2C11　半導体量子ドット複合体におけるオージェ効果を活用した効率的電子移動

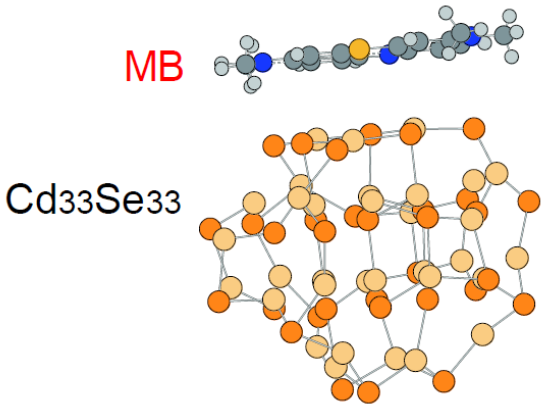
(京大院理[1]、ロチェスター大学[2])　金賢得[1]、Prezhdo Oleg V.[2]

Auger-assisted Electron Transfer from Photoexcited Semiconductor Quantum Dots

(Kyoto University[1], University of Rochester[2]) Kim Hyeon-Deuk[1]、Oleg V. Prezhdo[2]

We calculate a real-time electron transfer (ET) involving an Auger hole excitation. The hole excitation accompanies the ET from the quantum dot (QD) to the adsorbed methylene blue(MB) due to the Auger effect. Strong interaction between an electron and a hole stems from a quantum confinement effect of the carriers in the nanoscale QD.

The Auger-assisted ET mechanism is supported by direct time-domain *ab initio* simulation on a CdSe QD/MB complex. The simulation is performed using time-domain density functional theory combined with nonadiabatic molecular dynamics. The system comprises a $Cd_{33}Se_{33}$ QD in contact with the MB molecule. Photo-excitation promotes an electron from the QD HOMO to the QD LUMO, leaving a hole in the HOMO orbital. In traditional ET, nuclear vibrational motions accommodate the excess energy lost by the electron, as it moves from the donor to the acceptor species. In the current case, quantum confinement of the charge carriers in the QD enhances their Coulomb interaction, allowing effective electron-hole energy exchange of Auger-type, and opening up an alternative pathway for the energy flow. The energy lost by the electron is taken up by the hole, which is promoted from the HOMO to lower energy VB orbitals of the QD.



During ET, the electron energy decreases on a picosecond timescale, in agreement with the experimental data. At the initial stage the energy lost by the electron is gained exclusively by the hole, confirming the proposed Auger-assisted ET mechanism. The ET rate varies little along the donor-acceptor energy gap and reaches a plateau at the high energy gap. If the ET followed the Marcus theory, the rates would have changed by two orders for the high energy range. The *ab initio* results confirm that the Auger excitation of the hole eliminates the Marcus inverted region, suggesting efficient ET over wide energy range.

The maximum energy reached by the hole during the ET correlates with the energy

gap. The more energy the electron loses, the higher maximum energy the hole gains.

【参考文献】

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