Display, Lighting, Bioimaging

Synthesis and functionalization of colloidal quantum dots

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Abstract

Semiconductor nanoparticles (quantum dots) are chemically synthesized colloidal fluorophores characteristic of monochromatic luminescence. Both particle size and composition affect the energy structure and emission color of quantum dots, attracting significant attention as new materials for future optical devices. Aiming at quantum dot devices, we continue the development of environmentally benign quantum dots and functionalization by designing the surrounding environment.

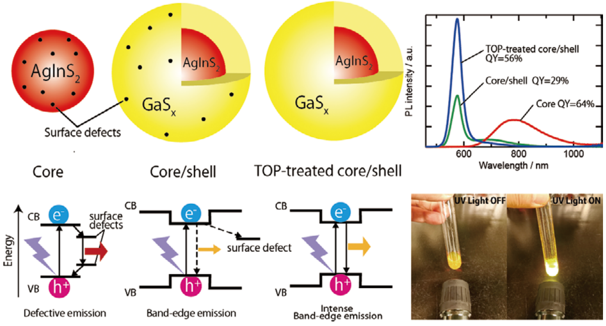
Background & Results

Embedding quantum dots in matrices is essential for demonstrating high-performance quantum dot devices by facilitating the injection of charge and energy. In particular, with the ongoing development of quantum dot LEDs, which are self-luminous devices similar to OLEDs, new ideas for utilizing quantum dots without compromising their properties are strongly required.

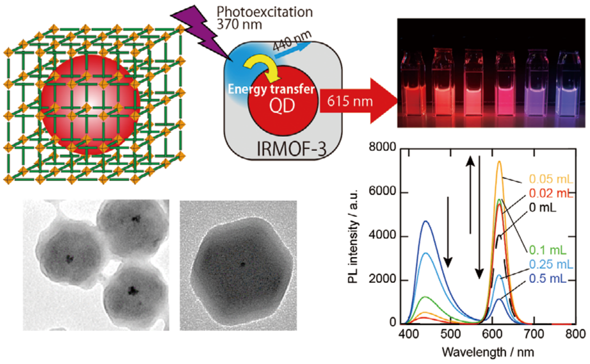
Significance of the research and Future perspective

The development of quantum dots originates from a weak fluorescence from nano-sized semiconductors over 30 years ago. Since then, the quantum dots have been improved to the point that they are used in commercial displays as wavelength conversion materials. Until a few years ago, the optically superior quantum dots were cadmium compounds, but their use in consumer products has encouraged the development of low-toxic alternatives. Our group developed photoluminescent $AgInS_2$ quantum dots and their analogs composed of three or more elements. However, the obtained photoluminescence was “defect emission” with a broad spectral width. Despite attempts by many researchers to narrow the spectral width, it had been impossible for several years, making researchers consider the band-edge emission hopeless. Under such situations, we focused on the surface condition and coated $AgInS_2$ quantum dots with gallium sulfide, an unconventional material as the shell. We eliminated surface defect levels and obtained a spectrally narrow band edge emission comparable to cadmium compound quantum dots in yellow region. We further demonstrated the green and red luminescence, which is more valuable as fluorescent materials, by alloying the core semiconductors to tune their bandgap.

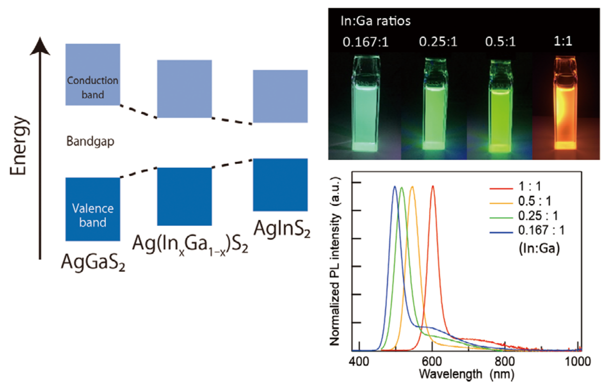
It is crucial to design matrices to manipulate the excitation energy of quantum dots. As a first step toward this goal, we used metal-organic frameworks (MOFs), which have a highly ordered structure and grow under mild conditions. Since the lattice size of MOFs is smaller than the diameter of quantum dots, we have attempted to create composite materials by growing MOFs from the surface of quantum dots. Using a typical MOF, ZIF-8, an ultimate structure that contains a single quantum dot at the center of a polyhedral MOF crystal was obtained. When a red-emitting quantum dot was incorporated in a blue-emitting IRMOF-3, the energy transfer from the MOF to the quantum dot occurred, doubling the photoluminescence intensity of the quantum dots.



Spectrally narrow band-edge photoluminescence from $AgInS_2$ quantum dots by the coating with GaS_x shell



Quantum dot–MOF complex fabricated by growing MOF from the surface of the quantum dots. Energy transfer was demonstrated by embedding red-emission quantum dot into blue-emission MOF.



Green-color emission from cadmium-free quantum dots by alloying $AgInS_2$ cores to increase their bandgap.

Patent	Japanese Patent Application No. 2017-037487, Japanese Unexamined Patent Publication No. 2018-044142, Japanese Patent No. 6464215
Treatise	Kumagai, Kohei; Uematsu, Taro; Kuwabata, Susumu et al. Photoluminescence Enhancement by Light Harvesting of Metal–Organic Frameworks Surrounding Semiconductor Quantum Dots, Chem. Mater., 2021, 33(5), 1607–1617. doi: 10.1021/acs.chemmater.0c03367 Watcharaporn Hoisang, Uematsu, Taro; Kuwabata, Susumu et al. Luminescent Quaternary $Ag(In_xGa_{1-x})S_2/GaS_y$ Core/Shell Quantum Dots Prepared Using Dithiocarbamate Compounds and Photoluminescence Recovery via Post Treatment, Inorg Chem, 2021, 60, 13101–13109. doi: 10.1021/acs.inorgchem.1c01513 Uematsu, Taro; Wajima, Kazutaka; Kuwabata, Susumu et al. Narrow Band-Edge Photoluminescence from $AgInS_2$ Semiconductor Nanoparticles by the Formation of Amorphous III-VI Semiconductor Shells, NPG Asia Materials, 2018, 10, 713–726. doi: 10.1038/s41427-018-0067-9
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Keyword	quantum dots, cadmium-free, fluorophores