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

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Abstract

A consistent calculation of resonant inelastic (Raman) scattering amplitudes for relatively large quantum dots, which takes account of valence band mixing, the 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.

The inelastic (Raman) scattering of light by a semiconductor quantum dot is an optical process which has proven to be very useful as an experimental technique for studying excited states [1, 2]. 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 [3, 4].

In the present paper, we address the question of resonant Raman scattering in a relatively large quantum dot, aiming at reproducing the main features of the Raman phenomenology by means of a transparent and consistent computational scheme. In particular, 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, and 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 also given.

Our starting point is the perturbation theory expression

$$A_{\mathrm{fi}} \sim \sum_{\mathrm{int}} \frac{\langle \mathrm{f}|H^{+}_{\mathrm{e-r}}|\mathrm{int}\rangle\langle \mathrm{int}|H^{-}_{\mathrm{e-r}}|\mathrm{i}\rangle}{h\nu_{\mathrm{i}} - (E_{\mathrm{int}} - E_{\mathrm{i}}) + \mathrm{i}\Gamma_{\mathrm{int}}}, \tag{1}$$

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for the amplitude of Raman scattering [5]. The kets $|\mathrm{i}\rangle$ and $|\mathrm{f}\rangle$ are written as: $|\mathrm{i}\rangle = |\psi_\mathrm{i}\rangle|N_\mathrm{i}\rangle$, $|\mathrm{f}\rangle = |\psi_\mathrm{f}\rangle|N_\mathrm{i} - 1, 1_\mathrm{f}\rangle$, where $|\psi_\mathrm{i}\rangle$, and $|\psi_\mathrm{f}\rangle$ are initial and final N-electron states, $|N_\mathrm{i}\rangle$ is a state with N_i photons of frequency ν_i, and $|N_\mathrm{i} - 1, 1_\mathrm{f}\rangle$ is a state with $N_\mathrm{i} - 1$ photons of frequency ν_i and one photon of frequency ν_f. On the other hand, the intermediate states are written as: $|\mathrm{int}\rangle = |\psi_\mathrm{int}\rangle|N_\mathrm{i} - 1\rangle$, where $|\psi_\mathrm{int}\rangle$ contains, besides the initial N electrons, an additional electron–hole pair. $H_\mathrm{e-r}$ is the electron–radiation interaction Hamiltonian, and $\Gamma_\mathrm{int} = 0.5$ meV—a phenomenological broadening.

Equation (1) shows the difficulties in computing A_fi for a dot containing dozens of electrons. One should construct approximations to $|\psi_\mathrm{f}\rangle$ in a 30 meV excitation energy interval, in which there could be hundreds of states, and approximations to $|\psi_\mathrm{int}\rangle$ 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 [6], for example, Raman intensities are almost identified with strength functions (modulated multipole strengths), in accordance to the interpretation given by the 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 $h\nu_\mathrm{i}$ 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 [1].

In our calculations, we start from the exact quantum-mechanical expression (1), and construct random-phase approximations to $|\psi_\mathrm{f}\rangle$, and Tamm–Dankoff approximations to $|\psi_\mathrm{int}\rangle$ [7]. Explicit formulae, which should be particularized to the pure electronic system, may be found in [8]. As the number of electrons in the dot is supposed to be relatively high, we expect that the insertion of the mean field functions $|\psi_\mathrm{int}\rangle$ and $|\psi_\mathrm{f}\rangle$ into (1) would lead to a qualitatively correct picture for the positions and intensities 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 for a 42-electron dot involves around 60 many-particle final states (for a given multipolarity and spin), and around 2000 intermediate states.

We first consider the extreme resonance condition, in which the incident laser energy is very close the effective band gap. In figure 1(a), the calculated Raman intensities (squared amplitudes smeared out by means of Lorentzians of width $\Gamma_\mathrm{f} = 0.5$ meV) for charge monopolar final states are shown. The dot is assumed to have a disk geometry with a 25 nm width. The lateral confinement is parabolic, with $\hbar\omega_0 = 6$ meV, reproducing the observed position of the Kohn mode in the dots studied in [2]. The magnetic field, equal to 1 T, is perpendicular to the dot plane. The final states considered in figure 1 have the same spin and angular momentum projection onto the magnetic field axis as the initial-state values. The nominal band gap is taken as 1560 meV. This gap is renormalized by Coulomb interactions to, approximately, 1567 meV. Raman amplitudes are computed in backscattering geometry. The initial and final light polarization vectors are parallel. The laser energy is swept from 1562 to 1590 meV, that is from below the effective band gap to 20 meV above it. The position of the collective monopolar state, which carries more than 99% of the energy-weighted sum rule, is indicated by a vertical line. In all of our calculations, the incident (and scattered) light form an angle of 30° with the dot normal, which means that the maximum transferred momentum of light

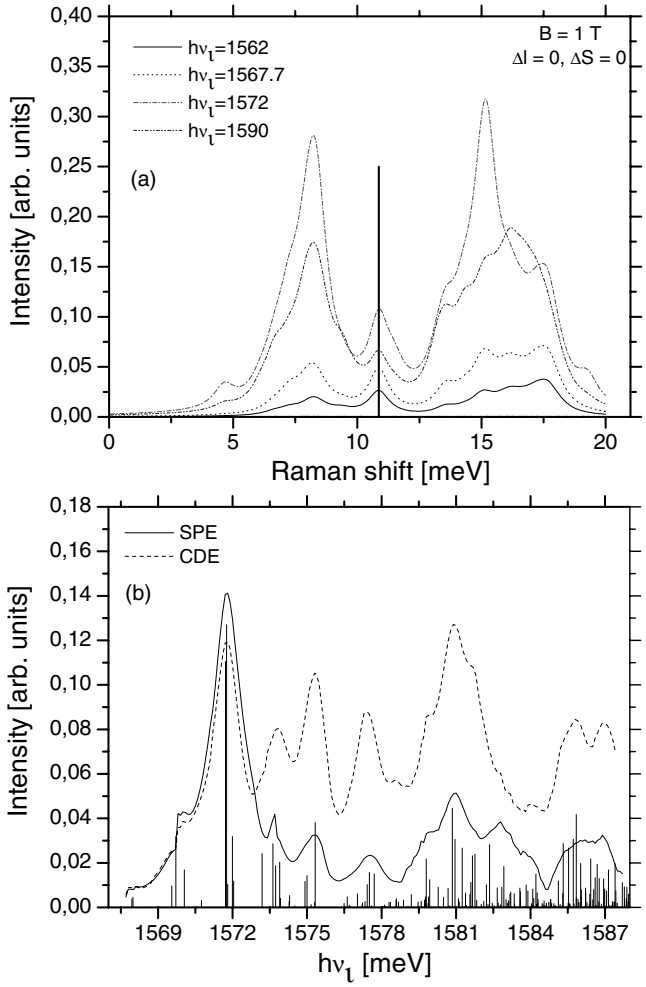

Figure 1. (a) Raman spectra for charge monopole final states at different frequencies of the incident light. In the present and next figure, $h\nu_{\rm i}$ is given in millielectronvolts. A vertical line indicates the position of the collective state. (b) Raman intensities of one SPE and the collective state as functions of $h\nu_{\rm i}$. The contribution of each intermediate state at resonance to the intensity of the SPE is also shown.

is $\Delta q_{\rm x} \approx 0.8 \times 10^5$ cm^{-1}. Under these circumstances, Raman spectra are dominated by the lowest multipolar final states [2].

Let us note that the collective monopolar state is seen as a distinct peak at any laser frequency. However, with final states carrying an almost zero fraction of the sum rule, for which reason we will call them single-particle excitations (SPE), are associated stronger, and wider, Raman peaks at energies below and above the collective state.

Figure 1(b) shows the Raman efficiency of two particular final states, that is their Raman intensities as a function of $h\nu_{\rm i}$. In general, the intensity of a given Raman peak in figure 1(a) is the result of three factors: (1) the number of final states contributing to it, (2) the Raman efficiencies of these states, and (3) interference effects.

Raman intensities corresponding to the collective monopolar state and to a SPE with Raman shift of 8.8 meV are shown in figure 1(b). The first interesting point, in qualitative

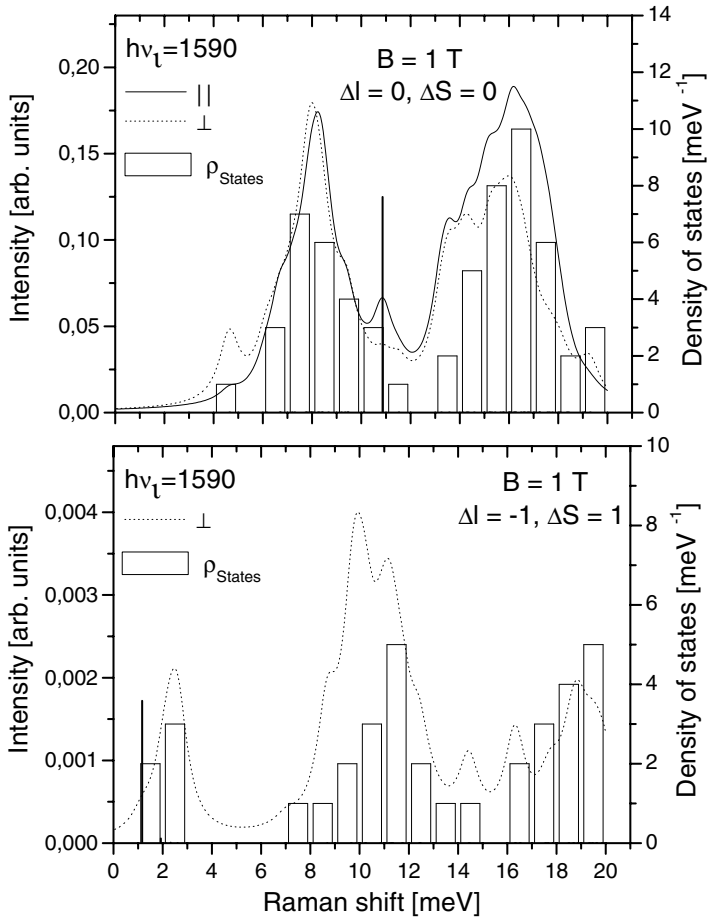

Figure 2. Upper panel: Raman intensities for charge monopole final states. Lower panel: Raman spectra in spin dipole states. The positions of collective excitations are indicated by dropped lines. The density of final-state energy levels is superposed in the figure.

accordance with the existing observations, is that SPE are enhanced when $h\nu_i$ is close to the band gap, whereas collective states are enhanced as $h\nu_i$ is raised. On the other hand, the contribution of each particular intermediate state at resonance, i.e. $|\langle f|H^{+}_{e-r}|int\rangle\langle int|H^{-}_{e-r}|i\rangle|^2/\Gamma^2_{int}$, to the Raman intensity of the SPE is also included in the figure (vertical lines) in order to allow evaluation of 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 neighbourhood 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 on the Raman spectra in figure 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 with bunches of energy levels. These findings are in qualitative agreement with the experimental results of paper [1].

In figure 2, curves labelled by a $\perp$ symbol represent a situation in which the incident and 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 in the

parallel-polarization geometry. In the lower panel, Raman intensities in spin excitation (SE) dipolar final states, are shown. By SE states we mean ones in which the total electronic spin projection is different from the initial-state value. The difference in magnitude for 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 the CE states, can possibly be measured. Notice also that the collective SE dipolar state, carrying more than 95% of the sum rule, and whose position is represented also by a dropped line, is observed only as a very small shoulder in the Raman spectrum.

Next, we consider the question of 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, such as those studied in [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, $\hbar\omega_0 = 3$ 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 $e^2 m_e^{1/2}/(\kappa\hbar^{3/2}\omega_0^{1/2})$ [10]. This means that the relative strength of Coulomb interactions is constant when the density is varied.

The results are shown in figure 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 ω_0 leads to a scaling of energies. For example, in figure 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 with this scaling, the density of energy levels decreases both in the intermediate and final states. Thus, the intensity of the Raman peaks should decrease by roughly a factor of 4 as ω_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 [2, 9]. This regime is characterized by the following properties: (a) an overall decrease of Raman amplitudes, (b) a reinforcement of the peaks associated with collective states, and (c) a suppression of the SP peaks, except at the lower edge of SP excitations [11].

In this case, it seems useful to distinguish the resonant and off-resonance contributions to the sum (1). 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 $\Gamma_{\rm int}$ is not a reasonable approximation. Indeed, one expects, for example, an increasing $\Gamma_{\rm int}$ as we move to intermediate states with higher excitation energies.

In figure 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 figure 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 more strongly suppressed than the collective one.

In general, the amplitude of the resonant contribution to (1) will decrease as $h\nu_{\rm i}$ rises. The reason is that both $\langle {\rm f}|H^+|{\rm int}\rangle$ and $\langle {\rm int}|H^-|{\rm i}\rangle$ decrease, while $\Gamma_{\rm int}$ 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 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

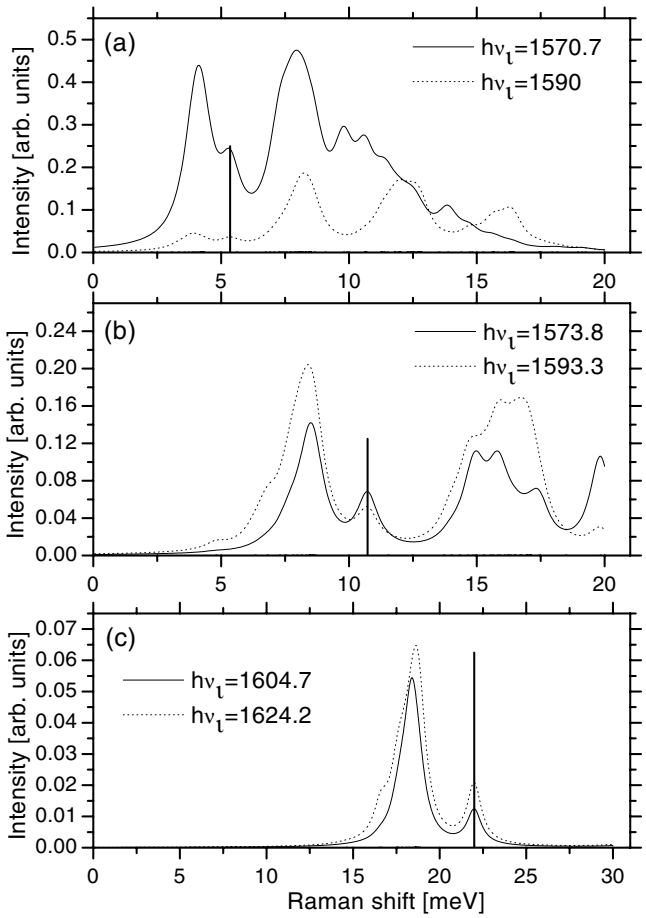

Figure 3. Raman spectra for charge monopole final states and parallel polarization. The magnetic field is set to zero. (a) $\hbar\omega_0 = 3$, (b) $\hbar\omega_0 = 6$, and (c) $\hbar\omega_0 = 12$ meV.

knowledge, the largest previous calculation [12] 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 with bunches of final-state energy levels and 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 SE peaks is shown to be one or two orders of magnitude weaker than the intensity of CE peaks for these laser energies. This means that only the lowest-energy spin excited states can possibly be measured. On the other hand, for $h\nu_{\mathrm{i}}$ 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 not covered; for example, clarifying 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

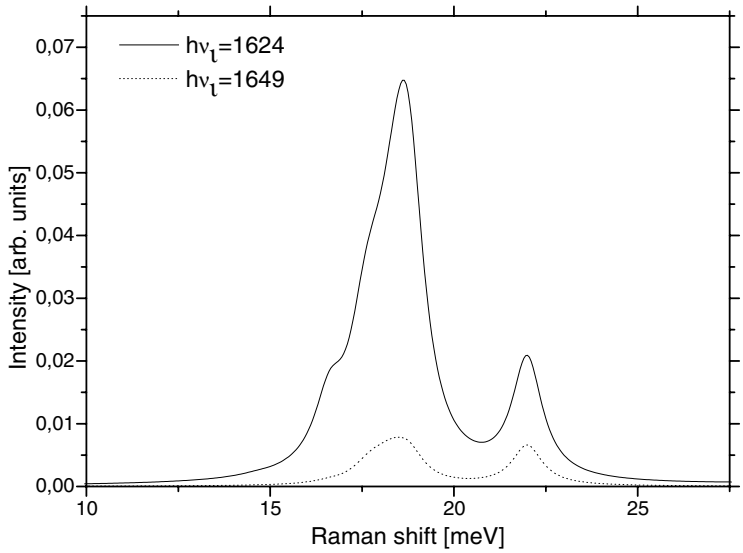

Figure 4. Raman spectra for laser energies 25 and 50 meV above the effective band gap. Only the off-resonance contribution to the latter is shown. See the explanation in the main text.

a series of 'excitonic' states [13]. These 'resonances' in intermediate states could be the reason for the 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 [2]. These, and other, non-covered aspects of Raman scattering in quantum dots indicate the need for more experimental and theoretical work on this subject.

Acknowledgments

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