Control of Emission Photon Statistics from a Single Quantum Dot Using Silver-Coated AFM Tip

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The emission photon statistics, i.e., single-photon and multiphoton emission from a single semiconductor quantum dot (QD) are one of the important emission behavior. Generally, the emission photon statistics of the QD are governed by the multiexciton dynamics based on the quantum confinement depending on the size, shape, and atomic composition of QDs themselves. We have demonstrated that the emission photon statistics from single QDs could be controlled by the interaction with the plasmonic nanostructures.[1-3] To elucidate the possibility of the control of the photon statistics using the plasmonic nanostructure, the best way is the direct observation of the emission behavior accompanying the interaction with the plasmonic nanostructure. In this work, the enhancement of multiphoton emission from a QD interacting with a plasmonic nanostructure was investigated using a silver-coated atomic force microscopy tip (AgTip) as the plasmonic nanostructure.[4] Using the AgTip, which exhibited a well-defined localized surface plasmon (LSP) resonance band, we controlled the spectral overlap and the distance between the single QD and the AgTip. The emission behavior of the single QD when approaching the AgTip at the nanometer scale was measured using off-resonance (405 nm) and resonance (465 nm) excitation of the LSP.

We directly observed the conversion of the single-photon emission from a single QD to multiphoton emission with reduction of the emission lifetime at both excitation wavelengths as the QD-AgTip distance decreased, whereas a decrease and increase in the emission intensity were observed at 405 nm and 465 nm excitation, respectively. By combining theoretical analysis and the numerical simulation of the AgTip, we deduced that the enhancement of the multiphoton emission was caused by the quenching of the single-exciton state due to the energy transfer from the QD to the AgTip and that the emission intensity was increased by enhancement of the excitation rate due to the electric field of the LSP on the AgTip. These results provide evidence that the photon statistics and the photon flux from the single QD can be manipulated by the plasmonic nanostructure through control of the spectral overlap and the distance.

References:

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