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Correlated electron transfer and nonlinear optical effects in QD sensitized solar cells[1] YURI DAHNOVSKY, GRIGORY KOLESOV, Department of Physics and Astronomy, University of Wyoming — Nonlinear optical properties in pump-probe experimental setups and photoelectric current are theoretically and computationally studied for both quantum dot sensitized solar cells (QDSSCs) and isolated quantum dots (QDs). A nonlinear polarization vector and correlated current are found from a solution of Kadanoff-Baym (KB) equations, which are solved by a novel efficient computational method based on a two-time spectral expansion (2TSE) of nonequilibrium Green's functions. In addition we study electron transfer within the uncorrelated Markovian approach. It is shown that differential transmission and electric current strongly depend on the value and the character of chemical bonding between a quantum dot and semiconductor surface. The higher the tunneling amplitude and bond width, the larger the differential transmission. The relaxation time for electronic states in a QD is also an important parameter indicating to the competition of two electron dynamics mechanisms, electron tunneling and electron relaxation. The higher the electron relaxation rate, the lower the discrepancy in the differential transmissions for QDSSCs and isolated QDs. The discrepancy between the markovian and KB approaches can reach as much as one order of magnitude.

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