.23)	i3
32	i1 (0.74)	gs	i1	i1	gs	i2 (2.70)
34 (D)	gs	i1 (5.59)	gs	gs	i1	i2 (8.77)

Tab. 1: Possible ground-state configurations (labelled "gs") and isomers (labelled "i" in sequence of lowest energy) for the clusters $N = 10$ to $N = 34$. Magic numbers of both harmonic oscillator ("HO") and hard-wall disk ("D") are indicated by bold numbers. The differences [in meV] in the relative total energies $E/N - t_z$ to the ground-state configuration are given below each classification.

It was pointed out in [12] that the transition from a soft and oscillator-like effective potential to a more hard-walled potential begins around $N = 24$. This agrees with the fact that $N = 24$ has a circular ground-state shape.

The particle numbers $N = 20$ and $N = 30$ correspond to closed shell

configurations in the two-dimensional harmonic oscillator, and we find a mixing of shell sequences in the intermediate range between a harmonic and hard-wall effective potential.

5 Conclusions

Motivated by the strong analogies between 3D Fermi liquids like nuclei and metal clusters, we studied free shape relaxations of quasi-2D jellium systems. Making use of the "ultimate jellium model" [1], we studied the shell structure and Jahn-Teller deformations [6] of "jellium slices" with up to $N = 34$ valence electrons, avoiding any symmetry restrictions by using a plane-wave basis for the iterative solution of the Kohn-Sham equations.

A map of shapes that most likely correspond to the ground-state configurations was given. From the systematics of the shape evolution it became clear that both the shell structure of the two-dimensional harmonic oscillator on the one hand, and a hard-walled disk potential on the other hand coincide in the region between $N = 12$ and $N = 34$.

Kohl *et al.* [12] pointed out that the planar confinement of the jellium clusters diminishes the shell effects strongly. They found that in the range between $N = 10 - 30$ the shell effects have a only a minor effect on the groundstate energies. This is confirmed by the present calculations, where no symmetry restrictions are imposed.

An extension of the present calculations to odd particle numbers and larger sizes, together with systematic study of the pronounced shape isomerism above $N = 34$, will be given elsewhere. Present calculations show that the potential energy surfaces of very large systems are far too soft, such that a clear decision on the ground-state shapes and form isomers is hard to make. Instead of few isomeric shapes, many configurations are found that rather resemble a ground-state landscape having more or less equal energy, than to individual Jahn-Teller shapes.

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