Colloidal nanosheets are two-dimensional nanomaterials dispersed in a solvent. These nanosheets usually exhibit anomalous properties different from those of bulk materials, making them promising candidates as next-generation building blocks for creating functional materials. Therefore, various colloidal nanosheets consisting of designed components with excellent functions have been synthesized so far. However, the precise control over the self-assembly of colloidal nanosheets for realizing emergent functionalities remains a challenge. To overcome this challenge, we have established a rational strategy to precisely control the interactions, especially the electrostatic repulsion, between colloidal nanosheets. This strategy has enabled us to construct spatially and temporally well-controlled self-assembly of colloidal nanosheets and to realize various dynamic functions.

(1) Dynamic photonic crystal with designable structural color

By maximizing the electrostatic repulsion between colloidal nanosheets in water, the distance between the nanosheets expands up to 675 nm, resulting in a largely periodic nanostructure, called a photonic crystal, that can selectively reflect a vivid structural color (*Nature Commun.* 2016). The structural color can be tunable by using various external stimuli. Furthermore, when the dynamic photonic crystal is annealed under homogeneous and gradient temperature, it exhibits single- and double-peak structural colors, respectively (*Angew. Chem. Int. Ed.* 2023).

(2) Anisotropic hydrogel with large mechanical anisotropy

By maximizing the electrostatic repulsion between colloidal nanosheets in a hydrogel, the mechanical anisotropy of the hydrogel is largely enhanced (*Angew. Chem. Int. Ed.* 2018). The resultant anisotropic hydrogel exhibits an anisotropic parameter of 85, where the elastic modulus in the vertical direction to the nanosheet plane is 85 times larger than that in the parallel direction. Furthermore, when the nanosheet orientation is rotated to 45°, the hydrogel exhibits mechanical nonreciprocity (*Science* 2023).

(3) Mechanically adaptive hydrogel with rapid and reversible mechanical tunability

By reversibly and largely modulating the electrostatic repulsion between colloidal nanosheets in water by using a thermal stimulus, an aqueous dispersion of colloidal nanosheets exhibits a reversible transition between a repulsion-dominant gel and an attraction-dominant gel (*Nature Commun.* 2020).

(4) Propagating wave with capability of mass transport

By increasing the electrostatic repulsion between colloidal nanosheets in water and then gradually decreasing it by using a chemical stimulus, colloidal nanosheets work collectively to generate a propagating wave that can transport microparticles to one direction (*Nature Commun.* 2021).

(5) Dynamic soft material that exhibits a reversible sol–gel transition

By tuning the electrostatic repulsion between colloidal nanosheets in water by using a thermal stimulus, an aqueous dispersion of colloidal nanosheets exhibits a reversible sol–gel transition (*ACS Appl. Mater. Interfaces* 2023).