**Sub-Picosecond Manipulation of Hot Carriers in Quantum Dots by Spin-Exchange Auger Interactions**

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The ability to effectively manipulate nonequlibirum ‘hot’ carriers could enable novel schemes for highly efficient energy harvesting and interconversion. One approach to harness a kinetic energy of hot carriers is through exploiting Auger interactions. In particular, using inverse Auger recombination or impact ionization, one can generate additional electron-hole pairs, which could, in principle, enhance efficiencies of photoelectrical and photochemical transformations [1]. The Auger interactions driving carrier multiplication (CM) compete with carrier cooling via phonon emission. In bulk semiconductors, the energy-gain rate due to Auger energy transfer (*r*g) is lower than the intraband cooling rate (*r*c) by at least of factor of 3. As a result, the energy required to create a new electron-hole pair (eh) is more than 3 bandgaps (eh > 3*E*g) instead of just *E*g, as defined by energy conservation. Due to relaxation of momentum conservation and reduced dielectric screening, Auger interactions are enhanced in zero-dimensional quantum dots (QDs). This leads to the enhancement of the CM effect manifested most prominently in the reduction of its threshold [2, 3]. However, as the rate of intra-band cooling is also enhanced, the *r*g/*r*c ratio remains around 0.3 and eh is still near 3*E*g, as in bulk systems.

Recently, we have discovered that contrary to this established paradigm, the energy-gain rate due to Auger interactions can exceed the rate of ‘nonproductive’ phonon-assisted energy losses in strongly confined magnetically doped QDs [4]. Specifically, we have demonstrated that by exploiting spin-exchange Auger interactions between manganese ions and intrinsic QD excitonic states, we can boost the Auger energy-gain rate almost ten-fold so the energy gain/loss-rate ratio becomes greater than 7, versus ~0.3 in undoped materials. The fact that spin-exchange Auger energy transfer outcompetes intraband cooling is indicated by observations of highly efficient Auger processes involving unrelaxed, hot carriers. In particular, we observe very fast (~140 fs) excitation of a magnetic ion by spin-exchange transfer from a hot QD exciton. Furthermore, we resolve an extremely fast (150 fs time scale) upconversion-like process wherein a hot photoinjected electron is intercepted by the spin-exchange Auger process and instead of relaxing to the band-edge is promoted to a ‘vacuum’ state outside the dot. The discovered effects are of great potential utility in photoconversion and photochemistry, electron photo-emission, and detection of energetic particles [5].

[1] J.H. Werner, S. Kolodinski, H.J. Queisser, Novel optimization principles and efficiency limits for semiconductor solar cells, Phys Rev. Lett., 72 (1994) 3851-3854.

[2] R.D. Schaller, V.I. Klimov, High efficiency carrier multiplication in PbSe nanocrystals: Implications for solar-energy conversion, Phys. Rev. Lett., 92 (2004) 186601-186601-186604.

[3] C.M. Cirloganu, L.A. Padilha, Q. Lin, N.S. Makarov, K.A. Velizhanin, H. Luo, I. Robel, J.M. Pietryga, V.I. Klimov, Enhanced carrier multiplication in engineered quasi-type-II quantum dots, Nat. Comm., 5 (2014) 4148.

[4] R. Singh, W. Liu, J. Lim, I. Robel, V.I. Klimov, Hot-Electron Dynamics in Quantum Dots Manipulated by Spin-Exchange Auger Interactions Nat. Nanotech., 14 (2019) 1035-1041.

[5] More energy instead of heat by planting magnetic spins into a quantum dot, SciGlow, October 7 (2019)

<https://sciglow.com/more-energy-instead-of-heat-by-planting-magnetic-spins-into-a-quantum-dot/>