We Glow Together: A Dialogue on Luminescent Compounds

espite the fact that the term "luminescence" was coined as early as 1888, this fascinating phenomenon continues to underpin tremendous research in areas such as lightemitting diodes (LEDs), phosphors, thermometry, thermoluminescence dating, anticounterfeiting, sensing, photodynamic therapy, and the nondisruptive observation of processes within a cell. It is no surprise therefore that research in this area is a major theme in both Crystal Growth & Design (CG&D) and Inorganic Chemistry (IC), as well as throughout the American Chemical Society journal portfolio. Between them, IC and CG&D published more than 500 articles in the general area of luminescence and luminescent materials in 2022 alone. As a result, we felt it appropriate to collect together some of the most intriguing examples of that work in a joint virtual issue. Expertise in this area is well represented on the journals' Editorial and Advisory Boards, and so as Editors-in-Chief we teamed up with Katja Heinze, Oliver Wenger, and Kenneth Lo from IC and Hai-Bo Yang, Shalini Singh, and Shung-Quan Zang from the CG&D side. Between the eight of us, we nominated our top picks of fascinating contributions in the luminescence arena from 2022, each of us focusing on a particular aspect of the topic. In the sections below, each member of our team gives their perspective on the subject and highlights the articles they have selected, taking us from luminescence phenomena in complexes, macrocycles, and cages via corresponding observations for metal clusters, nanocrystals, and quantum dots to organic luminescent materials.

MOLECULAR COMPLEX ES INVOLVING EARTH-ABUNDANT METALS WITH PHYSICAL PROPERTY SENSING APPLICATIONS: TEMPERATURE AND PRESSURE

Luminescent transition-metal complexes based on Earthabundant metal ions have gained tremendous interest—both for reasons of sustainability and of tackling fundamental scientific challenges. In particular, dⁿ valence electron configurations beyond the classical low-spin d⁶ and d⁸ electron configurations have emerged as new options. Naturally, the d¹⁰ electron configuration, lacking detrimental ligand field excited states, is most prominent, including copper(I) and also zinc(II) complexes. Exploitation of the thermally activated delayed fluorescence (TADF) phenomenon in copper(I) metal-to-ligand and ligand-to-ligand charge-transfer (MLCT, LLCT) emitters has been extremely successful to improve quantum yields and radiative rates (DOI: 10.1021/acs.inorg-chem.1c03371). Replacement of pyridines by phosphinines in pseudotetrahedral copper(I) complexes changes the emission

energy and the luminescence lifetime (DOI: 10.1021/acs.inorgchem.2c01145). Similarly, substituents at the Cp^R ligand in cyclic alkyl(amino)carbenes (CAAC) copper(I) half-sandwich complexes modulate the emission color from blue to orange (DOI: 10.1021/acs.inorgchem.2c02073), while negatively charged sulfonate groups impart water-solubility to phenanthroline copper(I) complexes (DOI: 10.1021/acs.inorgchem.2c02648). Intriguingly, long-range inductive effects can modulate the MLCT luminescence lifetime in homoleptic copper(I) complexes (DOI: 10.1021/acs.inorgchem.2c04315). Conversely to the d^{10} MLCT/LLCT emitters, a tetranuclear sulfido copper(I) cluster displays intense and long-lived phosphorescence from a ligand-to-metal charge-transfer (LMCT) state $[S^{2-} \rightarrow Cu_4]$ mixed with some metal-centered (ds/dp) character (DOI: 10.1021/acs.inorgchem.2c01707).

Moving over to zinc, pseudo-octahedral zinc(II) complexes of benzannulated N^N^O pincer-type ligands show long-lived green phosphorescence in the solid state (DOI: 10.1021/acs.inorgchem.2c02585), while tricoordinate zinc(II) carbene complexes carrying dianionic 1,2-benzene dithiolato or 1,2-benzene diolato ligands emit from yellow to the near-infrared (NIR) via TADF mechanisms according to quantum chemical calculations (DOI: 10.1021/acs.inorgchem.2c03301) and terpyridine zinc(II) complexes [Zn(tpy)₂]²⁺ with carbazole-type counteranions can form intracomplex donor/acceptor pairs showing TADF (DOI: 10.1021/acs.inorgchem.2c03804).

Complementary to the d¹0 electron configuration, the d⁰ configuration as found in Cp₂Ti¹V(CCAr)₂ allows for LMCT emission (DOI: 10.1021/acs.inorgchem.2c01746). Substituents at the remote ligand framework of scorpionate low-spin d⁵ iron(III) complexes retain their favorable ²LMCT luminescence characteristics (DOI: 10.1021/acs.inorgchem.2c02410). The geometry and electronic structure of potentially luminescent ²LMCT states of low-spin d⁵ iron(III) complexes with cyclometallating tridentate chelate ligands have been unveiled computationally (DOI: 10.1021/acs.inorgchem.2c04407). Beyond CT luminescence, spin-flip luminescence as shown by d³-chromium(III) complexes has been an increasingly deeply investigated topic, complementing [Ru-(bpy)₃]²+ photophysics and applications (DOI: 10.1021/jacs.1c05971). This includes the construction of binuclear

nonsymmetrical chromium(III) complexes showing intramolecular energy transfer (DOI: 10.1021/acs.inorg-chem.2c01940). Spin-flip emission from pseudo-octahedral d⁸ nickel(II) complexes on the other hand is much more challenging and has not yet been achieved (DOI: 10.1021/acs.inorgchem.3c00779). Beyond the luminescent complexes with abundant 3d elements, comparably abundant molybdenum(0) with a 4d⁶ electron configuration has been successfully developed into luminescent and photoactive materials (DOI: 10.1021/jacs.0c12805).

MOLECULAR COMPLEX ES INVOLVING PRECIOUS METALS

Molecular complexes based on precious metals with d⁶ and d⁸ valence electron configurations have remained popular, in particular with iridium(III), platinum(II), and gold(III). But new trends emerge. Dimer complexes and excimers, instead of the more frequently explored mononuclear compounds, are regaining interest, for example in work on platinum(II). Cooperative effects emerging from the close interaction between two metal complexes can lead to new properties not seen in mononuclear compounds (DOI: 10.1021/acs.inorgchem.2c02662), and the light-initiated formation of metalmetal bonds and associated coherence phenomena in platinum(II) dimers now attract attention (DOI: 10.1021/ acs.inorgchem.1c02469). With mononuclear complexes, a potentially attractive research direction involves the exploration of metals in less common oxidation states and coordination geometries, such as the investigation of octahedral platinum(IV) compounds instead of square-planar platinum(II) (DOI: 10.1021/acs.inorgchem.2c04116). Similarly, rhenium(I) is part of well-known luminophores with MLCT excited states, and the search for luminescent LMCT excited states looks very attractive (DOI: 10.1021/acs.inorgchem.2c03193). With well-established iridium(III) emitters, high-throughput synthesis and luminescence-based screening approaches now permit rapid identification of effective photocatalysts (DOI: 10.1021/jacs.1c12059).

MOLECULAR COMPLEXES ON THE BIOLOGICAL SIDE: APPLICATIONS AS SENSORS AND FOR PROTEIN BINDING

Luminescent transition-metal complexes have emerged as promising candidates for theranostic agents with "two-inone" capabilities: therapeutic and diagnostic applications. These complexes possess attractive photophysical and photochemical properties, including large Stokes shifts, long emission lifetimes, high photostability, and efficient photosensitization of reactive oxygen species. The large Stokes shifts minimize possible self-quenching effects, whereas the long emission lifetimes allow increased detection selectivity when using time-resolved spectroscopic and imaging techniques. The high photostability enables continuous exposure of the complexes to irradiation, allowing real-time monitoring of biomolecules and biological events at higher resolution with super-resolution microscopy. Many transition-metal complexes exhibit high singlet oxygen-photosensitization efficiencies, demonstrating excellent use as photodynamic therapeutics.

DNA is a vital molecule that plays an essential role in many important biological processes and has been a major molecular target for both imaging and therapy. For example, osmium(II) complexes comprising extended intercalating ligands show

effective DNA binding, which turns on their emission in the NIR region and thus enables cellular imaging (DOI: 10.1021/acs.inorgchem.2c01231). Bichromophoric ruthenium(II) complexes containing one or two nitro-1,8-naphthalimide moieties have also been reported to display strong DNA binding affinities coupled with interesting light-switch properties (DOI: 10.1021/acs.inorgchem.2c00064). Integration of a ruthenium(II)-based DNA light switch with a G-quadruplex-selective ligand yields a theranostic complex; this complex exhibits high affinity and specificity for G-quadruplex DNA and disrupts the quadruplex structure upon binding (DOI: 10.1021/acs.inorgchem.2c03903).

Current research focuses on the design of metal complexes with improved therapeutic properties for clinical translation. One major area of research involves the design of complexes with excitation and emission at longer wavelengths, which minimizes phototoxicity and allows compatibility for use in the biological window. For example, a dinuclear ruthenium(II) complex coordinated with a 2,7-bisbenzoimidazolyl-naphthyridine ligand has been reported for use in NIR imaging and for treating platinum-resistant cancer (DOI: 10.1021/acs.inorgchem.2c00714). Similarly, an oxygen-responsive iridium(III) complex featuring far-red emission with a long emission lifetime and high singlet oxygen photosensitization efficiency for oxygen sensing in vitro and in vivo, and for efficient photodynamic therapy in cancer cells, has also been reported (DOI: 10.1021/acs.inorgchem.2c02854). New strategies have been proposed for effective manipulation of the excited-state properties of iridium(III) complexes, resulting in emission colors ranging from yellow to deep red with high emission quantum yields (DOI: 10.1021/acs.inorgchem.2c03597).

Furthermore, the development of luminescent nonprecious transition-metal complexes for various biological applications has become an area of increasing interest due to their high abundance, low cost, and good biocompatibility. For example, cobalt(III) complexes coordinated with acetylacetonate-linked boron-dipyrromethene ligands have been developed as bioimaging and phototherapeutic agents (DOI: 10.1021/acs.inorgchem.2c00150). The successful development of highly luminescent binuclear neutral manganese(II) complexes as scintillators for X-ray imaging also highlights their potential in imaging applications (DOI: 10.1021/acs.inorgchem.3c00273).

Despite the promising applicability of luminescent transition-metal complexes, there are still challenges that need to be overcome to further amplify their diagnostic and therapeutic capabilities while minimizing off-target effects. By incorporating specific functional groups into these complexes, we can enhance their targeting capabilities, enabling them to be directed toward specific cell types and subcellular organelles. This targeting property paves the way for more accurate diagnosis and improved treatment outcomes. The clinical potential of these multifunctional complexes as theranostic agents is indeed exciting. It is expected that continued innovative designs will spearhead medical advancements, contributing to enhanced health and quality of life.

MACROCYCLES AND MICROCAGES WITH APPLICATIONS IN CATALYSIS

Luminous materials utilize signal changes of their optical properties to detect, modulate, and convert energy or frequency signals under external fields, such as light, electricity, heat, force, and magnetism. Coordination-driven self-assembly is an example of a powerful strategy for constructing discrete

two-dimensional metallacycles and three-dimensional metallacages with well-defined sizes and shapes, which enables the transformation of different components into predesigned assemblies. The realization of specific functions of these assemblies is an important driving force for self-assembly research. Supramolecular metal-organic assemblies have received extensive attention and have achieved breakthroughs in many fields because of their flexible designability and controllability. Among them, luminescent metallacycles and metallacages have been attracting research interest for their applications in photocatalysis, sensing, biological applications, renewable energy, and more. Examples include sensitive detection of picric acid (DOI: 10.1021/acs.inorgchem.1c03527), cations (DOI: 10.1021/acs.inorgchem.3c00437 and DOI: 10.1021/acs.cgd.1c00895), mechanical force (DOI: 10.1021/acs.inorgchem.1c03563 and DOI: 10.1021/acs.inorgchem.1c00233), and temperature (DOI: 10.1021/acs.inorgchem.2c02718), achieving efficient photocatalysis (DOI: 10.1021/acs.inorgchem.2c01869 and DOI: 10.1021/acs.cgd.0c01375) and host-guest loading (DOI: 10.1021/acs.inorgchem.2c02061). Nowadays, four main methods have been developed to construct luminescent metallacycles or metallacages: (1) Using fluorescent elements as a skeleton; (2) introducing luminescent primitives into the assembly by precovalent modification; (3) wrapping fluorescent elements through host and guest interaction; (4) using luminous metals such as ruthenium, iridium, rhodium, and europium.

METAL CLUSTERS FOR CATALYSIS, OPTICAL APPLICATIONS, AND MEDICAL DIAGNOSTICS

Metal clusters, which consist of three or more metal atoms or ions forming a skeletal structure, often exhibit fascinating properties that surpass those of monomeric metal complexes. The clusters often have structural diversity and exhibit unique and tunable luminescent properties, making them valuable for applications in areas such as optoelectronics, sensing, imaging, and light-emitting devices.

The eye-opening core—shell crystals that incorporate binary or ternary clusters including Ni₉W₆, Co₉W₆ Ni_xCo_{9-x}W₆, and Mn₉W₆ via epitaxial growth represent a great advance in cluster-based heterostructure through a controllable synthesis (DOI: 10.1021/acs.cgd.2c00216). The synthesis methods can tune structural diversity, for example with silver clusters, although they are coprotected by the same diphosphine and thiol ligands, and can exhibit interesting luminescence (DOI: 10.1021/acs.cgd.2c00744).

As excellent candidate phosphors, the cuprous iodide cluster with polytropic configurations and responsive luminescence, including solvato-, thermo-, and mechanochromism (DOI: 10.1021/acs.inorgchem.1c03876) furthermore showed photocatalytic activity in azide—alkyne cycloaddition reactions (DOI: 10.1021/acs.cgd.2c00476). Crystalline hybrid zinc complexes often emit different colors (DOI: 10.1021/acs.cgd.2c00117), including even in the near-infrared range, and the photoluminescence color and quantum yield of the rare trinuclear Zn(II) cluster depend on their substituents (DOI: 10.1021/acs.cgd.1c01319).

The different packings of Re(I) clusters emit phosphorescence originating from the electrostatic interactions between the charge distributions on neighboring cluster molecules inside the crystal, providing a new and powerful method to tune emission energy (DOI: 10.1021/acs.cgd.1c01278). In

addition, heterometallic clusters, such as Au-Cu and Au-Ag (DOI: 10.1021/acs.inorgchem.2c01474), Ln-Ag (DOI: 10.1021/acs.inorgchem.2c02991), and Cd-Tb clusters (DOI: 10.1021/acs.inorgchem.2c00393) possess the additional features of a synergistic effect between different metal atoms that can modulate emission, resulting in more intriguing luminescent properties.

(MULTI-)METALLIC CLUSTERS AND NETWORK COMPOUNDS

Luminescence is a key property of many inorganic compounds—across all length scales. Regarding clusters and extended networks, many of them involve 4f metal ions, which bring in their luminescent properties through their inherent 4f-5d and 4f-4f electronic transitions, similar to those observed in extended solids that incorporate lanthanide metal ions. This has been realized in lanthanide-doped polyoxometalates (DOI: 10.1021/acs.inorgchem.1c03073) or in lanthanide-based metal-organic frameworks (DOI: 10.1021/acs.inorgchem.1c03661), both of which can be used for (main group) metal-ion sensing or bioimaging (DOI: 10.1021/cr900362e). Similar luminescent metal-organic framework compounds were also reported without 4f metal ions, such as (chiral) zirconium-based materials, which serve to detect and recognize tryptophan enantiomers (DOI: 10.1021/acs.inorgchem.2c00991). In other cases, lanthanide ions perform in concert with transition-metal ions, such as in luminescent lanthanide-titanium oxo clusters (DOI: 10.1021/acs.inorgchem.2c01411) or in Ln-Zn clusters in which emissive properties are additionally combined with magnetism (DOI: 10.1021/acs.inorgchem.1c03359).

While 4f metal ions play a predominant role in emissive materials, there are cluster and network compounds that also exhibit luminescent properties owing to a specific interplay between the involved atoms. One leading group of emissive polynuclear aggregates involves coinage metal atoms (DOI: 10.1021/acs.inorgchem.2c01474 and DOI: 10.1021/acs.inorgchem.2c00366), which can also assemble to form luminescence sensors, for example for metal ions (DOI: 10.1021/acs.inorgchem.1c03049) or organic acids (DOI: 10.1021/acs.inorgchem.1c03527).

The development of materials for photovoltaics naturally leads to the discovery of luminescent compounds. This includes compounds comprising polynuclear complexes, for instance, halido metalate clusters of transition-metal ions (DOI: 10.1021/acs.inorgchem.2c01581 and DOI: 10.1021/acs.inorgchem.2c00169). In such studies, lifetimes, emission efficiencies, and bandwidths are particularly important. Last but not least, clusters based on main-group atoms can be luminescent. This is well-known from basic chemistry classes when it comes to the famous detection of tin (as Sn²⁺) by a bright blue emission from HCl solution. More complex tin compounds can show long-lived photoluminescence (DOI: 10.1021/acs.inorgchem.2c00182).

NANOCRYSTALS AND QUANTUM DOTS WITH APPLICATIONS IN DISPLAYS AND DRUG DELIVERY

Within the topic of nanostructured luminescent inorganic semiconductor materials, the materials represented in this issue range from classical CdSe quantum dots to III—V nanocrystals, lanthanide doped up-conversion nanocrystals, and advanced

perovskite crystals. In view of optoelectronic applications, tuning the properties of excitons and charge carriers by crystal engineering has always been a significant area of research. For instance, introducing ns² metal ions in an inorganic halide perovskite matrix can lead to improved emission efficiency, such as a facile solvothermal approach to design indium-based double perovskite crystals (DOI: 10.1021/acs.inorg-chem.1c03176). Further, the emission efficiency in these crystals was greatly boosted by breaking the parity-forbidden transition through antimony doping. In the same vein, centimeter-sized sodium-doped lead halide perovskite crystals have been reported, with enhanced emission owing to self-trapped exciton emission (DOI: 10.1021/acs.cgd.2c00526).

Furthermore, in perovskite research, a well-known toxicity problem as a barrier in the synthesis and processing of lead halide perovskite nanocrystals for industrial scale applications has been addressed. The result is environmentally benign syntheses of self-healed and printable CsPbBr₃ nanocrystals in polar nontoxic solvents with long-term stability and high photoluminescence efficiency (DOI: 10.1021/acs.inorg-chem.2c01113).

Beside perovskite nanocrystals, InP quantum dots have emerged as important bright and stable cadmium-free emitters for display and LED technologies. However, the conventional synthesis approaches cause defects due to surface oxidation, which introduces nonradiative recombination pathways that quench the bright photoluminescence. In this regard, InP/ZnSe core—shell quantum dots with metal oxide interfaces via atomic-layer deposition techniques have been designed (DOI: 10.1021/acs.inorgchem.3c00161). This interface engineering resulted in increased photoluminescence and narrowing of the emission line widths of the quantum dots. This research paves a new direction toward leveraging the structural complexity of colloidal nanocrystals for optoelectronic properties enhancement

Recent advances in lanthanide ion-doped upconversion luminescence materials are also highlighted. The impact of judicious doping of lanthanide ions in alkaline-earth metal silicate fluorides, oxides, and scandate phosphors has been demonstrated, which can dramatically improve the emission properties and temperature sensing behavior for anticounterfeiting, white LED, and optical thermometry applications (DOI: 10.1021/acs.inorgchem.1c03672 and DOI: 10.1021/acs.inorgchem.1c03190). Moreover, the synthesis of a series of redemitting Eu³⁺ doped oxide phosphors and their application in plant growth has been reported (DOI: 10.1021/acs.inorgchem.1c02836).

ORGANIC SUPRAMOLECULAR LUMINESCENT MATERIALS

Metals, be they transition-metal complexes, lanthanides, or metallic nanoparticles underpin a great deal of the research in luminescence. As a journal that encompasses both inorganic and organic systems, CG&D highlights some less-common examples of luminescent organic, hybrid, or supramolecular materials, particularly those in which emissive properties depend on the crystal environment or changes in solid-state conditions.

In the case of a donor- π -acceptor dye based on pyrene-pyrazole-aldehyde, there is a marked correlation of structure and emissive properties as a function of pressure as the balance between C-H···O hydrogen bonds, C-H··· π interactions, and

antiparallel $\pi-\pi$ stacking changes all the way up to 6 GPa. The shift is even accompanied by a visible color change (DOI: 10.1021/acs.cgd.2c01106). Adding an aromatic chromophore can also work well in hybrid materials. A report of a hybrid perovskite 4,4'-(anthracene-9,10-diylbis[ethyne-2,1-diyl])bis-(1-methyl-1-pyridinium) bismuth iodide (AEPyBiI) with a particularly large and excitable organic cation is a nice illustration of the approach. Its intense emission is an order of magnitude larger than high quality InP epilayers, and with its lead-free composition the material is aimed at application as a component of environmentally friendly light-emitting devices (DOI: 10.1021/acs.cgd.2c01005).

The solid-state environment can have a considerable role in modifying emissive properties. The red fluorescence of an octahydropyridoquinoline dicarboxylate is markedly enhanced in the solid state. This versatile compound is elastically bendable, and the emission responds to acid/base vapor and copper(II) (DOI: 10.1021/acs.cgd.2c00045). Multistimuli response materials of this type are particularly topical. Charge-transfer cocrystals of 4,4′-bis(9*H*-carbazol-9-yl)-1,1′-biphenyl (CBP) with various acceptors emit upon applying pressure to a mixture of a donor and acceptor, and the emission color gradually changes according to pressure covering the whole visible region. The materials have been used to make anticounterfeiting paper (DOI: 10.1021/acs.cgd.1c01208).

Mechanical properties such as elasticity in crystals continue to be highly topical, and it is remarkable how much mechanical mistreatment some organic solids can handle without losing crystallinity. Such materials are of interest in progressing toward flexible lightweight single-crystal optoelectronics. A Schiff base derivative forms brittle, plastic, and elastic crystals and undergoes transformation from elastic to plastic on desolvation, all signaled by luminescence changes (DOI: 10.1021/acs.cgd.2c00225). Similarly, the luminescence of thienylbenzothiadiazole derivatives is dependent on both mechanical and thermal responses, showing a stepwise switching of their near-infrared emission—that is, mechanochromism (DOI: 10.1021/acs.cgd.1c01128). Mechanochromism is also exhibited by the remarkable Form II of flufenamic acid, which shows bending and twisting behavior that in turn gives it mechanofluorochromism as the emission moves to higher energy ion bending (DOI: 10.1021/acs.cgd.1c01271).

Luminescent extended framework materials are particularly popular in sensing, and highlighted here is a luminescent uranyl sensor based on two-dimensional nanosheets of a hydrogen-bonded framework involving the classic eightmembered carboxylic acid ring supramolecular synthon (DOI: 10.1021/acs.cgd.2c01335). Perhaps one of the most interesting aspects of luminescence is the phenomenon of aggregation-induced emission (AIE) in which a nonluminescent compound becomes emissive because the solid state restricts deactivating conformational motion. An especially impressive example is a mechanochemical reaction involving norborn-2-en-7-one that releases CO accompanied by bright blue AIE giving a turn-on fluorescence readout when the reaction occurs (DOI: 10.1021/jacs.1c12108).

From all the wonderful articles that were published in *Inorganic Chemistry* and *Crystal Growth & Design* in this context, we can only highlight a few here—knowing that this collection is far from comprehensive. The selected articles are meant to illustrate the spectrum of different compounds featuring luminescence and to demonstrate that synergistic

theoretical and experimental studies on luminescent materials give us an opportunity to tune their optoelectronic properties. We wish everyone an enjoyable and inspiring read!

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