Auger Recombination in Semiconductor Quantum Dots

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INTRODUCTION
A quantum dot (QD) is a nanometer-sized piece of material, usually a semiconductor, with unique electronic and optical properties. Typical sizes of QDs are 2–10 nm in diameter, containing 100–10,000 atoms. They have few conduction electrons and the reduced size leads to quantum confinement and the appearance of atom-like energy levels as shown in Figure 1.

Semiconductor QDs have been researched extensively for their applications in lasers and solar cells. The advantageous properties of QDs over bulk semiconductor materials include a “tunable” bandgap energy [1], and a high carrier density resulting in multielectron production from a single photon [2]. The former allows lasers to emit at specifiable wavelengths (see Figure 2), and the latter the construction of extremely efficient solar cells.

The pathway of recombination of holes and electrons (excitons) has, in particular, attracted a lot of attention. Experimental evidence [3] shows that nonradiative Auger recombination is the dominant mechanism of electron relaxation. Auger recombination (illustrated in Figure 3) occurs when an exciton recombines and instead of emitting light, transfers the energy to a nearby electron (or hole), creating a “hot” electron (hole). In QD lasers, nonradiative processes are to be minimized, while in PV cells, exciton recombination detracts from the overall quantum yield. Hence, an accurate description of the relaxation time (the inverse of recombination rate) is essential to increasing the effectiveness of QDs.

We present a derivation that obtains agreement with the known bulk rate and modify it to account for the discrete atom-like energy levels in the QD.

METHOD
We used second-quantized methods to write the interaction operator and solved for the transition matrix element assuming Bloch wave-functions (valid for crystals sufficiently many atoms across). Using Fermi’s Golden Rule we then obtained a rate expression in terms of a sum over initial and final wave-vectors of this matrix elements and occupation probabilities (given by Boltzmann statistics; valid in this exponentially prohibited reaction) with a delta function enforcing energy conservation. The sum can be converted to an integral and evaluated using saddle point methods, and gives the correct size and exponential temperature dependence (Eq. 1).

Derivation of Rate in Bulk
We used second-quantized methods to write the interaction operator and solved for the transition matrix element assuming Bloch wave-functions (valid for crystals sufficiently many atoms across). Using Fermi’s Golden Rule we then obtained a rate expression in terms of a sum over initial and final wave-vectors of this matrix elements and occupation probabilities (given by Boltzmann statistics; valid in this exponentially prohibited reaction) with a delta function enforcing energy conservation. The sum can be converted to an integral and evaluated using saddle point methods, and gives the correct size and exponential temperature dependence (Eq. 1).

Figures 1-4. Length scales governing quantum confinement: Lp is the temperature length and L is the physical size. The dot result is an extrapolation, not the result of a derivation.

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EQUATIONS

Figure 1. Continuous energy levels in bulk (left) give way to discrete energy levels in QDs (right).

Figure 2. CdSe QDs of different sizes can fine-tune optical absorption properties.

Figure 3. Auger recombination schematically in terms of energy bands. Two electrons and a hole (left) recombine to form a “hot” electron (right), with time constant τe.

Figure 4. Quantum Dot Rate
It has been postulated that the exciton pair immediately relaxes to its first excited state before undergoing Auger recombination. Therefore, we take the probability of occupation to be 1 for lowest-lying states and 0 otherwise. We also assume that the length scale corresponding to the bandgap energy is much less than the size of the dot given that typical bandgap energies correspond to Angstrom length scales and typical QDs have sizes of greater than a few nanometers. With carrier kinetic energies much less than the required bandgap energy, we must broaden the optical absorbtion properties.

REFERENCES

ACKNOWLEDGEMENTS
We would like to thank Prof. Walter Lambrecht for his invaluable input and Prof. Jie Shan for procuring the funding. This research was made possible in part by the NSF.