第230回 OPERA研究交流セミナー
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Electron-responsive and Conductive Supramolecular Materials: Towards Chiroptical Switches for the Control of CPL Emission

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The incorporation of photo/redox-active units based on N,N'-disubstituted 4,4'-bispyridinium salts, commonly known as viologens, in the structure of supramolecular assemblies has been shown to confer interesting electron-accepting and photo/electrochromic properties to the materials. These responsive units have already been used as key elements for the development of electrochromic devices, molecular machines and organic batteries. Recently, there has been a renewed interest in the use of viologens as redox-responsive components in molecular junctions showing great promises for uses as conductive material in molecular electronics. Our research efforts in this field have led us to focus on molecular and supramolecular architectures involving viologens (4,4'-bipyridinium salts) as key electron-responsive building elements. In particular, we have developed different strategies allowing to exploit the ability of viologen derivatives to form π-dimers in their reduced state to achieve a remote control over their organization within supramolecular assemblies. Among our recent achievements, we have reported a sol/gel transition triggered by light-irradiation of a viologen-based coordination polymer formed in the presence of palladium ions. We now report the synthesis and detailed characterizations of supramolecular gels obtained by self-assembly of a dicationic low molecular weight gelator based on viologen. These molecules have been shown to self-assemble in pentanol to form chiral hollow core-shell cylinders eventually yielding dendritic clusters inducing gelation. We also showed that the optical, rheological and electrical properties of the gels can be tuned by addition of ionic additives. Careful control of the formation of charge-transfer complexes between viologens and iodides have led to a robust, transparent, conductive and chiral gel. Properties of this responsive material can be modulated when submitting it to electrical or light stimulation. Recently, we have also started to work on the development of switchable materials for circularly polarized luminescence (CPL). Preliminary results on new chiroptical fluorophores based on previously reported boron-based emitters will be described. We plan to use these new structures to develop bistable CPL-active materials for which stimulation changes the organization in supramolecular monolayers on surfaces. Analyses of such materials will be studied by a combination of electrochemical scanning tunnelling microscopy (EC-STM) at the liquid/solid interface and CPL confocal microscopy.

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