Oscillation Modes of Two-Dimensional Nanostructures within the Time-Dependent Local-Spin-Density Approximation

Antonio Puente and Llorenç Serra
Departament de Física, Universitat de les Illes Balears, E-07071 Palma de Mallorca, Spain
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We apply the time-dependent local-spin-density approximation to describe ground states and spin-density oscillations in the linear response regime of two-dimensional nanostructures of arbitrary shape. For this purpose, a frequency analysis of the simulated real-time evolution is performed. It is shown that the recently proposed spin-density waves in the ground state of certain parabolic quantum dots lead to the prediction of a novel class of excitations, soft spin-twist modes with energies well below that of the spin dipole oscillation.

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Recent advances in semiconductor technology nowadays allow the fabrication of nanostructures with many different shapes. In these systems the electrons, which are laterally confined at the semiconductor boundary, form a two-dimensional quantum dot with a shape which, to a certain extent, follows that of the nanostructure. This opens up the exciting possibility to produce and study an enormous variety of quantum dots, or artificial atoms as they are often called. For instance, it has been shown that the electronic structure in the small vertical quantum dots of Ref. [1] is given by the successive filling of shells obeying Hund’s rules as in atoms. Very relevant information about electronic excitations in quantum dots is also presently obtained from sophisticated far-infrared absorption [2] and light scattering experiments [3].

Up to now, the great majority of experimental and theoretical efforts were focused on quantum dots with circular symmetry. Many of the properties of circular dots are well reproduced by considering the electrons as confined by a parabolic potential, or by a simple jellium disk. To treat the electronic interactions, besides exact diagonalization for very small dots [4], the most successful approaches have been mean field theories like Hartree-Fock (HF) [5] and density functional in the local-spin-density approximation [6,7] (LSDA).

The latter ones have been extended using the random-phase approximation (RPA) to analyze collective excitations [5,8]. To our knowledge, all theoretical approaches addressing collective excitations in 2D quantum dots are limited from the start by the circular symmetry assumption. In this Letter we show how LSDA can describe both ground state and linear response of 2D quantum dots of arbitrary shape by using, respectively, energy minimization and real-time simulation of the spin-density oscillations as basic principles. We will show how from the response frequencies in the different channels (density, spin, and free responses) it is possible to gain information about the system deformation in a quantitative way. Besides, we will also analyze the effect on the response of the recently proposed spin-density waves (SDW’s) in the ground state of particular parabolic quantum dots. They could generate soft spin-twist modes, at energies well below that of dipole spin oscillation.

Several authors have recently addressed the problem of describing quantum dot ground states within LSDA. In particular, in Ref. [6] the single particle Kohn-Sham equations for electrons in a parabolic potential were solved avoiding any symmetry restriction by using a plane-wave basis. We use here the same LSDA functional of Ref. [6]; however we employ a different technique based on the discretization of the $xy$ plane in a grid of uniformly spaced points. For each spin ($\eta = \uparrow, \downarrow$) the Kohn-Sham equations read

$$\left(-\frac{1}{2}\nabla^2 + v^{(\text{eff})}({\bf r})\right)\varphi_{i\eta}({\bf r}) = \epsilon_{i\eta}\varphi_{i\eta}({\bf r}),$$

where $v^{(\text{eff})} = v^{(\text{conf})} + v^{(H)} + v^{(xc)}$ contains the confining, Hartree, and exchange-correlation potentials, the latter given by the local energy density as $v^{(xc)} = \partial\Xi_{xc}(\rho, m)/\partial\rho_\eta$. We have defined total density and magnetization, in terms of the spin densities $\rho_\eta({\bf r}) = \frac{1}{2}\sum_i |\varphi_{i\eta}({\bf r})|^2$, as $\rho = \rho_\uparrow + \rho_\downarrow$ and $m = \rho_\uparrow - \rho_\downarrow$, respectively.

As a test of the numerical code using the $xy$ grid we have checked that for a circular dot (namely, the parabolic one confined by $\frac{3}{2}m_0\hbar^2 r^2$, with $r$ the radial coordinate, $\omega_0 = 0.25\hbar^2$ [9] and $N = 20$ electrons) we find the same solution that is obtained by solving only the radial equation and imposing $e^{-iH_0t}$ as the angular part of the single particle wave functions. Next, we have considered different confining geometries for dots with $N = 20$ electrons. In particular, we present here results for a deformed parabola $\frac{1}{3}m(\omega_0^2 x^2 + \omega_0^2 y^2)$ with $\omega_x = 0.75 \omega_z = 0.22\hbar^2$; for a square jellium with $r_s = 1.51$ that corresponds to side length $L = 11.96a_B$; and for a rectangular jellium with the same $r_s$ and sides $L_y = 0.75 L_x = 10.37a_B$. We leave for future work a more systematic investigation of ground states and will concentrate here on the spin-density oscillations.

The previously mentioned ground states are shown in Fig. 1. In the deformed parabola, the density shows an
ellipsoidal shape with an aspect ratio similar to $\omega_y/\omega_x$. Three rows of local maxima can be seen in the inner part of the dot, aligned with the long axis. For the square jellium we obtain a rather abrupt electron density, with maxima at the corners and four additional inner maxima following the square symmetry. A similar structure is seen for the rectangle. Quite interestingly, while for the deformed parabola the magnetization vanishes everywhere; for the square and rectangle there is a magnetization wave in the ground state. The amplitude of this wave is approximately 15% and 25% of the maximum density, respectively. This finding is similar to the SDW predicted by Ref. [6] in some circular parabolic dots.

The description of spin-density oscillations in quantum dots has raised great interest, mainly due to the manifestation of these modes in far-infrared absorption and in Raman scattering experiments [2,3]. We refer here to general spin-density oscillations. When both spin components oscillator in phase they produce density modes and when they are out of phase, spin modes. In circularly symmetric dots, density modes have been studied using the Hartree [10] and Hartree-Fock [5] methods. More recently the LSDA to density functional theory has been used in circular dots to describe density and spin channels [8], taking into account the coupling between both. All these methods are based on the RPA perturbative treatment of the response by diagonalizing the residual interaction within a space of particle-hole excitations. They share as an essential ingredient the angular momentum selection rules given by the circular symmetry.

To describe spin-density oscillations in an arbitrary structure the RPA approach becomes practically unfeasible due to the lack of symmetry, which forces one to deal with enormous matrices $\chi(x, y, x', y'; \omega)$ in the formal RPA equation $\chi = \chi^{(0)} + V_{ph} \chi$, where $\chi^{(0)}$ is the independent particle correlation function and $V_{ph}$ is the residual particle-hole interaction. The calculation is also complicated due to the breaking of degeneracies with deformation that greatly increases the number of different particle-hole pairs contributing to $\chi^{(0)}$. An alternative approach that permits one to overcome these problems is based on real-time methods. These originate in fact from the time-dependent HF theory, and have been applied with success to nuclear [11] and to cluster physics [12]. In what follows we briefly comment on this approach and show how it applies to 2D nanostructures.

In the small amplitude limit, it is well known that both real-time (TDHF, TDLSDA) and RPA methods based on the corresponding ground states become equivalent.

We have performed TDLSDA calculations by integrating the time-dependent Kohn-Sham equations

$$i \frac{\partial}{\partial t} \varphi_{i\eta}(\mathbf{r}, t) = h_{\eta}[\rho, m] \varphi_{i\eta}(\mathbf{r}, t),$$

where $h_{\eta}$ is given by the square bracket in Eq. (1). We have integrated Eq. (2) using the Crank-Nicholson algorithm (for time step $n$ to $n + 1$)

$$\left(1 + \frac{i \Delta t}{2} h_{\eta}^{(n+1)}\right)\varphi_{i\eta}^{(n+1)} = \left(1 - \frac{i \Delta t}{2} h_{\eta}^{(n)}\right)\varphi_{i\eta}^{(n)}.$$ (3)

This is an implicit problem for $\varphi^{(n+1)}_{i\eta}$, since $h_{\eta}^{(n+1)}$ depends on the orbitals through the density and magnetization. In practice this forces one to proceed by iteration: with $h_{\eta}$ from the previous time step, a first guess of the new wave functions is obtained solving (3). These are then used to build a new $h_{\eta}$ and restart iteration. Using a small $\Delta t$ it is enough to make a double solution for each time step. If $h_{\eta}$ were constant in time, the algorithm would be exactly unitary. We have found that with small $\Delta t$ norm conservation is fulfilled with excellent accuracy.

In order to excite the oscillation modes of the system an initial perturbation of the wave functions is needed. Physically, this corresponds for instance to the interaction with a short laser pulse or with an appropriate projectile. In the calculation, it can be mimicked simply by a rigid translation of the wave functions with the operator $T(a) = e^{-i a \cdot \mathbf{r}}$ or by an initial impulse with $\Pi(q) = e^{-i q \cdot \mathbf{r}}$. When either $a$ or $q$ is small these perturbations induce dipole oscillations predominantly and the system’s response is restricted to the linear regime. Total density and spin modes are obtained with the rigid translations $a_1 = a_1 = a$ and $a_1 = -a_1 = a$, respectively, and analogously with impulse conditions. After the initial perturbation we keep track of the time-dependent dipole moments $d_i = (\mathbf{e} \cdot \mathbf{r})_i$ for density modes and $(\mathbf{e} \cdot \mathbf{r} \sigma_z)_i$ for spin ones. Here $\mathbf{e}$ corresponds to the direction of the initial perturbation given by $a$ or $q$. 

![Figure 1](image-url)
A frequency analysis of the dipole signal \(d_t\) gives the response frequencies of the system. Fourier transform methods can be used for this purpose. However, we have found more efficient a method of direct peak fitting to the simulated signal. We perform a least squares minimization of 
\[
\chi^2 = \sum_i [d_i - D(t)]^2 ,
\]
where \(D(t)\) is given by
\[
D(t) = \sum_{n=1}^{N} A_n \cos(\omega_n t) + B_n \sin(\omega_n t) .
\] (4)

In Eq. (4) a fixed number of frequencies is assumed. The minimization yields the amplitudes \(A_n, B_n\), and frequencies \(\omega_n\). Of course, it must be checked that the number \(N\) of frequencies is large enough to provide a good reproduction \(D(t)\) of the time series \(d_t\) and that convergence with increasing \(N\) has been reached. From the fitted \(D(t)\) we obtain \(D(\omega)\) as a discrete set of Dirac delta functions. In practice, these are smoothed into Lorentzians and the response strength is obtained as \(S(\omega) = |D(\omega)|\), or the power spectrum as \(P(\omega) = |D(\omega)|^2\).

We have performed the response calculation in real time for the same dots with 20 electrons discussed above. An excellent agreement with the RPA calculation was obtained for the circular parabola. Figure 2 shows the three responses for the deformed parabola, for which the magnetization \(m(r)\) is vanishingly small. The density response has only two peaks that coincide with the parabola frequencies \(\omega_x = 0.29H^*\) and \(\omega_y = 0.22H^*\). This is showing that TDLSDA satisfies the generalized Kohn’s theorem for a deformed parabola. Since the density dipole operator couples only to the center of mass motion, absorption in this channel can only take place exactly at the frequencies \(\omega_x\) and \(\omega_y\). The situation is completely different for the free and spin responses. They are rather fragmented and lie at lower energies. Peaks associated to oscillation along different axes are shown in different line types. The free response corresponds to keep the effective confining potential \(v^{(\text{left})}\) in (2) fixed to its static value. It thus models the oscillation of noninteracting particles in the static mean field. By comparing free, density, and spin responses we see the different nature of the residual interaction in both channels: weakly attractive in the spin response and repulsive in the density one. Figure 2 also shows the simulated time series in each case, as well as the fitted signal which on the plot scale superposes to the simulated one.

Figure 3 shows the corresponding results for the \(N = 20\) electron dots in the square and rectangular jellium. The density response of the square is still characterized by a very dominant peak that, nevertheless, is slightly fragmented. The free and spin responses are more fragmented. Quite interestingly, the spin response of the square shows four groups of peaks with an almost constant separation of \(0.05H^*\). In the rectangle case, the density response is clearly showing different oscillation frequencies in \(x\) and \(y\) directions. The same fact is an additional source of fragmentation for the free and spin channels.

In circular parabolic dots LSDA predicts spontaneous symmetry breaking ground states, of the type of SDW’s, for particular sizes [6,13]. This LSDA spin-density wave is more pronounced in quasi-one-dimensional rings [14].

FIG. 2. Simulated and fitted time series (left), and strength functions in arbitrary units (right) for the deformed parabola corresponding to free, density, and spin excitations. See text.

FIG. 3. Strength functions in arbitrary units for a square and a rectangular jellium. Free, density, and spin responses are shown. For the square, only the total spectrum is displayed since \(x\) and \(y\) oscillations are essentially degenerate.
In Ref. [15] similar SDW’s, as well as Wigner crystallized ground states, have been very recently reported in circular parabolic dots using an unrestricted Hartree-Fock approach. There is, however, a continuing discussion about the interpretation and possible relevance of these states [6,13,15].

We present in the rest of this Letter the TDLSDA result for the linear oscillations of a dot with \( N = 24 \) electrons in circular parabolic confinement (\( \omega_0 = 0.24H^* \)), for which there is a static SDW in the LSDA ground state [6]. We have found that the dipole oscillations of spin densities are quite similar to those obtained from a fully circular model [8]. However, the SDW can sustain a new type of oscillation. It is given by an alternating rotation of both spin densities in opposite directions, i.e., a spin twist of the static wave excited with the rotation operator \( \mathcal{R}(\theta,\nu) \), with \( \theta_1 = -\theta_1 \) being opposite rotation angles for each spin. The frequency of this mode is obtained analyzing the time evolution of the circular currents that appear after an initial rotation with \( \mathcal{R}(\theta,\nu) \). Figure 4 shows the strength of the spin-twist mode, in comparison with the normal spin dipole mode. Spin-twist modes are very soft, with energy well below the spin dipole one. They could signal the existence of static SDW’s in circular dots [16]. The spin-twist frequency is reflecting the curvature of the energy minimum corresponding to the symmetry broken ground state with respect to the circular one. We expect that circular systems having strong SDW’s in their ground states will also exhibit enhanced spin-twist modes. This happens in circular parabolic dots with increasing \( r_s \) for a fixed \( N \) [6] (the \( N \) systematics with fixed \( r_s \) is less clear) or in quasi-one-dimensional rings [14].

In conclusion, we have shown that TDLSDA can be applied to obtain the oscillation frequencies of nanostructures with arbitrary shape. It leads to the prediction of soft spin-twist modes in dots with circular parabolic confining and having static SDW’s in their ground state.

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[9] We use effective atomic units corresponding to GaAs values, i.e., \( H^* \approx 12 \) meV, \( \omega_0 = 98 \) Å and \( \tau^* = 55 \) fs as energy, length, and time units.
[16] Different from the circularly symmetric confining potential, in noncircular confinements (deformed harmonic oscillator, square, rectangle, ...), spin-twist modes will exist irrespective of the existence of a SDW in their ground state. This is because in deformed systems the circular symmetry is obviously broken.