Multipole excitations in quantum dots

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(March 24, 2022)

Abstract

We have employed time-dependent local-spin density theory to analyze the multipole spin and charge density excitations recently found in GaAs-AlGaAs quantum dots [C. Schüller et al, Phys. Rev. Lett 80, 2673 (1998)]. The overall agreement between theory and experiment is good, identifying the angular momentum of the modes observed in the experiment. We have found that high multipolarity spin density edge modes originate from interband transitions instead that from intraband transitions, as it happens in the dipole case.

PACS 73.20.Dx, 72.15.Rn
The characteristic single particle and collective excitations of typical quantum dots (QD) are known to lie in the far-infrared (FIR) energy region, i.e., they have energies that, depending on the size of the dot, span the range from a few tens of meV to a fraction of meV. Experimental information about FIR spectra was first obtained from photon absorption experiments on InSb and on GaAs quantum dots [1,2]. Since the confining potential for small dots is parabolic to a good approximation, and in the FIR regime the dipole approximation works well, the absorption spectrum is rather insensitive to the number of electrons in the dot, measuring to a large extent only the center-of-mass excitations, which at non zero magnetic fields (B) correspond to the two allowed dipole transitions arising from each of the two possible circular polarizations of the absorbed light. Two limitations of the absorption process, namely that it is dominated by the L = 1 multipole of the incoming electromagnetic wave, and its insensitivity to the electronic spin degree of freedom, have motivated that theorists have been mostly concerned with the study of dipole charge density excitations (CDE), although calculations of higher multipolarity density modes exist in the literature (see for example Refs. [3–6]).

The situation is changing with the use of inelastic light scattering to experimentally study QD excitations [7–9]. In a way, these studies complement the similar ones carried out in the past on the two dimensional electron gas (2dEG) [10]. Besides opening the possibility to study the wave vector dispersion, inelastic light scattering allows to disentangle charge from spin density (SDE) and single particle excitations (SPE), and to observe them all in the same sample.

Very recently, the B dispersion of CDE’s and SDE’s of different multipolarities has been experimentally determined in GaAs-AlGaAs quantum dots [11]. We present here a theoretical interpretation of these results based on the time-dependent local-spin density theory (TDLSDT) which addresses for the first time the description of high multipolarity spin modes.

To this end, we have obtained the ground state (gs) of an N = 200 electron dot confined by a uniform, positively charged disk of R = 120 nm solving the appropriate Kohn-Sham (KS) equations. These values correspond to a quantum dot thoroughly studied by Schüller et al [11]. The exchange-correlation energy density $\mathcal{E}_{xc}(\rho, m)$, where $\rho$ is the electron density and $m$ the spin magnetization, has been constructed from the results of Ref. [12] on the nonpolarized and fully polarized 2dEG using the two dimensional von Barth and Hedin prescription [13] to interpolate between both regimes. The only free parameter in the calculation is the number of positive charges in the disk, which has been set to $N^+ = 404$ to reproduce the dipole SDE at $B = 0$. The range of $B$ values investigated in this work corresponds to filling factors larger than 3.

Once the KS gs has been worked out, we have determined the induced densities originated by an external multipole field employing linear-response theory. Since we have described at length the dipole longitudinal response in dots [14], we give here only a few details for presentation purposes. For independent electrons in the KS mean field, the variation $\delta \rho^{(0)}_\sigma$ induced in the spin density $\rho_\sigma$ ($\sigma \equiv \uparrow, \downarrow$) by an external spin-dependent field $F$, whose non-temporal dependence we denote as $F = \sum \sigma f_\sigma(\vec{r}) |\sigma\rangle \langle \sigma|$, can be written as
\[
\delta \rho^{(0)}_\sigma(\vec{r}, \omega) = \sum_{\sigma'} \int d\vec{r}' \chi^{(0)}_{\sigma\sigma'}(\vec{r}, \vec{r}'; \omega) f_{\sigma'}(\vec{r}') ,
\]

where \(\chi^{(0)}_{\sigma\sigma'}\) is the KS spin density correlation function for independent electrons. In this limit, the frequency \(\omega\) corresponds to the harmonic time dependence of the external field \(F\) and of the induced \(\delta \rho^{(0)}_\sigma\). Eq. (I) is a 2\(\times\)2 matrix equation in the two-component Pauli space. In longitudinal response theory, \(F\) is diagonal in this space, and its diagonal components are written as a vector \(F \equiv \begin{pmatrix} f^\uparrow \\ f^\downarrow \end{pmatrix}\). We consider the external \(L\)-pole fields

\[
F^{(\rho)}_{\pm L} = r^L e^{\pm iL\theta} \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad \text{and} \quad F^{(m)}_{\pm L} = r^L e^{\pm iL\theta} \begin{pmatrix} 1 \\ -1 \end{pmatrix}
\]

which cause, respectively, the charge and spin density \(L\)-modes. For the monopole \(L = 0\) mode, these fields are simply taken proportional to \(r^2\). To differentiate the induced densities of each excitation channel they will be labelled with an additional superscript as \(\delta \rho^{(0,\rho)}_\sigma\) or \(\delta \rho^{(0,m)}_\sigma\).

The TDLSDT induced densities are obtained solving the equation

\[
\delta \rho^{(A)}_\sigma(\vec{r}, \omega) = \delta \rho^{(0,A)}_\sigma(\vec{r}, \omega) + \sum_{\sigma_1 \sigma_2} \int d\vec{r}_1 d\vec{r}_2 \chi^{(0)}_{\sigma\sigma_1}(\vec{r}, \vec{r}_1; \omega) K_{\sigma_1\sigma_2}(\vec{r}_1, \vec{r}_2) \delta \rho^{(A)}_{\sigma_2}(\vec{r}, \omega) ,
\]

where either \(A = \rho\) or \(A = m\), and the kernel \(K_{\sigma\sigma'}(\vec{r}, \vec{r}')\) is the residual two-body interaction.

Equations (II) have been solved as a generalized matrix equation in coordinate space. Taking into account angular decompositions of \(\chi^{(0)}_{\sigma\sigma'}\) and \(K_{\sigma\sigma'}\) of the kind \(K_{\sigma\sigma'}(\vec{r}, \vec{r}') = \sum_{\ell} K_{\sigma\sigma'}^{(\ell)}(r, r') e^{i\ell(\theta - \theta')}\), it is enough to solve this equation for each multipole separately because only modes with \(\ell = \pm L\) couple to the external \(L\)-pole field. One has

\[
K^{(\ell)}_{\sigma\sigma'}(r, r') = \frac{2}{\pi^{3/2}} \frac{\Gamma(\ell + 1/2)}{\Gamma(\ell + 1)} \frac{r^{\ell}}{r^{\ell} + 1} K_{\ell}(r) + \frac{\partial^2 E_{xc}(\rho, m)}{\partial \rho_\sigma \partial \rho_{\sigma'}} |_{gs} \frac{\delta(r - r')}{2\pi r} ,
\]

where \(K_n(x)\) is given by the hypergeometric function \([13] \frac{\pi}{2} \, {}_2F_1(1/2, n + 1/2; n + 1; x^2)\).

For a polarized system having a non zero magnetization in the gs, the \(\pm L\) modes are not degenerate and give rise to two excitation branches with \(\Delta L_z = \pm L\), where \(L_z\) is the gs orbital angular momentum. The induced charge or magnetization densities corresponding to density and spin responses are given by \(\delta \rho^{(A)} = \delta \rho^{(A)}_\uparrow + \delta \rho^{(A)}_\downarrow\) and \(\delta m^{(A)} = \delta \rho^{(A)}_\uparrow - \delta \rho^{(A)}_\downarrow\). From them, the dynamical polarizabilities in the density and spin channels are respectively given by

\[
\alpha^{(\rho)}_{\rho\rho}(\ell, \omega) = \int drr^{\ell+1} \delta \rho^{(\rho)}_\ell(r) \\
\alpha^{(m)}_{mm}(\ell, \omega) = \int drr^{\ell+1} \delta m^{(m)}_\ell(r) .
\]

For each \(L\) value, taking into account both \(\pm L\) possibilities we define \(\alpha^{(L)}_{AA}(\omega) \equiv \alpha_{AA}(L, \omega) + \alpha_{AA}(-L, \omega)\). Their imaginary parts are proportional to the strength functions \(S^{(L)}_{AA}(\omega) = \text{Im}[\alpha^{(L)}_{AA}(\omega)]/\pi\).
Figures 1 and 2 represent the spin and charge strength functions for $L = 0$ and 2, respectively. In the quadrupole case we have indicated with a $+(-)$ sign the SDE's arising from the $+L(-L)$ component of the $F^{(m)}$ operator in Eq. 2 (the low energy CDE is always a $+$ type excitation, whereas the high energy CDE are $-$ type excitations arising from the corresponding component of $F^{(o)}$). We have found that the spin peaks are rather fragmented, especially in the monopole case. However, they still are collective modes, with energies redshifted from the free electron ones due to the attractive character of the exchange-correlation vertex corrections.

We would like to draw the attention to the $-$ type, low energy quadrupole SDE which is seen in Fig. 2 to carry an appreciable strength at $\omega \sim 3.1$ meV and $B = 2$ T. When a magnetic field is perpendicularly applied to a QD, it is well known that the low energy excitation modes in the density channel are dipole edge CDE’s arising from intraband transitions while bulk, interband transitions lie higher in energy. That may change with increasing $L$, and it is particularly easy to see that this is the case if one looks at the SDE’s. An inspection to the KS single electron energies shown in Fig. 3 reveals that at high $L$’s, interband electron-hole excitations are at lower energies than intraband ones. Since the residual electron-hole interaction is weak in this channel, we have found that at $B = 2$ T, the lowest energy octupole SDE is indeed a mode built from interband electron-hole excitations. Still, this is an edge mode, as its existence is only possible because of the finite size of the system.

Figures 4 and 5 display the $B$ dispersion of the more intense SDE’s and CDE’s, respectively. The solid symbols represent the experimental data [11]. It can be seen from these figures that the overall agreement between theory and experiment is good. In both spin and charge density channels, TDLSDT reproduces the weak $B$ dependence of the $L = 0$ mode found in the experiment at small $B$ values. Our calculations confirm the $L = 0$, 1, and 2 multipolarity assigned in the experiment to the lower SDE’s, but cannot identify the origin of the higher SDE. Ruling out the possibility that it is an $L = 3$ or 4 SDE (see Fig 4), it might correspond to a $L < 3$ mode which gets some strength when the dot is probed by an excitation operator carrying a finite momentum $q$ on the dot plane, such as a Bessel function $J_L(qr)$ instead of the $r^L$ multipole we have been using. Indeed, the signal of that peak is weak and broad, as mentioned in Ref. [11].

At $B = 0$, the energies of the $L > 0$ spin density excitations follow the simple rule $E_L \sim LE_1$. We attribute this to the weakness of the residual interaction in the spin channel. The prominent role played by the strong residual interaction in the charge density channel causes that rule to fail for CDE’s.

As a general trend, the strength carried by the positive $B$ dispersion branch corresponding to the high $L$ spin density excitations diminishes as $B$ increases. We have also found that the spin strength becomes more fragmented with increasing $L$, whereas the bulk and edge magnetoplasmons associated with the $\pm L$ excitations are well defined modes.

The positive $B$ dispersion branches of the CDE’s reveal a complicated pattern at intermediate $B$ values, quite different from the expected classical one holding up to $B \sim 2$-3 T, but that however fits a large set of the experimental modes. The behavior of these branches has an interesting quantal origin, namely the formation of well defined Landau bands for magnetic fields larger than a critical value. Above it, the more intense high energy collective peaks mostly arise from transitions between Landau bands whose index $M$ differs in one unit, $\Delta M = 1$. Since these bands are made of many single electron states with different $\ell$
values and energies rather \( \ell \) independent if \( B \) is high enough [16], this explains the otherwise striking quasi \( L \)-degeneracy of the plasmon energies, only broken by finite size effects and the \( L \) dependence of the residual interaction. Other modes with \( \Delta M = 2 \) build branches satellite of those formed by the more intense \( L \)-peaks, which are clearly seen in the calculation. Satellite branches of this kind appear even in the dipole case [2,17], and are a clear signature of nonparabolic confinement.

This work has been performed under grants PB95-1249 and PB95-0492 from CICYT, Spain, and 1998SGR00011 from Generalitat of Catalunya. A.E. and M. B. (Ref. PR1997-0174) acknowledge support from the DGES (Spain).
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FIGURES

FIG. 1. Monopole strength function in arbitrary units as a function of energy. The thick solid line represents the charge density strength, the dashed line the spin density strength, and the thin solid line the free electron strength.

FIG. 2. Same as Fig. 1 for the quadrupole mode.

FIG. 3. Single electron energies as a function of orbital angular momentum $\ell$ for $B = 2$ T. The horizontal line represents the electron chemical potential. Full, upright triangles correspond to $\sigma = \uparrow$ states, and the empty, downright triangles to $\sigma = \downarrow$ states. Interband and intraband transitions with $\Delta \ell = 2, 3$ and 4 are represented to illustrate the energy crossing discussed in the text.

FIG. 4. Energy of the more intense SDE’s as a function of $B$. The lines are drawn to guide the eye, and the solid symbols represent the experimental data [11].

FIG. 5. Same as Fig. 4 for the more intense CDE’s.
$\omega$ (meV)

$L=0$

$B = 2\ T$

$1.5\ T$

$1\ T$

$0.5\ T$

$0$
