Power dependence of the photocurrent lineshape in a semiconductor quantum dot

A. Russell and Vladimir I. Fal’ko
Department of Physics, University of Lancaster, Lancaster LA1 4YB, United Kingdom

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We propose a kinetic theory to describe the power dependence of the photocurrent lineshape in optically pumped quantum dots at low temperatures in both zero and finite magnetic fields. We show that there is a crossover power \( P_c \), determined by the electron and hole tunneling rates, where the photocurrent no longer reflects the exciton lifetime. For \( P > P_c \), we show that the photocurrent saturates due to the slow hole escape rate, whereas the linewidth increases with power. We analyze the spin-doublet lineshape in high magnetic fields and determine to what measure it reflects the degree of circular polarization of incident light. © 2007 American Institute of Physics.

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In the recent years, advances in experimental techniques have allowed for optical studies on single self-assembled quantum dots (QDs), e.g., by using spatially resolved photocurrent (PC) spectroscopy. Typically in these experiments, QDs were embedded in biased p-i-n junctions, permitting to control the tunneling rates of optically generated electrons and holes from the dot and, therefore, the photocurrent formed by electrons and holes escaping in opposite directions. In principle, measurements of photocurrent can be used as a detection method to study spectral properties of a dot. In this letter, we analyze the kinetics of an optically pumped QD (e.g., InGaAs/GaAs) and study the power dependence of the photocurrent. We show that at low powers, the PC linewidth manifests the exciton lifetime in the dot, at high powers it becomes strongly influenced by dot kinetics and no longer reflects the properties of the dot. The crossover power between these two regimes is found to depend on both the electron and hole tunneling rates, as well as the efficiency with which photons are absorbed by the QD. The analysis is also extended onto the case of high magnetic fields and takes into account the degree of circular polarization of the incident light.

To model the power dependence of the photocurrent \( I_{PC} \) generated when a QD is optically pumped with monochromatic light of frequency \( \omega \) and power \( P \), we consider the processes shown in Fig. 1. Here, a pair of an electron and heavy hole in the lowest confined states in the dot (exciton) is created by either a \( \sigma^+ \) or a \( \sigma^- \) photon, followed by their escape to an electrode. In large electric fields \( F \), the dominant escape mechanism from the dot for the electron and hole is by tunneling, with the power-independent rates \( \gamma_{e,h}/h \propto \exp(-\sqrt{2m_{e,h}}\Delta V_{r}/3\hbar F) \). Due to a smaller electron mass, it is natural to assume that the hole tunnels slower than the electron (\( \gamma_e \gg \gamma_h \)). The escape of a photoinjected electron into the bulk semiconductor (e.g., GaAs) permits slightly off-resonant excitation of the dot, where the energy of the absorbed photon \( h\omega \) slightly differs from the exciton energy \( E_0 \). Note that in such a case, energy is conserved by the electron tunneling into the bulk semiconductor continuum of energy states, with the exciton appearing on the dot only virtually. As a result, the QD tends to be positively charged, blocking the absorption of another photon until the hole escapes and thus limiting the size of the photocurrent (which is generated by the repetition of such processes). Blocking of the next interband transition on the dot happens due to (a) the Fermi blocking for the transition in the same circular polarization and (b) the strong shift of the interband transition energy for a transition in the opposite polarization exciting the dot into a charged exciton state (as compared to the neutral one).

Below we develop a theory for the PC spectroscopy of dots in both zero and finite magnetic fields. The latter gives rise to an exciton Zeeman splitting \( \pm \gamma_z \), making the dot sensitive to the polarization degree of the optical pump \( \sigma \), where \( \sigma = \pm 1 \) corresponds to fully polarized \( \sigma \) light. The projection of the photon angular momentum onto the \( z \) axis, defined as the growth direction of the dot, is conserved in the system. Due to this, \( \sigma^\pm \) photons can only excite spin \( S_z = \pm 1/2 \) electrons and spin \( J_z = \pm 1/2 \) holes, forming excitons of energy \( E_0 \pm \gamma_z \) (Fig. 1). The excitation rate of the empty dot into a positively charged state with a \( J_z = \pm 1/2 \) hole takes a resonant form,

\[
\tilde{w}_e = \frac{1 \pm \sigma}{2} \frac{\gamma_e \alpha P}{\left( \delta \pm \frac{1}{2} \gamma_z \right)^2 + \frac{1}{4} \gamma_c^2}
\]

FIG. 1. (Color online) On the left, a schematic of the dot showing the processes involved in the rate \( w_e \). The dot is excited by a \( \sigma^+ \) photon, generating an exciton consisting of a spin \( J_z = +1/2 \) hole and a spin \( S_z = -1/2 \) electron, which quickly tunnels to the contacts resulting in the dot being positively charged. On the right, the same but for the rate \( w_- \) corresponding to excitation by a \( \sigma^- \) photon.

aElectronic mail: a.russell2@lancaster.ac.uk
have the following form;

\[ \frac{\partial}{\partial t} n^\pm + \gamma_h n^\pm = \frac{e}{\hbar} \left( w_+ + w_- \right) n, \]  

(4)

Using Eq. (1) we can now describe the photocurrent as a function of the electron and hole tunneling rates \((\gamma_e, \gamma_h)\), and the power, frequency, and polarization degree of light \((P, \omega, \sigma)\). In zero magnetic field, the photocurrent, given by Eq. (4), loses its \(\sigma\) dependence and only has one resonance at \(\delta=0\),

\[ I_{PC} = \frac{e \gamma_e P}{\delta^2 + 1^2}, \]  

(5)

where, typically for two-level systems,\(^1^9\)

\[ \Gamma = \frac{1}{2} \gamma_e \sqrt{1 + \frac{P}{P_c}}, \]  

(6)

where \(P_c = \gamma_e \gamma_h / 4 \alpha \hbar\). The lineshape of the dot as manifested in the photocurrent [Eq. (5)] is shown in Fig. 2 for various different powers. It is characterized by the linewidth \(\Gamma\) and integral strength \(A = \int I_{PC} d\omega\),

\[ A = \frac{2e\pi \alpha P}{\hbar \sqrt{1 + P/P_c}}. \]  

(7)

(The latter should not be confused with the integral photocurrent generated by white light.\(^2^0\))

The properties of the QD spectrum measured using photocurrent can be described by considering the two power regimes \(P \ll P_c\). In low powers, \(P < P_c\), the peak height, \(I_{PC}(\delta=0)\), increases proportionally to power, and the linewidth \(\Gamma \sim 1/\gamma_e\), reflects the lifetime of the exciton in the dot. When \(P > P_c\), the linewidth becomes strongly power dependent: \(\Gamma \sim 1/\gamma_e \sqrt{P/P_c}\). This result is similar to the phenomenological ansatz made in Ref. 11 and agrees with the recent observations in Ref. 9. Simultaneously, the integral strength increases at high powers, \(A = \sqrt{P}\), whereas the peak height saturates at \(e\gamma_e / \hbar\), indicating that the slow hole tunneling rate limits the magnitude of the photocurrent (as observed in Refs. 3,9,11).

In the presence of a magnetic field, photoexcitation of the dot with fully polarized light \((\sigma = \pm 1)\) results in the same photocurrent properties as described above, since only an exciton of one polarization can be excited into the dot. For \(\sigma \neq \pm 1\), the photocurrent takes the bimodal form shown in Fig. 3 for \(\sigma = -0.6\). In Fig. 3(a), the Zeeman splitting is small,
and even at low powers the two peaks overlap. In the high power limit, \( P \gg P_c \), the photocurrent behaves in a similar fashion as in the zero magnetic field case.

If \( E_Z^2 \) is large as compared to a typical linewidth, the two PC peaks become distinct even at high powers [Fig. 3(b)] and both display similar properties as the single peak described in Eq. (5). The value of \( \sigma \) determines how often one electron-hole spin configuration is excited into the dot as compared to the other. Thus one may expect that the polarizaton degree of light is reflected in the magnitude of the photocurrent at the energies \( \delta \pm \frac{1}{2} E_Z \), as is the case for low powers seen in Fig. 3(b). However, for high powers \( P \gg P_c \), the peaks both saturate at \( e \gamma_0 / h \) as described previously, and therefore their heights no longer reflect the value of \( \sigma \). Instead, it is now manifested by their respective linewidths which reflect the size of the area under the corresponding line.

To conclude, we have shown that there is a crossover power \( P_c \) beyond which the linewidth of the photocurrent no longer manifests the lifetime of electron states on the dot. At low powers, the dot (such as an InGaAs dot in a GaAs matrix) is nearly always empty and the photocurrent is proportional to the excitation rate, with broadening then reflecting the lifetime of the exciton. At high powers, the dot is more often occupied by a hole (therefore it is positively charged) and the height of the photocurrent peak saturates at \( e \gamma_0 / h \), determined by the slower hole escape rate. The cross-over power \( P_c = \gamma_c \gamma_0 / 4h \alpha \) depends on both the electron and hole escape rates, as well as the efficiency with which the dot absorbs photons incident on the dot. The proposed theory of PC lineshape at low temperatures explains the recent experimental observations of PC measurements on optically pumped quantum dots.3,9,11

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13We ignore the small effects on \( \gamma_{\pm} \) due to the electron and hole Zeeman energies, so that \( \Delta \gamma_{\pm} \) is independent of the electron/hole spin.
15In Ref. 3, a kinetic model has been used which considered resonant excitation of the dot, where the dot can be occupied by both a photoexcited electron and a hole. Such a model is formally applicable only when inelastic (e.g., electron-phonon) interactions make it possible for the optical absorption of a nonresonant photon, which may be the case at high temperatures. Also, the analysis in Ref. 3 does not give the power dependence of the lineshape of the photocurrent. In contrast, the analysis presented in this paper, which focuses on the low temperature regime, is applicable to the description of the PC generated by slightly off-resonant photons. This is because, in the presented analysis, an exciton occupies the dot only virtually, with energy conserved by the electron tunneling into the spectral continuum of the bulk semiconductor. As such our theory not only describes the saturation of the peak height but also predicts the lineshape of the photocurrent, and the power broadening, as experimentally shown in Refs. 9 and 11.
18Double occupation of the dot by two holes is prohibited in our model by a large Coulomb energy \( E_c = 1/2 e^2 / \epsilon a T \), where \( e \) is the dielectric constant and \( a \) is the size of the dot.
20The integral photocurrent is generated by nonmonochromatic light which one would attribute to a QD assembly, pumping the quantum dot with photons of various frequencies. In such a case, the excitation rate in Eq. (1) is given by the integral \( w_D = (1 + \sigma) / 2 \int \rho = a \rho(\omega) [1 + (1 + \sigma)^2] d\omega / (1 + \sigma) \). Here, \( \rho(\omega) \) is the spectral density of photons arriving with frequency \( \omega \), with the units of rate. Integrating and substituting into Eq. (4), we arrive at \( I_{\omega} = 2 \sigma a P / (1 + \sigma) / (1 + \gamma P / P) \), in agreement with Refs. 3, 9, and 11.