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Non-equilibrium carriers in type-II quantum dots

A. Shik†, H. Ruda† and E. H. Sargent‡
† Energenius Centre for Advanced Nanotechnology,
University of Toronto, Toronto M5S 3E4, Canada
‡ Department of Electrical & Computer Engineering,
University of Toronto, Toronto M5S 3G4, Canada

Abstract. The spatial distribution and recombination of nonequilibium electrons and holes in a system of type-II quantum dots is considered theoretically. In this system, contrary to the well-known type-I quantum dots system, one type of carrier is confined to the dot whereas the other type is localized in a Coulomb potential well outside the dot. The alters the recombination processes dramatically and results in new intensity and temperature dependencies in the photoconductivity. Possible device applications of type-II quantum dots, in particular for photovoltaic structures, are discussed.

Properties of non-equilibrium carriers in quantum dots (QD) have been actively studied in recent years, especially in connection with QD lasers. These investigations concerned QDs which employed type-I heterojunctions, wherein the narrow-gap dot material presents a potential well for both electrons and holes. There exists, however, another group of semiconductor heterojunctions, so-called type-II junctions, where the band diagram has a staggered character so that the material with lower potential energy for electrons has higher energy for holes and vice versa (Fig. 1(a)). This type of band diagram is realized in Ge_xSi_{1-x} alloys, in a series of III-V-compounds, particularly, $In_{1-x}Ga_xAs_{1-y}Sb_y$, and in AlAs/GaAs nanostructures with very thin GaAs layers. The problem of type-II QDs has become especially urgent in the recent years in connection with the creation and investigation of polymer-nanocrystal composites for light-emitting and photovoltaic devices [1, 2] since conducting polymers tend to have low electron affinities and form type-II junctions with the majority of semiconductors and dielectrics.

In the present work we consider the optical generation, spatial separation and recombination of non-equilibrium carriers in a system of QDs forming type-II heterojunctions with the matrix semiconductor. We discuss the prospects of using these structures in photovoltaic and light-emitting devices.

In the system considered, Coulomb effects resulting from charge separation play a crucial role, in contradistinction with the case of type-I QDs. One type of carrier (usually electrons) is captured into the quantum well of the QD, whereas the other type is concentrated mostly outside the QD in the potential well formed by Coulomb forces (Fig. 1(b)). First, we calculate self-consistently the potential profile of the system and find its main parameters: the band bending V_0 and the QD charge Q for different electron and hole Fermi quasi-levels F_n and F_p characterizing non-equilibrium carriers. We assumed $Q/e \gg 1$ which allows us to use the quasi-classical approximation in our calculations. At a very high excitation intensity both electron and hole gases become degenerate, and the problem requires solution of the corresponding Thomas–Fermi equation.

We proceed to calculate the recombination rate in a type-II QD given Q and V_0. Since electrons and holes are spatially separated, recombination requires either tunneling or ther-

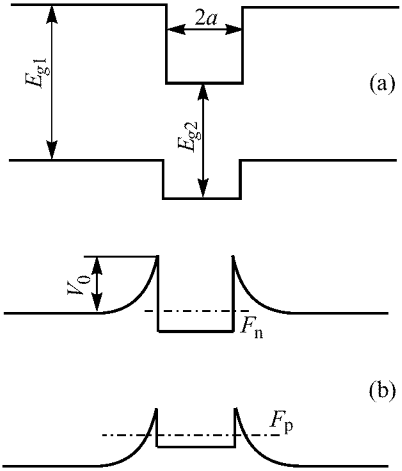

Fig. 1. Band diagram of an undoped type-II quantum dot in equilibrium (a) and under illumination (b).

mal activation. For III–V semiconductor heterosystems the main recombination mechanism at moderate temperatures is connected with the tails of electron wave functions in the matrix material. For spherical QDs, the corresponding radiative transitions occur between the states with equal angular momenta but have no selection rules in terms of the principal quantum number. This results in a broad spectrum of emitted radiation. The recombination rate depends weakly on the quasi-level positions and on the band bending V_0. In systems with a dramatic mismatch of wave functions at the QD interface (in particular, for polymer-based mixtures) tunneling effects are weak and recombination is related mostly with thermal activation. In this case the recombination rate is an exponential function of F_n, F_p and V_0. If the QD has a smaller band gap than the matrix (as in Fig. 1), F_p may cross the valence band inside the QD under high excitation. Holes will then enter the QD and recombination becomes activationless.

The calculations of recombination rate allow us to express the quasi-level energies F_n and F_p in terms of the light intensity I and, ultimately, calculate photoelectric phenomena in composite media containing type-II QDs. It is shown that the separation of non-equilibrium carriers in these systems increases the hole component of photoconductivity, decreases the electron component, and results in a substantial increase of the total photoconductivity $\Delta\sigma$ [3]. The effect is observed experimentally in a wide range of polymer-based composites with disparate inclusions [4–6]. When recombination has an activation (rather than tunnel) character, the modulation of V_0 by the light intensity results in a strongly sublinear character of the $\Delta\sigma(I)$ dependence.

In practical designing of QD-based light-emitting and photovoltaic devices the problem of optimal spatial distribution of QDs in the active region, $N(x)$, arises. In ordinary semiconductors with high carrier mobilities F_n and F_p are constant throughout the region and the profile of this distribution does not play a role. However, in polymer-nanocrystal mixtures, due to very low mobilities in conducting polymers, the processes of carrier separation and recombination are determined by their transport in the matrix and depend on the $N(x)$ profile. The complete theoretical description includes the equations connecting Q, V_0 and the recombination-generation rate with local carrier concentration, the continuity equations for electrons and holes in the matrix, and the Poisson equation describing the electric field distribution in the active region [7]. This system was solved for a light-emitting diode and its current-voltage characteristics and quantum yield η were calculated at different $N(x)$. It was shown that to maximize η, QDs must be concentrated in a thin layer shifted towards the contact with lower injection effectiveness. The theoretical results are in agreement with the experimental data, both from literature [1, 2, 4] and obtained in our laboratory for the PPV-CdS composites.

Type-II QDs are particularly promising for use in photovoltaic devices. In type-I systems photogenerated electrons and holes are concentrated in the dot regions and their effective separation requires additional excitation into the matrix material, which may suppress the effectiveness of photovoltaic devices. In type-II systems, the interface barriers do not prevent non-equilibrium carriers from effective separation.

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