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Are Commonplace Chiral Metal Complexes Unsuitable for Metamaterials?

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Abstract

Among typical metamaterials, we focus on the light "function" and chiral "element" in this chiral light metamaterial minireview. The authors are interested in inorganic materials, especially transition metal complexes. Metal complexes are compounds that have been studied for a long time for their optical properties (light absorption) and chirality (chiroptical spectroscopy, structural crystal chemistry, and asymmetric catalytic reactions). However, since the unit is a "molecule", few examples as (optical) metamaterials have been reported to date. Is it possible to add a function as a composite material using one method? No, the functions resulting from the nano pattern are very difficult to retrofit. Is there a way to fuse the rich compounds at the boundary between inorganic and organic with state-of-the-art optical metamaterials?

Keywords: Metamaterials, Optical materials, Hybrid materials, Chirality, Metal complexes.

Abbreviations: MOFs-Metal Organic Frameworks, TMDs-Transition Metal Dichalcogenides, CMs-Chiral Metamaterials, CD-Circular Dichroism, PT- Parity Time, RCP- Right Circularly Polarized, LCP- Right Circularly Polarized, SEMWs-Surface Electromagnetic Waves.

Introduction of Optical Metamaterials

We must apologize first. To tell the truth, we have no idea why we (conventional inorganic coordination chemists who are interested in chirality and optical spectroscopy) were invited to this Special Issue on the latest topics such as "optical metamaterials". We thought that such new topics of nanomaterials based on physics should involve conventional fields related to metal complexes such as organic–inorganic hybrid materials in molecular chemistry. Therefore, we composed this review article as a non-specialist literature survey, and we contrasted its relevance with our background research areas established classically. At the early stage of optical metamaterial research, Shalaev played an important role in presenting prospects and introducing reviews. With the advent of metamaterials, the spatial distribution of permittivity ε and permeability μ can be adjusted, and the flow of electrical energy can be controlled. This makes it possible to control light, which is not possible with materials that exist in nature.

In general, light propagates such that the optical path given by the product of physical length and refractive index is minimized; thus, by giving a complex distribution to the refractive index n, the optical path can be made almost arbitrary in complexity. You are then able to bend it; by creating the desired distribution of ε and μ, i.e., the distribution of the refractive index n, the space of light can be "curved" almost arbitrarily, such that the light is not only in the opposite direction (when n is negative), but also almost arbitrarily. It can then be propagated along all curves. Conversion optics using metamaterials are considered to be revolutionary materials in the field of photochemistry, enabling many exciting applications that were un-imaginable until recently [1].

Metamaterials, which are reasonably designed artificial materials, have been found to exhibit unprecedented electromagnetic properties not found in naturally occurring substances. The reason is that the two components existing in light, the electric field and the next component, can be bonded to the meta-atom, and this property enables completely new optical properties and exciting applications that never existed before. The unique electromagnetic properties provided by metamaterials have attracted a great deal of attention from various fields such as physics, material science, engineering, and chemistry. This encourages the combination of different disciplines, which is thought to have led to tremendous progress in metamaterial research. One of the attractive properties of metamaterials is their negative index of refraction. Refraction is considered to be one of the basic characteristics of how light propagates in a substance. The use of metamaterials with a negative index of refraction may lead to the development of a super-lens that can capture objects and fine structures that are much smaller than the wavelength of light. In addition, there are many possible applications using metamaterials that can artificially adjust light, such as antennas with excellent characteristics, optical nanolithography, nanocircuits, and meta-coatings that obscure objects. In addition, we propose the future of research involving metamaterials that still have a lot of room for development, such as 3D optical metamaterials, nonlinear metamaterials, and "quantum" perspectives of metamaterials. Expectations are also gathered for a typical outlook [2,3].

Inorganic Materials and Metamaterials

There are rich compounds at the boundary between inorganic and organic considered state-of-the-art metamaterials, namely, metal complexes and inorganic solid materials of small ligands (almost purely inorganic ones). To date, many materials that form frameworks have been identified, and, as their properties have been clarified, their properties that cannot be obtained with conventional inorganic materials have been revealed. These materials, which have unusual physical and chemical properties, include ubiquitous Metal Organic Frameworks (MOFs), covalent organic frameworks, high-density coordination polymers, and molecular frameworks. Many of these show various structural reactions and changes in properties. Unfortunately, known metamaterial MOFs are not associated with optical functions (**Figure 1**). The main properties of framework materials are negative compressibility, complete auxiliaries, negative thermal expansion, and negative gas adsorption. Many of the substances classified as meta-MOFs have common characteristics in the composition and topology of the ligand, which guides the design of unusual reactions. This is a phenomenon not reported in many MOFs with similar structural features. The reason is considered to be that the size of the crystal has a great influence on the flexibility. In addition, structural change kinetics are also said to affect the reactivity of these materials.

Furthermore, it is said that the arrangement and defects in the crystal structure have a great influence, but the truth is uncertain. Indeed, in the field of complex chemistry, MOFs are currently the most promising nanomaterials. Nanometer-scaled pores and two-dimensional layered structures create diverse functions. However, even though it may have a solid band electronic structure such as a molecular crystal, the interaction is limited on the scale of the wavelength of light. Even with nanostructures, light absorption is basically due to organic ligands and metal ions. Crystals of molecules composed of atoms do not optically change visible light, even if they can be analyzed by X-ray diffraction. Therefore, it is natural that a metal complex alone rarely exhibits properties as a metamaterial with respect to light. But can we say that there is no contribution to give useful functions to any method or any metamaterial? For example, many synthetic polymers such as optical fibers and plastic lenses are known to be useful as optical materials [4].

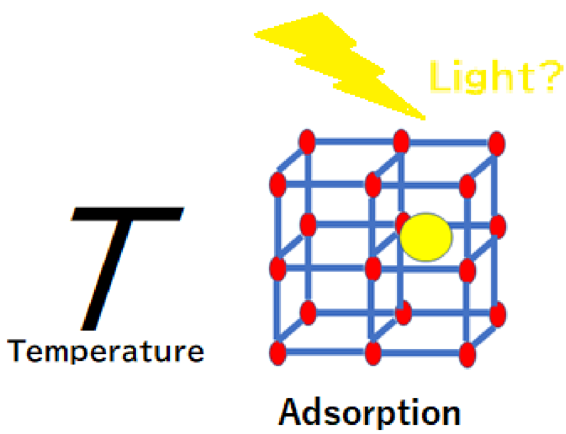

Figure 1: External physical stimulation for response of MOFs. Red points and blue lines denote metal ions and organic bridging ligands in MOFs. The yellow circle represents the adsorbed gas molecules. Mechanical adsorption of guest molecules (unexpected rheological transformation) and temperature (negative thermal expansion of volume) have been reported, although optical metamaterials are few.

On the other hands, some inorganic compounds may be potential materials for nanoscale two-dimensional materials, including optical metamaterials. The ability to extract substances a few atoms thick has led to the discovery of two-dimensional substances such as graphene and single layer Transition Metal Dichalcogenides (TMDs). The aim is to study the one-dimensional edges of these two-dimensional materials and control the edge plane ratio. Edges exhibit unique characteristics that are clearly different from planes and bulks, and, by controlling the edges, it is possible to design TMD metamaterials that have adjustable characteristics. However, technology for guiding such metamaterials with high accuracy has not yet been developed. Etching the TMD along a specific crystal axis results in sharp, zigzag edges at the near-atomic level. This results in a hexagonal nanostructure that combines order and complexity, such as nanoribbons and nano-junctions as thin as a few nanometers. This method enables the study of a wide range of TMD metamaterials by precisely controlling the structure at the atomic level [5,6]

Chiral Metamaterials

Chiral metamaterials are said to have many properties not found in conventional materials. The main characteristics and applications are described below. Firstly, when the chirality is strong enough, a negative index of refraction can be achieved in the microwave, terahertz, and optical regions even if neither ε nor μ is negative. Secondly, due to their extremely strong optical activity, it is possible to realize a compact wideband circularly polarized light element and asymmetric transmission. Thirdly, the super-chiral electromagnetic field in chiral plasmonic nanostructures is expected to significantly enhance the interaction between light and chiral molecules by orders of magnitude, opening new avenues for chiral detection with unprecedented sensitivity. Chiral metamaterials are being studied for many applications in the optical field, including sensors and nonlinear optics. If active control of metamaterial chirality can be realized, it may become an important element of future optical systems. To actively control chiral metamaterials, the metamolecule needs to be reconstituted from left-handed enantiomers to right-handed enantiomers and vice versa. Enantiomer switching is realized by selecting the deformation direction, and the polarity of the optical activity can be changed without changing the spectral shape. It has been confirmed that, when a phase-changing material whose refractive index can be changed by temperature, voltage, or optical pulse is integrated with a chiral metamaterial, the characteristics of the metamaterial are dynamically controlled. By cascading an active right-handed chiral dimer and a passive left-handed chiral dimer, thermal switching of handedness at a constant wavelength was achieved [7].

In the case of (Moiré) Chiral Metamaterials ((M) CMs), it is necessary to mention Circular Dichroism (CD) spectrum. For example, Wu et al. reported tunable chiroptical response of ultrathin active chiral metalmaterials [8]. They designed a MCM as a model metamaterial with the aim of demonstrating an active chiral metamaterial mediated by adjustable near-field coupling. This is the same Au layer in which dielectric spacer layers are stacked in a Moiré pattern. This significantly improved the spectral shift and line shape changes of the MCM's CD spectrum. In addition, there are reports showing actively adjustable near-field binding and chiroptical response of silk MCM using silk fibroin thin film as the spacer layer of MCM. In order to fully grasp the characteristics of metamaterials, it is necessary to describe the phenomena in the fields of optics and electromagnetics. Optical systems with gains and losses that respect Parity Time (PT) symmetry can have real eigenvalues, even though they are non-Hermitian. Circularly polarized waves occur in the chiral system, but this circularly polarized wave does not seem to be incompatible with the system with PT symmetry because the handedness is not maintained even if the space and time are inverted. However, this study shows that PT-symmetric permittivity, permeability, and chirality are possible in a particular configuration. Furthermore, even if the chirality greatly exceeds the PT symmetry, the eigenvalues of the real numbers are maintained. In one study, it was shown that chirality can be adjusted independently of permittivity and permeability by obtaining three constructive parameters of realistic chiral metamaterials from simulation and retrieval [9].

When a simple one-dimensional chiral PT symmetry system was investigated, it was confirmed that, in the case of a normal incident wave, the PT symmetry and chirality-related characteristics could be adjusted independently and superposed almost freely. On the other hand, in the case of oblique incidence, chirality affects all PT-related properties, resulting in unprecedented new ways of transmitting waves such as asymmetric transmittance, asymmetric optical activity, and ellipticity. Thus, the characteristics of an image were seen. Because these properties are highly controllable by both chirality and angle of incidence, PT symmetric chiral metamaterials offer a variety of possibilities (such as circularly polarized wave lasers and absorbers) and approaches for polarization control. This is considered to be useful for tunable polarizing filters, among other applications. Furthermore, the metamaterial functions as an electromagnet and is composed of an artificial structure arranged periodically. Conventional metamaterials require negative μ to achieve negative refraction. Chiral metamaterials are a new class of metamaterials that achieve negative refraction in a simpler way. We studied the characteristics of wave transmission in chiral metamaterials and showed that negative refraction can be realized in chiral metamaterials with strong chirality. In this case, negative μ is no longer necessary [10,11].

Chiral metamaterials are a special class of material that blocks the transmission of incident light and exhibits different optical responses depending on the interaction with Left and Right Circularly Polarized light (LCP, RCP). Research on this Chiral Metamaterial Absorber (CMMA) is being actively conducted. By achieving CMMA's compatibility with various physical, chemical, and biomolecular systems, we created many highly novel and specialized applications. Much research has emerged over the last decade, including studies focused on CMMA design, optimization, and manufacturing, as well as demonstrating a variety of applications. CMMA has the property of sufficiently absorbing light, but research has not yet been conducted to further utilize the absorbed light. Regarding chiral materials, research on Surface Electromagnetic Waves (SEMWs) of isotropic chiral metamaterials has been reported. The SEMW phase diagram is derived by combining Maxwell's equations and constructs including chirality. Numerical calculation results of the phase diagram of the surface of the chiral metamaterial in vacuum were shown as a function of permittivity, permeability, and chiral parameters. From the results, it was clarified that chirality tends to change the localized length of SEMWs and strongly changes the characteristics of the electromagnetic field near the surface [12,13].

Optical Chiral Metamaterials

Sensing devices may be useful examples of applications of optical chiral materials in general, although both the scientific principle and the practical strategy related to optical function should be developed before applied studies. After predicting the strong optical activity that causes negative refraction and negative reflection, more artificial chiral metamaterials have been designed and manufactured for use in different frequency domains from microwaves to light waves. Under such circumstances, there is a need for a method to easily and surely obtain effective configuration parameters of chiral metamaterials. An example of combining a chiral metamaterial and a nonlinear material has also been reported, opening up new possibilities in the field of nonlinear chirality, and it is thought that the foundation of a device with switchable chirality was successfully constructed. Furthermore, by combining chirality and hyperbolicity, it is possible to realize materials exhibiting new functionality such as photonic topological insulators. Much effort has been made in both top-down and bottom-up approaches in the process of creating chiral metamaterials. Recently, the production of 3D visualization chiral metamaterials has progressed using technologies such as the laser direct drawing method and block copolymer self-assembly method. However, due to the limited experimentally achievable chiral design and incomplete control of material properties such as loss and refractive index changes, the control of optical activity and circular dichroism at visible wavelengths remains difficult [14-16].

The MCMs proposed by Wu et al., [17,18] may be promising materials for optical functions. Chiral metamaterials are expected to have various applications due to advances in nano-processing technology and the elucidation of the mechanism of chirality effects in the optical field; however, two or more examples with different lattice constants and relative spatial displacements were recently proposed. They are expected to be used for applications of chiral metamaterials using the Moiré laminating method, which superimposes their periodic patterns. Moiré chiral metamaterials utilize grid-dependent chirality, are more cost-effective than traditional chiral metamaterials, and are flexible and highly reconstructible. Expectations are gathering for their conversion. A new type of chiral metamaterial in which two layers of the same achiral gold nanohole array are laminated in a Moiré pattern has been reported. Moiré chiral metamaterials can precisely adjust the optical activity due to chirality through in-plane rotation between two layers of nanohole arrays. Furthermore, Moiré-type chiral metamaterials have also been applied to label-free asymmetric identification of biomolecules and drug molecules at the pg level. Moiré chiral metamaterials are promising for various photonic and optoelectronic activities due to their ultrathin thickness (about 70 nm, which is only 1/10 of the operating wavelength), strong chirality, and high adjustment ability. Needless to say, nanoscale lattices for Moiré patterns are not at the "molecular level"; therefore, stacking of two-dimensional MOFs may not be appropriate for such applications **(Figure 2).** Of course, they are completely incompatible with the wavelength and diffraction of light [17,18].

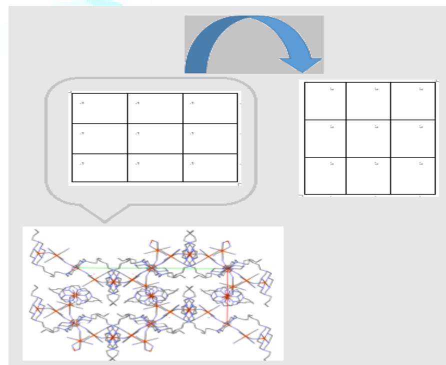

Figure 2: Virtual concept of MCMs composed of two-dimensional MOFs with stacking of two "nets" at different angles. Two-dimensional nets of MOFs are stacked perpendicularly in this figure.

Summary and Perspectives (Hybridization of Metal Complexes)

The development of MOFs can also incorporate the very promising direction of using chemical methods to obtain metamaterials. By shifting from a physical manufacturing method to a chemical manufacturing method, it is possible not only to simplify the process for obtaining a specific material, but also to diversify the final structure. Accordingly, it seems very promising to chemically develop MOFs that have the properties required for metamaterials. As a new method that can surely form a higher-resolution quantum/nonlinear metamaterial, the capability of achieving the necessary structural scale and availability is required. The most promising method is to utilize highly porous structures based on organometallic complexes that have unique properties in the field of nonlinear optics. Special attention should be paid to the combination of metals and ligands when using an organic metal framework. In order to further improve the optical properties of the materials, various parameters such as ligand length, degree of substitution, conjugate in the system, and the use of guest molecules with different properties need to be adjusted [19].

It is necessary to be able to create "nanostructures" shorter than the wavelength of light (metal nano, plasmons, etc.). Unfortunately, substances with $\varepsilon<0$ are less common than those with $\mu<0$. Combining materials can affect their refractive index, e.g., in a polymer matrix. Establishing a negative refractive index involves the use of nanostructures shorter than the wavelength of light (e.g., microwave).

Inorganic crystals and chiral substances change the speed of light, e.g., via "birefringence". In fact, quartz has distinct properties when exposed to left and right circularly polarized light related to birefringence and optical rotation. It would be difficult to realize an optical metamaterial with a single crystal of a metal complex. Instead, composites including an optical metamaterial and chiral complex (only for the purpose of adjusting properties) or including metal nanoparticles and a chiral complex can be used. Surface plasmon resonance of gold nanoparticles and nanorods allows generating a resonator with a fine metal structure that resonates with light.

A single resonator is called a meta-atom, and a substance in which meta atoms are dispersed in a liquid is called a meta-fluid. Composite materials with these substances are possible. Metal complexes are groups of compounds that have been studied for a long time for their optical and chirality such as chiroptical and emission spectroscopy, as well as asymmetric reactivity. However, since the unit is a "molecule", few examples as (optical) metamaterials have been reported so far. Is it possible to add a function as a composite material using one method? Is there a way to fuse the rich compounds at the boundary between inorganic and organic with state of the art optical metamaterials? One answer may by the “luminescence of a transition metal complex inside a metamaterial nanocavity” reported by Connell et al. [20] (**Figure 3** (left)). The large spin–orbit interaction of heavy metal atoms results in an excited state with remarkable magnetic dipole properties. The authors described the production of a metamaterial composed of nanowires separated from a metal mirror by a polymer thin film doped with a luminescent organometallic iridium (III) complex. The metamaterial nanostructure architecture supports two different optical modes and realizes their assignment with the help of a numerical simulation. This is one of the few research examples that combined an optical metal complex with a metamaterial.

Indeed, in conventional fluorescent or other spectroscopic studies for metal complexes in both solids and solutions, the refractive index is not really a problem. The combination of optical materials and refractive index of media may attract new attention in the future. Although we would like to introduce or discuss other studies, there are few examples of metal complex-based metamaterials at present. Generally speaking, well-designed composite materials of metal complexes are known to be useful functional materials that combine the advantages of multiple components that would be infeasible when using the individual components (**Figure 3** (right)). For example, in a PMMA polymer matrix, azobenzene can be directly aligned via the irradiation of linearly polarized UV light due to the Weigert effect, and (chiral) Schiff base metal complexes also increased their optical anisotropy observed with polarized UV-Vis or FT-IR spectroscopy. In order to experimentally interpret the interaction of light and compounds, as well as the intermolecular interaction of the photofunctional hybrid materials, computational chemistry methods such as TD-DFT calculations and other theoretical interpretations assuming dipoledipole interactions or statistical distributions based on certain physical properties have been developed [21-23].

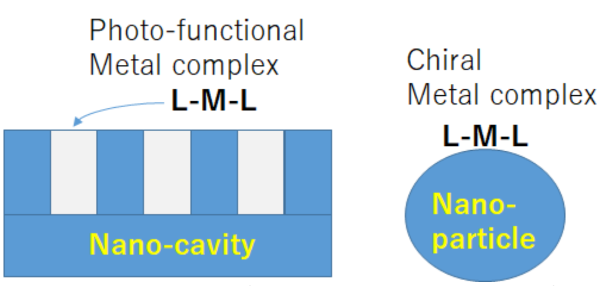

Figure 3: Example of optical metamaterials composed of nanomaterials of metal complexes (L–M–L denotes the general representation of Metal (M) and Ligand (L) complexes). References are presented in the text.

In recent years, this has allowed investigating photo functional hybrid materials of (chiral) Schiff base metal complexes and their interaction with several types of topological lights such as linearly polarized UV light, circularly polarized UV light, plasmons on the surface of metal nanoparticles, infrared free-electron lasers, and optical vortices or polarized UV light generated by a synchrotron facility.

Author Contributions

Conceptualization TA, writing original draft preparation, YS, DN, and TA. All authors read and agreed to the published version of the manuscript.

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