Circularticlely polarized luminescence of agglomerate emitters

Chong Zhang1 | Si Li1 | Xi-Yan Dong1,2 | Shuang-Quan Zang1

1 Green Catalysis Center, Henan Key Laboratory of Crystalline Molecular Functional Materials, Henan International Joint Laboratory of Tumor Theranostical Cluster Materials, and College of Chemistry, Zhengzhou University, Zhengzhou, China
2 College of Chemistry and Chemical Engineering, Henan Polytechnic University, Jiaozuo, China

Correspondence
Xi-Yan Dong and Shuang-Quan Zang, Green Catalysis Center, Henan Key Laboratory of Crystalline Molecular Functional Materials, Henan International Joint Laboratory of Tumor Theranostical Cluster Materials, and College of Chemistry, Zhengzhou University, Zhengzhou 450001, China. Email: zangsqzg@zzu.edu.cn; dongxiyan0720@hpu.edu.cn

Abstract
Circularticlely polarized luminescence (CPL) originates from the chiral emissive excited states. CPL materials have promising applications in 3D optical displays, encryptions, biological probes, chiral photoelectric devices, and CPL switches, most of which require excellent CPL performances including bright luminescence and high luminescence dissymmetry factor ($g_{\text{lum}}$) in the agglomerate state. This review systematically summarizes the progress about CPL of aggregate and solid materials, such as organic materials, metal-organic materials (such as coordination polymers, organic-inorganic metal halides, metal clusters, and cluster-assembled materials), the assembled materials by supramolecular interactions, and liquid crystals. We also present the current challenges and a future perspective of chiral emitting agglomerate materials.

KEYWORDS
agglomerate emitters, aggregate science, circularly polarized luminescence, CPL measurement method

INTRODUCTION
Light, including sunlight, moonlight, and lighting equipment, is constituted by all possible vibration directions perpendicular to the direction of the light propagation. And polarized light is divided into linearly polarized (LP), elliptically polarized, partially polarized, and circularly polarized (CP) light in terms of the difference of the light vibration direction vector perpendicular to the direction of light propagation.

Particularly, CP luminescence (CPL) has gained increasing interests recently due to its benefits for eye health and its potential applications, such as 3D displays, optoelectronic devices, nonlinear optics, spintronics devices, optical sensors, optical information storage and processing systems, CPL lasers, and liquid crystal (LC) display panel. Compared with the method of using a linear polarizer and a quarter-wave plate, the chiral materials can directly emit right- and left-CPL and thus avoid ca. 50% light loss.

Hence, scientists have been developing CPL-active materials with great enthusiasm. Considering the importance of CPL-active agglomerate materials in practice, we will focus on solid-state organic materials, supramolecular assembly materials, LCs, metal-organic materials (such as coordination polymers, hybrid organic-inorganic metal halides and perovskites, metal clusters, and cluster-based materials) (Figure 1). Luminous organic materials generally possess good optical performance in the solvent, while the performance in aggregation or solid state is limited. Particularly, with the development of organic science, many organic materials in agglomerate state could emit light. Especially, benefited from the discovery of aggregation-induced emission (AIE) phenomenon, AIEgens could be modified and regulated their self-assembly form in agglomerate aggregates to obtain the excellent optical properties. When AIEgens are bounded with chiral moieties, they may show good CPL signals in the aggregate state. In the aggregate state or solid state, chiral couplings between the transition dipole moments (electric dipole moment in most cases) operate, which could enhance the CPL property. Because of these advantages, multitudinous chiral AIE systems with CPL have emerged. Besides, axial chiral organics with CPL have also been reported. Crystalline metal organic materials, among which all atoms of metal organic crystalline materials are spatially long-range ordered, are also a class of important research material, and the associated CPL activities have emerged and increased sharply. Moreover, there are some reports on other crystalline metal organic materials with CPL characters such as coordination polymers, hybrid organic-inorganic metal halides, metal clusters, and cluster-based materials in the solid state, which still widens the choice of materials for CPL devices. Moreover, self-assembly is associated with...
the spontaneous formation of stable, well-defined structures driven by noncovalent interactions under certain conditions. Benefited from this, self-assembled supramolecular co-gels cannot only impart achiral luminophores with CPL, but also significantly enhance the luminescence dissymmetry factor ($\delta_{\text{lum}}$) of systems. Also, LCs, that is, the mesophases between the crystal phase and the fluid phase, have been commercialized in LC displays (LCDs). Therefore, it provides an opportunity for the application of CPL LCs, and the LCs system with CPL has made a progress recently. From the above, the study of CPL for the agglomerate materials is flourishing nowadays.

Currently, there are only several review articles related to CPL-active AIE materials. Zheng’s group reviewed the synthesis and application of chiral AIEgens. Similarly, Pasini’s group also summarized the emerging optical technologies based on AIE-based materials. Besides, Liu’s group also reported a review about CPL in nanomaterials. Zang’s group reviewed the progress of metal clusters possessing AIE and CPL. However, to date, there are no reviews that focus on agglomerate emitters with CPL and the CPL measurement methods that are important for the researchers in this field.

This review aims to provide (i) a basic introduction of chiroptical properties and detection method of solid-state CPL-active materials, (ii) a systemic summary of the progress of diverse agglomerate with CPL activities, (iii) amplification approach of CPL-active materials in the agglomerate state, and (iv) the outlook on the potential applications of agglomerate emitters with CPL.

**DEFINITION AND DETECTION METHODS OF CPL**

Generally, the CP light is produced by LP light passing through a quarter-wave plate, nevertheless, causing the ca. 50% light loss during the process of both traversing the plates and converting unpolarized light into LP light (Figure 2A). Also, CP light can enhance the image contrast and protect vision health. CPL refers to the emission of chromophores with a preferential handedness after they are excited by light, which is divided into propagation in a left- or right-handed spiral mode in space. While chiral non-racemic luminescent enantiomers enabling to directly generate CPL are more valuable in practical application. Notably, enantiomers have exactly identical physical and chemical properties. Thereat, a deeper understanding of a chiral substance cannot only require X-ray determination of its structure, but also study its optical activity by circular dichroism (CD) and CPL spectroscopy.

CD reflects the chiral ground state conformation through detecting the difference between left- and right-handed absorption. The measurement of CD is the common pathway for the absolute configuration of chiral substances when it comes to characterizing chiral substances. During the testing process, when the alternative left- and right-CP lights go through the chiral substances, they will exhibit different absorption capacities for left- and right-CP light (Figure 2B). And the generation of Cotton effect is accompanied by absorption spectra. Generally, the CD dissymmetric degree can be quantified to the absorptive asymmetric factor ($g_{\text{abs}}$), such as the Equation (1):

$$g_{\text{abs}} = \Delta \epsilon / \epsilon = 2 (\epsilon_L - \epsilon_R) / (\epsilon_L + \epsilon_R)$$

where $\epsilon_L$ and $\epsilon_R$ represent the molar absorption coefficients of left- and right-handed CP light, respectively. Enantiomers have opposite CD signals, and a negative CD signal refers to that right-handed CP light absorbed more than the left-handed CP light, and vice versa. And racemic configurations exhibit CD-silent.

CPL reflects the excited state information of chiral luminescent substances. When the chiral substances are excited by unpolarized light, the emissive intensities of generating left- and right-CPL are different, which is always accompanied by the emission spectra (Figure 2C). In spectroscopic methods, the difference of the emissive intensity ($\Delta \lambda$) is defined as the Equation (2):

$$\Delta \lambda = \lambda_L - \lambda_R$$

where $\lambda_L$ and $\lambda_R$ represent the intensity of left- and right-CP, respectively. Owing to the extreme difficulty of direct testing, the common parameter for evaluating CPL activities is defined as the Equation (3):

$$g_{\text{lum}} = 2 (\lambda_L - \lambda_R) / (\lambda_L + \lambda_R)$$

where $g_{\text{lum}}$ is referred to the luminescent dissymmetry factor. When the value of $g_{\text{lum}}$ is $\pm 2$, it represents a complete CPL signal. Nevertheless, when the $g_{\text{lum}}$ is 0, it means there is no CPL signal. Theoretically, $g_{\text{lum}}$ is derived from Equation (4):

$$g_{\text{lum}} = -4 |P_{ab}| |M_{ab}| \cos \theta_{ab} / (|P_{ab}|^2 + |M_{ab}|^2)$$

where $|P_{ab}|$ and $|M_{ab}|$ are the electric-dipole transition moment and the magnetic-dipole transition moment,
**FIGURE 2** (A) The schematic diagram of left- and right- CP light generation. (B) The schematic illustration of (i) CD and (ii) CPL tests respectively. Therefore, through rationally adjusting $|M_{ab}|$ and the $|P_{ab}|$ vectors of chiral molecules, materials with high $g_{\text{lum}}$ values will be promising. Mentioned above are the basic principles of detecting the chiroptical signals.

In short, high-performance CPL has always been the pursuit of scientist. In the process of realizing high-performance CPL, quantifying the overall efficiency of CP emitters plays a vital role in developing CPL materials. Arrico et al proposed the most useful metric for determining the quantity of CPL, that is the brightness for CPL ($B_{\text{CPL}}$) defined as Equation (5):

$$B_{\text{CPL}} = \varepsilon_{\text{abs}} \times \varphi \times \frac{|g_{\text{lum}}|}{2}$$

where $\varepsilon_{\text{abs}}$ and $\varphi$ represent the molecular extinction coefficient measured at the excitation wavelength, and the emission quantum yield (QY), respectively. The concept of the brightness for CPL ($B_{\text{CPL}}$) has guiding significance for the development of high-performance CPL materials.

Although the study of CPL is very active nowadays, to some extent, it is still in its infancy. Firstly, in past decades, CPL spectra in literature were almost recorded on the designed and laboratory-built spectrometer, whose construction requires basics of photophysics. For most chemistry researchers, this is a great challenge. Until the early 21st century, CPL spectrometer is commercialized, but it is high-cost. Secondly, although many CPL-active materials are prepared, the in-depth understanding of CPL properties is still lacking. Thirdly, for the applications of CPL materials, such as data storage, optical information processing, magnetic chiroptical fields are much less explored. Finally, the considerable difficulty in correctness of CPL signals also hinders the development of the CPL field. Generally, CPL measurement is accurate when the orientational distribution of the samples is isotropic. However, for practical device applications, how to measure CPL of materials in the condensed phase, especially the agglomerate state, may be more of a challenge due to the influence of artifacts caused by macroscopic anisotropies—the linear birefringence, linear dichroism, and LP luminescence. And since the $g_{\text{lum}}$ values for chiral materials are generally small and less than $\pm 0.01$ except for partial LC systems and rare earth complexes, the high sensitivity and accuracy of CPL spectrometers are the prerequisites for artifact-free spectra. Hereat, some improvements of solid-state CPL measurements...
have been made to remove artifacts resulted from macroscopic anisotropies. (i) The instrument (JASCO, CPL-300) was designed that the samples can be placed on a horizontal plane to greatly reduce the influence of artifacts, just utilizing unpolarized excitation light to measure.\(^\text{[50]}\) (ii) And equipped with a double-prism monochromator, it can also reduce the influence of stray light, and so on.\(^\text{[51]}\) (iii) Using a specially finetuned CPL spectrophotometer with correctly adjusted optical axes of the analyzer and a collimator lens with low intrinsic birefringence.\(^\text{[49]}\) (iv) In the process of measuring CPL spectra of agglomerate state materials, to eliminate the influence of macroscopic anisotropy caused by sample preparation to some extent, samples dispersed in KBr pellet, Nujol, and isotropic film are available since the finely dispersed sample may be considered of isotropic nature.\(^\text{[51–53]}\) Furthermore, the method that the ground agglomerate samples are sandwiched between two quartz slides is also reported.\(^\text{[54]}\)

Moreover, although the above-mentioned improvement of the instrument and the sample preparation methods can effectively reduce the effect of artifacts, CPL signals are also susceptible to the experimental deviations. The agglomerate samples could be divided into two types, that is, (a) highly ordered samples including single crystals and LCs, and (b) randomly oriented samples, such as dispersed in KBr pellet, Nujol, and isotropic film. For type (a), measuring CPL spectra with a dedicated CPL spectrometer based on the Stokes-Mueller matrix analysis is necessary.\(^\text{[49]}\) For type (b), to obtain the true CPL spectra, one should better assure that signals associated with anisotropy are excluded as much as possible by rotating the sample around the excitation light propagation axis and analyzing the collected CPL spectra from different angles.\(^\text{[51]}\) In all, multiple measurements of CPL spectra are necessary for an authentic signal. In addition, these techniques for CPL tests are also applicable for CD measurements of agglomerate samples.\(^\text{[52]}\) With the broader attention of CPL field, the test methods of CD and CPL of agglomerate materials will be further improved.

**ORGANIC MATERIALS**

Since organic materials have the advantages of high photoluminescence (PL) QY and easy derivation, they are excellent optical materials, which are widely applied in various systems all the time,\(^\text{[55–58]}\) and thus, they are easier to realize low-cost and good CPL. Combining chiral moieties with luminophores through covalent bonds is the most common and direct way to prepare organic CPL materials. As expected, chiral luminescent molecules with CPL activities have grown enormously over the years.\(^\text{[59–62]}\) However, aside from liquid crystalline conjugated polymers\(^\text{[63,64]}\) and some self-assemblies from single-source,\(^\text{[66]}\) most of them with small \(g_\text{ lum}\) values (10\(^{-5}\)–10\(^{-2}\)) were investigated in the solution.\(^\text{[65–71]}\) From solution to condensed phase to solid state, the performance of organic materials becomes worse even disappeared due to common aggregation-caused quenching (ACQ) effect. From the view of practical application, high PLQY, \(g_\text{ lum}\), and stability in agglomerate state are exceptionally necessary.\(^\text{[50,72,73]}\) Therefore, how to use universally available chiral organic dyes that commonly suffer from the ACQ effect to achieve bright solid-state CPL-active organic materials remains a great challenge.

An exactly opposite phenomenon to the ACQ effect, namely, AIE, was observed by Tang and coworkers,\(^\text{[74]}\) bringing the light for CPL of organic chromophores in the agglomerate state. They have made a substantial contribution to small molecule AIEgens with CPL properties, whose method was appropriate welding of chiral moieties and AIE-active luminophore (Figure 3A). In 2012, as a proof of concept, a chiral silole derivative bonded with mannosse units as side chains (TPS-ABM) was synthesized (Figures 3B and 3C).\(^\text{[58–59]}\) As expected, when TPS-ABM was completely dissolved in pure dichloromethane (DCM) solution, no fluorescence emission was observed because of the free intramolecular rotation. After this, with the addition of nonsolvent of TPS-ABM, the DCM solution could form a suspension, that is, molecular aggregation, simultaneously triggering the CD and fluorescence. Notably, the AIE effect enhanced the PLQY (0.6%, solution; 81.3%, solid state), which significantly inhibited the ACQ effect of conventional luminous molecules. Moreover, organic TPS-ABM preferred to emit right-handed CPL with extraordinarily high \(g_\text{ lum}\) values (-0.32). Subsequently, they successively reported a series of chiral silole and tetraphenylethylene (TPE) derivatives bearing chiral moieties such as L-valine, etc., which showed distinct CPL signals in the aggregate state.\(^\text{[58,75–77]}\)

In 2019, Cai’s group synthesized a series of novel chiral TPE-modified 1:1 α/sulfoono-γ-AAApeptides exhibiting CPL and typical AIE effect in the aggregate state (Figure 3D).\(^\text{[78]}\) In this report, the structure of I was successfully determined, showing that it adopted right-handed helical conformation. It was noted that I exhibited fluorescence even when completely dissolved in water due to the restriction of helical molecular scaffold. Notably, TPE moieties precisely anchored the right-handed helical framework, resulting in a large \(g_\text{ lum}\) of 1.2 × 10\(^{-2}\) in the aggregate state. Additionally, atomically precise chiral AIEgens with mid-molecular-weight (1000–3000 g mol\(^{-1}\)), especially showing CPL, were rare. Using the molecular design strategy, chiral AIEgens with CPL activities have been reported a lot.\(^\text{[50,67–73,74,79,80]}\)

Aside from the above aggregates, some CPL-active organic molecules have been reported in a KBr pellet-dispersed state or a crystalline state.\(^\text{[81]}\) Also in 2019, Kawai and Mietivier et al designed and prepared two chiral difluoro-boron β-diketone (DFB)-based compounds as well as further studied their mechanochromic fluorescence (Figure 3E).\(^\text{[81]}\) The DFB derivatives ((R/S)-DFB-Hex, (R/S)-DFB-PhEt) both exhibited CPL activities in the solution and the solid state. Later, the CPL properties were studied from three different forms of existence in the solid state. Notably, both compounds showed poor CPL signals in the as-deposited (AD) state, that is, mostly amorphous whereas the CPL intensity was much enhanced and \(g_\text{ lum}\) values reached an order of 10\(^{-2}\) after the thermal annealing (TA state), which was caused by chiral arrangement of chromophores. Most interestingly, (R/S)-DFB-Hex in the smeared state (SM) exhibited mechanically induced inversion of the CPL, whose reason might be a switching between monomer and excimer emission.

Except for the above-mentioned chiral organic molecules grafted by chiral functional groups, there is a kind of organic molecules of axial chirality, which also show CPL activities in the solid state.\(^\text{[82–84]}\) Through dispersing the samples in a PMMA film or a KBr pellet, Imai and co-authors reported an interesting phenomenon that CPL of axially chiral
binaphthyl fluorophores was reversed. Moreover, a series of unprecedented axially chiral peri-xanthenoxanthenes (PXXs) 2–5 were synthesized based on 1,1′-bi-2-naphthol (BINOL) without optical resolution by Ema and coworkers in 2019. Among the axially chiral PXXs, 5, adopting a herringbone structure and further avoiding $\pi\cdots\pi$ stacking mode, showed intense fluorescence (PLQY, 0.13) and CPL ($g_{\text{lim}} = \pm 4.8 \times 10^{-3}$) in the solid state (Figure 3F). Thereinto, the methylenedioxy-bridged conformation of PXX 5 regulated the angle between the vectors of the magnetic and electric transition dipole moments, which was essential for achieving good performance of CPL activity, evidenced by DFT calculations.

Organic materials with intrinsic room temperature phosphorescence (RTP) have significantly gained more interests because of the potential applications of organic optoelectronics. However, to date, CP organic RTP is rarely reported in the solid state. In 2019, Zheng’s group...
prepared four RTP materials through integration of carbazole unit directly onto the [2.2]paracyclophane unit, showing excellent CPL ($g_{\text{lum}}$, -1.2 × 10$^{-2}$) in toluene solution, which was a pity that CP organic RTP in the solid state was not studied\[87\]. Shortly after, Huang et al. presented a one-stone-two-birds strategy to integrate CPL into RTP/ultralong RTP (OURTP) molecules by direct bonding a flexible chiral ester chain into phosphor (carbazole unit) for (i) chirality transfer and (ii) aggregation coupling under photon irradiation of the resultant aggregates. Notably, the G-Py and (i) chirality transfer of the self-assembly has been widely reported in recent years.\[10,29,90–93,95,96\] The chiral matrix can co-assemble with achiral but also achiral chromophores with CPL activity (Figure 4A). The chiral matrix can co-assemble with achiral chromophores, subsequently inducing the achiral component to show color- or intensity-tunable CPL signals, which were H-aggregation and intra/intermolecular interactions of the chiral molecules. And notably, $g_{\text{lum}}$ values (±2.3 × 10$^{-3}$) of the OURTP and the pristine RTP emission were almost identical. Finally, they constructed a logic device based on photo/thermal-responsive reversibility of CP-OURTP/RTP.

Considerable progress has been made in low-molecular weight organic molecules with CPL activities. However, organic high-molecular-weight macromolecule has been less studied.\[159\] We will not go into details hereat, as some excellent reviews are already available.\[29,88,89\]

SUPRAMOLECULAR ASSEMBLY MATERIALS

Supramolecular assembly is a well-defined molecular complex, formed by noncovalent interactions, containing ion–dipolar interactions, hydrogen bonds, π–π stacking, and electrostatic interactions, etc. They possess the characteristics of flexibility, biocompatibility, and easy-processing.\[12,90–93\] Chiral supramolecular assemblies significantly have a chiral amplification effect on the circular polarization degree of some CPL-active materials.\[10,29\] Moreover, self-assembly strategies can endow not only chiral chromophores but also achiral chromophores with CPL activity (Figure 4A). The chiral matrix can co-assemble with achiral chromophores, subsequently inducing the achiral component to show color- or intensity-tunable CPL signals, which has been widely reported in recent years.\[10,29,90–93,95,96\] Notably, the reasons of chirality transfer of the self-assembly system might be the dipole-dipole interactions between transition moments of the achiral components and those of chiral hosts in terms of the Kirkwood-Tinoco equation.\[97\]

Back in 2011, Okano et al. demonstrated a novel CPL emissive system that was established by a stir-induced strategy, in which an achiral green luminescent dye (Rhodamine B) embedded into a stir-induced chiral hydrogel system and shows induced CPL.\[98\] However, extrinsically chiral hosts hinder the development of self-assembly materials with CPL in a way. To sweep this obstacle, Ihara and co-workers presented a novel self-assembly system based on electrostatic interactions, which was described for generating bright and color-tunable CPL emissions through chiral induction in nonchiral, organic, different fluorophores by chiral nanotemplate systems (Figure 4B).\[199\]

In this context, three amphiphilic molecules (L-glutamic acid derivatives, i.e., G-Py$^+$, G-IQ$^+$, G-NH$_3^+$) were selected as chiral templates. When the achiral anionic dye (NK-2012) was mixed with chiral templates, the electrostatic interaction promoted the formation of co-assemblies, in which co-assemblies of G-Py$^+$ and G-IQ$^+$ formed well-developed fibrillar aggregates whereas that of G-NH$_3^+$ showed globular aggregates. Notably, the G-Py$^+$/NK-2012 co-assemblies exhibited a higher PLQY (0.075) than that of NK-2012 molecule (0.0016) due to high rotational resistive microenvironment. Interestingly, as NK-2012 was achiral, the induced chirality of supramolecular assemblies was evidenced by temperature-dependent CD measurement. And the result indicated that the co-assemblies were constructed through $H$-stacked aggregates. Apart from the enhancement of the fluorescence quantum yield, remarkable CPL signals, a maximum $g_{\text{lum}}$ of ±0.102, of NK-2012 dye was brought about by the electrostatic interaction and enhanced chirality of the nanotemplate.\[100,101\]

In addition, many studies about self-assemblies are reported by Liu and Duan’s group.\[10,91–93,95–96,102–108\] For example, in 2017, based on previous literature, they utilized the host gelator 1,3,5-benzenetricarbonyl L-glutamate diethyl ester\[109\] to mix with different AIEgens, and then forming nanoassemblies, which showed enhancement in emission intensity and bright CPL ranging from 425 to 595 nm with $g_{\text{lum}}$ values of around 10$^{-3}$.\[102\]

Perovskites have attracted much attention because of their good photoelectric properties. However, CPL-active perovskites have not been reported until 2018.\[103\] Using a self-assembly method, Liu and coworkers successfully endowed perovskite nanocrystals (NCs) with CPL (Figure 4C). Firstly, CsPbX$_3$ (X = Cl, Br, I) perovskite NCs were obtained by a simple approach. And the colors of bright PL emission of CsPbX$_3$ changed by adjusting the halide ion. Considering that oleic acid and oleylamine were capped on the surface of perovskite NCs, amine-containing lipid was selected as chiral host to partly replace some of oleylamine on the surface of perovskite NCs and thus promote the construction of assemblies. Through such congelation, the perovskite NCs could inherit the chirality of host, resulting in color-tunable CPL ($g_{\text{lum}}$, ca. 10$^{-3}$). It is a pity that the CD spectra of the resultant perovskite NCs were not obtained. Furthermore, such co-assemblies could be processed into a film and further facilitate flexible CPL devices. In their other study, quantum dots (QDs) with color-tunable and white CPL were designed and prepared by a simple method that co-assembly of chiral gelators and achiral QDs.\[104\] In particular, materials with white CPL are in high demand for promising applications in lighting.\[110,111\] Also, Feng’s group also presented the full-color and white CPL nanoassemblies in 2020, which were used to fabricate CP organic light-emitting devices (CP-OLEDs) (Figure 4D).\[111\]

In recent years, a variety of cases based on supramolecular assembly method have emerged.\[65,67,112–118\] For instance, chiroptical interaction of achiral thioflavin T bound to chiral insulin amyloid fibrils has been evidenced by CD and CPL for the first time.\[119\] Feng’s group published a paper that using chiral phenylalanine-based gelator and achiral coumarin-based emitters could form supramolecular assemblies with unique CPL properties.\[90\] Furthermore, Longhi’s group reported a case that achiral polystyrene made optically
active (by electronic circular dichroism (ECD), vibrational circular dichroism (VCD), and CPL) through insertion of chiral molecules (carvone and other natural products).\textsuperscript{[120]} Also, mixing chiral binaphthyl sulfonates (BNS) and achiral bipyrene-based pyridinium (BPP), Cheng and Zhu et al synthesized a chiral supramolecular assemblies (BNS–BPP), exhibiting strong CD and CPL signals.\textsuperscript{[65]} And using TPE-derivative as a silicon (Si) precursor, Yang and Li et al also successfully prepared a pair of homochiral spiral TPE-Si nanotubes via the supramolecular templating approach.\textsuperscript{[118]}

In 2019, Tang et al also used special chiral hosts to endow achiral luminophores with CPL activities.\textsuperscript{[112]} In this work, enantiomeric pairs of Au compounds (S/R)-1 were prepared by hybridizing chirality with aurophilic interactions based on binaphthol skeleton, featuring an AIE property (Figure 5A). More interestingly, owing to aurophilic interactions, the obtained chiral Au complexes were found to undergo hierarchical self-assembly processes, and the difference of instant morphologies corresponded to the different stages of the assembly process, respectively. During the hierarchical evolution processes, real-time CD monitoring showed an impressive chiral inversion and amplification of chiral systems. Thus, the chiral Au(I) systems with well-defined architectures were also ideal chiral templates. Then, the achiral luminophores, such as 9,10-bis(phenylethynyl)anthracene (BPEA), TPE, and 2,3,5,6-tetrakis(4-methoxyphenyl)pyrazine (4PP-4 M) were selected to act as guest molecules. Through co-assembly, the resulting supramolecular assembly systems successfully showed bright CPL with $g_{\text{Lum}}$ values of around $10^{-3}$. Moreover, in 2016, Kumar et al reported a supramolecular polymerization system possessing extraordinarily strong $g_{\text{Lum}}$ value ($\pm 1.45$).\textsuperscript{[121]} They utilized intrinsically favorable properties of M$^+$[Eu($\pm$-hfbc)$_4]$\textsuperscript{3+} (M = Cs, Rb, K, Na; hfbc, heptfluorobutyl camphorate) to tune their chiroptical properties in polymers (Figure 5B). Thereinto, varying metals
(Na, K, Rb, Cs) led to the different lengths and extensions of twist in the aggregates, further affecting the value of $g_{\text{gum}}$.

During the co-assembly process, achiral guest luminescent materials can inherit chirality from the chiral host, resulting in CPL activities. If the chiral host is Luminous and its emission overlaps well with the absorption of the guest, the energy transfer may occur in co-assembly system. This is an interesting phenomenon for co-assemblies with CPL. To this end, in 2017, Liu and Duan’s group presented an innovative research that chirality and energy transfer amplified CPL by co-assembly process (Figure 5C).\[105\] The chiral AIE-based gelator could self-assemble into nanohelix with bright blue emission through noncovalent interactions, including the hydrogen bond and $\pi\cdots\pi$ stacking. Subsequently, when achiral acceptors (BPEA) were added into the self-assembly system, the co-assemblies would be formed instantly. Moreover, the co-assembly process resulted in that the BPEA inherited the chirality of chiral nanohelix, which was evidenced by CD measurement. Notably, the $g_{\text{gum}}$ of the CPL signals of the acceptor by exciting the nanohelix (donor) at 320 nm was $3 \times 10^{-3}$, which were larger than around 2.5 folds than by directly exciting the BPEA (acceptor) at 400 nm ($g_{\text{gum}} = 1.2 \times 10^{-3}$). Thus, energy transfer indeed amplified CPL. Apart from the above example, similar studies are also reported by their group. They constructed a chiral supramolecular co-assembly system, possessing cooperative effect and sequential energy transfer.\[106\] The cyanostilbene-appended glutamate compound (CG) could form a hydrogel, showing both CD and CPL. The blue-emissive thioflavin T (ThT) and yellow-emissive acridine orange (AO) were added into the self-assembly system to construct the co-assemblies, thus resulting in the acceptors could inherit the chirality of CG host. Especially, the CPL of two acceptors, ThT and AO, could be amplified in sequence due to continuous energy transfer from CG to ThT then to AO. In another latest work, according to the positive-/negative-charge type of acceptors, the single self-assembled system successfully integrated a selective chirality and energy transfer causing an enhanced CPL signal.\[116\]

Certainly, there are also other co-assembly systems, where chiral templates of achiral luminescent materials endowed with CPL activities are protein nanocage, DNA, etc.\[122,123\]

**LCs**

LCs are an important class of optoelectronic materials, typically composed of organic matters, and have some characteristics of both liquid and crystal, showing some unique properties, which are extensively applied in LCDs.\[124\] To fit for the practical application, many CPL-active LC systems have been explored.\[10,29\] One unique and key optical property of the chiral nematic LCs (N*LCs) is the selective reflection of CPL and thus combining excellent

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**FIGURE 5** (A) The evolution process of (S,R)-1. Reproduced with permission.\[112\] Copyright 2019, American Chemical Society. (B) The structure, CD, UV, and CPL spectra of $M^+\{\text{Eu(}+\text{-)}\text{-hfbc}\}_4\}^+$ ($M = \text{Cs, Rb, K, Na}$). Adapted with permission.\[121\] Copyright 2016, Royal Society of Chemistry. (C) Schematic diagram of energy transfer amplified CPL. CPL and $g_{\text{gum}}$ spectra of co-assemblies. Adapted with permission.\[105\] Copyright 2017, Springer Nature.
luminescent molecules with LC supramolecular organization is a good option for CPL-active LCs.\(^{[10]}\) Generally, preparation strategies of N*LCs with CPL are as outlined below (Figure 6): (i) adding the chiral dopants (chromophores) into achiral N*LCs, the resultant N*LCs system will generate enhanced CPL signals, which is one of the most effective methods.\(^{[125–129]}\) (ii) adding the achiral dopants (chromophores) into chiral N*LCs, the resultant N*LCs system can determine and adjust the emission intensity.\(^{[130–133]}\) (iii) another approach is to directly synthesize LC materials with CPL.\(^{[134,135]}\) Since some reviews and accounts are already available, and emphasis of this review is forced on the CPL of solid substances, we will not discuss about CPL of LCs here detailedly.\(^{[10,29]}\)

**METAL ORGANIC MATERIALS**

Developing crystalline metal-organic materials with CPL is a better method, but there are still great challenges. On the one hand, such crystalline metal-organic materials commonly have high PLQYs owing to decreased nonradiative decay, low optical loss, and enhanced photostability, etc.\(^{[136–140]}\) Besides, the structures of the crystals can be well solved by X-ray diffractometers, and then their structures-properties relationships can be studied deeply, which is instructive to the design of materials with excellent performance.\(^{[136–137,141]}\) But on the other hand, they are difficult to cultivate, especially chiral crystals. Currently, in aggregated state or crystalline state, three main kinds of materials could exhibit CPL, including metal clusters and cluster-based materials, coordination polymers as well as hybrid organic-inorganic metal halides.

**Coordination polymers**

In past decades, coordination polymers, especially star subclass that MOF with tunable pores, have made considerable progress due to extensive applications for gas storage/separation,\(^{[142–145]}\) catalysis,\(^{[146–148]}\) luminescence,\(^{[149–151]}\) proton conduction,\(^{[152]}\) and drug delivery.\(^{[153,154]}\) However, coordination polymers with CPL remain still quite scarce to date, although some synthetic strategies have been reported (Figure 7A). For example, in 2017, Yan and coworkers synthesized 3D Ln-BTC MOFs, crystallizing in a chiral space group \(P4_122\) (Tb-BTC, Eu@Tb-BTC), through a solvothermal process of Ln\(^{3+}\) and BTC in N,N-dimethylformamide (DMF) and H\(_2\)O solutions.\(^{[136]}\) Ln-MOFs with high PLQYs not only showed an optical waveguide effect in the propagation process, but also exhibited CPL properties (\(g_{\text{lum}}\)), \(-3.32 \times 10^{-3}, -2.56 \times 10^{-4}\), which correspond to the spectral range of the \(5D_4 \rightarrow 7F_5\) and \(5D_0 \rightarrow 7F_2\) transitions for Tb-BTC and Eu@Tb-BTC, respectively (Figure 7B).

In 2020, Liu and Zhu’s group prepared CPL-active materials based on \(\gamma\)-CD-MOF (\(\gamma\)-cyclodextrins-metal organic framework) with unique cubic chirality and size effect for the first time (Figure 7C).\(^{[155]}\) In this report, \(\gamma\)-CD-MOF was self-assembly formed by the six \(\gamma\)CDs, and its pore size is 1.7 nm, whereas the intrinsic void of \(\gamma\)CD is 0.95 nm.
FIGURE 7  (A) The construction strategies of chiral coordination polymers and MOF-based composites with CPL. (i) Introduction of the chiral linker. (ii) Chiral MOF induces guest molecules to generate CPL. (iii) Metal node and achiral linker assembled the chiral MOF with CPL. (iv) Chiral guest molecules induce MOF to generate CPL. (B) The structure and CPL spectra of Ln-BTC. Adapted with permission, [136] Copyright 2017, John Wiley and Sons. (C) The schematic diagram of lum@CD-MOF. And the impact of size effect on induced CPL. Adapted with permission, [155] Copyright 2020, John Wiley and Sons. (D) ORTEP views, PL, and CPL spectra of [Eu(+/−tfc)(dpbb)]n in the solid state. Adapted with permission, [156] Copyright 2018, Royal Society of Chemistry. (E) The structure, CD, and CPL spectra of 1D-coordination polymers (R/S-L-Cd). Adapted with permission, [141] Copyright 2019, Royal Society of Chemistry. (F) CPL spectra and enantioselective recognition of chiral ZIFs. Adapted with permission, [158] Copyright 2019, John Wiley and Sons. (G) Schematic diagram of synthesis of CPL-active MOFs. Adapted with permission, [159] Copyright 2020, American Association for the Advancement of Science.
The luminophore@γCD-MOF of host-guest interaction was fabricated by in situ encapsulation that γCD-MOF directly integrated achiral luminophore including different charges (neutral, positive, and negative), ACQ, and AIE organic matters, which varied in size from 0.5 to 2.1 nm. More interestingly, luminophore@γCD-MOF showed determined and strong negative CPL when the size of the guest luminophore was close to the pore size of γCD-MOF, that is, stronger host-guest interaction. While, sizes of guest luminophore that was smaller than a cubic void of γCD-MOF and larger than a void of γCD, it showed uncontrollable CPL signals, that is, sometimes positive or negative CPL. Furthermore, the small-sized guests preferred to be confined into the intrinsic void of γCD.

Nevertheless, compared with single-handed or uncontrollable CPL, one pair of mirrored and well-regulated CPL signals are of significance much more. In 2018, Hasegawa et al synthesized chiral spiral Eu(III) coordination polymers ([121,157] which indicated that the chiroptical activity could be amplified through forming helical structure by polymerization of mononuclear complexes in the solid state. In 2019, Cao’s group cleverly utilized chiral ligands and cadmium(II) nitrate to achieve novel enantiomeric 1D compounds (R/S-L-Cd), showing second harmonic generation activity, two-photon excited luminescence as well as good CD and CPL signals ($g_{\text{lum}} = 0.17$) in comparison with those of mononuclear counterpart (0.36). They showed good CD and CPL properties ($g_{\text{lum}} = 0.17$) in comparison with those of mononuclear counterpart in the solid state,[121,157] indicating that the chiroptical signal could be amplified through forming helical structure by polymerization of mononuclear complexes in the solid state. Shortly after that, Duan’s team presented a design strategy for first chiral zeolitic MOF (R/S-ZIF) nanomaterials exhibiting enhanced and inverted CPL, which fabricated by the ligand-exchange approach that the ingenious design of a pair of chiral functional imidazole-based emitters (R/S-1) replaced achiral ligand, i.e., 2-methylimidazole.[158] Interestingly, compared with the chiroptical properties of synthesized R/S-1, R/S-ZIF nanomaterials not only exhibited an elevated PLQY (0.3 to 0.4), but also significantly amplified the $g_{\text{lum}}$ ($10^{-2}$ to $10^{-3}$), which were caused by the restriction of chiral emitters in Zn–ligand complex formation and well-ordered arrangement of R/S on the framework of ZIF-8 after ligand-exchange, respectively (Figure 7F). Moreover, the chiral ZIF nanoparticles could be used to enantioselective recognition through the difference of emission intensities of chiral ZIFs treated with α-hydroxycarboxylic acids. Most importantly, the authors solved a persistent problem that the large $g_{\text{lum}}$ always accompanied with small photoluminescent quantum yield. Subsequently, based on previous work, using chiral ZIFs, constructed by simultaneously mixing 2-methylimidazole and L-/D-histidine, as host for encapsulating dyes, QDs, and upconversion nanoparticles, respectively, Liu and Duan’s group reported color-tunable, white CPL-active, and upconverted CPL-active ZIFs (Figure 7G).[159]

As we know before, although MOFs as chiral hosts have successfully induced achiral luminophores to generate CPL, the helical channels of chiral MOFs have not been used for endowing guest with CPL, which could be deemed an efficient strategy to amplify $g_{\text{lum}}$. Herein, Zang’s group prepared enantiomeric pairs L/D-CMOF ⊃ fluorophore composites (Figure 8A).[110] Achiral organic fluorophores were confined in nanometer-sized helical channels, which efficiently overcame the detrimental ACQ effect, and at the same time, they adhered to the inner surface of the helical channels via non-covalent interactions, successfully resulting in some helical arrangements. As a result, crystalline L/D-CMOF ⊃ fluorophores displayed color-tunable and white CPL with a PLQY (30%). Also, a white CP light-emitting diode was fabricated by L/D-CMOF ⊃ fluorophores. More importantly, the $g_{\text{lum}}$ values of a series of L/D-CMOF ⊃ fluorophores composites are consistent with the change trend of their interaction energies, resulting in an enhanced $g_{\text{lum}}$ ($±0.0115$), which was observed for the first time in host-guest systems, revealing the origin of confinement in the nanometer-sized helical channels. This strategy paves a new way for realizing universal chiroptical materials and enhancing $g_{\text{lum}}$ value through helical channels of chiral MOFs. Meanwhile, a host (MOF)-guest (organic molecule) system was designed by Zeng et al., in which the achiral organic molecule ((4-p-((dimethylamino)styryl)-1-methylpyridinium iodide [DSM]) was encapsulated in the chiral channel of enantiomeric Ln-MOFs that are P-((+)-/M-((−)-)TbBTC (BTC: 1,3,5-Benzeniccarboxylic acid) showed a good CPL ($g_{\text{lum}} = 10^{-3}$) and a high PLQY (10%), and enabled efficient energy transfer from P-((+)-/M-((−)-)TbBTC to DSM along with the emission color change (Figure 8B).[160] And the CPL of the microarea of the single crystal was further studied.

Recently, Liu et al further extended their work on the construction of CPL-active MOF with precise structure.[161] In 2020, they prepared a pair of homochiral AIEgen MOFs with excellent CPL performance ($g_{\text{lum}} \approx ±10^{-2}$) (Figure 8C). The CPL activities of AIEgen MOFs originated from luminescence and chirality of tetrakis(4-pyridylphenyl)ethylene (TPyPE). The reason of such bright luminescence was that the intramolecular vibration and rotation of TPyPE were weakened by the restraints of the enantiomeric MOF skeleton. Moreover, the achiral TPyPE inherited the chirality of chiral donors (camphoric acid) through-space chirality transfer. Interestingly, the enantiomeric MOFs possessed the ultrasonication/grinding-switchable CD, FL, and CPL properties, which were caused by subtle change of microenvironment (rotation or vibration) of AIEgen rotors in the homochiral MOF skeleton under external mechano-stimuli. This laid the foundation for the development of CPL-active smart materials.

From the foregoing, CPL of guest molecules induced by the chiral MOFs has been reported, while the CPL of MOFs induced by the chiral guest molecules is rare. In 2019, Zang et al synthesized mesoporous crystalline silver-chalcogenolate cluster-assembled materials through a system of achiral luminous host (MOF)-chiral guest molecules (L/D-menthyl), namely, 1 ⊃ L/D-MT, showing an excellent mirror image Cotton effect ($g_{\text{lum}} = ±1.1 \times 10^{-3}$) and CPL emission ($g_{\text{lum}} = ±1.5 \times 10^{-3}$) (Figure 8D).[162]
Figure 8  (A) Schematic synthesis and molecular simulation of L/D-MOF ⊃ fluorophores. Photographs of the fabricated WLED. Adapted with permission,[110] Copyright 2020, John Wiley and Sons. (B) An achiral stilbazolium dye (DSM) achieved induced molecular chirality by the host-guest method. Microarea CPL of the single crystal measurement. Adapted with permission,[160] Copyright 2020, Royal Society of Chemistry. (C) Schematic diagram of the chiral reticular self-assembly strategy. CPL and $\g_{\text{sum}}$ spectra of D-MOF under different mechno-stimuli. Adapted with permission,[161] Copyright 2020, John Wiley and Sons. (D) The chemical structure of chiral guest molecules (L/D-menthol). A schematic diagram of the tailoring luminescence in AIE ligand-based mesoporous MOF by encapsulation of chiral guest molecules. CPL spectra of 1 ⊃ L/D-MT. Adapted with permission,[162] Copyright 2019, Chinese Chemical Society.
Some good work is being reported successively. Ma’s group achieved the CF phosphorescence in MOF at room temperature.\[163\] Li and co-workers prepared a series of CPL-active MOFs through subtle changes of ligands.\[164\]

Hybrid organic-inorganic metal halides

In recent years, hybrid organic-inorganic metal halides with the excellent luminescent properties, representing a star class of molecular crystalline material, are extensively applied in light-emitting diodes, luminescent solar concentrators, photovoltaic modules, and lasers, etc. due to intrinsic mechanical flexibility, structural tunability, and easy processability.\[137,165,166\] Why hybrid organic-inorganic metal halides possess luminescent properties of interest? The reasons, bright PL, narrow light emission, balanced charge carrier mobility, and tunable excitation binding energy, are presented here.\[167,168\] However, with the exception of carrier mobility, and tunable excitation binding energy, metal halides possess luminescent properties of interest? The (PEA)₂PbI₄ (PEA = \(\text{C}_8\text{H}_9\text{NH}_3\)) were successfully obtained, that is, originated their chiral space groups \(\text{Cc}⁻\text{3}⁻\text{m}\). Also the chirality of chiral arylamines (R/S-MBA) was verified by vibrational CD. (R,S)-MBA-based perovskites showed crystalline powder nonlinear optical (NLO)-inactivity, whereas (R)- and (S)-MBAPbX₃ demonstrated crystalline powder NLO-activity. Notably, (R)- and (S)-MBAPbBr₃ showed excellent CPL signals with a short PL lifetime (Figure 9D). The single crystal has good CPL properties, which lays a foundation for the design of miniature chiral optoelectronic devices.

As a further development, utilizing the above chiral ligand (R/S-MBA), Dang et al synthesized enantiomeric pairs of homochiral (R)- and (S)-MBAPbX₃ (X = Br, I) single crystals by a simple approach, that is, regulating the ratio of reactants and cooling rate.\[180\] As expected, (R)- and (S)-MBAPbX₃ (X = Br, I) single crystals exhibited obvious bathochromic-shift and mirror image CD responses in comparison with those of their crystalline powder, whereas there were no CD responses in the racemic (R, S)-MBA-based perovskites. In addition, the racemic (R, S)-MBA-based perovskites showed crystalline powder nonlinear optical (NLO)-inactivity, whereas (R)- and (S)-MBAPbX₃ demonstrated crystalline powder NLO-activity. Notably, (R)- and (S)-MBAPbBr₃ showed excellent CPL signals with a short PL lifetime (Figure 9D). The single crystal has good CPL properties, which lays a foundation for the design of miniature chiral optoelectronic devices.

After that, an unprecedented example that organic-inorganic hybrid metal halides, (KC)₂MnX₄ (X = Cl, Br) with achiral space group (Cc) showed CPL activities, was reported by Zang’s group (Figure 9E).\[137\] The (KC)₂MnCl₄ single crystals possess 1D morphology, high crystallinity, and bright emission with high PLQYs (7.79%, 38.5%), which showed excellent optical waveguide phenomenon. Notably, it is worth noting that although the point group \(m\) (\(C\)) is achiral, single crystals crystallized in this point group could theoretically produce chiroptical activity in terms of previous literatures.\[140,181\] For this reason, the author has carried on the verification. More excitingly, (KC)₂MnBr₄ metal halides crystallized in the space group of \(Cc\) that was a subset of the achiral point group \(m(C)\), which provided theoretical support for studying CPL. In the structure of (KC)₂MnX₄, the luminous units, that is, racemic tetrahedral [MnX₄]²⁻ units, were arranged along with right- and left-handed rotating optical axes, hence probably enabled achiral crystals with remarkable CPL activities. Subsequently, they performed CPL testing on (KC)₂MnCl₄ in powder samples and the single crystal to verify the theory.

Benefited from the developments of luminous ferroelectric materials, the field of ferroelectric emission display has made great progress.\[182–185\] Particularly, structurally designable molecular ferroelectrics are more attractive and are conductive to synthesis of CPL-active ferroelectric materials. In 2020, Fu’s group cleverly designed and synthesized the enantiomeric organic-inorganic perovskite ferroelectrics, \(R/S-3\)-(fluoropyrrolidinium)MnBr₃, showing CPL activity for the first time. (Figure 9F).\[186\] The structures of enantiomeric organic-inorganic perovskites were determined by single crystal X-ray diffraction, and their chiral space groups \(P2\) and Flack values elucidate the enantiomorphous nature. Also the chirality of \(R/S-3\)-(fluoropyrrolidinium)MnBr₃ was verified by vibrational CD. Notably, \(R/S-3\)-(fluoropyrrolidinium)MnBr₃ not only had ferroelectric properties, but also possessed prominent optical properties such as strong PL and corresponding mirror image CPL \((\g\text{Lum.} \pm 6.1 \times 10^{-7})\). Most importantly, the introduction of chiral organic cations resulted in chiral arrangements of [MnBr₃]⁻⁻⁻⁻ chain, hence leading to the emergence of CPL activity in enantiomeric organic-inorganic perovskite ferroelectrics. The combination of ferroelectricity and CPL enriches the populations of CPL.
FIGURE 9  (A) Synthetic route of homochiral hybrid organic-inorganic metal halides with CPL. i. The hybrid organic-inorganic metal halides, including right (red)/left (blue)-handed of achiral cations by the slide and mirror symmetry in the m point group, could emit CPL. ii. The introduction of chiral cations causes hybrid organic-inorganic metal halides to produce CPL. (B) The crystal structures and CPL spectra of (R- and S-MBA)2PbI4. CD spectra of (R-, S-, and rac-MBA)2PbI4 films. Adapted with permission. (C) The crystal structures and CPL spectra of (R/S)-α-(PEA)2PbI4 and C6H6I4PbI4. Adapted with permission. Copyright 2019, American Chemical Society. (D) CD and CPL of (R)-/S)-MBAPbBr3 single crystals. Adapted with permission. Copyright 2020, American Chemical Society. (E) The crystal structure and analysis of (KC)2MnCl4 compound. The microarea CPL of the (KC)2MnCl4 single crystal measurement. Adapted with permission. Copyright 2019, American Chemical Society. (F) Packing views of (S)- and (R)-3-(fluoropyrrolidinium)MnBr3. Insert curves represent the corresponding ferroelectricity. CPL emission spectra vs wavelength curves. Adapted with permission. Copyright 2020, American Chemical Society. (G) The generation and measurement method of one-photon or two-photon excited PL in perovskites. And one-photon or two-photon excited CPL spectra of perovskite nanocrystals. Adapted with permission. Copyright 2019, American Chemical Society.
Except for atomically precise perovskite materials with CPL, in another work, using the chiral capping ligands, namely, R/S-α-oxalocarbamoylamine to modify the surface of two-photon absorption active cesium lead bromides (CsPbBr3) with high PLQYs, the first case of chiral perovskite NCs (R/S-Pero-NCs) possessing two-photon absorption upconverted CPL was prepared by Liu and Duan’s group.[187] After the modification, enantiomer R/S-Pero-NCs exhibited mirror-image Cotton effect around 504 nm, which indicated that CsPbBr3 core inherited the chirality of (R/S-α-oxalocarbamoylamine) through the interaction between chiral capping ligands and the CsPbBr3 NC’s surface.[188] That is, in the R/S-Pero-NCs, the modified R/S-α-oxalocarbamoylamine could disturb the surface pattern of NCs effectively, as illustrated in Figure 9G, which could lead to the chiral transfer from the shell to the core. Moreover, under the excitation of both 375 nm CW-laser (PLQY, 60%–70%) and 800 nm laser, chiral perovskite NCs emitted bright PL emissions with general $\delta_{\text{lim}}$ Values (6.5 × 10^{-4} and -1.0 × 10^{-3}; 3.5 × 10^{-3} and -2.3 × 10^{-3}).

**Metal clusters and cluster-based materials**

Atomically precise metal nanoclusters have gained extensive interests due to intriguing their graceful structures and fascinating chemical/physical properties.[189–202] Meanwhile, one of the most charming and significant properties of metal clusters is luminescence, whereas it remains a challenge that obtaining metal clusters with bright luminescence at room temperature due to the instability and their sensitivity to the external environment.[191,203,204] Thus, aiming to the difficulty, some strategies that thermally assisted delayed fluorescence (TADF), AIE as well as crystallization-emission (CIE) have been proposed and validated.[205–207] Aside from the bright PL, chirality is another prerequisite for constructing metal clusters with CPL activities. Recently, introducing chiral ligands or resolution, CPL-active metal clusters, especially in their crystalline state, have been reported successively.[209–211] In 2017, Tang et al showed enantiomeric pairs of homochiral Au clusters (R/S-Au3) with CPL-activities by introducing chiral ligands.[209] The structure determined from single crystal X-ray diffraction contained three gold atoms arranged in a equilateral triangle. Furthermore, viewed from a perspective perpendicular to Au3 plane, the peripheral ligands that coordinated with the Au atoms were arranged in a left or right helical mode (Figure 10C). Enantiomeric Au clusters dispersed in dichloromethane showed mirror image CD signals ($\delta_{\text{lim}}$, 7 × 10^{-3}) and didn’t emit luminescence at all. After adding n-hexane (70%) into the above system, the enantiomeric Au clusters were easily self-assembled to nanocubes that exhibited a cubic morphology. Strikingly, these cluster assemblies showed bright PL (PLQY, 3.6%) and CPL responses ($\delta_{\text{lim}}$, ±7 × 10^{-3}) in n-hexane (70%). The results reveal that strong intermolecular C-H⋯π interactions and ordered structure enable the emergence of CPL-active cluster assemblies.

In 2020, Li et al elaborately presented a two-step approach to prepare silver clusters with CPL. In the first step, they used chiral amino acids to replace part of NO3⁻ of the surface of (CO3)2Ag24(SBu10(NO3)8(DMF)6 cluster and then obtained homochiral (CO3)2Ag24(SBu10(L/D-Proline))3(NO3)2(NO3)2 (namely, L/D clusters) that are CD-active and CPL-inactive. Subsequently, the residue NO3⁻ of L/D clusters was further thoroughly substituted by achiral luminophors, that is, 4,4′-bis(2-sulfonato styryl)biphenyl disodium salt (Na2CF351) and N, N′-di(ethan sulfonic acid)-3,4,9,10-perylene tetracarboxylic diimide (H2TauPDI), which formed the Ag24 clusters (CO3)2Ag24(SBu10(L/D-Proline))3(CF351)2 (abbreviated as L/D-CF351) and (CO3)2Ag24(SBu10(L/D-Proline))3(TauPDI)2 (abbreviated as L/D-TauPDI), respectively, exhibiting mirror image CD responses and good CPL properties in the solid state. Their corresponding $\delta_{\text{lim}}$ approached to ±5 × 10^{-3} and ±4 × 10^{-3}, respectively (Figure 10D).[212,213] This method provides a new sight for the synthesis of multifunctional clusters. Subsequently, this laboratory constructed two enantiomeric pairs of Au clusters with atomic precision through a facile way, Au4(PL/PD)4 and {Au4(L4/D4)}n, which were successfully assembled into a CP-OLED for the first time.[214] Bulky substituents in the shell layer in Au4(PL/Au4PD4 segregated the quaternary Au4 AIEgens, resulting in the inherent high-energy cyan CPL with PLQY = 14.4% in the aggregated state, while a thinner shell caused the Au4 AIEgens to be linked together by intercluster Au–Au interactions, giving rise to a green CPL that had a much higher PLQY of 41.4%.
FIGURE 10  (A) Synthetic route of homochiral metal clusters with CPL. (B) The structure, CD, and CPL spectra of enantiomeric Ag₆L₆/D₆ and Ag₆PL₆/PD₆; Theoretical calculation, optical absorption, and electron transition of Ag₆L₆. Adapted with permission,[208] Copyright 2020, The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. Distributed under a Creative Commons Attribution No Commercial License 4.0 (CC BY-NC) http://creativecommons.org/licenses/by-nc/4.0/. (C) The overall structures of R/S-Au₃. SEM (upper) and TEM (down) images of R/S-Au₃ assemblies. Emission spectra (upper) of R/S-Au₃ in dichloromethane with different n-hexane contents. CPL spectra (down) of R/S-Au₃ assemblies in 70% n-hexane. Adapted with permission.[209] Copyright 2017, John Wiley and Sons. (D) Stepwise achievement of CPL of L/D-CF351 and L/D-S-Au₃. Cluster. Adapted with permission.[213] Copyright 2020, John Wiley and Sons.
and $g_{\text{av}} = \pm 7 \times 10^{-3}$ (Figure 11A). For the first time, the AIE$_{\text{g}}$-based AIEgens were fabricated into CP-OLEDs with nearly equal to $\pm g_{\text{av}}$.

Very recently, one racemate of silver clusters, composed of chiral Ag$_{10}$[Cu$_5$(C$_2$B$_{10}$H$_{12}$S$_4$)$_4$Dppm$_6$ (abbreviated as Ag$_{10}$-rac; 8,9,12-trimercapto-1,2-closo-carborene, C$_2$B$_{10}$H$_{12}$S$_4$; bis(diphenylphosphino)methane, Dppm), was prepared and characterized by their group.$^{[215]}$ More interestingly, spontaneous resolution driven conglomerate crystallization of the racemate could be realized by a facile approach of only dissolving in dimethylacetamide (DMAc), resulting in racemic conglomerates (R/L-Ag$_{10}$), where each single crystal was homochiral (Figure 11B). Moreover, due to their red-emitting, CPL of single crystal was carried out ($g_{\text{av}} = \pm 7 \times 10^{-3}$).

In addition to the above noble metal nanostructures, the mushroom growth of Cu(I) clusters probably benefited from its high PLQYs and polynuclear configurations, which are worth grafting CPL properties.$^{[216]}$ In 2019, Zang and coworkers obtained a new chiral, luminescent, anionic, thiolated copper cluster $[\text{Cu}_{12}(\text{S}^\prime\text{Bu})_6]^-$ in the crystalline state.$^{[217]}$ When achiral [K-(CH$_3$OH)$_2$(18-crown-6)] was used as a cation, a racemic Cu$_5$:CH$_3$:OH was obtained (Figure 11C). Interestingly, thermally induced CH$_3$:OH release led to structure transformation from the racemic Cu$_5$:CH$_3$:OH to mesomeric Cu$_5^-$, in which a stacking rearrangement of $[\text{Cu}_{12}(\text{S}^\prime\text{Bu})_6]^-$ was induced. So far, both 1-CH$_3$:OH and 1 are CPL-silent. When cationic D/L-valinol was introduced into D/L-Cu$_{14}$V$_{\text{cyt}}$, the symmetry of structure was broken by hydrogen bond, and hence the racemates Cu$_5$:CH$_3$:OH clusters were eventually converted to optically pure homochiral clusters, which evidenced by single-crystal structures and mirror CD signals. Excitingly, the enantiomeric pair D/L-Cu$_{14}$V$_{\text{cyt}}$ in the solid state exhibited bright red CPL with a reasonably high $g_{\text{av}} = (\pm 9.77 \times 10^{-3})$ value centered at 660 nm. In all, introducing chirality of counter ions is a simple way to construct CPL-active metal clusters with precise structure.

Not long after, Zang’s group cleverly designed and synthesized enantiomeric Cu(I) clusters (R/S-Cu$_{14}$) triggered by AIE and CIE using purely alkynyl ligands.$^{[218]}$ In R/S-Cu$_{14}$ clusters, a chiral metal core Cu$_{14}$ originated from chiral alkynyl ligands. R/S-Cu$_{14}$ could be well dispersed in dichloromethane (CH$_2$:Cl$_2$), resulting in symmetric CD signals and no CPL activities (Figure 11D). Notably, the CPL intensities started from nothing and subsequently gradually increased with the addition of nonpolar n-hexane, whose reason was AIE effect, that is, PL intensity increased as the enhancement of aggregation degrees. From the above experimental results, the AIE effect triggered the CPL responses ($g_{\text{av}} = (\pm 1.0 \times 10^{-2})$) of resultant aggregated states of R/S-Cu$_{14}$ clusters. It was worth noting that R/S-Cu$_{14}$ clusters also displayed red emissive CPL responses ($g_{\text{av}} = (\pm 3.0 \times 10^{-3})$) in the crystalline state at room temperature. Subsequently, their group synthesized enantiomeric pairs of trinuclear Cu(I) clusters with AIE character, which showed CPL signals ($g_{\text{av}} = (\pm 2 \times 10^{-3})$) around 610 nm in the aggregate state (Figure 11E)$^{[219]}$. While, in glycerol with high viscosity, R/S-Cu$_{3}$ showed yellow-emissive CPL due to further restrictions of intramolecular rotations.

Several days later, enantiomeric tetranuclear copper(II) clusters, (R/S-MBA)$_4$Cu$_4$I$_4$, with intense orange CPL (PLQY, $\approx 60\%$), were easily designed and obtained by Tang’s group.$^{[214]}$ Enantiomeric (R/S-MBA)$_4$Cu$_4$I$_4$ showed mirror image CD signals and CPL signals ($g_{\text{av}} = (\pm 1 \times 10^{-2})$) in the crystalline state (Figure 11F). In addition, further extended work was presented by Zhu et al, they assembled the thiolated [Au$_1$Ag$_{22}$-(S-Adm)$_1$]$_2$$^{[2+]}$ (SC, where S-Adm = 1-adamantaneethiol) superatom cluster into a framework material through inorganic SbF$_6$− linkers and thus constructed SCIF-1 and SCIF-2 (Figure 11G)$^{[218]}$. Interestingly, the obtained SCs precursor was chiral. And utilizing the chiral SCs precursor, SCIF-1 and SCIF-2 by adjusting the amount of SbF$_6$− could be prepared. SCIF-1 crystallized in an achiral space group, forming a racemic superstructure, which led to CPL-silent. However, unlike SCIF-1, SCIF-2-Left and SCIF-2-Right enantiomers were completely separated, leading to larger channels and CPL signals ($g_{\text{av}} = (\pm 3 \times 10^{-3})$).

Shortly after, a pair of Ag$_{10}$ clusters (2-(S/R)) have been constructed by Zhao’s group, using (S)/(R)-3-ethyl-5,5',6,6',7,7',8,8'-octahydro-1,1-binaphthyl-2,2'-diamine, 2-(pyridin-2-yl)-1H-benz[d]imidazole (PBZ), and AgClF$_3$SO$_3$ (Figure 11H) $^{[219]}$. And structural analysis indicated that the peripheral chiral ligands, the helical arrangement of PBZs, and the spirocyclic Ag$_9$ core collectively contributed the chirality of 2-(S/R). Moreover, Ag$_9$ cores are coordinated with Ag10 through the pendant NH$_2$ group of each chiral ligand, resulting in an infinite chain. Interestingly, the aggregations of 2-(S/R) could cause fluorescence-to-phosphorescence switching. Remarkably, 2-(S/R) triggered outstanding CPL signals with high $g_{\text{av}}$ of $\pm 0.16$ in solid film. Such $g_{\text{av}}$ value lays the foundation for CPL-active metal clusters in practical application.

Other crystalline materials

In 2011, Tsumatori et al, for the first time, obtained the CPL spectra of Eu(III) complexes based on single crystal and then demonstrated that CPL varied according to its lattice plane. Furthermore, their CPL spectra in the crystals and solution were different.$^{[220]}$ More recently, Kawai’s group reported enantiomers, [(R)- or (S)-Pr-pybox)$_8$(LnIII)$_8$(THP)$_8$ (Ln = Eu and Tb, THP = tri-anionic tris-β-diketonate and Pr-pybox = chiral bis(4-isopropyl-2-oxazolinyl)pyridine), exhibiting remarkable CPL signals with extraordinary $g_{\text{av}}$ factors ($\pm 1.25; \pm 0.25$) in chloroform.$^{[221]}$ Particularly, circular Eu$^{III}$ helicates dispersed in the PMMA films, and KBr pellets also showed similar CPL nature. And large $g_{\text{av}}$ values enabled eye distinguishable circular polarization. Hereafter, Ono et al demonstrated the preparation and optical resolution of the dinuclear triple-stranded helicates, showing mirror images CD and CPL signals ($g_{\text{av}} = (10^{-3})$ order) with multi-color PL in the solid state or dichloromethane.$^{[222]}$

CPL is a unique excited-state luminescent property, which is conducive to dig deep for excited state information compared with CD.$^{[10]}$ However, owing to the lack of further study and commercial instruments for CPL measurements in the early days, numerous atomically precise enantiomer crystalline materials with remarkable Cotton effect didn’t explore their excited state information, which severely hinders applications of CPL-active enantiomer crystalline materials.$^{[173,212,223-231]}$ Notably, the representative examples detailed above clearly exhibited the potential
Figure 11. (A) The coordination modes of the Au₄ unit and chiral ligands. CPL spectra of (Au₄L₄)₄/(Au₄D₄)₄ in aggregate state and solid state. Prototype and CPEL spectra of CP-OLED devices. Adapted with permission. Copyright 2020, Tsinghua University Press and Springer-Verlag GmbH. Germany, part of Springer Nature. (B) Schematic synthesis and spontaneous resolution driven conglomerate crystallization of the racemic Ag₃0 rac clusters (L/R, left/right). Adapted with permission. Copyright 2020, American Chemical Society. (C) Schematic diagram of cations manipulating the chiral assembly of chiral luminescent [Cu₅(S'Bu)₆]⁻ into racemic, mesomeric, and homochiral crystals. CD and CPL spectra of D/L-Cu₅V in the solid state. Adapted with permission. Copyright 2019, John Wiley and Sons. (D) Schematic diagram of assembly process and evolution of chiroptical properties of R/S-Cu1₄ clusters. Adapted with permission. Copyright 2019, John Wiley and Sons. (E) The structures, CD, and CPL spectra of R/S-Cu3. Adapted with permission. Copyright 2020, American Chemical Society. (G) The crystal structure and CPL spectra of SCIF-1, SCIF-2-Left, SCIF-2-Right. Adapted with permission. Copyright 2020, John Wiley and Sons. (H) The crystal structures, CPL spectra (solid line), and g熳 curves (dotted line) of 2-(S/R). And a zigzag infinite coordination chain in 2-(S). PL spectra of 2-(S) with AIE feature. Adapted with permission. Copyright 2020, John Wiley and Sons.
of chiroptical materials in the crystalline state. And the rational design and synthesis of CPL-active materials in the agglomerate state are certainly an area of opportunity and are expected to be “fertile ground” in the next phase of development for crystalline metal organic materials.

AMPLIFICATION APPROACH OF CPL MATERIALS IN AGGLOMERATE STATE

It is well known that the pursuit of a high \( g_{\text{lum}} \) factor is one of the keys to develop CPL-active materials. In fact, however, most reported CPL in various agglomerate emitters (vide supra) suffered from small \( g_{\text{lum}} \) factors, generally \( 10^{-3} \) or even less and is hard for practical applications.\(^{[82–84,136,160,180,213]} \) Theoretically, \( g_{\text{lum}} \) that is, the degree of CPL signals can be predicted according to Equation (4). Besides, the fact that \( |M_{ab}|^2 \) is negligible compared with \( |P_{ab}|^2 \) in the event of electric-dipole-allowed transitions, and thus Equation (4) can evolve to Equation (5).\(^{[6,10,29]} \)

\[
g_{\text{lum}} = \frac{4|M_{ab}|}{|P_{ab}|} \cos \theta_{ab} \tag{5}
\]

Based on this formula, a conclusion that \( g_{\text{lum}} \) is proportional to \( |M_{ab}| \) and inversely proportional to \( |P_{ab}| \) can be drawn. Consequently, a high \( g_{\text{lum}} \) value generally requires electric-dipole-forbidden transition and magnetic-dipole-allowed transitions. This is why chiral lanthanide(III) complexes possess the advantages in terms of \( g_{\text{lum}} \) factors, in which large \( g_{\text{lum}} \) factors can be obtained owing to the magnetic-dipole-allowed f-f transitions,\(^{[232–233]} \) whereas small electric-dipole transitions always result in a lower PLQY that is characterized by the other factor of CPL performance.\(^{[240]} \) Hence, it remains a challenge to achieve excellent CPL performance, that is, brightness for CPL (\( B_{\text{CTM}} \)). Currently, several strategies aimed at the amplification of \( g_{\text{lum}} \) values of agglomerate emitters have been reported,\(^{[10,234]} \) including aggregation-amplified CPL,\(^{[59,210–211]} \) assembly-amplified CPL,\(^{[76,232,233,235–237]} \) Förster resonance energy transfer (FRET)-amplified CPL,\(^{[105–106,110,116]} \) well-ordered arrays of chiral emitters-amplified CPL,\(^{[158]} \) LC-amplified CPL,\(^{[126–129]} \) achiral component-amplified CPL,\(^{[93]} \) helicity-amplified CPL (Figure 12).\(^{[110,160]} \) Here, we will not review the detailed examples in this section since they are scattered in previous chapters.

CONCLUSION AND OUTLOOK

Agglomerate CPL materials are emerging and will gain more attentions. Nevertheless, there remain many difficulties and challenges in this field. (i) Improving the accuracy of CPL signals of solid-state samples directly from CPL-spectrometers needs more technological progress and supports.\(^{[47–50]} \) The commercial CPL-spectrometers are relatively new. And CPL measurement based on them is accurate when the orientational distribution of the samples is isotropic, such as solvent samples. Therefore, the CPL signals of solid-state materials are susceptible to macroscopic anisotropy, which would result in some undesirable artificial effects.\(^{[47]} \) (ii) It remains a considerable challenge to amplify \( g_{\text{lum}} \) values in a controllable way. The approach that helical assembly of luminescent cores may be expected to enlarge the \( g_{\text{lum}} \) of the agglomerate materials. (iii) The thermal stability of CPL characteristics is also an important index to evaluate whether it can be applied in practice since high temperatures are inevitable in the operation of electronic devices. Good CPL characteristics can be maintained at high temperature, that is, high temperature tolerance of CPL,\(^{[55]} \) which will be the direction of scientist’s efforts and also a challenge.

Great efforts based on the synthesis of CPL-active agglomerate materials have been made and exploiting their potential applications in displays, sensing, biomedicine, switches, and encryption are emerging. The construction of CPL devices with practical applications has attracted attention and been paid to extensive research. CPL devices including CP-OLEDs,\(^{[6,37]} \) CP-LCDs,\(^{[133]} \) CP laser emission,\(^{[17–18]} \) photonics, and CPL detectors,\(^{[170–171]} \) will be extensively researched. Especially, CP-OLEDs, based on CPL-active materials, are widely studied owing to simple device architectures, high efficiencies, low energy consumption, and tunable colors, which are regarded as the most promising candidates for the next generation of the display and lighting facility.\(^{[36–37,238–263]} \) Besides, CPL is also beneficial for developing chiroptical sensors.\(^{[117–249]} \) And CPL switches are also very interesting.\(^{[253–254,256,258]} \) Furthermore, due to the unique polarization properties of CPL, the materials with CPL can be applied to biomedicine (cell imaging).\(^{[210]} \) In terms of encryption and decryption, a logic device integrating CPL and lifetime encoding also realizes multi-informational data encryptions.\(^{[186]} \) In addition, new applications of CPL-active materials, such as data storage, optical information processing, magnetic chiroptical fields,\(^{[10,36,29,132]} \)

We hope that this review will give inspiration for more general design principles of efficient CPL-exhibiting materials in agglomerate state. We believe that the future is bright in this field, and the exciting new materials will be reported.
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ORCID
Shuang-Quan Zang https://orcid.org/0000-0002-6728-0559

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Xi-Yan Dong is Associate Professor in the College of Chemistry and Chemical Engineering, Henan Polytechnic University. She pursued her postgraduate study at Zhengzhou University and received her PhD degree of Chemistry under the supervision of Prof. Thomas C. W. Mak and Prof. Shuang-Quan Zang. Her research interest focuses on metal clusters, luminescent crystalline materials, and circularly polarized luminescence.

Shuang-Quan Zang received his PhD degree in Chemistry from Nanjing University in 2006 under the supervision of Prof. Qingjin Meng. After postdoctoral research with Prof. Thomas C. W. Mak at The Chinese University of Hong Kong, he joined the College of Chemistry of Zhengzhou University. He received The National Science Fund for Distinguished Young Scholars in 2018. He is serving as Dean of College of Chemistry and Green Catalysis Center at Zhengzhou University. His current scientific interests focus on atomically precise metal clusters, cluster-assembled materials, functional metal-organic frameworks, and chiroptical materials.

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AUTHOR BIOGRAPHIES

Chong Zhang received his MS degree from Guilin University of Technology in 2018. He is currently pursuing a PhD degree at the College of Chemistry, Zhengzhou University under the supervision of Prof. Shuang-Quan Zang and Yun-Yin Niu. His research direction is in the design of chiroptical materials based on metal clusters and MOFs.

Xi-Yan Dong is Associate Professor in the College of Chemistry and Chemical Engineering, Henan Polytechnic University. She pursued her postgraduate study at Zhengzhou University and received her PhD degree of Chemistry under the supervision of Prof. Thomas C. W. Mak and Prof. Shuang-Quan Zang. Her research interest focuses on metal clusters, luminescent crystalline materials, and circularly polarized luminescence.