Single-particle-like states in few-electron quantum dots

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We investigate theoretically Raman spectra of few-electron quantum dots. Spectra obtained by an exact many-body treatment and by a time-dependent local-density approximation are compared. We show that single-particle-like excitations can be expected for systems with only six electrons. The energies of these excitations are close to the Kohn-Sham level spacings.

Resonant Raman scattering by low-dimensional electron systems is still controversially discussed. Since the pioneering work of Pinczuk *et al.*,[1] one usually refers to the electronic excitations as charge-density (CDE's), spin-density (SDE's), and the so-called single-particle excitations (SPE's). These three types of excitations are experimentally distinguished by polarization selection rules and have been measured in two-dimensional (2D),[1] 1D,[2,3] and 0D systems.[4–6] Quasiparticle excitations have been known for a long time from experiments on bulk semiconductors,[7] and consequently the energy of the broad peak, lying energetically between the SDE and the CDE of a GaAs-$Al_xGa_{1-x}As$ quantum well, was identified with the single-particle subband spacing. Excitations that showed all the characteristics of SPE's were also found in 1D and 0D systems, even though it is known that in the extreme limits of a 1D Luttinger liquid[8] and a two-electron quantum dot[9] no single-particle-like features occur in the spectra. It is therefore of interest to examine the scattering mechanism of the SPE's, and the question if there are any single-particle-like excitations in the many-body spectrum of low-dimensional systems.

According to the theory of Blum[10] there are two important ingredients for the calculation of the resonant Raman cross section: the two-particle correlation function and the valence-band structure. The correlation function captures all the many-body effects of the electron system in the conduction band whereas the energies of the valence-band states determine the resonant enhancement of the scattering amplitudes. Recent work concentrated on the Raman scattering of 1D systems[8,11] and on the effect of the resonant enhancement in quantum dots.[12] In calculations within the local-density approximation[12] (LDA) it was shown that in extreme resonance additional modes appear in the spectrum of a 12-electron quantum dot. These excitations were found to lie energetically close to the Kohn-Sham single-particle energy differences and were therefore called SPE's. Many-body effects were treated within the LDA (Refs. 13–16) of the density-functional theory.[17–19]

In this paper we concentrate on the other aspect of resonant Raman scattering, namely, the many-body electron-electron interaction. We will show that single-particle-like excitations can be expected in systems with only six electrons. Exact numerical diagonalizations are compared to the results of an LDA calculation. The calculated polarized and depolarized spectra are quantitatively very similar, which proves that the SPE-like excitation is not an artifact of the LDA.

We consider a circular symmetric parabolically confined GaAs quantum dot at a magnetic field of $B=0$ T. The extent of the electronic wave function in growth direction is neglected. Throughout the paper we use the following parameters: $\hbar\omega_0=6$ meV for the confinement energy of the dot, $m^*=0.07m_0$ for the effective mass, and $\epsilon=12.53$ for the dielectric constant. Then the many-body Hamiltonian in second quantization reads

$$H=\sum_{im\sigma}\epsilon_{im}a^\dagger_{im\sigma}a_{im\sigma}+\frac{1}{2}\sum_{i_1i_2i_3i_4m_1m_2m_3m_4\sigma\sigma'}\times\langle i_1m_1i_2m_2|V|i_3m_3i_4m_4\rangle a^\dagger_{i_1m_1\sigma}a^\dagger_{i_2m_2\sigma'}a_{i_4m_4\sigma'}a_{i_3m_3\sigma}, \tag{1}$$

where i and m label the radial and angular momentum quantum numbers, respectively, and σ labels the spin projection. The one-body term is already diagonal with the energies

$$\epsilon_{im}=\hbar\omega_0(2i+|m|+1). \tag{2}$$

$a^\dagger_{im\sigma}$ and $a_{im\sigma}$ are creation and annihilation operators of an electron in a single-particle state with spin projection σ and the orbital wave function

$$\psi_{im}(\mathbf{r})=\langle\mathbf{r}|im\rangle=R_{im}(r)e^{im\varphi}. \tag{3}$$

The radial wave functions are given by

$$R_{im}(r)=\frac{(-1)^i}{l_0}\sqrt{\frac{i!}{\pi(i+|m|)!}}e^{-r^2/2l_0^2}\left(\frac{r}{l_0}\right)^{|m|}L_n^{|m|}\left(\frac{r^2}{l_0^2}\right), \tag{4}$$

where $l_0=\sqrt{\hbar/(m^*\omega_0)}$ is the characteristic length of the harmonic oscillator and $L_n^{|m|}$ are the Laguerre polynomials. The Coulomb matrix elements are given by

$$\langle i_1m_1i_2m_2|V|i_3m_3i_4m_4\rangle=\frac{e^2}{4\pi\epsilon\epsilon_0}\int d^2r\,d^2r'\frac{\psi^*_{i_1m_1}(\mathbf{r})\psi^*_{i_2m_2}(\mathbf{r}')\psi_{i_3m_3}(\mathbf{r})\psi_{i_4m_4}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}. \tag{5}$$

For the calculation of the low-lying eigenstates of H we employ standard diagonalization techniques as described in Refs. 20–29. The LDA calculations are described in Ref. 12. In a second step the nonresonant Raman cross section at $T=0$ K is calculated from[10]

$$\frac{d^2\sigma}{d\Omega d\omega}=\hbar\frac{\omega_F}{\omega_I}\sum_F |M_{FI}|^2\delta(E_F-E_I-\hbar\omega), \tag{6}$$

where $|F\rangle$ is an exact many-particle state and $|I\rangle$ is the ground state. In the nonresonant case the transition matrix element M_{FI} is given by

$$M_{FI}\propto\sum_{i_1m_1i_2m_2\sigma}\langle i_1m_1\sigma|e^{i\mathbf{q}\cdot\mathbf{r}}|i_2m_2\sigma\rangle\langle F|a^\dagger_{i_1m_1\sigma}a_{i_2m_2\sigma}|I\rangle \tag{7}$$

for the CDE's and

$$M_{FI}\propto\sum_{i_1m_1i_2m_2\sigma}\langle i_1m_1\sigma|\sigma_z e^{i\mathbf{q}\cdot\mathbf{r}}|i_2m_2\sigma\rangle\langle F|a^\dagger_{i_1m_1\sigma}a_{i_2m_2\sigma}|I\rangle \tag{8}$$

for the SDE's. $\mathbf{q}=\mathbf{k}_i-\mathbf{k}_s$ is the wave vector which is transferred in the inelastic light scattering process when $\mathbf{k}_i$ and $\mathbf{k}_s$ are the wave vectors of incident and scattered light, respectively.

Figure 1 compares the energy levels obtained within the LDA with the exact many-body energies. The left-hand side shows the self-consistent potential and the three lowest electronic shells of a six-electron dot. The degeneracy of the third shell is lifted due to the electron-electron interaction. The figure on the right-hand side displays the many-body energies. The ground state is a spin-singlet state with the total angular momentum $M=0$. We consider excited states with $M=1$ and $S_z=0$. For $S=1$ states, S_z can be $+1$, 0, or -1, while M is fixed. But for the case of zero magnetic field, which we consider here, the exact many-particle states with $S_z=\pm1$ are energetically degenerate with the state $S_z=0$. Therefore, we consider the $S_z=0$ states only. We find two low-lying singlet and two triplet states. The transition to the $S=0$ state with an excitation energy of $\Delta E=6$ meV is the Kohn's mode, which is usually referred to as a collective CDE. It is shifted to higher energies compared to the single-particle energy differences. The lowest-energy state with $M=1$, which is an $S=1$ state, is identified as the collective SDE. The interesting result is that we find exactly two additional many-body states that have excitation energies very close to the Kohn-Sham level spacings. The higher-energy state has $S=0$, the lower-energy state $S=1$.

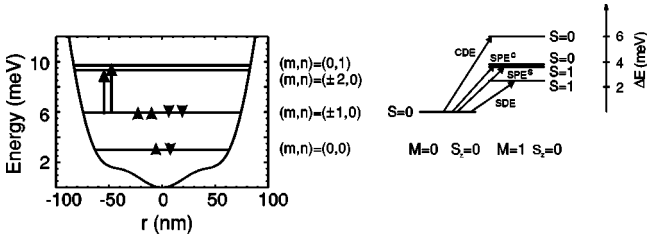

FIG. 1. Single-particle (left figure) and many-particle (right figure) states of a six-electron quantum dot. M is the total angular momentum, S the total spin, and S_z the spin projection in z direction. The single-particle quantum numbers are m and n where m is the single-particle angular momentum and n is the radial quantum number.

The spectra in Fig. 2 are calculated from Eq. (6). The upper figure shows spectra obtained from the exact diagonalization, the lower figure spectra which are calculated within the time-dependent LDA (TDLDA). In both treatments we find the collective CDE, the Kohn's mode, at the energy of the external potential ($\hbar\omega_0=6$ meV). The energy of the collective SDE is shifted to lower values in the TDLDA compared to the exact calculation. This is due to the lack of correlations in the LDA ground state. As discussed in Ref. 21 for the quantum-dot helium, angular correlations are more important for the spin-singlet ground state than for the $S=1$ excited state, which is better described by the LDA. As a result the energy of the ground state is strongly decreased by the correlations and the SDE is higher in energy in the exact treatment. The important result is that single-particle-like excitations are found in both treatments with comparable energies and excitation strengths. The energies of these modes are close to the Kohn-Sham single-particle level spacings and carry only small contributions from collective dy-

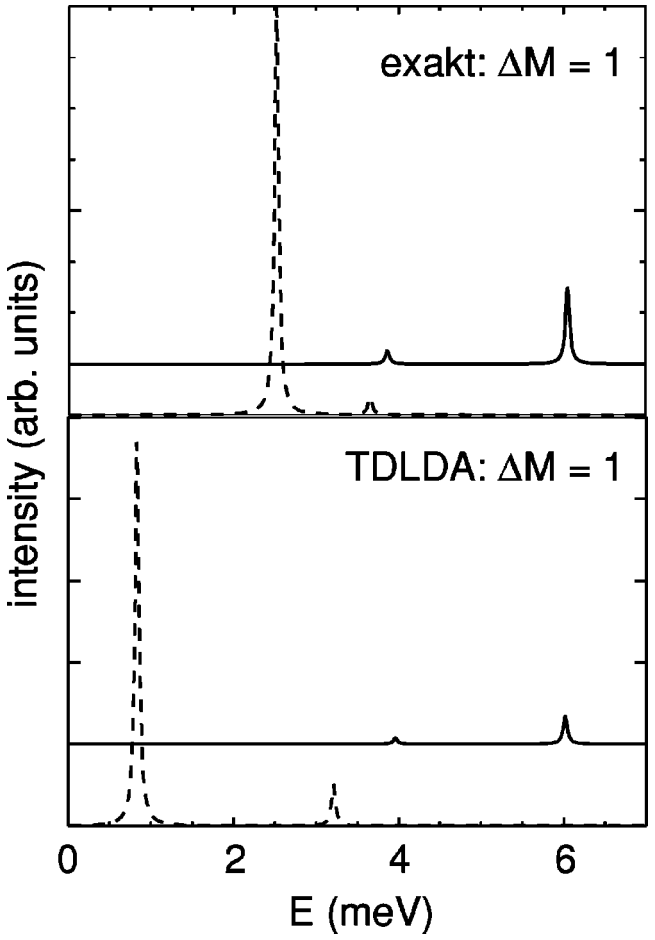

FIG. 2. Calculated nonresonant Raman spectra for a six-electron quantum dot. The upper figure shows spectra obtained within an exact calculation the lower figure those of a time-dependent LDA treatment. The solid and dashed lines represent polarized and depolarized spectra, respectively. All spectra are calculated for a lateral wave-vector transfer of $q=1\times10^6$ cm^{-1} and a change in the angular momentum of $\Delta M=1$.

namical shifts. The LDA result for a six-electron dot is very similar to that of a 12-electron system. The only difference is that in the case of three filled electronic shells, two SPE's show up in the spectra for each polarization. For a growing number of occupied shells these excitations emerge to the broad peak, which is usually observed in quantum dots with many electrons.

In summary we have theoretically analyzed the many-body excitations of a six-electron quantum dot. We have shown that, besides the well-known SDE's and CDE's, single-particle-like excitations can be expected in systems with electron numbers as low as six. The energies of these excitations are very close to the self-consistent Kohn-Sham level spacing. We therefore call these excitations SPE-like. We conclude that these SPE's are accurately described by a TDLDA. However, the energy of the collective SDE is underestimated within the TDLDA.

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