Photoluminescence of a Quantum Dot Hybridized with a Continuum of Extended States

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We calculate the intensity of photon emission from a trion in a single quantum dot, as a function of energy and gate voltage, using the impurity Anderson model and variational wave functions. Assuming a flat density of conduction states and constant hybridization energy, the results agree with the main features observed in recent experiments: non-monotonic dependence of the energy on gate voltage, non-Lorentzian line shapes, and a line width that increases near the regions of instability of the single electron final state to occupations zero or two.

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Semiconductor quantum dots (QD’s) have attracted much attention recently and have been proposed for numerous applications. It has been established that they are promising candidates in quantum information processing, with spin coherence times of several microseconds [1, 2] and fast optical initialization and control [3, 4, 5]. QD’s are also of interest in the field of spintronics (electronics based on spin) [6, 7, 8]. Optical means of manipulation [8] and detecting [7] the spin were proposed. QD’s might also be used as semiconductor optical amplifiers [9]. Most of the above cited works involve optical transitions which create or destroy excitons. In particular, when a positive gate voltage is applied, configurations with one more electron can be stabilized, and the optical transition takes place between a state with the valence band full and only one electron in the conduction band, and a state which consists of two conduction electrons and a hole in the valence band (a trion) [3, 4, 5, 10, 11, 12, 13]. The gate voltage $V_g$ allows to control the spin dynamics and the optical emission $10, 11, 12, 13$.

An interesting aspect of the photoluminiscence (PL) decay of the excitons and trions, is the manifestation of the hybridization of the spin localized in the QD with a continuum of extended states, and the related Kondo effect [12, 13, 14]. This hybridization can be described by the impurity Anderson model [15] [see Eq. 7 below]. In the limit of small hybridization and odd number of electron in the QD, this model reduces to the Kondo model, in which the localized spins have an exchange interaction with the spin of the extended states [12]. The Kondo physics is also clearly present in transport properties of QD’s [16, 17].

In recent experiments, the PL that results from the decay of the $X^1−$ trion has been measured as a function of $V_g$ [13]. The PL is a consequence of an optical transition from the trion to a state with one localized electron in the QD, hybridized with a continuum. As a consequence of this hybridization, the PL is broad (half width at half maximum $w \sim 1$ meV) near the low $V_g$ limit of stability of the trion (for lower $V_g$, the neutral exciton $X^0$ is stable). This limit is slightly below $V_g^0$, where we call $V_g^n$ the gate voltage for which the configurations with $n$ and $n+1$ electrons in the QD, and no holes in the valence band, are degenerate. The striking observed behavior can be summarized in three main results: i) as $\Delta V_g = V_g - V_g^0$ increases from negative values, the PL first blueshifts and then redshifts, ii) the PL line shape is non-Lorentzian, with a low energy tail, iii) while $w \sim 1$ meV for $V_g \sim V_g^0$ or $V_g \sim V_g^1$ (near the limits of stability of one electron in the QD), the PL line is much narrower for intermediate values of $V_g$. This has been noted before [3]: while at intermediate gate voltages Atatüre et al. succeeded to prepare a spin state with more than 99.8 % fidelity, the coupling of the spin with the reservoir at the edges ($V_g \sim V_g^0$ or $V_g \sim V_g^1$) spoils this efficiency.

The line shape has been quantitatively fit only for $V_g \sim V_g^0$ using a one-particle approach with additional questionable assumptions [13]. In this Letter, we calculate the PL on the whole range of gate voltages using many-body variational wave functions (VWF’s) [17, 18]. Our results provide an explanation of the three main experimental results mentioned above and show that they are a consequence of strong correlations. We also show that the shift of the PL line is directly related with its asymmetry by a simple sum rule.

Using Fermi’s golden rule, the PL intensity at zero temperature is given by

$$ I(\omega) = \frac{2\pi}{\hbar} \sum_f |\langle f | H_{LM} | i \rangle|^2 \delta(h\omega + E_f - E_i), \quad (1) $$

where $\omega$ is the PL frequency, $|i\rangle$ is the initial (trion) state, $|f\rangle$ denotes the possible final states, $E_f$ is the energy of the state $|j\rangle$, and the relevant part of light-matter interaction can be written as

$$ H_{LM} = (A \sum_{\sigma} d_{\sigma}^\dagger p_{\sigma} + H.c.), \quad (2) $$

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where $A$ is proportional to the vector potential, $d_{i}^{\dagger}$ creates an electron at the QD (with spin $\sigma$), and $p_{r}$ annihilates a valence electron at the QD, with the same symmetry as that of the corresponding absorbed photon.

The experimental results indicate that the hybridization of the trion state with the continuum is small. In any case the hybridization between holes and extended states favors a singlet state. Taking the limit of vanishing hybridization of the VWF of Varma and Yafet [13] for this case [in analogy to Eqs. (8), (9) and the discussion below them], one obtains [19]

$$|i\rangle = \frac{1}{\sqrt{2}} \sum_{\sigma} c_{kF\sigma}^{\dagger} d_{i}^{\dagger} |S\rangle,$$

where $c_{k\sigma}^{\dagger}$ creates a conduction electron in a spherical wave with wave vector $k$, $k_{F}$ refers to the Fermi wave vector and $|S\rangle$ is the many-body state with all valence states of the dot and all conduction states below the Fermi energy occupied. Replacing Eq. (3) in Eq. (11), the intensity becomes

$$I(\omega) = \frac{2\pi}{\hbar} |A|^{2} \rho_{1}(E_{i} - \hbar\omega),$$

where

$$\rho_{1}(\omega) = \sum_{f} |\langle f |1\rangle|^{2} \delta(\hbar\omega - E_{f})$$

is the spectral density of the many-body state

$$|1\rangle = (d_{i}^{\dagger} c_{kF\uparrow}^{\dagger} - d_{i}^{\dagger} c_{kF\downarrow}^{\dagger}) |S\rangle \sqrt{2},$$

obtained after the application of $H_{L,M}$ [Eq. (2)] on $|i\rangle$ [Eq. (3)].

Since in the possible final states $|f\rangle$, the holes are absent and do not play any role, our task reduces to calculating $\rho_{1}$ for the Hamiltonian of the Anderson model, which reads

$$H = E_{d} \sum_{\sigma} d_{i}^{\dagger} d_{i} + U d_{i}^{\dagger} d_{i}^{\dagger} d_{i} + \sum_{k\sigma} \epsilon_{k}\epsilon_{k\sigma} c_{k\sigma}^{\dagger} c\sigma + V_{d}\delta_{k\sigma} + H.c.,$$

where $E_{d} = E_{d}^{0} - eV_{g}/\lambda$ (with $\lambda \simeq 7$ [13]) is controlled by the gate voltage $V_{g}$.

It is easy to see that in the non-interacting case ($U = 0$), $\rho_{1}$ is given essentially by the spectral density of $d$ electrons, which is a symmetric Lorentzian for the usual assumptions of constant $V_{k}$ and constant density of extended states per spin $\rho(\omega)$ [20]. Therefore, to explain the observed behavior within a one-electron picture, some structure in either $V_{k}$ or $\rho(\omega)$ has to be assumed [13]. Instead, as we show below, the three main experimental results mentioned above appear naturally when the interactions are properly taken into account.

We solve the problem using many-body VWF's [13, 18]. Similar VWF's were used successfully to study the interaction between spins of two QD's [21]. We extend previous approaches to the case of finite $U$. To simplify the discussion, we begin assuming $U \to \infty$, which is a good approximation when $E_{d}$ is near the Fermi energy $\epsilon_{F} = \epsilon_{kF}$, which we take as zero in what follows. For $U \to \infty$, the VWF proposed by Varma and Yafet is a good approximation for the correlated ground state $|g\rangle$ of $H_{L,M}$ [15, 18]. Similar VWF's were used successfully to study the interaction between spins of two QD's [21].

### Solution

$$|g\rangle = \alpha|F\rangle + \sum_{k\sigma} \frac{\beta_{k}}{\sqrt{2}} d_{i}^{\dagger} c_{k\sigma}|F\rangle,$$

where $|F\rangle = c_{kF\uparrow}^{\dagger} c_{kF\downarrow}^{\dagger} |S\rangle$. Assuming constant $V_{k}$ and $\rho(\omega)$, the resulting approximate ground state energy $E_{g}$ comes from the solution of the equation

$$E_{d} - \frac{\beta}{\sqrt{2}} (2\Delta/\pi) \ln [(W + \delta)/\delta] = 0,$$

where $\beta = E_{d} - E_{g} > 0$, $\Delta = \pi\rho V^{2}$ is the resonant level width and $-W$ is the bottom of the band of extended states [22]. Note that in the limit $V \to 0$ when $E_{d} < \epsilon_{F}$ Eq. (12) gives $\rho(\omega)$ [1]. Therefore, the state $|g\rangle$ can be regarded as the limit of the Kondo ground state, when the Kondo temperature $T_{K} \sim \exp(\pi E_{d}/2\Delta) \to 0$.

In the thermodynamic limit, $|g\rangle$ is orthogonal to the state $|1\rangle$. However, it is easy to see that excited states can be obtained making electron-hole excitations on $|g\rangle$, which have energy near $\langle 1|H|1\rangle = E_{d} [22]$ and hybridize with $|1\rangle$. This hybridization leads to a broadening of $\rho_{1}(\omega)$, in a way analogous to a resonant level model, in which a localized state is mixed with a continuum [20]. Specifically we consider the states

$$|k\rangle = \alpha^{k}\epsilon_{k\uparrow} + \sum_{k} \beta_{k}^{k} y_{kk\uparrow},$$

where

$$|\epsilon_{k}\rangle = \frac{1}{\sqrt{2}} (c_{kF\uparrow}^{\dagger} c_{kF\downarrow}^{\dagger} - c_{kF\downarrow}^{\dagger} c_{kF\uparrow}^{\dagger}) |S\rangle,$$

$$|y_{kk\uparrow}\rangle = \frac{1}{\sqrt{2}} \sum_{\sigma} d_{i}^{\dagger} c_{k\sigma}^{\dagger} c_{k\sigma}^{\dagger}. $$

Here the subscript $k$ ($k'$) denotes extended states below (above) the Fermi energy. It is easy to see that optimization of the coefficients for each $k'$ (minimizing the energy), leads to $\alpha^{k} = \alpha$, $\beta_{k}^{k'} = \beta_{k}$, independently of $k'$. In addition, $||1|H||2^{2} = \alpha V_{k}^{2} = (1 - n_{d})|V_{k}^{2}|^{2}$,
where \( n_d = \langle g | \sum_{\sigma} d_\sigma^\dagger d_\sigma | g \rangle \) is the electronic occupation at the QD. From the effective resonant level model, one obtains a broadening of the PL line given approximately by \( w = (1 - n_d) \Delta \). This is a simple, elegant result which agrees qualitatively with experiment near the intermediate valence regime (IVR) \( |E_d - \epsilon_F| \sim \Delta \), and predicts an exponentially small width in the Kondo regime \( \epsilon_F - E_d, E_d + U - \epsilon_F \gg \Delta \). However, since double occupancy is neglected, this result is clearly wrong in the other IVR \( |E_d + U - \epsilon_F| \sim \Delta \), while in the Kondo regime, virtual fluctuations through double occupied states were neglected.

To improve this calculation, we first replace the states \( |y_{kk'}\rangle \) in Eq. (11) by others \( |\tilde{y}_{kk'}\rangle \), which take into account double occupancy

\[
|\tilde{y}_{kk'}\rangle = \gamma |y_{kk'}\rangle + \sum_{q\sigma} \frac{\beta_{q\sigma}}{\sqrt{2}} d_\sigma^\dagger c_{q\sigma} |y_{kk'}\rangle, \tag{12}
\]

where the coefficients are determined minimizing \( \langle \tilde{y}_{kk'} | H | \tilde{y}_{kk'} \rangle \), for fixed \( k \) and \( k' \). Second, in a similar way to the resulting states \( |k'\rangle \), we construct linear combinations \( |k\rangle \) appropriate for the other IVR \( |E_d + U - \epsilon_F| \sim \Delta \). This is done easily using a symmetry property of \( H \):

\[
d_1^\dagger \rightarrow d_1, \ c_{k\uparrow}^\dagger \rightarrow -c_{k'\downarrow}, \ d_1^\dagger \rightarrow -d_1, \ c_{k'\downarrow}^\dagger \rightarrow c_{k\uparrow}, \text{ with } \epsilon_{k'} = -\epsilon_k \text{ (for } \epsilon_F = 0)\], which changes \( -E_d \) into \( E_d + U \) and therefore interchanges IVR’s. For example using this transformation \( |e_{k'}\rangle \rightarrow d_1^\dagger d_1^\dagger \sum_{\sigma} c_{k\sigma}^\dagger c_{k\sigma} |S\rangle /\sqrt{2} \). Third, since the states \( |k'\rangle \) and \( |k\rangle \) are not orthogonal, we discretize them and use a symmetric Löwdin procedure to orthogonalize them \[23\]. Finally, including the state \( |1\rangle \), we numerically diagonalize \( H \) and calculate the spectral density using Eq. (13).

The parameters were chosen as follows: \( \Delta \) is our unit of energy. It should be of the order of 1 meV to give the observed PL width for \( E_d \sim \epsilon_F \). Guided from experiment \[13\], we took \( U = 24\Delta \). Finally we took a broad flat band symmetrically placed around \( \epsilon_F \) of width \( 2W = 100\Delta \). The results depend only logarithmically on \( W \) [see for instance Eq. (9)] and the band edges are very far form the region of interest. Therefore there are no particular features of the band or hybridization that can affect the results. To discretize the states \( |k'\rangle \) and \( |k\rangle \), the band was divided in at least 2000 points with decreasing energy separation towards the Fermi energy [24].

In Fig. 1 we represent \( \rho_1(\omega) \) as a function of frequency \( \omega \) for various values of \( E_d \). This density with the frequency axis inverted and shifted by the energy of the initial state \( E_i = E_S \) [22] corresponds to the PL spectrum [see Eq. (1)]. Due to our choice of a symmetric band, and the above mentioned symmetry property of \( H \), the densities for \( E_d \) and \( -E_d - U \) are identical. Therefore, one can restrict the study to \( E_d > -U/2 = -12\Delta \). Note that in most curves an asymmetry with a high energy tail (corresponding to a low energy tail in the PL) is evident, in agreement with experiment. We return to this point below. For \( E_d = 0 \) or slightly negative, there are two relative maxima in \( \rho_1(\omega) \). This structure is already present using the simplest VWF’s given by Eqs. (10) and (11). The peak at smallest energies corresponds to excitations near the ground state. At low energies, one expects that \( \rho_1(\omega) \) has a power-law behavior characteristic of x-ray edge singularities [14, 26] which is not captured by our variational approach. This structure might be washed by a temperature or other broadening effects not included in our calculations. This is likely the reason why a structure with two peaks has not been so far reported. Note that while the density near the ground-state energy can be described more accurately with the numerical renormalization group [14, 25], this technique does not resolve accurately the structure at higher energies [25, 27].

One striking feature observed in Fig. 1 is the dra-
matic narrowing of the peak as the system leaves the IVR and enters the Kondo regime. In Fig. 2 we represent the half width at half maximum as a function of the dot level, which is related to the gate voltage by $E_d = -e\Delta V_g/\hbar$. The slight increase of $w$ for $\Delta V_g = 0$ (and also $E_d = -24\Delta$) might be related to the two-peak structure of $\rho_1(\omega)$ near these values of $E_d$ and we believe it is not too significative. Instead, for the symmetric case $E_d = -12\Delta$, we obtain $w = 5 \times 10^{-4}\Delta$, corresponding to a decrease of the line width in three orders of magnitude. This is consistent with different experiments [3,11,12].

In Fig. 2 we plot $E_M$, the energy $\hbar\omega$ for which $\rho_1(\omega)$ reaches its maximum value as a function of $E_d = -e\Delta V_g/\hbar$. The shift $E_M - E_d$ is always negative (the peak is displaced towards the ground state energy). This means that the energy of the final state decreases and therefore, the position of the PL peak is always blueshifted with respect to the situation without hybridization. However, the non-monotonic behavior observed in Fig. 2 implies that as $\Delta V_g$ increases starting from negative values, the PL peak first blueshifts, but after $\Delta V_g = 0$, the peak redshifts, in agreement with experiment [12]. The fact that the maximum shift is obtained near the IVR’s might be expected from the fact the maximum energy gain in the ground state is obtained at those regimes.

It is easy to see from Eq. 3 that $\int d(\hbar\omega)\rho_1(\omega) = \langle 1|H|1 \rangle = E_d$ [22]. In words, the center of gravity of the density states remains at $E_d$ when the QD electron hybridizes, while its maximum displaces at a lower energy $E_M$. Therefore, to satisfy the sum rule, $\rho_1(\omega)$ should be asymmetric with a high energy tail, and the degree of asymmetry is directly proportional to the magnitude of the shift $|E_M - E_d|$.

In summary, using an impurity Anderson model to describe the final states of a photoluminescence experiment in a quantum dot, with the simplest possible assumptions for the density of extended states and their hybridization with the dot electrons, we obtain a blueshift and correlated asymmetry of the line shape, and a line width, which explains the main features of recent experiments in the whole range of applied gate voltage.

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