The combination of multiple functional molecules is of great importance for the creation of hybrid nanomaterials with new desired functions. Among functional materials, colloidal semiconductor nanocrystals, so-called quantum dots (QDs), have attracted much attention because of their outstanding photoluminescence properties, such as a high photodurability, sharp PL spectra. Therefore, the emission and optoelectronic properties in the dispersed conditions have been actively studied. In addition, understanding the physical properties derived from inter-QD interactions observed in the solid condition of QDs is quite important for the development of QD-based materials science. However, the individual QDs are unsuitable for self-assembly in solution due to the high dispersibility in organic solvents. Therefore, the construction of assembled structures composed of multiple QDs is still challenging (Review: self-assembly of QDs, Ref 26, *Chem. Eur. J.*, **2020,** *26* ,7176).

In contrast, the self-assembly of organic π-conjugated molecules is a powerful tool to create various extended nanostructures, such as nanofibers and nanosheets (Ref 16, *Chem. Eur. J.*, **2017***, 23,* 5270). When this prominent self-assembly is applied to the QD system, indirect QD self-assembly can be realized. Recently, we proposed a new strategy to form supramolecular structures of QDs by using the self-assembly of low molecular weight organic molecules (Ref 20, *Chem. Eur. J.*, **2019***, 25,* 167). Furthermore, we succeeded in the construction of highly ordered QD arrangement structures and their photocontrol (Ref 31, *Angew. Chem. Int. Ed.,* **2021**, *in press*, attached paper).

1) QD arrangement based on self-assembly of PBI

First, we selected a perylene bisimide (PBI) dye with adhesion moieties to CdSe QDs as the self-assembling molecule, and conducted the mixing of QD and PBI in non-polar solvents. Upon mixing, QD and PBI coaggregate into a unique nanostructure composed of arranged QDs along a two-dimensional (2D) sheet of PBI, as confirmed by transmission electron microscopy (TEM) observation. To obtain insight into the mechanism of QD-PBI coaggregate formation, we studied the kinetics of time-dependent coaggregation using UV/vis absorption spectroscopy and TEM observation. From the in-depth analysis, the QD-PBI mixture just after mixing formed an amorphous coaggregate without QD arrangement through a strong face-to-face (H-type) π–π stacking between PBI. After one hour, the amorphous aggregates transformed into the well-defined coaggregates with QD arrangement via the rearrangement of stacking style of PBI.

2) Photocontrol over highly ordered QD supramolecular structures

The use of photochromic molecules such as azobenzene is an essential strategy for the creation of photoresponsive materials (Ref 8, *Nature Commun.,* **2015**, *6*: 8936; Ref 23, *Angew. Chem. Int. Ed.*, **2019**, *58*, 14173). In order to realize a photoresponsive QD system, we synthesized an azobenzene derivative (Azo) with an adhesion moiety to CdSe QDs and attempted the coaggregation of QD and Azo in non-polar solvents. As a result, we succeeded in the formation of highly ordered hybrid structures with QD arrangement, in which the energy transfer between QDs occurs effectively. Furthermore, upon photoirradiation, the highly ordered structure transferred into ill-defined coaggregates showing a lower efficiency of energy transfer, which is based on the photoisomerization of Azo. Over one day, the irradiated coaggregates underwent recovery of the PL properties corresponding to the arranged QDs through thermal isomerization.