Inelastic light scattering and the excited states of many-electron quantum dots

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A consistent calculation of resonant inelastic (Raman) scattering amplitudes for relatively large quantum dots, which takes account of valence-band mixing, discrete character of the spectrum in intermediate and final states, and interference effects, is presented. Raman peaks in charge and spin channels are compared with multipole strengths and with the density of energy levels in final states. A qualitative comparison with the available experimental results is given.

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The inelastic (Raman) scattering of light by a semiconductor quantum dot is an optical process which has proven to be very useful as a experimental technique to study excited states. The interpretation of a resonant Raman experiment requires, however, a big experimental and theoretical effort. The theoretical description is often so complicated that consistent calculations have been carried out only for the smallest dots.

In the present paper, we address the question about resonant Raman scattering in a relatively large quantum dot, aimed at reproducing the main features of the Raman phenomenology by means of a transparent and consistent computational scheme. Particularly, we focus on topics such as the character (single-particle or collective) of the Raman peaks, the role of interference effects, Raman peaks for spin-excited final states, the modifications of the spectrum as the background electron density is changed, or the evolution of the spectrum as the incident laser energy moves from close to the effective band gap to well above it. Results of calculations are presented for GaAs dots with AlGaAs barriers. A qualitative comparison with the available experimental results is given.

Our starting point is the perturbation-theory expression

\[ A_{fi} \sim \sum_{int} \frac{|f| H_{e-r}^\dagger |int| (int|H_{e-r}^\dagger)|i}\rangle}{\hbar \nu_i - (E_{int} - E_i) + i\Gamma_{int}}, \] (1)

for the amplitude of Raman scattering. The kets \(|i\rangle\) and \(|f\rangle\) are written as: \(|i\rangle = |\psi_i\rangle|N_i\rangle\), \(|f\rangle = |\psi_f\rangle|N_i-1,1_f\rangle\), where \(|\psi_i\rangle\) and \(|\psi_f\rangle\) are initial and final N-electron states, \(|N_i\rangle\) is a state with \(N_i\) photons of frequency \(\nu_i\), and \(|N_i-1,1_f\rangle\) is a state with \(N_i-1\) photons of frequency \(\nu_i\) and one photon of frequency \(\nu_f\). On the other hand, the intermediate states are written as: \(|int\rangle = |\psi_{int}\rangle|N_i-1\rangle\), where \(|\psi_{int}\rangle\) contains, besides the initial \(N\) electrons, an additional electron-hole pair. \(H_{e-r}\) is the electron-radiation interaction hamiltonian, and \(\Gamma_{int} = 0.5 \text{ meV} - \) a phenomenological broadening.

Eq. (1) shows the difficulties in computing \(A_{fi}\) for a dot containing dozens of electrons. One should construct approximations to \(|\psi_f\rangle\) in a 30 meV excitation energy interval, in which there could be hundreds of states, and approximations to \(|\psi_{int}\rangle\) in a 30 meV energy interval above the band gap. In the latter situation, hole-band mixing should be taken into account in order to describe scattering to spin-excited final states. Notice that interference effects may come out from the sum over intermediate states. The 30 meV upper bound in final states is a typical threshold for phonon excitations.

Commonly, one avoids computing the intermediate states by approximating the whole expression for \(A_{fi}\). In Ref. (2), for example, Raman intensities are almost identified with strength functions (modulated multipole strengths), in accordance to the interpretation given by authors of paper (2) of their results. This approximation to \(A_{fi}\) neglects contributions from single-particle final states and interference effects from intermediate states. It is supposed to be valid for laser energies well above the effective band gap. A second common approximation to \(A_{fi}\), which also neglects interference effects, is the so-called extreme resonance condition, in which \(h\nu_i\) is very close to the band gap. In this case, Raman intensities are almost identified with excited-state luminescence peaks, i.e. with the peaks in the density of final-state energy levels. This interpretation was used by the authors of Ref. (1).

In our calculations, we start from the exact quantum-mechanical expression (1), and construct Random-Phase approximations to \(|\psi_f\rangle\), and Tamm-Dankoff approximations to \(|\psi_{int}\rangle\). Explicit formulæ, which should be particularised to the pure electronic system, may be found in (2). As the number of electrons in the dot is supposed to be relatively high, we expect that the insertion of the mean field functions \(|\psi_{int}\rangle\) and \(|\psi_f\rangle\) into (1) would lead to a qualitatively correct picture for the positions and in-
tensities of Raman peaks. The Hartree-Fock (HF) basis is used throughout. For holes, the HF equations include the electron mean field and the heavy-light hole mixing, treated by means of the Kohn-Luttinger Hamiltonian. A typical calculation in a 42-electron dot involves around 60 many-particle final states (for a given multipolarity and spin), and around 2000 intermediate states.

The dot is assumed to have a Lorentzians of width $\Gamma$. The incident laser energy is very close to the effective intensity of the SPE is also shown. Around 2000 intermediate states. (b) Raman intensities corresponding to the collective monopolar state and to a SPE with Raman shift of 8.8 meV are shown in Fig. 1(b). The first interesting remark, in qualitative accordance with the existing observations, is that SPE are enhanced when $\hbar \nu_i$ is close to the band gap, whereas collective states are enhanced as $\hbar \nu_i$ is raised. On the other hand, the contribution of each particular intermediate state at resonance, i.e. $|\langle f|H^+_{\mu}|int\rangle\langle int|H^{-}_{\nu}|i\rangle|^2\Gamma_{int}^2$, to the Raman intensity of the SPE is also included in the figure (vertical lines) in order to evaluate interference effects. Peaks in the Raman efficiency are related to particular intermediate states giving strong contributions to the sum (1). In the figure, weak constructive or destructive interference in the neighborhood of these intermediate states can be appreciated.

The density of final state energy levels, computed from the Random Phase approximation to $|\psi_f\rangle$, is superposed to the Raman spectra in Fig. 2 in order to show its correlation with Raman peaks. The shape of the Raman spectrum depends on the frequency, but it is apparent that strong Raman peaks are associated to bunches of energy levels. These findings are in qualitative agreement with the experimental results of paper [1].

In Fig. 2 curves labelled by a $\perp$ symbol represent a situation in which the incident scattered light polarization vectors are orthogonal. For charge excitation...
(CE) channels, in the upper panel, the Raman spectrum in the orthogonal-polarization case is similar to that one in the parallel-polarization geometry. In the lower panel, Raman intensities in spin-excitation (SE) dipolar final states, are shown. By SE we mean states in which the total electronic spin projection is different from the initial state value. The difference in magnitude of peaks related to CE and SE in the orthogonal polarization would mean that SE peaks will be, in general, washed out, and only the lowest-energy SE levels, which are shifted to the left of CE states, have a chance to be measured. Notice also that the collective SE dipolar state, carrying more than 95% of the sum rule, and which position is represented also by a drop line, is observed only as a very small shoulder in the Raman spectrum.

Next, we consider the question about the effect of the density of the electronic cloud on the Raman spectra. In our 42-electron dot, we can control the density by varying the confinement strength: \( \rho \sim N^{1/2} m_e \omega_0 / \hbar \). Notice, for instance, that the density of larger dots with around 200 electrons, as those studied in Ref. 2, is similar to the density of our 42-electron dot when the parameter \( \hbar \omega_0 \) is doubled from 6 to 12 meV. Calculations were done also for a smaller frequency, \( \hbar \omega_0 = 3 \text{ meV} \), with the purpose of obtaining the whole picture. In our calculations, we fixed the Coulomb-to-oscillator ratio of characteristic energies, given by the parameter \( e^2 m_1^{1/2} / \left( \kappa \hbar^3 / 2 \omega_0^{1/2} \right) \). It means that the relative strength of Coulomb interactions is kept constant when the density is varied.

The results are shown in Fig. 3. For each value of \( \hbar \omega_0 \), Raman spectra are computed for laser energies 5 and 25 meV above the effective band gap. The curves can be qualitatively understood on simple grounds. An increase in \( \omega_0 \) leads to a scaling of energies. For example, in Fig. 3 (c), only the first SP and the first collective peaks are seen, the rest of the spectrum is moved to higher energies. In accordance to this scaling, the density of energy levels decreases both in intermediate and final states. Thus, the intensity of the Raman peaks should decrease by roughly a factor of 4 as \( \omega_0 \) is doubled. The relative intensity of peaks depends on the Raman efficiency, as mentioned above.

Finally, we want to discuss the situation in which the incident laser energy is well (around 50 meV) above the effective band gap. This regime is characterized by the following properties: (a) an overall decrease of Raman amplitudes, (b) a reinforcement of the peaks associated
to collective states, and (c) a suppression of the SP peaks, except at the lower edge of SP excitations. In this case, it seems useful to distinguish the resonant and off-resonance contribution to the sum. Only the latter can be evaluated within our computational scheme because, for excitation energies around 50 meV, the intermediate states are doubtfully described by a simple Tamm-Dankoff approximation, and a constant $\Gamma_{\text{int}}$ is not a reasonable approximation. Indeed, one expects, for example, an increasing $\Gamma_{\text{int}}$ as we move to intermediate states with higher excitation energies.

In Fig. 4, the off-resonance contribution to the Raman spectrum for laser energy 50 meV above the band gap is drawn, along with one spectrum, already shown in Fig. 3 (c), corresponding to an incident photon energy 25 meV above the band gap. The off-resonance curve is computed from a sum which includes, as before, intermediate states with excitation energies below 30 meV. The smaller amplitude of this curve is due to the big energy denominators. It is apparent, however, that the SP peak is stronger suppressed than the collective one.

In general, the amplitude of the resonant contribution to (1) shall decrease as $h\nu_i$ rises. The reason is that both $\langle f|H^+|\text{int}\rangle$ and $\langle \text{int}|H^-|i\rangle$ decrease, while $\Gamma_{\text{int}}$ increases in this case. The reinforcement of collective states could be the effect of resonances in the intermediate states, but this is a question that requires a further work.

In conclusion, we presented calculations for the amplitudes of resonant Raman scattering in 42-electron GaAs quantum dots based on the exact perturbation-theory formula (1). To our knowledge, the largest previous calculation considered a 12-electron dot, only one valence hole sub-band (the heavy hole), and assumed a spin unpolarized HF ground state.

Features related to SP (dominant) and collective final-state excitations are apparent when the incident laser energy is varied in a 20 meV energy interval above the effective band gap. These features may be correlated to bunches of final-state energy levels and to particular intermediate states giving strong contributions to the sum (1). Weak constructive or destructive interference effects can be appreciated in this regime. The intensity of spin-excitation peaks is shown to be one or two orders of magnitude weaker than the intensity of charge-excitation peaks for these laser energies. It means that only the lowest-energy spin excited states have a chance to be measured. On the other hand, for $h\nu_i$ well above the band gap, the off-resonance contribution to the Raman spectrum shows a strong suppression of SP peaks. These results are in qualitative agreement with the observations.

There are many interesting points still uncovered. For example, to clarify the properties of the intermediate states giving a strong contribution to (1). In quantum wells and for Raman shifts above 30 meV, peaks in the Raman efficiency of collective SE are shown to correspond to the absorption or emission of photons of particular frequencies, which are identified in PLE as a series of “excitonic” states. These “resonances” in intermediate states could be the reason of strong enhancement of collective SE in quantum dots for $h\nu_i$ well above the band gap. Indeed, SE collective peaks were observed in this regime. These, and other, uncovered aspects of Raman scattering in quantum dots indicate the need for more experimental and theoretical work on this subject.

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