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Introductory Chapter: Chirality

Takashiro Akitsu

1. Introduction

Chirality is a concept that is related not only to organic chemistry but also to each field of natural science in a narrow sense, and it is considered that awareness of hierarchy is important for universal and comprehensive understanding. For example, on a small scale, from the decay of elementary particles, the angular momentum of atoms and light or gravity, organic molecules, biochemical or organic-chemical reactions, crystal structures, supramolecular composite structures, and living organisms, and even galaxy vortices should be considered in this context. Here are some dreams (hypothesis) and excellent examples of interdisciplinary application of “chirality”.

2. Angular momentum arising chirality in molecular level

A few years ago, in order to overcome such hierarchy, it was merely a “thought experiment” and a “working hypothesis,” but I applied the force of a classical magnetic field to a molecule on a quantum mechanical scale. Like “molecular machines” attracting recent attention, mechanical laws and mechanical properties at the level of molecules and aggregates that are originally below the nanometer scale are used as if they were parts on the centimeter scale, or they are excluded. It can be said that chemistry and material science, which are controlled by the field, are new fields that can be expected to develop in the future. Expanding from a quantum chemistry point of view to a slightly macroscopic point of view, if a molecule is treated as “a kind of rigid body with a certain size and shape” in the framework of classical mechanics, “torque” and “rotational inertia” are among the mechanical properties. Focusing on “momentum”, it is expected to discuss the transmission of rotation and the interaction with the orbital angular momentum, which are useful for nanotechnology.

Apart from the control of electronic states and molecular structures by the external field in the conventional quantum chemistry framework, polarized ultraviolet light and magnetism are used as means of external field control, and metal complex molecules, metal complexes-high realization of control of droplets (microdroplets) in molecular/protein composite materials and microfluidics containing composite materials, and mechanical properties such as “torque” and “rotational inertia momentum”, which are indicators of the control, are used in molecular design. The goal of this “dream” is to establish a new concept of rotational motion with chirality to be introduced as a new parameter.

There is a magnetic field effect on red blood cells as a phenomenon that has been known for a long time in a similar substance system [1]. Hemoglobin, in which heme iron is paramagnetic and the protein site is diamagnetic, receives resistance from the fluid as it flows through the blood vessels, but when a specific magnetic field acts, blood flow is promoted. This is because not only the paramagnetic part where a direct

magnetic force is expected, but also the diamagnetic part that occupies most of it undergoes an orientation change that lowers the energy of the magnetic moment due to the repulsion from the magnetic field, making it easier to move (**Figure 1**).

At this time, if the erythrocytes themselves, which are nanoscale molecular aggregates, are regarded as rigid bodies having a constant mass, size, and flat shape (momentum of inertia),

1. external magnetic force
2. magnetic momentum
3. Lorentz force
4. rotational momentum of inertia
5. rotational torque
6. fluid resistance reduction

In order to design a material that is easy to rotate (prone to generate the same torque), the classical mechanical “momentum”, how to incorporate the external field response site in the molecule, and the mass-distance distribution from the center of gravity to the end of the molecule, it should be necessary to introduce new molecular parameters (not found in conventional quantum chemistry, *etc.*).

For the optimized structure of the complex after photoisomerization, “the sum of the products of the atomic weight of each atom and the distance between the center of gravity of the molecule (which may be defined as the center of paramagnetic metal)” will be calculated. This is a quantity corresponding to the rotational moment of inertia of classical mechanics, and is newly introduced as a parameter on the molecular side in order to discuss the correlation with the quantity corresponding to the torque realized by the change in external field orientation.

Ferrofluids have long been known as functional materials related to the dynamics of fluids and continuums. However, it was expanded from the fact that the magnetic material is a uniformly dispersed droplet, starting from the (single molecule) “rotational moment of inertia” and “rotational torque”, which are important parts of this research. It is significant in proposing parameters for molecules that are completely different from the concept of discussing the propagation of effects to composite materials and droplets in microspace.

Further, as a precedent example in which a rotational motion is caused by light, there is a circularly polarized laser irradiation on metal nanoparticles, metal nanorods, etc., but the size of the target substance composed of metal atoms is too large. Physical quantities such as angular momentum and torque may be able to challenge the limits of size and hierarchy reflected in the chirality of molecules and aggregates.

Figure 1.
Schematic representation of the concept of hemoglobin in blood flow (arrows) under magnetic field (**H**).

Circularly polarized light with spin angular momentum results in a spiral orientation with chirality. In recent years, with “optical vortex” having orbital angular momentum, dynamics such as torque and light pressure when a spiral structure is transferred to a circular region centered on a singular point on the material surface. Then I noticed that the current situation is that the chemical understanding of “treating molecules classically” may be inadequate. In addition, when attempting to grow a single crystal of a complex-encapsulating protein in a droplet, the “force acting on an object” including gravity is referred to by referring to the protein crystal growth and molecular orientation under zero gravity or strong magnetic field conditions. I came up with the idea that the analogy to molecules and nano-aggregates would be effective.

Unfortunately, verification research of this hypothesis has not yet been conducted to date.

3. Chiral-induced spin selectivity (CISS) effect for hybrid materials

By the way, as a chirality phenomenon that can still cross the hierarchy, “chiral-induced spin selectivity (CISS)” proposed by Professor Ron Naaman is one of the most notable concepts at present [2]. The information gained through the experiment will be meaningful to the science, though this description is merely a “proposal” level.

As mentioned below, injecting electrons into oxidoreductase through oligopeptide which works as a spin filter is the first ever experiment, and it is crucial to cooperate with Dr. Naaman for this research. They believe several years of experience on metal complexes and profound knowledge about the CISS Dr. Naaman has can lead to a promising outcome. One of results they already expect to have is that if they detect the change of chiral property affected by the subtle transformation of metal complex and oligopeptide, they can observe and analyze the electrochemical change.

Their cooperative research covers the fields of the surface physics and the biochemistry, and their results of fixing the electron transfer pathway between electrode and enzyme, and of control of magnetism that metal complex has by the external magnetic field, would be applied to other fields. Also, the CISS effect will offer new possibilities for a better understanding of process of spin selection in the biology and spin electronics applications.

The injection of electrons into laccase using the CISS effect is the first trial of its kind. If this experiment was conducted successfully, the results would contribute to other fields including the spin electronics.

Then they were planning to apply the CISS effect to metal complex to magnetize spins from outside to control them while performing electrochemical measurement. The CISS effect is that in chiral molecules only spins which are orientated in a certain direction can be transferred smoothly. This effect has been studied a lot in recent years with Dr. Naaman, as the leading researcher. With Dr. Naaman's cooperation, who is the leading researcher of this effect, it is particularly noteworthy that difference in chirality shows different spin polarization, which suggests that this effect is applicable to a system where laccase is used (**Figure 2**).

Prepare a measuring device described, then inject spin polarized electrons into laccase where metal complex is present. Place oligopeptide and the metal complex between the electrode and the laccase. They use the oligopeptide and the metal complex, which let spin polarized electrons be transferred smoothly, so that the increase of amperage should be expected. They also analyzed how spin polarized electrons affect the way electron-transfer to the enzyme is occurred. As

Figure 2.
Proposed of measuring system for CISS effect of redox proteins (laccase).

a preliminary experiment, they conducted electrochemical measurement using the above method without the metal complex. The result shows that the chirality of peptide affects the electron-transfer to the laccase. Following this outcome, they expect the follows. If they insert the metal complex between the oligopeptide and laccase, with the metal complex being fixed inside the hydrophobic pockets of laccase, the metal complex would be fastened firmly to the laccase. Then the electron-transfer to the Cu-T1, which proceeds oxygen reduction reaction, can be carried out selectively. In addition to that, they will examine how the electron-transfer would change when creating an external magnetic field, and they will also analyze it by comparing the use of ferromagnetic or photoresponsive metal complexes.

Some reports have been submitted regarding the injection of electron spins only into membrane protein or oligopeptide in those days, but there has not been any report on the injection of the electron spins into oxidoreductase. The results from the preliminary experiments suggest there is a possibility of realizing the electron-transfer without being influenced by the external magnetic field. Their idea has an intension of examining the electron-transfer in depths from a new perspective by injecting the electron spins into laccase using the CISS effect to elucidate the difference electrochemically between other mediate complexes. The analysis about behavior of the electrons will contribute to the development of biochemistry and a field of spintronics device.

Unfortunately, actual research using laccase only has not yet been published to date.

4. Concluding remarks

In this way, “chirality” may be found in many fields of natural sciences and technology potentially. The main purpose of this plan about the CISS effect study was just to improve electron transfer between the electrode and the enzyme in enzymatic biofuel cell using the CISS effect. Along with that, they are also planning to reveal from a spin-polarized point of view how magnetic field and chiral property affect the electron transfer reactions. Also, if we achieve the implementation of the CISS effect, the result would be applied “chirality” for other areas such as energy materials science and medical biochemistry.

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Author details

Takashiro Akitsu
Department of Chemistry, Faculty of Science, Tokyo University of Science,
Tokyo, Japan

*Address all correspondence to: akitsu2@rs.tus.ac.jp

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