

Optical resonators are minute objects that can confine light inside their body by the total internal reflection and thereby transmit resonated or monochromatic light. The resonators have been intensively studied in the field of optics as an essential component for spectrometers and laser emitters. The resonators attract renewed attentions in the last decade in the fields of chemistry and biology, where they can be utilized as miniaturized optical sensors and tags that are capable of tracking chemical and biological events in the living organisms and ambient environments. However, the applicability of the resonators to these soft targets has been limited because most of the resonators are made from inorganic materials that are processed by top-down methods.

Organic compounds have been barely utilized in the field of resonator optics due to their less optical durability and performance, but they are highly promising in terms of the human- and environment-friendliness, stimuli-responsiveness, functional tunability, and the virtually infinite palette of their structure and function. We envisaged that optical resonators made from functional organic molecules will be a potential candidate that can overcome the current limit of the optical resonators. Through the research described below, we establish a self assembly-based methodology for making resonators from functional organic compounds and, additionally, found unique optical functionalities that stems from the chemistry of the constituent organic compounds.

1. Self-assembly of symmetric micro-aggregates from organic molecules and polymers

Optical resonators should be highly symmetric and smooth for the efficient light confinement. Photolithography is a powerful tool to curve the morphology but is typically inapplicable to functional organic compounds. We tried to achieve such sophisticated morphologies by the self-assembly. Particularly intriguing morphologies achieved so far are the largest metal–organic nanotube (*Science* **2014**, *J. Am. Chem. Soc.* **2015**), thermally robust yet self-healable porous crystal (*Science* **2018**, *ChemComm* **2020**), and concave micro-crystals (*Science* **2022**). Not only the morphologies but the basic underlying supramolecular chemistry behind the assembly is also our research target. We have elucidated general and fundamental issues on molecular self-assembly including (1) how discrete molecules spontaneously pack into a porous framework via van der Waals forces (*Commun. Chem.* **2021**, *ChemComm* **2022**), and (2) how concave crystals grow in a synchronous and uniaxial fashion (*Science* **2022**).

2. Optical resonance and lasing with self-assembled organic particles

In comparison with inorganic ones, organic compounds are advantageous for mechanical flexibility, chemical affinity, processability, and stimuli-responsiveness. Self-assembly methods written in the above section allows us to shape organic compounds with these functions into optical resonators. Representative achievements include energy-harvesting dendrimer lasers (*Angew. Chem. Int. Ed.* **2020**), an angularly anisotropic circularly polarized luminescence emitter (*J. Am. Chem. Soc.* **2021**), a mechanically flexible and optically tunable single-crystalline resonator (*Adv. Opt. Mater.* **2021**), optical logic gates

based on a photo-isomerization reaction (*Adv. Funct. Mater.* **2021**), a laser emitter from biological proteins (*Mater. Chem. Front.* **2021**), and nano-porous highly sensitive VOC sensor (*ACS Appl. Polym. Mater.* **2022**). The resonance and lasing performances reported in these works are highly influenced by the chemistry of the host organic molecules and, thereby, realize unprecedented stimuli-responsive optical functions.