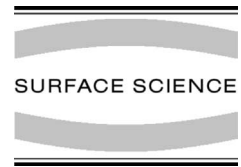




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Quantum size and correlation effects in the double excitation spectrum of a quantum dot

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Abstract

This work deals with the double excitation of electronic states of semiconductor quantum dots as induced by photoabsorption. The prime focus is put on the manifestation of quantum size and correlation effects in the two-particle excited spectrum. The photon field is treated within the dipole approximation whereas the Green function method is employed to describe the excited two-particle spectrum. Our numerical simulations demonstrate that the dimensionality of the dot as well as the amount of inter-electronic correlation have a marked effect on the dot's excitation spectrum. The underlying physics of these effects can be understood from the functional structure of the inter-electronic interaction in a many-electron system as well as from the characteristic change of the electronic states when varying the size of the dot from the strong localization regime to an extended structure. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Nano and mesoscopic semiconductor quantum dots (QDs) are systems in which the charge carriers are confined in all directions to characteristic lengths comparable with their de Broglie wave length [1]. The extremely versatile nature of QDs make them an appealing candidate both for applied and fundamental research. For example, parameters such as particle number and charge density can be experimentally varied in regions not accessible in atomic or molecular systems which renders possible a detailed study of correlation and confinement effects. Experimentally, the properties

of QDs can be explored by optical methods such as multiexciton [2] and single photoemission spectroscopy [3]. In this theoretical work we propose the one-photon double emission (DPE) of QDs as a tool to investigate correlation and quantum size effects. This method is particularly suitable since the DPE process is inhibited in absence of inter-particle interactions [4,5]. In the DPE experiments one resolves simultaneously the energies and emission angles of two excited electrons for well defined properties of the photon field and the QD under study. For atomic and molecular systems the DPE technique have been exploited extensively [6] whereas for extended systems the first experiments has been reported recently [7]. In a previous work [4,5] we derived the form of the transition amplitude of the DPE for extended systems. The practical calculations of these amplitudes requires

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however certain approximation for the many-body excited state. In Refs. [4,5] we suggested a non-perturbative method in which the DPE transition amplitude is determined by the overlap of the excited and ground two-particle states.

In this work we evaluate the DPE transition amplitude using the two-particle Green operator to propagate the two-particle ground state to the photoexcited one. The dipole approximation for the photon field is employed. To clarify the role of confinement, we probe simple models of a parabolic QD and a thin film. In the case of a film electrons have discrete energy levels in one direction and are free to move in the other two. In the case of a QD the electrons are confined in all three directions and have only discrete bound states. To explore the effect of the size of the system on the DPE spectrum we consider QDs with sizes ranging from tens to hundreds of angstroms. Since the DPE process is highly sensitive to the electronic interactions we treat the inter-electronic interaction between the two photoelectrons to all orders and include the effects of all the other electrons within the static-screening concept.

2. Formal development

We consider a process in which a photon with energy $\hbar\omega$ is absorbed by an electronic system S . Subsequently two electrons are emitted from S and their kinetic energies E_1 and E_2 and their emission solid angles Ω_1 and Ω_2 are determined at the same time, i.e. wave vectors \mathbf{k}_1 and \mathbf{k}_2 of these two photoelectrons are measured. In what follows we assume that the photon field affects only the degrees of freedom of the two emitted electrons, which will be termed ‘active’. The other ‘passive’ ground state electrons of S are not influenced by the photon field but they determine nevertheless the yield of DPE though the amount of screening of the electron–electron interaction of the two active electrons. For example, for extremely strong screening the DPE diminishes rapidly. In the dipole approximation for the photon field, the velocity-form transition-matrix element is then given by the golden rule

$$T = \langle \Psi_{12} | \hat{\epsilon} \cdot (\nabla_1 + \nabla_2) | \Phi_{12} \rangle. \quad (1)$$

Here $|\Psi_{12}\rangle$ and $|\Phi_{12}\rangle$ are the final and initial states of the two active electrons whereas $\hat{\epsilon}$ is the photon polarization vector and $\nabla_{1/2}$ are the single particles velocity operators. There are numerous studies on the ground-state electronic structure of QDs [1]. This work focuses on the treatment of the highly excited states where electronic correlation is known to be prominent. The excited state of the dot at an energy given by the frequency of the absorbed photon is generated by propagating the initial state as

$$\Phi_{12}(\mathbf{r}_1, \mathbf{r}_2) = G_{12} \phi_{12}. \quad (2)$$

Here the ground state ϕ_{12} is constructed as an antisymmetrised product of single-particle states of the system S (spin–orbit interaction is neglected and the spin part is factored out)

$$\phi_{12}(\mathbf{r}_1, \mathbf{r}_2) = \Phi_1(\mathbf{r}_1)\Phi_2(\mathbf{r}_2) \pm \Phi_2(\mathbf{r}_1)\Phi_1(\mathbf{r}_2). \quad (3)$$

The single particle states Φ_1 , Φ_2 are derived by solving the time-independent Schrödinger equation with an appropriate potential of the dot, as will be explained in more details later on. The Green operator in Eq. (2) is given by the integral equation

$$G_{12} = G_0 + G_0 V_{12} G_{12}. \quad (4)$$

Here, G_0 is the free Green operator and V_{12} is the (re-normalized) Coulomb interaction between the two excited electrons. We note here, that the potential of the dot does not enter in this expression for the Green operator, for the single-particle QD parts of the total Hamiltonian have already been accounted for by employing in Eq. (2) the single particle states $\Phi_{1/2}$ (which are eigensolutions of the single-particle dot’s Hamiltonian).

As we are dealing in this work with parabolic QDs with a confining potential being of a short-range character, we assume the emitted photoelectrons to occupy plane waves (labeled by \mathbf{k}_1 and \mathbf{k}_2) in the vacuum, i.e. $\langle \Psi_{12} | \approx \langle \mathbf{k}_1, \mathbf{k}_2 |$. This approximation is reasonable for relatively high energies of the photoelectrons (a few times the ionization potential).

To deduce the optical double transition amplitude (1) it is advantageous to transform to the center-of-mass coordinates of the electron pair

$$T \propto \int \langle \mathbf{K}^+ \mathbf{K}^- | \hat{\epsilon} \cdot \nabla_{\mathbf{P}^+} | \mathbf{P}^- \mathbf{P}^+ \rangle \langle \mathbf{P}^- \mathbf{P}^+ | G_{12} | \mathbf{Q}^- \mathbf{Q}^+ \rangle \langle \mathbf{Q}^- \mathbf{Q}^+ | \phi_{12} \rangle d^3 \mathbf{P}^+ d^3 \mathbf{P}^- d^3 \mathbf{Q}^- d^3 \mathbf{Q}^+ \\ \propto \int \hat{\epsilon} \cdot \mathbf{P}^+ \delta(\mathbf{P}^+ - \mathbf{K}^+) \delta(\mathbf{P}^- - \mathbf{K}^-) \langle \mathbf{P}^- \mathbf{P}^+ | G_{12} | \mathbf{Q}^- \mathbf{Q}^+ \rangle \langle \mathbf{Q}^- \mathbf{Q}^+ | \phi_{12} \rangle d^3 \mathbf{P}^+ d^3 \mathbf{P}^- d^3 \mathbf{Q}^- d^3 \mathbf{Q}^+ \quad (5)$$

$$= \hat{\epsilon} \cdot \mathbf{K}^+ \int \langle \mathbf{K}^- \mathbf{K}^+ | G_{12} | \mathbf{Q}^- \mathbf{Q}^+ \rangle \langle \mathbf{Q}^- \mathbf{Q}^+ | \phi_{12} \rangle d^3 \mathbf{Q}^- d^3 \mathbf{Q}^+ \quad (6)$$

$$= \hat{\epsilon} \cdot \mathbf{K}^+ \int \langle \mathbf{K}^- | G_{12} | \mathbf{Q}^- \rangle \langle \mathbf{Q}^- \mathbf{K}^+ | \phi_{12} \rangle d^3 \mathbf{Q}^-. \quad (7)$$

$\mathbf{K}^+ = \mathbf{k}_1 + \mathbf{k}_1$ and $\mathbf{K}^- = (\mathbf{k}_1 + \mathbf{k}_1)/2$. Upon substituting Eq. (2) in Eq. (1) we deduce

In Eq. (5) we inserted a complete set of plane waves $\int | \mathbf{P}^- \mathbf{P}^+ \rangle \otimes \langle \mathbf{P}^- \mathbf{P}^+ | d^3 \mathbf{P}^+ d^3 \mathbf{P}^-$ and a further equivalent set labeled by $| \mathbf{Q}^- \mathbf{Q}^+ \rangle$. Furthermore in Eq. (7) we exploited the fact that G_{12} acts on the state $| \mathbf{Q}^- \rangle$ only. This is due to the special choice of the electron-electron interaction. It is straightforward to transform the expression (7) back to the single particle coordinates, in which case we arrive at

$$T \propto \hat{\epsilon} \cdot (\mathbf{k}_1 + \mathbf{k}_2) \int d^3 \mathbf{p}' \langle \frac{1}{2}(\mathbf{k}_2 - \mathbf{k}_1) | G_{12} | \frac{1}{2}(\mathbf{k}_1 + \mathbf{k}_2) - \mathbf{p}' \rangle \langle \mathbf{p}', \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{p}' | \phi_{12} \rangle. \quad (8)$$

Here $| \mathbf{p}' \rangle$ is a single-particle state labeled by the wave vector \mathbf{p}' . The expression (8) exhibits the DPE selection rule $\hat{\epsilon}(\mathbf{k}_1 + \mathbf{k}_2) \neq 0$, which implies that the electron pair forms a dipole and the photon electric field acts on the center of mass of this dipole (i.e. of the electron pair). The matrix element of the Green's function [8] embodies the dynamical correlation effects in that it dictates how a particular momentum component of the initial state ($\langle \mathbf{Q}^- \mathbf{K}^+ | \phi_{12} \rangle$) is propagated to eventually reach the detector states $| \mathbf{K}^- \mathbf{K}^+ \rangle$. From Eq. (7) it is clear that if G_{12} were the free propagator, only the momentum components of the initial state determines the detected DPE signal and this initial momentum distribution is not distorted by the electronic correlation. In Eq. (7) the extension of the system enters through the Fourier transform of the initial state that is indeed strongly dependent on the system size, as will be shown below.

3. Quantum size effects in the angular correlation of the electron pair

The parabolic potential well is a frequently used model to mimic the confining potential of a QD (cf. [1] and references therein). In the absence of magnetic fields the potential reads

$$V(x, y, z) = \sum_{i=x,y,z} \frac{1}{2} m_i^* \omega_i^2 x_i^2 - V_0 \leq 0, \quad (9)$$

where V_0 is being the bottom of the potential well. The positions of the electrons are denoted by x_i and the effective mass is m^* . The length scale $l_0 = \sqrt{\hbar/(m^* \omega_0)}$ associated with the confinement determine the parameter $\lambda = l_0/a_B$ (a_B is the effective Bohr radius) which is a measure of the coupling strength.

The non-relativistic eigenvalues and eigenfunctions of the Schrödinger equation involving the finite parabolic potential well (9) are obtained numerically.

The single-particle density of states is given by the delta functions $\rho(E) = \sum_i \delta(E - E_i)$, where i counts all the (degenerated) levels. The sixfold DPE differential cross section is

$$\frac{d\sigma}{dE_1 dE_2 d\Omega_1 d\Omega_2} \propto \int |T|^2 \delta(E_i - E_f) \rho(E_a) \rho(E_b) dE_a dE_b, \quad (10)$$

where E_a and E_b are the energies of the electrons in the initially bound states and E_i and E_f are the total energies of the initial and the final state, respectively, i.e.

$$E_i = E_a + E_b + \hbar\omega, \quad E_f = \frac{k_1^2 + k_2^2}{2}. \quad (11)$$

The DPE angular correlation discussed in this section is an angular distribution of one of the electrons over two angles θ and φ while the emission angles of the second electron are fixed and the direction of the polarization vector $\hat{\epsilon}$ is well defined. For space limitations we present here only one typical example. We consider a potential of a constant depth ($V_0 = 300$ meV) and a radius ranging from 7 to 400 Å. Suppose the photoemission occurs from the lowest level. This means that the initial state is a singlet and the total final state energy $E_1 + E_2$ is thus fixed by the photon energy. Fig. 1 shows the case of $E_1 = 1$ eV, $E_2 = 19$ eV, $\theta_1 = 0$, $\phi_1 = 0$, the vector $\hat{\epsilon}$ is perpendicular to the emission direction of the first electron (Fig. 1). The angle $\phi_2 = 0$, and the DPE intensities are plotted as functions of θ_2 . The minimum at $\theta_2 = 0$ arises due to the DPE selection rule exposed above: the two electrons cannot be emitted with a total wave-vector perpendicular to the field. The three curves (1,2,3) correspond to the QD radii 20, 135 and 400 Å, respectively. If the dot is small enough (as in the case of curve 1), the initial state

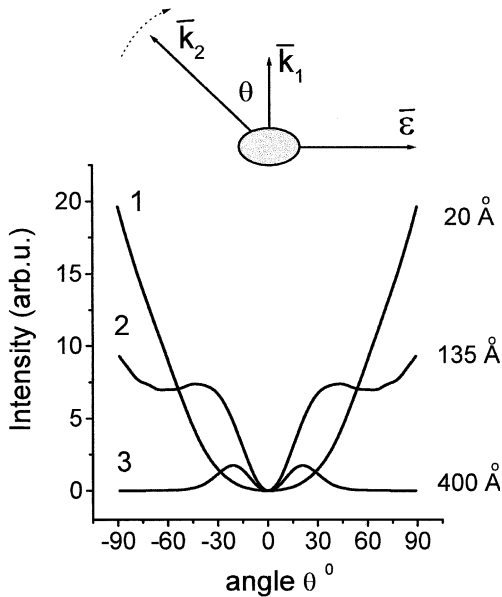


Fig. 1. Experimental geometry and DPE angular distributions from spherical quantum dots of different sizes. Angles of emission are $(\theta_1, \phi_1) = (0, 0)$ and $(\theta_2, \phi_2) = (\theta, 0)$, kinetic energies $E_{f1} = 1$ eV, $E_{f2} = 19$ eV. Curves 1,2,3 correspond to dot radii 20, 135 and 400 Å, respectively.

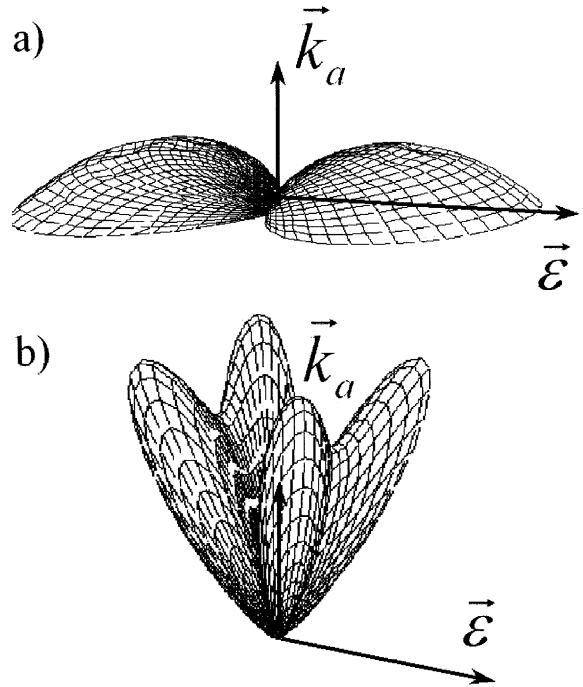


Fig. 2. 3D angular distributions for the same conditions as (a) curve 1, and (b) curve 3 of Fig. 1.

is strongly localized. It's Fourier transform $\langle \mathbf{p}', \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{p}' | \Phi_{12} \rangle$ (which enters in T) is delocalized and smooth in a large domain of angles (θ_2, ϕ_2) . This can be seen in Fig. 2(a) which shows the three-dimensional plot of the case of curve 1 (Fig. 1(a)). Since $E_2 \gg E_1$, the sum $(\mathbf{k}_1 + \mathbf{k}_2)$ is almost parallel to vector \mathbf{k}_2 . Since the DPE is proportional to $\hat{\epsilon}(\mathbf{k}_1 + \mathbf{k}_2)$ we notice a continuous increase of the curve 1 of Fig. 1 towards the edges. The shape of Fig. 2(a) can be understood as follows: since $E_2 \gg E_1$ it is expected that the photon is absorbed mostly by the fast electron, which undergoes a dipole transition from the initial isotropic state with a weak coupling to the second electron. This result is a *p*-type distribution of the fast emitted photoelectron.

For larger sizes of the QDs the function $\langle \mathbf{p}', \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{p}' | \Phi_{12} \rangle$ is rather localized at certain angles (more spread in configuration space). This results is squeezing effect of the angular distribution of the photoelectrons as can be seen in Fig. 2(b).

4. Effect of electronic correlation strength: role of screening

We now discuss film systems. These are systems which are confined in one direction (say the z -direction) and largely extended and jellium-like in other x, y -directions. Thus the single particle states can be written as

$$\begin{aligned}\Phi_{\mathbf{p}^{\parallel}}(\mathbf{r}) &= e^{i\mathbf{p}^{\parallel}\cdot\mathbf{r}} \cdot \Phi^{\perp}(z), \\ \langle \mathbf{q} | \Phi_{\mathbf{p}^{\parallel}}(\mathbf{r}) \rangle &= \delta(\mathbf{q}^{\parallel} - \mathbf{p}^{\parallel}) \cdot \langle \mathbf{q}^{\perp} | \Phi^{\perp} \rangle,\end{aligned}\quad (12)$$

where \mathbf{p}^{\parallel} is a component of the wave vector of the electron parallel to the surface, which is used as a quantum number denoting the state. The perpendicular component is confined from both sides of a layer, so that the total energies in the initial and final states are:

$$\begin{aligned}E_i &= -E_a^{\perp} - E_b^{\perp} + \frac{p_a^{\parallel 2} + p_b^{\parallel 2}}{2} + \hbar\omega_{ph} - 2V_0 \quad \text{and} \\ E_f &= \frac{k_1^2 + k_2^2}{2},\end{aligned}\quad (13)$$

where all energies (measured relative to the vacuum level) should be understood as absolute values and negative values are indicated explicitly when necessary. $E_{a,b}^{\perp}$ are the eigenenergies of the electrons a and b associated with the motion in the z -direction, V_0 is the jellium zero, initial state wave vectors are labeled by letters, final state by numbers.

The charge and spin fluctuation screens the mutual Coulomb interaction between an electron pair. The simplest model to account for this shielding effect is given by the Thomas–Fermi theory which derives the electron–electron potential as

$$\begin{aligned}V_{12}(r_{12}) &= \frac{\exp(-\lambda r_{12})}{r_{12}}, \\ V_{12}(q) &= \frac{4\pi}{q^2 + \lambda^2}.\end{aligned}\quad (14)$$

The screening length λ indicates the decay strength of the two-body interaction and is determined by the mean electronic density.

To a first-order the Green's function associated with V_{12} is given by $G_{12} \approx G_0 V_{12} G_0$ (the zero-order

term $G_{12} \approx G_0$ reduces the problem to the single-particle one and T vanishes therefore). The z -component of the initial state is taken as a simple analytic one-parameter function, commonly used in the description of inversion layers, surface states, etc. [1,9]:

$$|\Phi^{\perp}\rangle = \begin{cases} \sqrt{\frac{b^3}{2}} \exp(-\frac{bz}{2}) & z > 0, \\ 0 & z < 0, \end{cases}\quad (15)$$

$$\langle q^{\perp} | \Phi^{\perp} \rangle = \sqrt{\frac{b^3}{2}} \frac{1}{(iq^{\perp} + b/2)^2}.\quad (16)$$

Using this initial state and property (12) one can reduce the integration in the transition amplitude (8) to one-dimensional (over perpendicular component of vector \mathbf{p}'), and finally the transition cross section takes form:

$$\begin{aligned}\frac{d\sigma}{dE_1 dE_2 d\Omega_1 d\Omega_2} &\propto |\hat{\epsilon} \cdot \mathbf{K}^+|^2 \int d^2\mathbf{p}_a^{\parallel} \int d^2\mathbf{p}_b^{\parallel} \left| \int d\mathbf{p}'^{\perp} \right. \\ &\times \left. \left\langle \frac{1}{2}(\mathbf{k}_2 - \mathbf{k}_1) | G_0 V_{12} G_0 | \frac{1}{2}(\mathbf{k}_1 + \mathbf{k}_2) - \mathbf{p}'^{\perp} - \mathbf{p}_a^{\parallel} \right\rangle \right|^2 \\ &\cdot \left| \langle \mathbf{p}'^{\perp}, k_1^{\perp} + k_2^{\perp} - \mathbf{p}'^{\perp} | \Phi_{12}^{\perp} \rangle \right|^2 \delta(E_i - E_f) \delta(\mathbf{k}_1^{\parallel} + \mathbf{k}_2^{\parallel} \\ &- \mathbf{p}_a^{\parallel} - \mathbf{p}_b^{\parallel}),\end{aligned}\quad (17)$$

with E_i and E_f from Eq. (13). Integration over \mathbf{p}_a^{\parallel} can be further removed due to the δ -function $\delta(\mathbf{k}_1^{\parallel} + \mathbf{k}_2^{\parallel} - \mathbf{p}_a^{\parallel} - \mathbf{p}_b^{\parallel})$ in Eq. (17), but we leave it in this form to underline the conservation of parallel component of the total momentum of the electronic pair upon transition. From Eq. (14) it is clear that the unscreened Coulomb interaction ($\lambda \rightarrow 0$) favors small momentum transfer $q \rightarrow 0$, i.e. large-distance scattering. This situation changes when the interaction is shielded. In the extreme of very strong shielding $\lambda \gg 1$ only very large momentum transfer $q \geq \lambda$ (hard collisions) do contribute to T . The appropriate geometry to trace this aspect is shown in Fig. 3(b). Both electrons have equal energies and equal angles with respect to the surface normal. The polarization vector is normal to the surface and parallel to $\mathbf{k}_1 + \mathbf{k}_2$. Let us increase both angles θ_1, θ_2 gradually and simultaneously from 0° to 90° , so that $\mathbf{k}_1 + \mathbf{k}_2$ remains always perpendicular to the surface and parallel to $\hat{\epsilon}$. Then the vector $1/2(\mathbf{k}_2 - \mathbf{k}_1)$, which

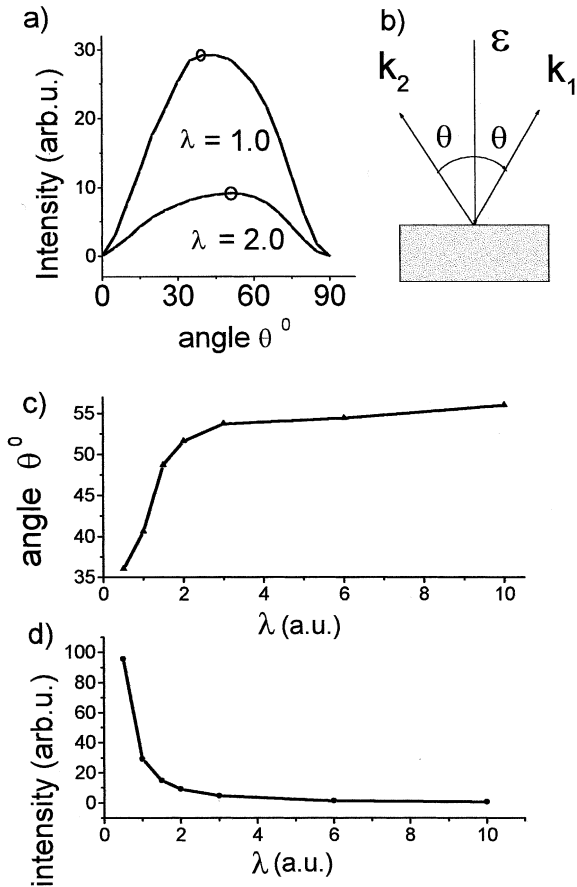


Fig. 3. The effect of screening in DPE angular distributions. Angles θ of two electrons are increased from 0 to 90° simultaneously, $\varphi_1 = 0^\circ$, $\varphi_2 = 180^\circ$, vector $\hat{\epsilon}$ is normal to the plane of the film. (a) DPE intensity as a function of $\theta_1 = \theta_2$, two curves taken at two values of the screening length, maxima are shown by the small circles; (b) sketch of experimental geometry; (c) angles of the maximum value as a function of screening length; (d) Intensity in the maximum as a function of screening length.

enters Eq. (17), continuously grows. It gives the amount of mean momentum transfer, which should happen between two electrons, having their initial state wave vectors uniformly distributed over a region, restricted by the screening length λ . Fig. 3(a) shows the DPE cross section as a function of $\theta_1 (= \theta_2)$. The two curves correspond to two screening lengths. We note at first that the intensity, in particular at the maximum, falls down with increasing λ . This is shown explicitly in Fig. 3(c) and is evident from the fact that the DPE process

depends on the strength of interaction between the electrons. Furthermore we notice that the angular position of the maximum in Fig. 3(a) grows with increasing λ (Fig. 3(d)). That is for more effective screening the photoelectrons escape at larger mutual angles. At $\theta_1 = 0$ no emission happens because electrons cannot follow the same direction with the same energies. At $\theta_1 = 90$ emission is impossible because of the DPE selection rules. The maximum is somewhere in between and finally seems to saturate with λ , which is a consequence of saturation of the potential form-factor itself.

5. Conclusion

The aim of this work has been to trace the footprints of the degree of localization and correlation in a confined system, a QD. As tool to realize this we proposed the one-photon two-electron excitation technique and developed the necessary theoretical and numerical concepts to evaluate the two-particle excited spectrum. The prime focus is put on the manifestation of quantum size and correlation effects in the two-particle excited spectrum. The photon field is treated within the dipole approximation whereas the Green function method is employed to describe the excited two-particle spectrum of the many-electron finite system. We discussed the DPE process in simple systems, ranging from the ‘atomic’ version (in a QD) to its ‘surface’ counterpart (in a thin film). Our numerical simulations demonstrate that the degree of confinement as well as the amount of inter-electronic correlation have a marked effect on the double excitation spectrum. The underlying physics of these effects can be understood from the functional structure of the inter-electronic interaction in a many-electron system as well as from the characteristic change of the electronic states when varying the size of the system.

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