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Article:

Basu, S. and Amdursky, N. (2025) Circularly polarized luminescence from assembled nanoscale particles. ACS Nano, 19 (38). pp. 33717-33733. ISSN: 1936-0851

<https://doi.org/10.1021/acsnano.5c10742>

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Circularly polarized luminescence from assembled nanoscale particles

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Abstract: Circularly polarized luminescence (CPL) from assembled nanoscale materials presents a rapidly advancing field with significant implications for optoelectronics, bioimaging, and chiral photonics. The ability to engineer CPL-active systems through the controlled assembly of nanoscale particles offers a versatile platform for tuning chiroptical properties beyond what is possible with individual components. This review provides a comprehensive overview of design strategies for achieving efficient CPL emission from nanoscale particles and their assemblies. We systematically differentiate between the design approach and the specific nanoscale entities employed, analyzing the benefits and limitations associated with each. A particular emphasis is placed on hierarchical chiral structures capable of exhibiting CPL activity, whether constructed from inherently chiral or achiral building blocks. For assemblies composed of achiral components, we delve into the physicochemical mechanisms underlying their emergent chirality and CPL behavior. In the final part of the review, we highlight recent advances and future prospects in the field, with a focus on the development of stable, high-performance CPL-active nanomaterials for applications in optoelectronics, bioimaging, and chiral photonics.

Keywords: circularly polarized luminescence, chirality, assembly, nanoscale particles, fluorescence

1. Introduction

Light, as an electromagnetic wave, is characterized by its polarization, which describes the orientation of its oscillations relative to its direction of travel. Polarization is typically defined by the behavior of the electric field vector, which classifies light into linear, elliptical, or circular polarization.¹ Circularly polarized light arises when the electric field vector rotates uniformly with a fixed phase difference while maintaining a constant magnitude. Depending on the rotation direction, circularly polarized light can be left-handed (counterclockwise) or right-handed (clockwise).

Although circularly polarized light can be artificially generated by combining two orthogonal linearly polarized beams or using a linear polarizer and a quarter-wave plate, these methods are inherently inefficient, with energy losses often exceeding 50%.² To address this, researchers have focused on developing chiral luminescent materials that can directly emit circularly polarized light, known as circularly polarized luminescence (CPL) materials, which efficiently produce CPL without external manipulation. This intrinsic optical property not only provides valuable insights into the excited-state structures of chiral materials but also opens up pathways to innovative applications, including 3D displays, CPL lasers, chiral sensing, optoelectronic devices, and asymmetric catalysis.³⁻⁸ As a result, CPL materials have become a prominent area of research, driving advancements in the field of chiral functional materials. The efficiency of CPL is typically evaluated using the dissymmetry factor (g_{lum}), which measures the degree of asymmetry between left- and right-circularly polarized emissions.² The g_{lum} is determined using the formula:

$$g_{lum} = 2 \frac{I_L - I_R}{I_L + I_R}$$

where, I_L and I_R denote the luminescence intensities of left- and right-circularly polarized light, respectively. A maximum g_{lum} value of 2 indicates entirely left- or right-circularly polarized emission, whereas $g_{lum}=0$ signifies unpolarized luminescence. Nevertheless, determining the optimal mathematical relationship between the parameters that define a CPL material and its performance remains challenging, making it difficult to identify the ‘ideal’ CPL material. Theoretically, the g_{lum} factor can be predicted by the equation:

$$g_{lum} = 4 \cos(\theta) \frac{|m||\mu|}{|m|^2|\mu|^2}$$

where m , μ , and θ represent the magnetic transition dipole moment, electric transition dipole moment, and the angle between them, respectively. This formula provides a clear guideline for designing CPL-active materials by enhancing m , reducing μ , and optimizing their angle θ .²

While aiming optimizing the CPL properties of materials, lanthanide complexes are promising candidates, attributed to their parity-allowed f-f transitions, which lead to strong magnetic dipole transitions and weak electric dipole transitions, and consequently, one of the highest g_{lum} values measured.^{9, 10} While these properties make lanthanide complexes attractive CPL materials, their practical application is often limited by factors such as low chemical

reactivity for post-synthetic modifications, moderate fluorescence quantum yield, and high production costs.¹¹

An alternative class of CPL-active materials consists of tailored chiral organic molecules exhibiting central, axial, planar, or helical chirality.¹¹⁻¹³ These molecules offer precise structural control, facile functionalization, and high quantum yields. However, their g_{lum} values typically range between 10^{-5} and 10^{-2} due to their electric-dipole-allowed but magnetic-dipole-forbidden transitions.¹¹ Moreover, their photo instability and susceptibility to degradation under prolonged irradiation remain significant drawbacks.

To address these challenges, recent efforts have focused on CPL-active nanoscale particles such as quantum dots (QDs),¹⁴⁻¹⁸ carbon dots (CDs),^{19, 20} and atomic nanoclusters.²¹⁻²⁴ These materials offer several advantages, including high quantum yields, cost-effectiveness, tunable surface functionalization, and superior photostability.²⁵⁻³³ However, individual nanoscale particles often exhibit low CPL efficiency due to weak chiral interactions at the molecular level.³⁴ To circumvent this limitation, researchers have turned to the controlled assembly of nanoscale particles, leveraging collective chiral effects to amplify CPL signals. By organizing nanoparticles into hierarchical chiral structures, such as helicoidal arrangements, superlattices, or supramolecular aggregates, stronger chiral optical responses can be achieved. This strategy not only enhances g_{lum} but also enables dynamic tunability of CPL properties through external stimuli, such as changes in solvent polarity, pH, or applied fields. Consequently, the assembly of nanoscale particles represents a promising avenue for developing next-generation CPL-active materials with enhanced efficiency and stability.

In this review article, we focus on design strategies for achieving efficient CPL emission from nanoscale particles and their assemblies. We differentiate between the design strategies and the chosen nanoscale particle for each strategy while discussing the advantages and disadvantages of each chosen entity. We further highlight one of the most important additional advantages of CPL-active hierarchical chiral structures, which is their ability to be constituted by either chiral building blocks or achiral ones, while still having CPL activity. For CPL-active structures composed of achiral nanoscale particles, we discuss the physicochemical origin of chirality and CPL-activity acknowledgements. In the second part of the review, we explore emerging trends and future directions in the development of stable, high-performance CPL-active nanoscale materials, emphasizing their potential applications in optoelectronics, bioimaging, and chiral photonics.

2. Chiroptical Assembly of Nanoscale Materials: Principles and Design Strategies

The assembly of nanoscale particles has been a popular choice for achieving superior physicochemical properties vis-à-vis individual constituents.³⁵⁻⁴⁵ Likewise, the controlled assembly of nanoscale particles into chiral architectures has emerged as a powerful strategy for enhancing chiroptical responses and achieving efficient CPL. Unlike molecular chiral luminophores, which rely on intrinsic asymmetry, chiroptical assemblies leverage nanoscale organization to amplify chiral optical effects through collective interactions.⁴⁶ By precisely

arranging nanoparticles into hierarchical structures, researchers could manipulate the interplay between electric and magnetic dipole transitions, thereby optimizing CPL efficiency. The design of such assemblies, however, requires a careful balance of interparticle forces, symmetry-breaking mechanisms, and external stimuli responsiveness to achieve high g_{lum} and robust optical activity. In this section, we explore the fundamental principles governing the chiroptical assembly of nanoscale particles and discuss key design strategies to maximize CPL performance, including self-assembly approaches, template-directed synthesis, and external field-induced structuring.

2.1. Fundamental principles of chiroptical nanoscale assembly

Before going into the various design principles for assembling chiroptical structures from nanoscale particles, we must first distinguish between the nanoparticles and the origin of chirality. We will start by discussing particles with an intrinsic chirality, i.e., from the particle's core structure, followed by strategies to induce chirality to particles lacking an intrinsic one.⁴⁷ While intrinsic chirality is inherent to the nanoparticle, stemming from factors such as atomic arrangements, crystallographic structures, shape asymmetry, or electronic transitions,⁴⁸ induced chirality results from interactions with chiral environments, including chiral ligands, solvents, or templating effects.⁴⁹⁻⁵³

2.1.1. Chirality at the nanoscale: Intrinsic chirality

When discussing the intrinsic chirality of nanoscale particles (**Figure 1**), we essentially refer to the structure of the nanoparticle. In this section, we will differentiate the origin of the intrinsic chirality of various structures, while differentiating between particles with an exact atomic arrangement, e.g., metal nanoclusters, and particles having a certain crystal structure, e.g., QDs.

Chirality at the nanoscale: Intrinsic chirality

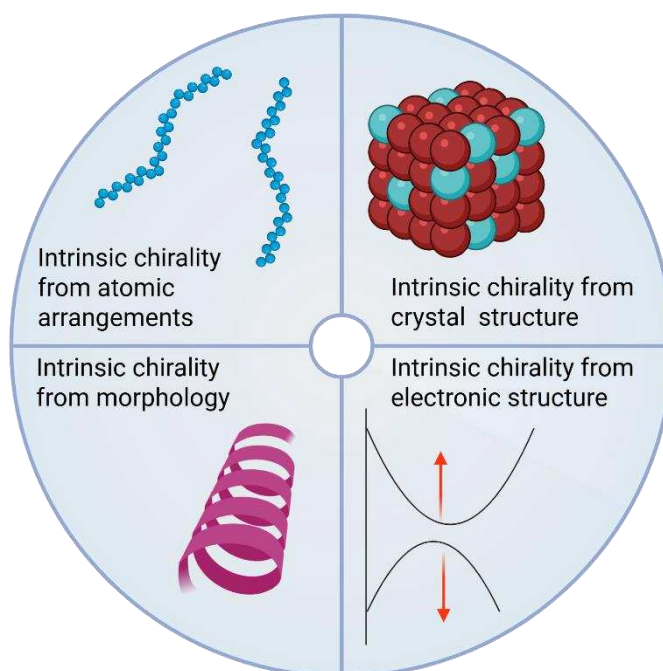


Figure 1. Schematic illustration of the various origins of intrinsic chirality at the nanoscale: the asymmetric arrangement of atoms, the random arrangement of atoms in nanocrystals, the morphology of the nanoscale particle, and the electronic structure of the nanoscale particles.

Intrinsic chirality from atomic arrangement – Some nanoparticles possess a precise atomic arrangement. These particles are commonly described as either metal nanoclusters or semiconductor nanoclusters. In such nanoclusters, the exact number of atoms and their arrangement are known. Accordingly, if the atomic arrangement of the nanocluster particles is chiral, i.e., they exhibit chiral atomic packing that is characterized by an internal structure that lacks mirror symmetry, this particle will have an intrinsic chirality.⁵⁴ In this context, it has been demonstrated that quantum size effects can distort atomic arrangements, resulting in an inherently chiral lattice structure.^{55, 56} An important example was presented in a theoretical study of gold nanocages, where a series of intrinsically chiral Au_{9n+6} clusters ($n \geq 2$) were predicted.⁵⁷ These structures consisted of helical Au_{9n} nanotubes capped with Au_3 units at both ends, with chirality originating from the helical arrangement of gold atom strands rather than from any chiral centers at the atomic level. The smallest stable intrinsically chiral nanocages, $\text{Au}_{42}(\text{C}_3)$ and $\text{Au}_{105}(\text{C}_3)$, were composed of repeating Au_7 “golden-eye” subunits and exhibited C_3 symmetry. Notably, the chiral Au_{42} cage was found to be energetically competitive with previously reported achiral isomers and capable of encapsulating smaller species, such as Au_6 , forming the stable complex $\text{Au}_6@ \text{Au}_{42}(\text{C}_3)$.

Intrinsic chirality from crystal structure arrangement – Nanoparticles lacking a precise atomic arrangement are usually characterized by a crystal structure, such as in semiconductor or metallic nanoparticles. Similar to the previous group, if the crystal structure exhibits a chiral lattice rearrangement, the nanoparticle will possess an intrinsic chirality. For example, the crystal structure of nanoparticles made of quartz, Te, and Se naturally possesses helical atomic stacking, resulting in chiral crystal lattices.⁵⁸ The handedness of these materials is dictated by the screw-axis symmetry within their crystal packing, leading to differential interactions with

circularly polarized light. For example, in a detailed structural analysis of quartz, the concept of continuity in symmetry and chirality was introduced as a novel, quantitative descriptor to assess the global chirality of extended crystal structures.⁵⁹ Multiple hierarchical units within low quartz - including the SiO₄ tetrahedron, the Si(OSi)₄ unit, and the SiSi₄ tetrahedron - were evaluated in terms of their chirality, tetrahedrality, and distortion from ideal symmetry. Among the various units, a four-SiO₄ segment helix was identified as the motif within the quartz lattice that is most probable to induce chirality. Chirality values were determined not only for low quartz, but also for high quartz and cristobalite, offering a comparative framework for understanding the intrinsic chirality embedded in crystal architectures. This study demonstrated that chirality in crystals may not only arise from macroscopic morphology or defects, but can be fundamentally encoded in the spatial arrangement of symmetric units across the lattice. Furthermore, intrinsically chiral nanocrystals of α -mercury sulfide (HgS), whose chirality arises from their non-centrosymmetric crystal structure, were synthesized via a colloidal precipitation reaction in which the chiral thiolate-containing molecule penicillamine was used.⁶⁰ This molecule was expected to bind strongly to Hg²⁺ ions and was found to direct the transformation of initially formed achiral β -HgS nanocrystals into the thermodynamically favoured chiral α -HgS phase. A gradual colour change from black to orange was observed during this transformation. The resulting α -HgS nanocrystals were obtained with high enantiomeric excess and exhibited a strong chiroptical response, several orders of magnitude greater than that observed for achiral semiconductor nanocrystals coated with similar chiral ligands.

Intrinsic chirality from crystal structure defects – Following the previous class, the intrinsic chirality of nanoparticles that are characterized by a crystal structure does not necessarily have to emerge from a chiral lattice rearrangement, but also from a defect in an achiral crystal rearrangement. Such a defect can introduce asymmetry, thus resulting in an intrinsic chirality and the nanoparticle's chiroptical response. For example, Te and Se nanowires with defects in their crystal structure (i.e., missing Te/Se atoms) display helical crystal symmetry, making them intrinsically chiral and strong CPL emitters.⁶¹⁻⁶³ In a notable example, it was shown that in CdSe nanocrystals, intrinsic chiroptical activity was attributed to chiral dislocations and point defects that disrupted local symmetry and introduced handedness into otherwise symmetric structures.⁶⁴ These defects were believed to have formed spontaneously during crystal growth and were proposed to exist not only in wurtzite CdSe but also in other lattice types, particularly in twinned nanocrystals. As both left- and right-handed variants were equally likely to form, racemic mixtures were typically produced, rendering their chiroptical signatures undetectable by ensemble circular dichroism measurements. Nonetheless, the presence of such defects indicated that many colloidal inorganic nanocrystals previously considered achiral may in fact have contained intrinsically chiral subpopulations.

Intrinsic chirality from morphology – The shape of the nanoparticle can also result in an intrinsic chirality. In this class, the nanoparticle's shape induces asymmetry and results in an anisotropic nanostructure. Nanoscale particles with non-centrosymmetric morphologies, such as twisted nanoribbons, helices, helicoidal nanowires, and asymmetric quantum dots, break mirror symmetry, resulting in preferential interactions with left- or right-circularly polarized

light. This effect is particularly evident in plasmonic nanoparticles, where structural asymmetry governs optical scattering and chiral plasmonic behavior.⁶⁵⁻⁶⁷ To this end, a compelling example was demonstrated in the case of rolled-up semiconductor nanoplatelets, where intrinsic chirality was introduced through controlled mechanical deformation.⁶⁸ Upon relaxation of strain and the subsequent formation of tapered nanoscrolls, the initially planar nanoplatelets were transformed into chiral structures, and their crystal lattices acquired a helical distortion. This deformation enabled optically active transitions by facilitating the mixing of electric and magnetic dipole moments within the nanocrystal's electronic subsystems. A simple physical model based on linear coordinate transformations was developed, allowing the nanoscroll to be mathematically unrolled into a flat, topologically distorted nanoplatelet, which made it possible to analytically evaluate its chiroptical properties. Using this approach, the total and differential absorption, as well as circular dichroism, were calculated and found to be largely independent of the nanoplatelet's lateral size in the winding direction. The model was applied to CdSe nanoscrolls, where a clear correspondence between circular dichroism and absorption spectral features was observed. The study elucidated that the optical activity of such nanoscrolls originated from the chiral lattice deformation, with well-defined selection rules for interband transitions. In a related study, chiroptical activity was investigated in three-dimensional CdSe nanotetrapods synthesized via a hot-injection method, which yielded structures with uniform morphology and distinct crystal phases in the core and arms, forming intracrystal heterojunctions.⁶⁹ Upon capping with enantiopure cysteine ligands, strong circular dichroism signals were observed, which were attributed not merely to surface interactions, but to intrinsic excitonic transitions influenced by the structural asymmetry of the tetrapods. The circular dichroism response was found to be highly sensitive to the aspect ratio, with the anisotropic g-factor reaching a maximum at an intermediate size, and then decreasing, indicating that morphology-controlled excitonic coupling between the core and arms played a central role in determining the optical activity.

Intrinsic chirality from electronic structure – The last mechanism to induce optical chirality of nanomaterial-based structures is based on exploiting the electronic transitions in certain nanoparticles.⁷⁰ Some QDs and semiconductor nanocrystals exhibit chiroptical properties due to their intrinsically chiral electronic wavefunctions, even in the absence of structural asymmetry. This is particularly evident in lanthanide-doped nanocrystals, where the interaction of electric and magnetic dipole transitions within the f-f orbitals results in strong CPL activity.⁷¹ Also, in the context of atomic clusters, a detailed computational investigation of the $[\text{Au}_{20}(\text{PP}_3)_4]\text{Cl}_4$ cluster revealed that its Au_{20}^{4+} core adopted an intrinsically chiral geometry, arising from a triblade 7-atom motif positioned asymmetrically atop a regular Au_{13} icosahedron.⁷² This chiral arrangement led to distinct splitting of the superatomic 1D and 1F orbitals, producing an unusual electronic configuration: $1\text{S}^2 1\text{P}^6 1\text{D}^4 2\text{S}^2 1\text{D}^2$. The optical activity in the visible range was dominated by electronic transitions within this superatomic framework and metal-to-ligand charge transfer (MLCT) bands. Comparison between the circular dichroism spectra of bare and ligand-protected clusters showed that the chiroptical response in the unprotected Au_{20}^{4+} and Au_{23}^{9+} clusters was governed by the intrinsic asymmetry of their electronic structure, while in ligand-protected systems, such as $\text{Au}_{38}(\text{SMe})_{24}$, the chiral arrangement of surface motifs dominated the circular dichroism features.

2.1.2. Chirality at the nanoscale: Induced chirality

As stated, unlike intrinsic chirality, many achiral nanoparticles can also acquire chirality through various interactions, a phenomenon known as induced chirality (**Figure 2**). Induced chirality can be a result of a wide variety of external influences, while below, we will

summarize the mechanisms that can induce chiroptical activity.⁷³⁻⁷⁵ It is worth-mentioning here that induced chirality at the nanoscale can manifest at two interconnected levels: within individual nanoparticles and through their collective organization into assemblies. At the single-particle level, chirality often arises from ligand functionalization, solvent interactions, or external fields, which introduce asymmetry in the particle's electronic environment. In contrast, when achiral nanoparticles assemble under the guidance of chiral templates or supramolecular cues, cooperative interactions lead to emergent chiroptical activity that is often stronger and more tunable than that of isolated particles. In the following discussion, we consider both approaches, with emphasis on how each contributes to enhancing CPL through either local perturbations or collective, assembly-driven effects.

Induced chirality of nanoscale particles from external cues

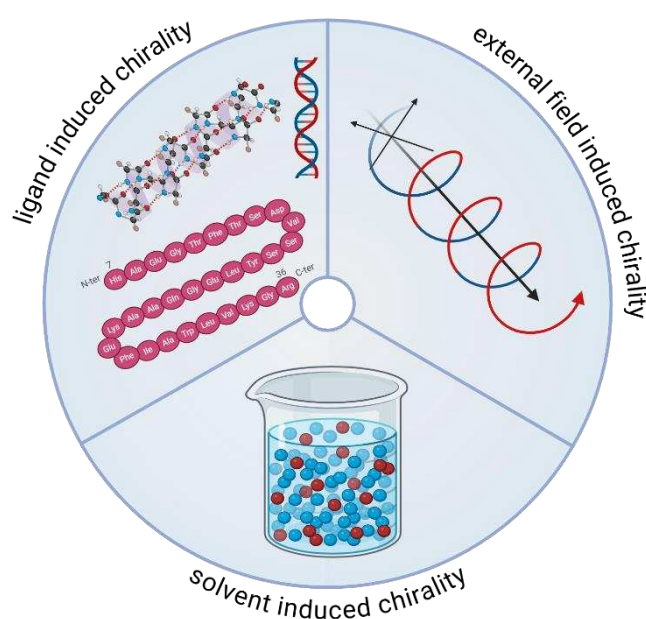


Figure 2. A Schematic representation of the various cues that can induce chirality at the nanoscale: ligand functionalization, external field (e.g., light, magnetic, or electronic), and solvent interactions.

Induced chirality from ligand functionalization - One of the most common approaches to inducing chirality is ligand functionalization, where chiral molecules such as amino acids, peptides, or DNA selectively bind to the nanoparticle surface, introducing asymmetry in electronic transitions and optical responses.^{76, 77} For instance, gold and silver nanoparticles modified with chiral thiols exhibit strong circular dichroism and CPL due to ligand-induced perturbations in their plasmonic properties. Another effective strategy is chiral template-assisted assembly, where preformed chiral structures, such as DNA origami, helical peptides, or liquid crystal phases, guide the organization of achiral nanoparticles into twisted architectures, amplifying their chiroptical response.⁷⁸⁻⁸⁰ A notable example for this strategy is the formation of three-dimensional plasmonic chiral architectures that were constructed by dressing DNA origami templates with gold nanoparticles at precisely defined positions.⁷⁸ The gold nanoparticles were first assembled along linear arrays on a two-dimensional rectangular

DNA origami sheet, with sub-10 nm spatial control. Upon programmed rolling of the DNA template, the gold nanoparticles were repositioned into a well-defined helical geometry, effectively transforming an initially achiral planar configuration into a chiral 3D nanostructure. Chiral organization was also induced through DNA-functionalized bimetallic nanostructures designed for enhanced optical response. In a recent study, gold–silver core–shell nanorods were synthesized and simultaneously functionalized with DNA, enabling their precise assembly onto DNA origami templates.⁸¹ The incorporation of a silver shell significantly improved the plasmonic performance of the nanorods compared to their gold-only counterparts, leading to a marked enhancement in circular dichroism signals. This strategy not only addressed the stability limitations of silver nanorods in aqueous and buffered environments but also demonstrated how ligand-driven self-assembly could be exploited to engineer robust and optically active chiral nanostructures. As an extension of this approach, gold nanobipyramids were assembled into both static and dynamic chiral architectures using DNA origami.⁸² These gold nanobipyramids dimers exhibited more pronounced and complex chiroptical responses than conventional gold nanorod-based dimers, highlighting how precise shape control and programmable DNA-guided assembly can be used to tune the optical properties of ligand-induced chiral plasmonic systems. While such assemblies are known for strong circular dichroism, they often suffer from low optical asymmetry due to suboptimal electric–magnetic dipole coupling and strong scattering effects. However, these limitations were recently overcome through the long-range, liquid-crystal-like organization of gold nanorods into helices mediated by human islet amyloid polypeptides.⁸³ This biomolecular templating enabled polarization-dependent spectral shifts and significantly reduced energy-state scattering, particularly for dipoles with antiparallel orientation. Remarkably, the optical asymmetry was enhanced by more than 4600-fold in these supramolecular helices, demonstrating that hierarchical organization driven by ligand interactions can dramatically enhance the chiroptical performance of plasmonic materials.

Induced chirality by solvent or external field - Additionally, solvent interactions and asymmetric confinement can influence nanoparticle behavior, with chiral nematic liquid crystals aligning QDs into ordered arrays that display chiroptical activity.⁸⁴ Beyond chemical interactions, external fields such as circularly polarized light, magnetic fields, or electric fields can dynamically induce chirality. CPL irradiation, for example, can selectively excite electronic transitions in nanoparticles, leading to preferential alignment and chiral emission. At the same time, magnetic and electric fields can manipulate dipole-dipole interactions or asymmetric charge distributions, further enhancing chiroptical effects.^{85, 86}

Unlike intrinsic chirality, which is fixed by the nanoparticle's structure, induced chirality is highly tunable and reversible, making it particularly advantageous for developing dynamic CPL systems, real-time sensing platforms, and stimuli-responsive photonic materials. By harnessing these strategies, researchers can design next-generation chiral nanoscale assemblies with tailored optical properties for a wide range of applications. However, it is also important to note that intrinsic chirality typically produces a stronger chiroptical response than ligand-induced chirality, as it arises from the collective contribution of all atoms within the chiral structure rather than from surface-bound asymmetry alone.

While the intrinsic origins of chirality at the molecular or nanoscale level provide the foundation for understanding chiroptical properties, these effects are often weak in isolation. A critical step toward achieving pronounced CPL is the induction and amplification of chirality through assembly formation, where individual chiral or achiral building blocks collectively generate stronger optical activity. Thus, the approach of assembly acts as the essential link between the origin of chirality and its practical utilization. In the next section, we focus on the diverse strategies employed to construct such assemblies, illustrating how these design approaches enhance CPL performance.

2.2. Design strategies for chiroptical nanoscale assemblies exhibiting CPL

Having examined how chirality can arise both within individual nanoparticles and through their assembly via external cues, the next step is to focus on the rational design principles that systematically guide the construction of chiral ensembles. The development of chiroptical nanoscale assemblies requires precise design strategies to control their structure, stability, and optical properties. Unlike isolated chiral molecules, where chirality is often inherent, nanoscale assemblies leverage collective interactions to amplify chiral optical effects, particularly CPL. Achieving high g_{lum} values and tunable chiroptical responses depends on the rational selection of building blocks, controlled assembly techniques, and external modulation strategies.

2.2.1. Self-Assembly of nanoscale particles endowed with CPL activity

Self-assembly, governed by weak intermolecular forces such as hydrogen bonding, π - π stacking, van der Waals interactions, and electrostatic interactions, provides a robust strategy for constructing chiroptical nanoscale materials. This approach has been particularly effective in assembling plasmonic, semiconductor, and hybrid nanomaterials into well-defined helical superstructures, thereby enabling CPL emission. The underlying chemistry of these self-assembled systems is crucial, as molecular interactions dictate the chiral organization and CPL properties of the final assemblies. In this section, we will detail the various nanoscale particles that were used for the self-assembly of CPL-active assemblies. Our primary differentiation here will be the used nanoscale particle: QDs, perovskites, CDs, and metal nanoclusters, while highlighting the key strategies and chemical interactions involved in the use of each nanoscale particle, as summarized in **Table 1**.

One of the most widely employed methods for inducing chiral self-assembly is ligand-mediated organization, where chiral organic ligands or biomolecules guide achiral nanoparticles into supramolecular chiral structures. DNA origami, peptides, and block copolymers serve as molecular scaffolds, imposing a chiral environment that amplifies the chiroptical response of the assembled nanomaterials. The chemical design of these ligands is fundamental in determining not only the stability of the self-assembled structures but also their optical properties. The electronic coupling between adjacent nanomaterials in such architectures is highly sensitive to ligand length, steric hindrance, and functional group interactions, affecting g_{lum} and emission wavelengths. In the following section, we will discuss

the four main design strategies, focusing on the specific nanomaterial in question, of how to utilize self-assembly to make higher-hierarchical assemblies with CPL activity (**Figure 3**).

Self-assembly of quantum dots for CPL activity - In QDs systems, supramolecular self-assembly has been successfully utilized to achieve CPL emission by embedding QDs within chiral organic matrices (**Figure 3a**).⁸⁷ Liu and co-workers have demonstrated that the use of 3-mercaptopropionic acid-capped QDs combined with chiral gelators has allowed for tunable CPL emission, where the molecular chirality of the gelator has dictated the handedness of CPL. The key chemical process governing this transformation involves hydrogen bonds between the achiral QDs and the chiral supramolecular matrix, which have facilitated the asymmetric spatial organization of the achiral QDs, based on a mechanistic study involving the spectral analysis of disassembled mixtures. In the disassembled state, where the QDs and gelator remained molecularly dispersed in a good solvent, strong photoluminescence was retained under UV excitation, but no CPL signal was detected. This absence of CPL, in contrast to the pronounced CPL observed in the coassembled gels, clearly indicated that the coassembly process played a critical role in the chirality transfer from the chiral gelator to the QDs. The well-ordered spatial arrangement of QDs along the chiral nanotubes formed by the gelator was identified as essential for the emergence of CPL. Furthermore, the electrostatic and hydrogen-bonding interactions between the 3-mercaptopropionic acid ligands on the QD surface and the amine groups of the chiral gelator were considered the primary driving forces for the coassembly, enabling the formation of the chiral supramolecular architecture necessary for CPL induction. Thus, in such QD-gelator systems, the origin of chirality is not intrinsic to the QDs themselves but rather induced through supramolecular co-assembly, wherein the chiral gelator matrix transfers its handedness to the otherwise achiral nanocrystals.

Self-assembly of perovskites for CPL activity - Perovskite nanomaterials have also been endowed with CPL activity via controlled supramolecular assembly (**Figure 3b**).⁸⁸ Wang and co-workers have shown that 2-dimensional chiral perovskite heterostructure films, composed of chiral (R-/S-MBA)₂PbI₄ perovskites coupled with CsPbBr₃ QDs, exhibited strong CPL signals due to energy transfer between the chiral perovskite layers and QD layers. The presence of carboxyl-functionalized chiral ligands in these perovskite structures has played a critical role in stabilizing the layered assembly, enabling efficient chirality transfer. The fine-tuning of anion exchange chemistry between chiral perovskites and CsPbBr₃ QDs has further provided a handle to adjust emission properties, demonstrating precise control over the CPL response via targeted chemical modifications. In this regard, a series of chiral perovskite heterostructure films was prepared by combining 2D chiral perovskites ((R-/S)-MBA)₂PbI₄ with CsPbBr₃ QDs. Strong circular dichroism signals were observed from these films, along with a clear CPL response under ambient conditions, with a maximum measured g_{lum} of 6×10^{-3} . Circularly polarization-resolved transient absorption spectra indicated that energy was transferred from the chiral perovskite layer to the QDs layer, resulting in the generation of spin-polarized excitons in the QDs, which in turn gave rise to the CPL signal. Additionally, the photoluminescence (PL), CD, and CPL spectra were modulated over a broad wavelength range by varying the thickness and composition of the chiral perovskite layer. This indicated that the extent of anion exchange between the chiral perovskites and QDs was dependent on these

structural parameters. Through this strategy, chiral perovskite films with enhanced CPL responses were fabricated, and important insights into the origin of CPL in such hybrid systems were provided.

Beyond 2-dimensional perovskites, Liu and co-workers have shown that achiral perovskite nanocrystals could be functionalized with CPL activity by co-assembling them with chiral gelators in nonpolar solvents.⁸⁹ The chirality transfer mechanism in these systems has relied on noncovalent interactions such as π - π stacking and hydrogen bonding between the perovskite nanocrystal surface and the chiral gelator, leading to the emergence of CPL with dissymmetry factors in the 10^{-3} range. The molecular design of these gelators, incorporating aromatic π -conjugated cores and amide functionalities, has ensured both strong CPL signals and stable self-assembled structures. To further investigate the mechanism of induced CPL in these coassemblies, time-resolved photoluminescence measurements were performed. The luminescence lifetime of perovskite NCs was found to increase significantly upon coassembly with the chiral gelator, indicating that nonradiative decay processes were effectively suppressed. In contrast, when the NC-gelator mixtures were examined in their disassembled state in a good solvent, no CPL signal was observed despite strong emission under UV excitation. This absence of CPL in the disassembled state, coupled with the intense CPL observed only in the coassembled structures, confirmed that the coassembly process played a critical role in chirality transfer. The well-ordered arrangement of perovskite NCs within the chiral supramolecular matrix was identified as essential for achieving induced CPL in these hybrid materials.

The aforementioned studies involve the generation of assemblies of perovskite nanocrystals for CPL emission; however, a fundamental distinction lies in the origin of chirality. In the case of 2D chiral perovskite-QD heterostructures, the chirality is intrinsic to the perovskite layers themselves, which transfer their chiral excited-state character to the coupled quantum dots through energy transfer pathways and spin-selective processes. By contrast, for assemblies based on achiral perovskite nanocrystals, chirality is externally induced at the supramolecular level, where co-assembly with chiral gelators modifies the nanocrystal surface and propagates molecular chirality throughout the network, thereby giving rise to CPL-active architectures.

Self-assembly of carbon dots for CPL activity - Owing to the rich surface chemistry of CDs, they have been utilized as a nanomaterial for making self-assembled structures featuring CPL activity. Lu and co-workers have shown that the introduction of hydrophobic aromatic ligands onto CDs could facilitate an ordered assembly via π - π interactions, while residual chiral amides have mediated hydrogen bonding, resulting in asymmetric bending of the nanostructures and strong CPL signals. The modulation of these surface functional groups has enabled full-color CPL emission and solvent-dependent CPL reversibility.⁹⁰ In one approach, cholesteryl-functionalized CDs have been synthesized by Li and co-workers using cholesteryl monoprotected ethylene diamine as a precursor, leading to polarity-sensitive self-assembly that has enhanced chiroptical signals⁹¹ (**Figure 3c**). The strong van der Waals interactions between the rigid cholesteryl units have dictated the chiral organization of the CDs aggregates,

amplifying CPL dissymmetry factors from 10^{-3} to 10^{-2} in solvent-free conditions.⁹¹ While the intrinsic chirality of the cholesteryl precursor was acknowledged to contribute to the optical activity, the pronounced CPL response in this system was primarily attributed to supramolecular self-assembly, in which the collective ordering of cholesteryl groups played the dominant role. Moreover, an intriguing approach has been developed by the combination of achiral CDs with solvent-induced chirality transfer to generate large supramolecular structures exhibiting CPL.⁹² In this system, CDs with bright luminescence and tunable surface moieties have been synthesized using different precursors. The effect of introducing a chiral solvent into aqueous dispersions of these CDs has been systematically investigated, revealing that only nitrogen-containing CDs with amine functional groups have exhibited CPL signals and self-assembled into large fibrillar structures. Mechanistically, two interrelated interactions were crucial: (i) interfacial recognition between the CD surface and the chiral solvent, and (ii) intermolecular interactions among CDs leading to assembly. The amine-functionalized CDs were drawn to the aqueous-limonene interface, likely due to lone pair- π interactions between the nitrogen atoms and the π -electrons of limonene. This interaction not only facilitated chirality induction but also positioned the CDs in close proximity. Once localized at the interface, hydrogen bonds between amine and carboxyl/amine groups on neighboring CDs promoted the formation of extended fibrillar networks, which supported the emergence of CPL. In contrast, non-doped CDs, despite interacting strongly with limonene via hydrophobic forces, lacked the necessary surface functionality for further assembly, while only sulfur-functionalized CDs exhibited poor limonene interaction due to surface oxidation, and failed to assemble or generate CPL. The combined requirement of interfacial localization and directional inter-CD interactions emphasizes the dual role of amine groups in mediating both chiral recognition and supramolecular assembly. These findings highlight how surface chemistry can be precisely tailored to enable the transformation of achiral fluorescent nanoparticles into hierarchically organized, optically active structures. This solvent-guided assembly strategy offers a powerful route toward the bottom-up construction of CPL-active chiral nanomaterials from simple, achiral building blocks. In this system, chirality was not derived from the carbon dots themselves but was induced through solvent-mediated interfacial recognition and supramolecular assembly, whereby the handedness of the chiral solvent was transferred into the emergent fibrillar nanostructures.

Self-assembly of metal nanoclusters for CPL activity - Metal nanoclusters have emerged as another class of CPL-active materials, where self-assembly strategies have played a pivotal role in modulating their chiroptical properties. To this end, Amdursky and co-workers fabricated supramolecular assemblies of atomic nanoclusters, achieved through a hierarchical process (**Figure 3d**).⁹³ At first, uniform achiral gold nanoclusters (AuNCs) that were ligated using mercaptopropionic acid were further complexed with Zn ions. The latter complexes were organized into fiber conformations via hydrogen bonding with Tween-20. These assemblies exhibited right CPL at 443 nm with a dissymmetry factor of 13×10^{-3} , while the other structures in the hierarchical assemblies remained CPL silent. Interestingly, the final fiber configuration displayed excitation-dependent luminescence, which again was attributed to hydrogen bonding. Notably, CPL signals appeared only when excited at the red edge of the circular dichroism spectrum, indicating that only specific chiral transitions contributed to CPL.

emission. It is worth mentioning that in this system, chirality was not intrinsic to the gold nanoclusters but was induced at the supramolecular assembly level, where hierarchical organization and hydrogen-bonding interactions transferred chiral information into the final fibrous structures that exhibited CPL.

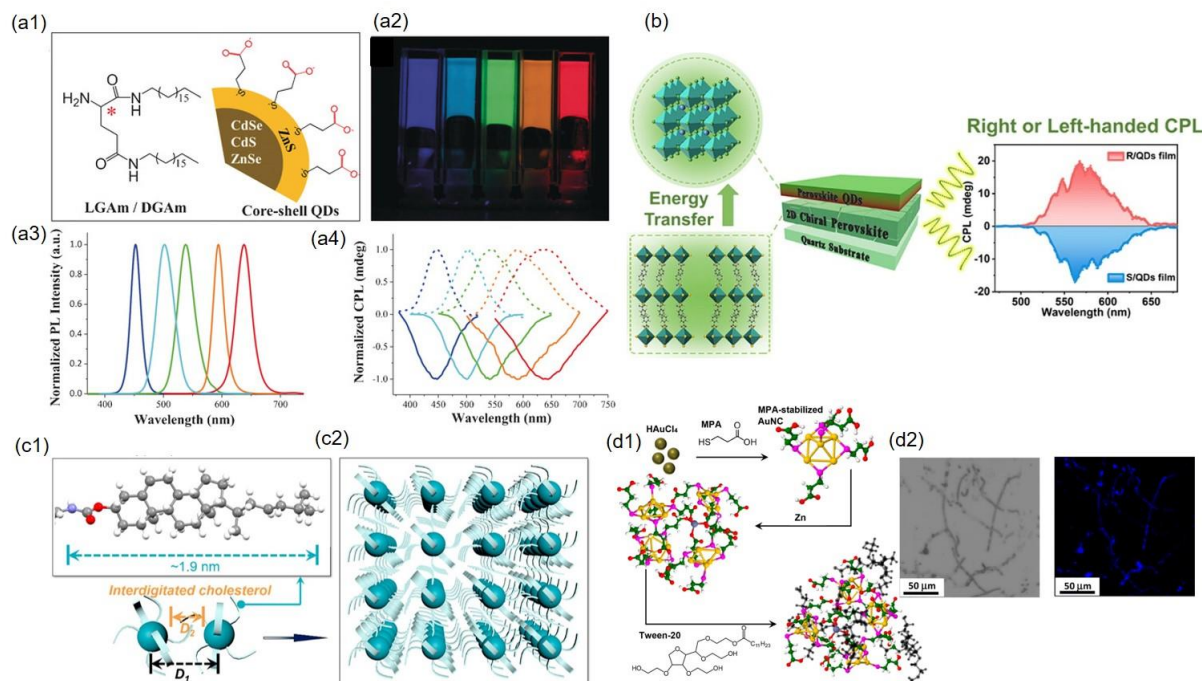


Figure 3. (a1) Molecular structures of the gelators and the QDs used to achieve CPL active self-assemblies. (a2) Digital photographs of self-assembled QDs under UV light. (a3) Fluorescence and (a4) CPL spectra of self-assembled QDs using chiral gelators. Reproduced with permission from ref⁸⁷. Copyright 2017 John Wiley & Sons. (b) Schematic representation of the fabrication process for a layered perovskite and QD heterostructure exhibiting CPL activity mediated by energy transfer. Reproduced with permission from ref⁸⁸. Copyright 2024 American Chemical Society. (c1 – c2) Illustration and structural packing parameters of CDs assembled into configurations exhibiting CPL activity. Reproduced with permission from ref⁹¹. Copyright 2023 American Chemical Society. (d1) Schematic depiction of the hierarchical organization of AuNCs facilitated by zinc ions and Tween-20, resulting in a CPL-active structure. (d2) Confocal laser scanning microscopy images showing the fluorescent fibrillar structures formed in the assembly. Reproduced with permission from ref⁹³. Copyright 2023 American Chemical Society.

A complementary approach to CPL engineering in metal nanoclusters has involved chirality modulation through ligand exchange chemistry by Liu and co-workers.⁹⁴ By designing Ag nanoclusters (AgNCs) stabilized by thiazithione- and oxazinthione-based ligands, researchers have achieved tunable CPL properties via ligand-dependent supramolecular interactions. Notably, methyl/phenyl ligand exchange has induced chirality inversion of the AgNCs assemblies, attributed to variations in inter-cluster interactions and supramolecular helical organization. This atomic-level control over chirality transmission has provided insights into the fundamental role of ligand chemistry in dictating CPL properties.

Liquid crystal (LC) matrices have emerged as a powerful medium for directing the self-assembly of CPL-active nanoclusters, providing a confined and dynamic environment for supramolecular chirality modulation. A recent breakthrough in this domain has involved the helical assembly of atomically precise achiral AuNCs within an LC template.⁹⁵ A major challenge in integrating AuNCs into LC phases has been their poor miscibility; this has been successfully addressed through dual-ligand functionalization, incorporating both dodecanethiol and a specially designed LC-like ligand. The tailored surface chemistry of AuNCs has enabled their controlled dispersion within the LC phase, leading to the formation of helical nanofilaments upon self-assembly. The structural organization of these LC-templated AuNC assemblies has played a crucial role in CPL generation. The hierarchical helical structuring, driven by intermolecular van der Waals and π - π interactions, has preserved the atomic precision of the nanoclusters while simultaneously tuning their luminescence properties. The introduction of thermal processing as a secondary control parameter has enabled researchers to fine-tune the optical properties of the assembled AuNCs, revealing the direct impact of hierarchical structuring on CPL activity. Importantly, in this system, chirality was not intrinsic to the AuNCs but was induced through the LC-templated supramolecular helical assembly, which transferred chiral organization into the final nanofilament structures responsible for CPL.

Self-assembly has thus remained a cornerstone strategy in engineering CPL-active nanomaterials, with chemistry-driven approaches playing a decisive role in defining chiroptical outcomes. From ligand-mediated QDs assemblies to perovskite nanocrystal gels, from hydrogen-bonded metal nanoclusters to LC-directed helical AuNCs, the precise control of molecular interactions has continued to unlock new possibilities in chiral optoelectronics. Future research in CPL-active nanomaterials will likely focus on enhancing stability, optimizing electronic properties, and integrating self-assembled systems into device architectures, ultimately driving innovations in quantum optics, asymmetric catalysis, and next-generation photonic technologies.

Table 1. Summary of the strategies employed for the self-assembly of CPL-active nanoscale particles.

Nanoscale particle	Key strategy	Chemical interactions involved	η_{lum}
QDs	Chiral gelator mediated co-assembly ⁸⁷	Hydrogen bonding and electrostatic interactions between the carboxylate groups of the 3-mercaptopropionic acid ligands on the QD surface and the amine groups of the chiral gelator.	2.5×10^{-3}
2D Perovskites and QDs	Formation of layered structure eventually leading to energy transfer from chiral perovskite layer to QD layer ⁸⁸	Electrostatic interactions between carboxylate groups of the ligands in the perovskites with the QDs	6×10^{-3}
Perovskite nanocrystals	Chiral gelator mediated co-assembly ⁸⁹	π - π stacking and hydrogen bonding between the perovskite nanocrystal surface and the chiral gelator	$\sim 10^{-3}$
CDs	Polarity sensitive self-assembly of cholesterol-based CDs ⁹¹	van der Waals interactions between cholesterol units	$10^{-3} - 10^{-2}$
	Chiral solvent mediated self-assembly ⁹²	(i) interfacial recognition between the CD surface and the chiral solvent due to lone pair- π interactions, and (ii) intermolecular interactions among CDs leading to assembly.	$\sim 10^{-2}$
Metal nanoclusters	Metal ion and polymer induced hierarchical assembly ⁹³	Complexation reaction between ligands stabilizing Au NCs, and (ii) hydrogen bonding between –OH groups of tween-20 and Zn Au NCs.	$\sim 10^{-2}$
	Ligand exchange reaction ⁹⁴	Opposite arrangement of methyl and phenyl groups of chiral ligands stabilizing the nanoclusters	2.77×10^{-3}
	Liquid crystal mediated helical assembly ⁹⁵	Van der Waals interactions between the long alkyl chains of the ligands stabilizing the nanoclusters	$\sim 10^{-1}$

2.2.2. Template-assisted assembly of nanoscale particles endowed with CPL activity

In addition to self-assembly, template-guided fabrication provides a powerful strategy for controlling the chiral organization of nanoscale particles. This approach involves pre-structured templates, either biological, soft, or hard templates, that impose a chiral spatial arrangement on the nanoparticles. Biological templates such as DNA, proteins, or viruses act as molecular scaffolds that guide the hierarchical organization of nanoparticles into helical or twisted structures.⁹⁶⁻⁹⁹ Soft templates, including micelles and block copolymers, enable dynamic chiral organization in solution-phase synthesis, offering tunability in chiral optical properties.¹⁰⁰ Hard templates, such as mesoporous silica or chiral nanocavities, provide structural rigidity and control over nanoparticle positioning, resulting in enhanced stability of the assembled chiroptical system.¹⁰¹ In this section, we will specifically highlight strategies that have successfully facilitated the assembly of nanoscale particles through template-guided approaches (**Figure 4**). In line with the previous section, also this section will be divided according to the nature of the nanoscale particle used for the template-assisted assembly: QDs, perovskites, and CDs, while distinguishing between the template that was used for each nanoscale particle and the chemical interactions involved in making the final assemblies, as summarized in **Table 2**.

Templates for CPL-active assemblies of nanoscale particles

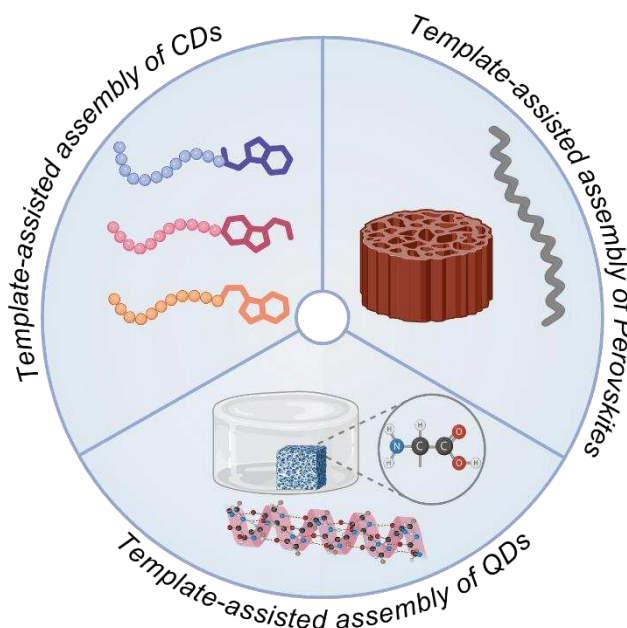


Figure 4. Schematic illustrations of the various templates used to achieve CPL active assemblies of nanoscale particles: Fmoc-peptides as templates for CDs (top left), peptide hydrogels for QDs (bottom), and silica scaffolds and nanohelices for perovskites (top right).

Template-assisted assembly of quantum dots for CPL activity - A compelling example of this strategy was demonstrated in an organic–inorganic co-assembled hydrogel system, where achiral QDs exhibited induced CPL upon encapsulation in a chiral peptide dendron hydrogel.¹⁰² The QDs, functionalized with 2-aminoethanethiol, were effectively captured by multiple

carboxyl groups in the hydrogelator N-behenic-1,5-bis(L/D-glutamic acid)-L/D-glutamic diamide (L-/D-BGAc), forming co-assembled hydrogels. The resulting structures displayed strong photoluminescence and intense CPL, with the handedness of the emission being dictated by the supramolecular chirality of the hydrogel. This study not only provided insight into chirality transfer mechanisms in hybrid hydrogel systems but also introduced a new approach for designing functional chiroptical materials with tunable CPL properties.

Another remarkable example of template-directed CPL materials was demonstrated using ferritin, an α -helix-rich rhombic dodecahedral protein, as a nanoreactor to synthesize CdS quantum dots.¹⁰³ The resulting S QDs exhibited left-handed CPL originating from both direct transitions and surface-trapping states, with relatively large g_{lum} values. Laser photoetching allowed precise modulation of the CPL properties, inducing a blue shift in the photoluminescence and CPL bands associated with surface-trapping sites while eliminating CPL from the direct transition band. This ability to control emission characteristics through templated synthesis and post-synthetic modifications opens new pathways for chiroptical memory, CPL-based light-emitting devices, and biomedical imaging using inorganic nanoparticles.

Template-assisted assembly of perovskites for CPL activity - Beyond organic templates, an inorganic-based CPL-active system was developed using a "chiral host-luminescent guest" strategy, where silica was employed as a chiral host to transfer chiroptical properties to achiral luminescent guests, including lead-halide perovskites.¹⁰⁴ Chiral silica scaffolds were chemically modified via silane coupling with amino or phenyl groups, which facilitated noncovalent interactions with luminescent guests. Upon confinement within the modified chiral silica, achiral perovskites exhibited apparent chiroptical activity in circular dichroism spectra and became CPL-active, despite lacking inherent chirality. The surface functionalization of silica played a crucial role in transferring chiral information, highlighting the significance of host-guest interactions in CPL modulation.

Further demonstrating template-assisted chiroptical induction in perovskite nanocrystals (PNCs), inorganic silica nanohelices with predefined right- or left-handed chirality were used as templates to induce optically active properties in CsPbBr₃ PNCs grafted onto their surfaces.¹⁰⁵ In solution, the PNC-functionalized nanohelices exhibited no detectable chiroptical properties. However, upon drying and film formation, the PNCs assembled into closely packed helical superstructures, resulting in significant circular dichroism and CPL signals with a dissymmetry factor of up to 6×10^{-3} . Structural investigations via grazing incidence X-ray scattering, tomography, and cryo-electron microscopy revealed a transformation from loosely organized assemblies in suspension to helically packed configurations in the solid state. Computational simulations based on the coupled dipole method demonstrated that the observed circular dichroism originated from dipolar interactions between the helically arranged PNCs and decreased with increasing interparticle distance. This study highlights the significance of spatial confinement and packing density in determining the chiroptical properties of perovskite-based CPL materials.

Template-assisted assembly of CDs for CPL activity - Chiral CDs can be used as templates for making advanced structures. This was demonstrated by guiding the supramolecular self-assembly of N-(9-fluorenylmethoxycarbonyl) (Fmoc)-protected glutamic acid, leading to the formation of chiral hydrogels with a helical nanostructure.¹⁰⁶ The co-assembled hydrogels functioned as powerful chiral templates, enhancing the luminescence g_{lum} of the CDs up to 10^{-2} . Furthermore, these hydrogels exhibited universal chiral transfer properties, inducing chirality in inserted dye molecules and enabling full-color CPL via energy transfer-mediated CPL. Energy transfer-mediated CPL was systematically explored through Förster resonance energy transfer (FRET) between achiral dyes embedded within the hydrogel matrix. Specifically, fluorescence and absorption spectral overlap among three dyes - thioflavin T (ThT), thiazole orange (TO), and pyronin Y (PY) - established the necessary criteria for sequential FRET. TO acted as a central energy relay, serving as an efficient acceptor for ThT and donor for PY. When ThT and TO were co-assembled within the hydrogel, increasing the proportion of TO led to a decrease in ThT's CPL emission at 490 nm and the emergence of a new CPL signal from TO around 550 nm, indicating effective energy transfer. Upon incorporation of the third dye, PY, a further red-shifted CPL signal at ~600 nm appeared while TO's emission diminished, confirming a two-step energy relay from ThT to TO and finally to PY. The energy transfer efficiencies reached up to 67% for the ThT/TO pair and 52% for the ThT/TO/PY triad at optimal dye ratios, affirming highly efficient chiral energy transfer within the hydrogel matrix. These results provide compelling evidence that the chiral hydrogel not only retains and amplifies the chirality of its CD-derived scaffold but also transfers this chiral information through excited-state interactions. The successful demonstration of multi-step CPL-FRET using achiral dyes highlights the hydrogel's robustness as a chiral excitation-state template and presents a promising strategy for constructing complex CPL-active systems with tunable emission profiles. Further, this CPL-active template, based on chiral CDs, expands the design strategies for chiral functionalized nanomaterials, demonstrating a versatile approach for template-guided assembly of CPL-active systems.

These studies highlight the versatility of template-guided strategies in designing CPL-active materials, demonstrating how molecular or nanoscale templates can effectively imprint chiral information onto inorganic and organic luminophores. By systematically tuning host-guest interactions, this approach offers a promising route for developing next-generation CPL materials with applications in optoelectronics, quantum optics, and chiral sensing.

Table 2. Summary of the strategies employed for template-assisted assembly of CPL active nanoscale particles.

Nanoscale particle	Template	Chemical interactions involved	ϕ_{lum}
QDs	N-behenic-1,5-bis(L/D-glutamic acid)-L/D-glutamic diamide (hydrogelator) ¹⁰²	Hydrogen bonding among carboxylate groups	$10^{-2} - 10^{-3}$
	Ferritin (α -helix-rich rhombic dodecahedral protein) ¹⁰³	Ion flow through specific channels within apoferritin subunits followed by QD formation and assembly assisted by glutamate residues	$10^{-3} - 10^{-4}$
Perovskites	Chiral silica ¹⁰⁴	Non-covalent interactions between the amino groups in modified chiral silica and achiral perovskites	2.0×10^{-3}
	Silica nanohelices ¹⁰⁵	Dipolar interactions between the helically arranged perovskite nanocrystals	6×10^{-3}
CDs	N-(9 fluorenylmethoxycarbonyl) (Fmoc)-protected glutamic acid ¹⁰⁶	Hydrogen bonding and π - π stacking between the template and the CDs	10^{-2}

2.2.3. External force-assisted chiral assembly

Beyond chemical and template-driven methods, external fields can also be employed to dynamically induce chirality in nanoscale assemblies. These external stimuli provide a means to modulate and reconfigure chiral nanostructures, offering adaptive and reversible chiroptical properties. CPL irradiation can selectively promote chiral reorganization by inducing asymmetric photochemical interactions in nanostructured films or liquid-phase colloidal systems. Magnetic fields influence the dipole-dipole interactions of magnetic nanoparticles, leading to chiral aggregation or twisting of colloidal assemblies. Electric fields can manipulate the alignment of anisotropic nanoparticles, introducing structural asymmetry and thereby enhancing CPL signals in optoelectronic materials. By leveraging these field-induced techniques, researchers can design stimuli-responsive chiral assemblies, paving the way for real-time tunable CPL devices and dynamic chiral sensors.

To this end, luminescent cholesteric LC elastomer films with mechanically tunable CPL were fabricated through LC-templated chiral self-assembly and in situ covalent cross-linking of photopolymerizable CsPbX₃ (X = Cl, Br, I) perovskite QDs nanoparticles (**Figure 5**).¹⁰⁷ The resulting films exhibited structural color in natural light and CPL emission (g_{lum} up to 1.5) under UV illumination. The CPL intensity and handedness were dynamically controlled by adjusting the helicoidal nanostructure and helical domains. Notably, a reversible CPL switch was achieved through biaxial stretching, enabling flexible anti-counterfeiting labels that revealed encrypted information under specific polarization states. This approach provided a new strategy for developing stimuli-responsive chiral perovskite materials for optical encryption and reconfigurable displays.

Further, a study on the magneto-chiral characteristics of two achiral, soluble, diamagnetic perovskite-type PbQDs demonstrated that applying magnetic fields of 158–198 mT in toluene at 25 °C produced magnetic CPL with magnitudes of approximately 10^{-3} in the 480–580 nm range.¹⁰⁸ The CPL strength increased with field intensity, and rapid, reversible switching between right- and left-handed CPL was achieved simply by reversing the magnetic poles. These findings revealed that the handedness and polarization of CPL from perovskites can be precisely and reversibly modulated by external magnetic fields, offering a promising route for dynamic control in advanced photonic applications. In this case, chirality was not intrinsic to the PbQDs but was induced through the application of an external magnetic field, which imparted a dynamic and reversible chiral bias to the otherwise achiral system.

Furthermore, electron-beam-induced CPL from chiral plasmonic nanostructures was also studied. Individual gold nanohelices were characterized using angle-resolved cathodoluminescence polarimetry within advanced analytical electron microscopy. Interestingly, a strong correlation was observed between the handedness of the nanohelices and both the dominant circularity and the directional intensity distribution of their far-field emission. Electron energy-loss spectroscopy and numerical simulations revealed that longitudinal plasmonic modes oscillating along the helical windings induced transverse dipolar modes at specific resonances, whose radiative decay generated CPL in the visible range. This

electron-beam-driven approach was shown to provide deep mechanistic insight into plasmonic chirality and to open avenues for the precise mapping, tuning, and using nanoscale CPL sources in nano-optics, metamaterials, and plasmonically enhanced chiral sensing.¹⁰⁹

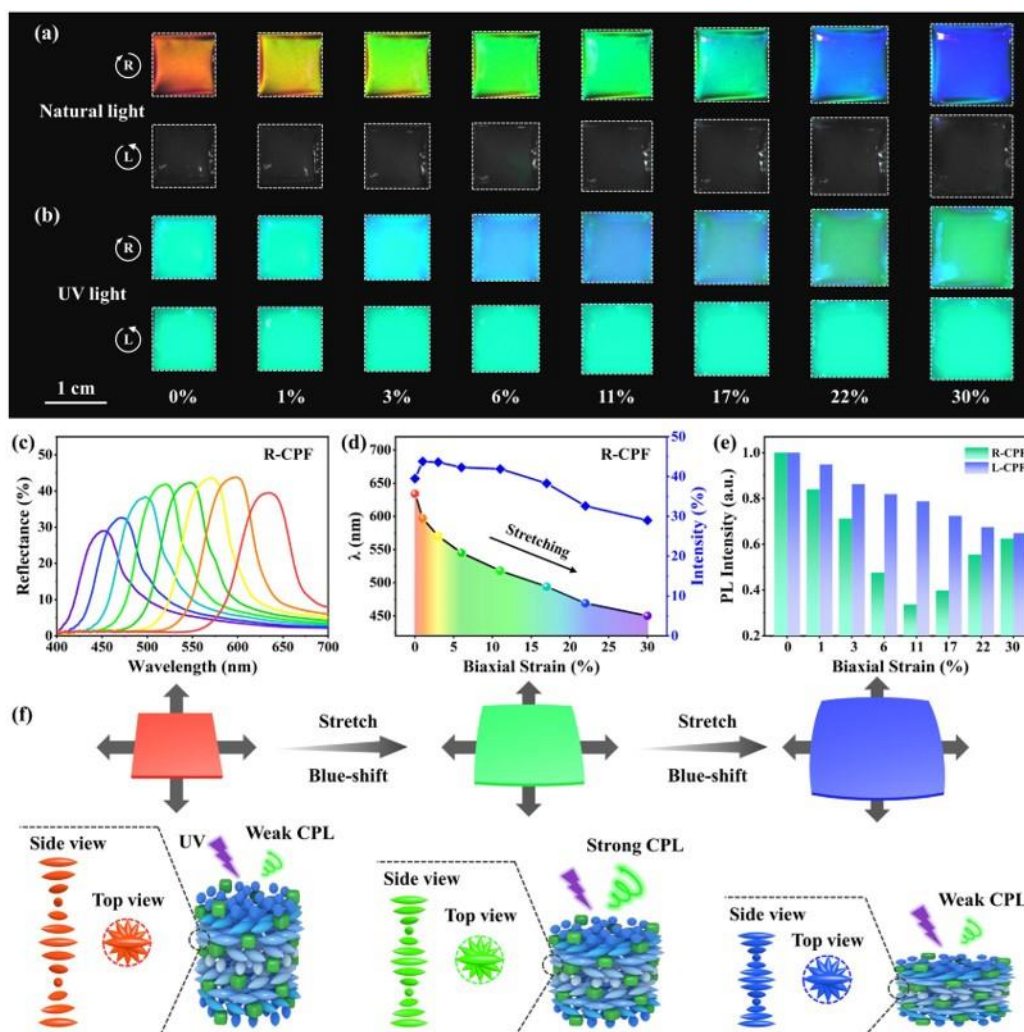


Figure 5. Digital images of self-assembled perovskite QDs under (a) natural light and (b) UV light. (c) Reflectance spectra under right-handed circular polarization across varying levels of biaxial strain. (d) Variation in peak reflection wavelength and reflectance with biaxial strain, recorded using a right-handed circular polarization filter. (e) Photoluminescence intensity trends under right- and left-handed circular polarization filters as biaxial strain changes. (f) Illustration depicting how biaxial stretching influences circularly polarized light. Reproduced with permission from ref¹⁰⁷. Copyright 2024 John Wiley & Sons.

2.2.4. Nonlinear and Nonconventional Routes to CPL active nanoassemblies

Upconverted circularly polarized luminescence (UCPL) has attracted considerable attention, as it enables the manifestation of chiroptical properties through nonlinear photophysical processes.¹¹⁰ Among these, two-photon absorption (TPA) - based UC-CPL (TP-UCPL) is particularly promising, since near-infrared excitation can induce upconverted CPL emission at shorter wavelengths. This approach offers several advantages, including reduced background autofluorescence, enhanced penetration depth, improved spatial selectivity, and tunable CPL

handedness, making it highly relevant for advanced photonic, optoelectronic, and bioimaging applications.

Recent advances highlight the versatility of this strategy. For example, the first demonstration of TP-UCPL was reported in chiral α -octylamine-modified CsPbBr₃ perovskite nanocrystals, which exhibited exceptionally large two-photon absorption cross-sections ($\sigma_2, 800\text{ nm} = 3.68 \times 10^4\text{ GM}$) and g_{lum} values up to 7.0×10^{-3} .¹¹¹ The handedness of the emission could be directly tuned by the molecular chirality of the surface ligands, yielding mirror-image CPL responses and offering a new perspective for the design of UCPL processes.

Similarly, π -extended nanographenes have emerged as efficient UC-CPL platforms. The enantiopure superhelicene nanographene, constructed from two saddle-shaped and one planar hexabenzocoronene units into an undecabenz[7]carbohelicene, represented the first fully π -extended [7]helicene. Racemic resolution enabled detailed chiroptical analysis, revealing dissymmetry factors of $\sim 2 \times 10^{-3}$ in both absorption and emission. Remarkably, nonlinear optical characterization demonstrated a two-photon absorption cross-section of 870 GM at 800 nm with excellent overlap between linear, nonlinear, and chiral emission channels.¹¹²

In another case, a distorted ribbon-shaped nanographene incorporating a saddle-shaped ketone and a [5]carbohelicene unit demonstrated a unique combination of TPA-based upconversion and CPL.¹¹³ The structural distortion induced by the edge defects endowed the system with strong chiroptical activity, while the extended π -conjugation and push-pull electronic geometry promoted efficient nonlinear absorption. This compound further displayed a long emission lifetime of 21.5 ns, as supported by electronic structure calculations.

Beyond photonic upconversion, recent progress has shown that thermal emission itself can become circularly polarized when chirality is engineered at the nanoscale.¹¹⁴ Although Planck's law does not predict such polarization, nanostructured filaments with twisted geometries (e.g., nanocarbon or metallic helices) have been shown to emit black-body radiation (BBR) with strong ellipticity across the 500–3000 nm spectral range. This effect arises because the submicrometer-scale chirality of the filaments satisfies fluctuation-dissipation constraints and enforces symmetry breaking in absorptivity and emissivity according to Kirchhoff's law. The resulting thermal CPL displayed emission anisotropy and brightness up to 10–100 times greater than conventional chiral emitters. Importantly, the helical architecture enabled precise spectral tuning of chiral BBR, while encapsulation in refractive ceramics yielded stable, durable, and highly efficient chiral light sources capable of operating at extreme temperatures.

Collectively, these advances demonstrate that both nonlinear optical excitation pathways (TPA-UCPL) and structural control of thermal emission (chiral BBR emitters) represent complementary strategies to expand the frontiers of CPL-active materials. By bridging molecular design, nanostructured geometries, and fundamental emission processes, they pave the way for highly efficient, tunable, and robust chiral light sources for next-generation optical technologies.

2.3. Application Potential of CPL-Active Nanoscale Assemblies

Till now, we have outlined the various design principles and strategies for the making of CPL-active materials using nanoscale materials. One of the primary driving forces behind the development of CPL-active nanoscale assemblies is their potential applications in photonic technologies, security encryption, optical sensing, and next-generation display systems. The ability to control and manipulate CPL at the nanoscale has led to the development of innovative materials with tunable emission wavelengths, high dissymmetry factors, and enhanced quantum yields. Various strategies, including supramolecular self-assembly, template-guided organization, and host-guest interactions, have been employed to achieve strong CPL responses, enabling the integration of these assemblies into functional optoelectronic devices. In this section, we will highlight a few groups of applications for various assemblies of CPL-active materials as a function of the nature of each structure.

A remarkable example of CPL-active assemblies involves chiral CDs synthesized from R-/S- α -phenylethylamine and 3,4,9,10-perylenetetracarboxylic dianhydride.¹¹⁵ These CDs exhibit bright, concentration-dependent luminescence with reversible morphological and spectral responses. Their aggregation into nanoribbons enhances CPL activity, achieving a dissymmetry factor (g_{lum}) of 2.18×10^{-2} at 709 nm, which is the longest CPL wavelength and highest g_{lum} reported for CDs. These unique optical properties have been harnessed for multilevel chiral logic gates in anti-counterfeiting encryption, flexible electronic printing, and solid-state CPL emission (**Figure 6a**). Furthermore, their integration into electroluminescent light-emitting diodes (LEDs) has resulted in deep-red CPL emission with an external quantum efficiency of 1.98%, marking the first demonstration of chiral electronic devices based on CDs.

Beyond carbon-based materials, hybrid photonic films have been designed to achieve dual CPL and circularly polarized room-temperature phosphorescence (**Figure 6b–c**).¹¹⁶ These films, co-assembled from cellulose nanocrystals (CNCs), poly(vinyl alcohol) (PVA), and CDs, provided a stable chiral environment for triplet excitons, enabling tunable CPL with invertible handedness and high dissymmetry factors (g_{lum} up to -0.27). By controlling the CNC/PVA ratio, photonic band gaps were tuned, leading to right-handed circularly polarized phosphorescence with extended phosphorescence lifetimes (up to 103 ms) and large phosphorescence dissymmetry factors (g_{phosp} up to -0.47 [to be distinguished from g_{lum}]). The development of patterned films with multiple polarization features further showcases their potential in optical encryption, photonic information storage, and advanced display technologies.

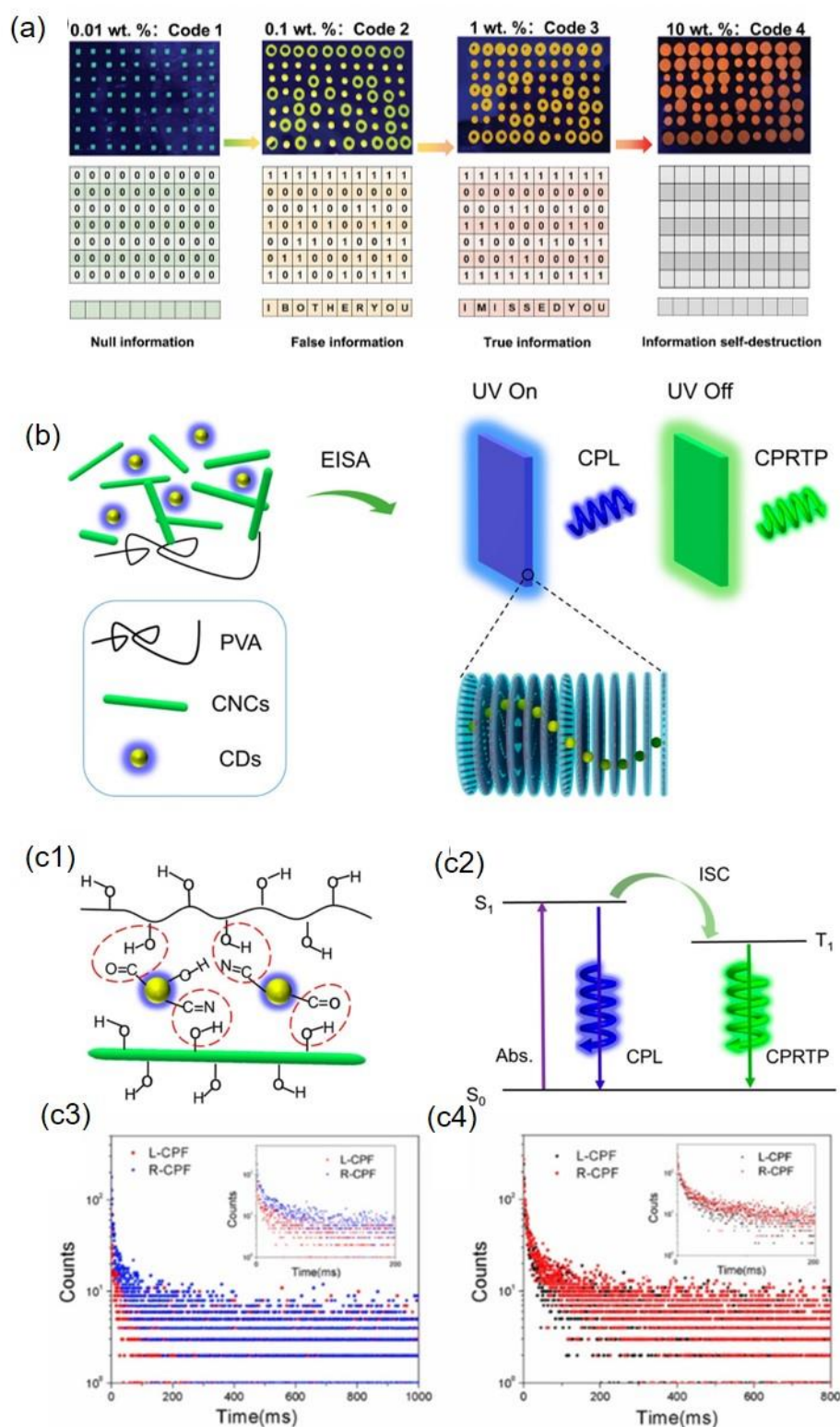


Figure 6. (a) Optical images demonstrating the dynamic information encryption and self-erasure functionality of CPL active assembly of CDs at varying concentrations. The top row displays visually encrypted data, the middle illustrates the applied coding scheme, and the bottom reveals the decoded content, highlighting the system's potential for secure data storage and responsive optical encryption. Reproduced with permission from ref¹¹⁵. Copyright 2024 John Wiley & Sons. (b) Schematic illustration of the formation of chiral photonic films composed of CNCs, PVA, and CDs through evaporation-induced self-assembly, resulting in

structures exhibiting CPL. **(c1)** Proposed chemical interactions between CNCs, PVA, and CDs leading to the formation of CPL-active hybrid structures. **(c2)** Illustration of the underlying mechanisms responsible for CPL and CP RTP in the chiral photonic films. Time-resolved phosphorescence of the hybrid chiral photonic films assembled with **(c3)** 0.01 g and **(c4)** 0.03 g PVA. Reproduced with permission from ref¹¹⁶. Copyright 2020 American Chemical Society.

In the realm of QD-based CPL-active assemblies, chiral QDs have been explored for security encryption and polarization detection.¹¹⁷ Multicolor chiral QDs were synthesized through surface oxidation tuning and phosphorus doping, achieving high fluorescence quantum yields (up to 84.63%) across near-UV to red wavelengths. By embedding these QDs into CNC matrices, CPL-active composite films were formed, exhibiting bright, multicolor CPL with exceptionally high dissymmetry factors ($g_{lum} = -0.24$ to -0.90). These films enabled the successful detection of visual polarization and the development of chiral devices for encryption, optical sensing, and next-generation display applications.

Furthermore, a series of stable CPL materials was fabricated by co-assembling high-quality, non-toxic QDs with liquid crystals using a helical self-assembly strategy.¹¹⁸ These CPL materials exhibited a high figure of merit, ensuring strong luminescence dissymmetry and quantum yield for anti-counterfeiting applications. In the designed hybrid system, two distinct contributions to the observed CPL were identified. First, the chiral organization of the inorganic quantum dots within the liquid crystal matrix was recognized as providing the structural basis for circular polarization. Second, a much larger g_{lum} was achieved when the photonic bandgap of the chiral liquid crystals was tuned to overlap with the emission peak of the quantum dots. This overlap was realized by adjusting the weight ratio of the chiral dopant to the nematic liquid crystal, through which selective reflection was optimized and photonic amplification of the circularly polarized emission was obtained. Thus, the system was shown to involve both the intrinsic contribution of nanoscale particle assembly and the extrinsic enhancement from photonic bandgap effects, which together yielded strong CPL. Multimodal-responsive security materials were developed, capable of encryption–decryption based on six different stimuli, including light activation, polarization, temperature, voltage, pressure, and viewing angle. Additionally, the CPL-active materials were integrated into fibers, enabling the fabrication of intelligent textiles with adaptive color-changing properties for enhanced security features.¹¹⁸

An additional example is the fabrication of highly chiroptical sensors via template-guided assembly. This was exemplified by gold–silver bimetallic nanoclusters (AuAgNCs) co-assembled with a helical G-quartet nanofiber structure to achieve CPL (**Figure 7**).¹¹⁹ In this system, adenosine 5'-monophosphate-templated AuAgNCs exhibited enhanced fluorescence with a quantum yield of 9.4% and a decay time of 3.8 μ s. Simultaneously, self-assembled helical G-quartet nanofibers (g-fibers) formed using guanosine 5'-monophosphate served as a chiral template for CPL emission. Notably, the handedness of CPL emission was dynamically modulated by K^+ ions, with a g_{lum} factor reaching 10^{-2} . This CPL-active assembly was leveraged as a chiroptical sensor for l-cysteine detection, exploiting the quenching effect on AuAgNCs. The sensor exhibited a detection limit of 95.7 nM over a concentration range of 0.1 to 8 μ M, demonstrating sensitivity comparable to conventional analytical techniques.

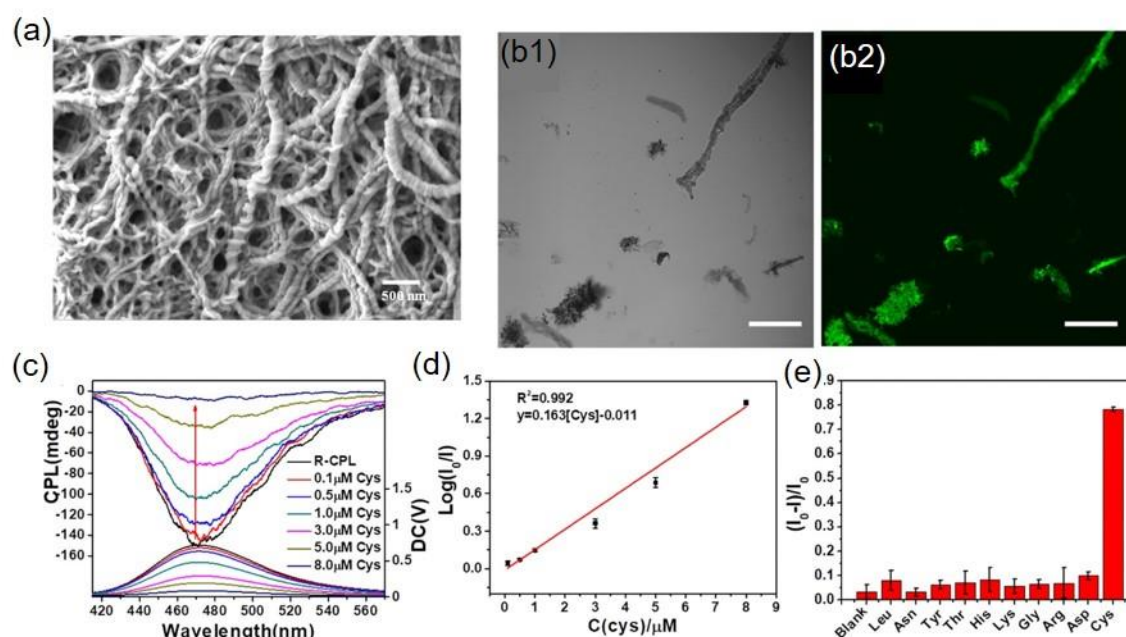


Figure 7. (a) Scanning electron microscopy (SEM) and (b1-b2) Confocal fluorescence microscopy images of AuAgNCs fibers. (c) CPL spectra of the AuAgNCs recorded at varying cysteine (Cys) concentrations ranging from 0.1 to 8 $\mu\text{mol L}^{-1}$. (b) Calibration curve illustrating the relationship between relative CPL intensity and Cys concentration. (c) Assessment of the sensor's selectivity toward Cys compared to other amino acids, demonstrating its specificity for target detection. Reproduced with permission from ref¹¹⁹. Copyright 2020 American Chemical Society.

The rapid advancements in CPL-active nanoscale assemblies underscore their immense potential for next-generation photonic and optoelectronic applications. From chiral CDs and QDs to hybrid photonic films and template-guided nanostructures, these systems have demonstrated remarkable tunability in emission properties, high dissymmetry factors, and functional adaptability. Their integration into security encryption, polarization detection, and optical sensing technologies highlights the broad scope of their practical applications. However, challenges such as improving long-term stability, scalability, and achieving even higher g_{lum} values remain critical for their widespread adoption. Moving forward, interdisciplinary efforts in nanomaterial synthesis, self-assembly strategies, and device integration will be essential to fully harness the capabilities of CPL-active nanoscale assemblies, paving the way for innovative solutions in advanced display systems, bioimaging, and quantum optics.

2.4. Challenges and future outlook

The field of CPL from nanoscale assemblies has seen remarkable progress, yet several challenges must be addressed to fully harness their potential in optoelectronics, quantum optics, and biomedical applications. The g_{lum} achievable in nanoscale particles and their assemblies is limited by a combination of intrinsic material properties and structural considerations. In most nanoscale emitters, the optical transition dipole moments couple only weakly to their chiral environment, yielding inherently low g_{lum} values, typically between 10^{-5} and 10^{-2} . This intrinsic limitation is generally inherited by assemblies formed from such nanoscale particles,

as the weak chiroptical response of the individual building blocks imposes an upper bound on the overall dissymmetry achievable. Furthermore, in chiral nanoassemblies, the presence of both left- and right-handed domains can lead to partial cancellation of CPL signals, thereby reducing the overall g_{lum} . Additional factors such as size and shape polydispersity, variations in assembly order, and random emitter orientations in bulk measurements also attenuate the measured dissymmetry. In the context of enhancing g_{lum} in chiral assemblies of nanoscale particles, one possible strategy is the suppression of racemate formation. As stated above, the fabrication of chiral assemblies often results in racemic mixtures containing both right- and left-handed CPL-active assemblies, whose opposing contributions partially or completely cancel each other, thereby reducing the observed g_{lum} . To address this, purifying the chiral assemblies or introducing a chiral bias during synthesis to preferentially form one enantiomer over the other can substantially increase the net g_{lum} by maximizing the contribution of the dominant enantiomer. Furthermore, assembling nanoscale particles with narrow size dispersion helps preserve coherent coupling between individual units and ensures better alignment of their optical resonances across the ensemble. Such uniformity minimizes inhomogeneous broadening, allowing the chiroptical response to add constructively rather than being averaged out, thereby supporting higher and more reproducible g_{lum} values. One of the key obstacles lies in understanding and controlling chirality transfer within these assemblies. While host-guest interactions, solvent effects, and molecular packing have been utilized to induce CPL, the precise mechanisms governing chiral propagation, particularly in inorganic and hybrid materials, remain elusive. Developing predictive models using quantum mechanical simulations and machine learning could provide valuable design principles for optimizing chiroptical properties. Another critical challenge is the low g_{lum} observed in most current systems, often ranging from 10^{-5} to 10^{-2} , which limits their practical application. Strategies such as enhancing excitonic coupling, tuning ligand chemistry, and leveraging plasmonic interactions have been explored to amplify chiroptical responses, but achieving a balance between optical efficiency, stability, and scalability remains an ongoing challenge. Colloidal stability is another primary concern, as many CPL-active assemblies, especially organic and hybrid systems, suffer aggregation and structural degradation under ambient conditions. The development of crosslinked supramolecular architectures, robust inorganic CPL materials based on stronger chemical forces compared to non-covalent interactions, and stimuli-responsive assemblies could enhance long-term stability and tunability. Furthermore, despite numerous proof-of-concept studies, integrating CPL assemblies into functional devices remains a bottleneck due to challenges in thin-film deposition, aggregation-induced quenching, and the need for scalable synthesis. Advancements in printing techniques, microfluidic-assisted assembly, and hybrid material processing could enable the development of circularly polarized LEDs, chiral photodetectors, and optical encryption systems. Looking ahead, expanding the scope of CPL materials beyond conventional organic and inorganic systems could open new possibilities. Emerging platforms such as MXenes, metal-organic frameworks, and covalent organic frameworks offer exciting prospects for designing robust CPL-active structures. Additionally, dynamic and switchable CPL systems incorporating photochromic, thermochromic, and mechanochromic elements could enable real-time control over optical chirality, which is highly desirable for applications in information storage, reconfigurable optics, and security features. The integration of CPL-active assemblies into biomedical

imaging, chiral drug delivery, and optogenetics remains largely unexplored, presenting an opportunity to develop next-generation bio-responsive chiral nanomaterials. Addressing these challenges will require a multidisciplinary approach, combining materials chemistry, supramolecular design, computational modeling, and device engineering to create CPL-active materials with enhanced performance and real-world applicability. By overcoming these limitations, nanoscale CPL assemblies could revolutionize quantum technologies, bioimaging, and advanced optical display systems, paving the way for innovative breakthroughs in chiral photonics.

2.5. Conclusion

The field of CPL-active materials has unlocked new possibilities in photonic technologies, bioimaging, and chiral optoelectronics. Yet, its full potential remains constrained by the limitations of conventional chiral luminophores: low dissymmetry factors, poor stability, and restricted tunability. To address these challenges, the assembly of nanoscale particles has emerged as a powerful strategy, leveraging supramolecular self-assembly, template-guided organization, and host-guest interactions to enhance CPL performance. In this review, we explored how the rational design of nanoscale architectures can amplify CPL signals, improve structural stability, and introduce tunability across emission wavelengths. By shifting the focus from individual luminophores to their hierarchical assemblies, we highlighted innovative approaches that push the boundaries of CPL-active materials. Additionally, we examined emerging trends, ranging from hybrid photonic systems to bioinspired designs, and emphasized their transformative potential in next-generation optoelectronics and sensing technologies. As the field advances, refining our understanding of nanoscale chiral interactions and optimizing CPL-active assemblies will be key to unlocking materials with unprecedented efficiency and functionality. With continued innovation, CPL-driven nanostructures can be integrated into various applications, including quantum computing, encrypted optical data storage, and real-time biological imaging.

Acknowledgements

SB thanks the Department of Atomic Energy (project no.: VS8/SIN/414), government of India, and Anusandhan National Research Foundation (Grant number: ANRF/ECRG/2024/000340/CS) for financial support.

Conflict of Interest

The authors declare no conflict of interest.

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