【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project：Paradigm shift in the method for observing non-equilibrium processes in real space: Elucidation of nucleation processes from solution by TEM

KIMURA Yuki
(Hokkaido University, Institute of Low Temperature Science, Associate Professor)

Research Project Number: 20H05657 Researcher Number：50449542
Keyword：Nucleation, Nanoparticle, Transmission electron microscopy, In-situ observation, Crystal growth

【Purpose and Background of the Research】

Nucleation is a process whereby particles are formed by the agglomeration of atoms or molecules. Because the nucleation process determines the size, crystal structure, number density, and other properties of the resulting particles, an understanding of its mechanism is crucial for the development of materials science. Nevertheless, our understanding of the physical and chemical processes involved in nucleation remains poor. In this project, our objective is not only to achieve an understanding of nucleation process of individual materials, but also to develop a method for determining the physical properties of nanoparticles and elucidating the roles of dehydration, viscosity, and dimer formation in the nucleation process. Consequently, our final objective is to identify the key factors that determine the nucleation route.

【Research Methods】

Our main method is the in-situ transmission electron microscope (TEM) observation of nucleation processes from aqueous solutions. In order to understand the role of the hydrated layer, we have also conducted nucleation experiments from ionic liquid solutions and the gas phase. To observe nucleation from solution samples, we use three techniques (a liquid cell holder (Fig. 1), a solution cell, and a graphene film). These techniques allow us to directly observe the growth rate, shape, assembly, arrangement, and size of the resulting crystals and to identify the phases by electron diffraction patterns. Here, we establish a novel dynamic observation method for non-equilibrium processes using machine learning to visualize the entire nucleation process from solution through precursors to crystal formation by in-situ observation using a TEM.

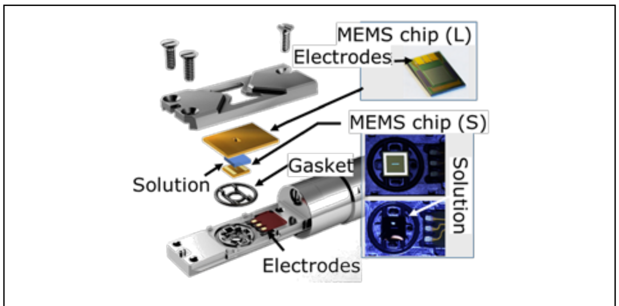

Figure 1 One of three techniques for introducing an aqueous solution into a transmission electron microscope. A liquid cell holder with a heating capability that encloses the solution between two plates with a window. The window is a 30 nm-thick amorphous silicon nitride film, through which electrons can transparent to observe the solution, and the two liquids can be mixed during heating up to 100°C during observation.

【Expected Research Achievements and Scientific Significance】

Because our aim is to identify the key factors that govern the nucleation process and then to construct a nucleation model, our project constitutes groundbreaking fundamental scientific research. After our project, we hope to see a new world in which bottom-up processes for producing nanoparticles and crystals from atoms and molecules can be designed. In addition, we hope to understand the processes involved in the formation of cosmic dust, which consists of nanoparticles with a size of less than 100 nm, and which is abundant in the universe in the gas outflow of dying stars. Furthermore, we will identify conditions that are conducive to the precipitation of metastable phases to facilitate the dissolution of medicines. Thus, the significance and impacts of the results are, potentially, extremely wide-ranging.

【Publications Relevant to the Project】

- T. Yamazaki, Y. Kimura, P. G. Vekilov, E. Furukawa, M. Shirai, H. Matsumoto, A. E. S. Van Driessche, K. Tsukamoto, Two types of amorphous protein particles facilitate crystal nucleation, Proceedings of the National Academy of Sciences of the United States of America, 114 (2017) 2154-2159.
- Y. Kimura, K. K. Tanaka, T. Nozawa, S. Takeuchi, Y. Inatomi, Pure iron grains are rare in the universe, Science Advances, 3 (2017) e1601992.

【Term of Project】 FY2020-2024

【Budget Allocation】 154,900 Thousand Yen

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project : Creation of a unified theory of global environmental changes and resource generation

KATO Yasuhiro
(The University of Tokyo, Graduate School of Engineering, Professor)

Research Project Number: 20H05658 Researcher Number : 40221882
Keyword : Resource exploration, seafloor mineral deposits, global geochemical cycles

【Purpose and Background of the Research】

The field of resource investigation has shifted from land to ocean; however, the aims of previous studies of seafloor mineral resources were limited to clarifying the ore genesis of individual resources, hindering attainment of a unified picture of the Earth system and ore genesis. Generation of resources in the oceans and associated environmental changes can be regarded as aspects of the global cycles of various elements and materials. The question “how did these resources form?” can be answered only through a quantitative analysis of global geochemical cycles (Fig. 1), including input of elements from the mantle to the ocean via volcanic and hydrothermal activities and output to the mantle through precipitation and deposition of elements to the seafloor and their subsequent subduction into the trenches. Pelagic clay is the key material for elucidating these global cycles. We will apply newly developed “chemostratigraphic probes” to pelagic clay cores collected from various oceanic regions, to map time-series information recorded in pelagic clays worldwide, and to elucidate the dynamics of global geochemical cycles.

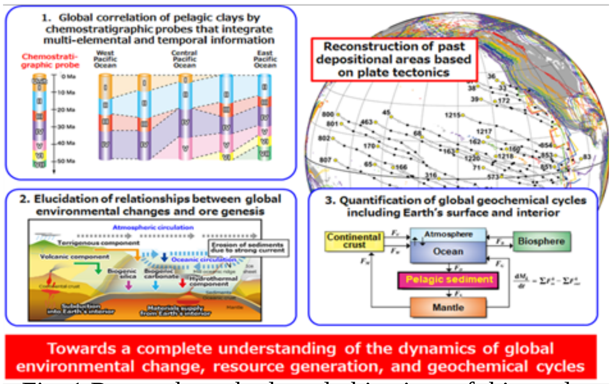

Fig. 1 Research methods and objectives of this study

【Research Methods】

“Chemostratigraphic probes”, which integrate multi-elemental and chronological information, will be used to correlate pelagic clays over a wide area of several oceans. First, we will perform high-precision chemical analyses on International Ocean Discovery Program core samples to construct a large-scale, multi-elemental dataset of pelagic clays. In addition, we will combine Os isotopic ratios and ichthyolith biostratigraphy to precisely determine depositional ages. Then, we will analyze the high-dimensional data covering global oceans by multivariate analyses (e.g., independent component analysis). The results will provide a comprehensive picture of temporal variations in pelagic-clay geochemistry, as well as their causes and a record of erosion of sedimentary layers. We will also construct a global geochemical cycle model incorporating all the geochemical and temporal information on pelagic clays to quantify the mass balance and flux variations between the atmosphere–ocean and solid earth systems. As part of this modeling, we will focus on the cycles of critical elements to elucidate the entire picture of ore genesis in the ocean.

【Expected Research Achievements and Scientific Significance】

Quantitative analysis of the mass balances of various elements on the Earth's surface, as well as the factors and processes controlling them, will provide a unified picture of the causal relationships between environmental changes and resource generation. This unified picture will facilitate novel, systematic exploration for promising resources. Furthermore, pelagic clays are the most important interface between the solid earth and the atmosphere–ocean systems, serving as a bridge between the disciplines of environmental and solid earth sciences. By obtaining a precise picture of this interface for the first time, we will create a theory to explain various phenomena on the Earth in a unified framework, from a panoramic view of global geochemical cycles.

【Publications Relevant to the Project】

・Kato, Y. et al. “Deep-sea mud in the Pacific Ocean as a potential resource for rare-earth elements.” *Nature Geoscience* **4**, 535-539 (2011).
・Takaya, Y. et al. “The tremendous potential of deep-sea mud as a source of rare-earth elements.” *Scientific Reports* **8**, 5763 (2018).
・ Ohta. J. et al. “Fish proliferation and rare-earth deposition by topographically induced upwelling at the late Eocene cooling event.” *Scientific Reports* **10**, 9896 (2020).

【Term of Project】 FY2020- 2024

【Budget Allocation】 156,900 Thousand Yen

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project： Development of atomic-resolution magnetic field imaging electron microscopy and its application to interface characterization in magnetic materials

SHIBATA Naoya
(The University of Tokyo, Institute of Engineering Innovation, Professor)

Research Project Number: 20H05659 Researcher Number：10376501
Keyword：Scanning transmission electron microscopy, electromagnetic field, interface, magnetic materials, steel

【Purpose and Background of the Research】

In this research, we will develop a new atomic-resolution electron microscopy method that realizes real-space observation of local atomic structures and related atomic-scale electromagnetic field distribution simultaneously, based on the atomic-resolution magnetic field-free electron microscope that the principal investigators succeeded in development for the first time in the world. We will realize direct observation of atomic magnetic moments, interfacial magnetic structures, electrical polarization, etc., which have been impossible to directly observe by electron microscopy. Furthermore, this method will be used for the interface studies of magnets, spin devices, steels, topological materials, ceramic materials, etc., aiming to understand the interaction mechanisms between the atomic-scale local structures and magnetisms. By doing so, we essentially elucidate the origin of functional properties in magnetic materials, and establish a new magnetic material design strategy based on the interface control. The ultimate goal of this research is to promote the development of functional magnetic materials and contribute to the sustainable growth of society and industry.

【Research Methods】

We will develop an ultra-high resolution electromagnetic field imagnig electron microscopy by fusing the atomic resolution magnetic field-free electron microscope with the differential phase contrast (DPC) scanning transmission electron microscopy (STEM) method. Specifically, we will develop a quantitative electromagnetic field observation method by detecting the center of mass of an electron diffraction pattern using segmented and pixelated detectors, and an electric / magnetic field separation method using a new specimen holder. Furthermore, the developed method will be applied for the interface studies of various magnetic materials (such as rare earth magnets, ferrite magnets, spin devices, electrical steels, topological materials and so on), in order to elucidate the fundamental mechanisms of the interaction between local atomic-scale structures and related magnetisms.

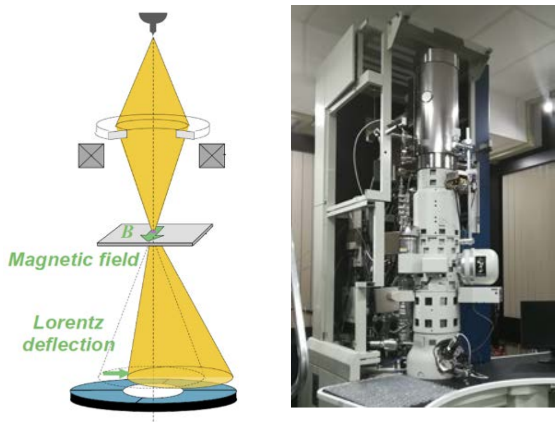

Fig.1 Schematic of DPC STEM and atomic-resolution magnetic field-free electron microscope

【Expected Research Achievements and Scientific Significance】

In order to understand the mechanism of functional properties in magnetic materials, it is essential to elucidate the local electromagnetic field structures induced by the local structure such as interfaces inside the materials. In conventional electron microscopy, direct observation of local electromagnetic fields at atomic resolution has been extremely challenging. In this study, a new electron microscopy method that enables simultaneous measurement of both the atomic-level local structures and the local electromagnetic fields induced by them will be developed. which is expected to open up a new stage of electron microscopy. Furthermore, this research is not limited to the development of microscopy techniques, but is aiming to apply this newly developed method to the interface studies of important magnetic materials. Through these studies, we aim to establish a new material interface design strategy for functional magnetic materials.

【Publications Relevant to the Project】

・ N. Shibata, T. Seki *et al.*, “Atomic resolution electron microscopy in a magnetic field free environment,” *Nature Comm.*, **10**, 2380 (2019).
・ N. Shibata, T. Seki *et al.*, “Direct Visualization of Local Electromagnetic Field Structures by Scanning Transmission Electron Microscopy,” *Acc. Chem. Res.*, **50**, 1502-1512 (2017).

【Term of Project】 FY2020-2024

【Budget Allocation】 148,300 Thousand Yen

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project： Terahertz dynamics of single molecule transistors and its application to quantum information processing

HIRAKAWA Kazuhiko
(The University of Tokyo, Institute of Industrial Science, Professor)

Research Project Number: 20H05660　　Researcher Number：10183097
Keyword：single molecule transistors, terahertz radiation, quantum information technology

【Purpose and Background of the Research】

In recent years, single molecules have attracted attention as devices that can apply molecular functions to electronics. Furthermore, molecular vibrations have properties as quantum mechanical oscillators and have a possibility of becoming a new medium for quantum information processing. Since the energies of elementary excitations such as quantum levels and molecular vibrations in such atomic-scale nanostructures corresponds to the photon energy of terahertz (THz) electromagnetic waves, useful information on the physics and dynamics of such quantum nanostructures can be obtained by THz spectroscopy.

Recently, we have formed electrodes with atomic-scale gaps onto a single molecule and used them as THz antennas to focus THz electromagnetic waves on a single molecule. It has become possible to measure extremely weak THz signals derived from excitation within a single molecule.

We think that research on the interaction between THz electromagnetic waves and quantum nanostructures has just entered a new phase. Without missing this timing, we should dig into the physics of the quantum nanostructures and investigate their applications as media for quantum information processing.

The purpose of this research is to further promote and deepen the new field of atomic-scale "terahertz nanoscience" that is currently emerging, and to explore the possibility of its applications.

【Research Methods】

1) Terahertz spectroscopy of single molecules: The molecular vibrations and conduction electrons are strongly coupled and, by coherently controlling the molecular vibrations with THz pulses, we will establish the basis for quantum control of electron conduction and explore the possibility of application to new quantum information processing.

2) Resistive detection of single molecule nuclear magnetic resonance (NMR): We will perform resistive detection of NMR signal of single atoms and molecules by using nanogap electrodes and explore a possibility of using it as a medium for carrying new quantum information.

3) Ultra-strong THz electric fields in the nanogap electrodes: Unprecedented “ultra-strong ac electric fields of ~GV/cm range in the nm region" generated by the field enhancement effect of the nanogap electrodes. We will elucidate novel electron dynamics in such ultrahigh ac fields.

4) Use of nanomechanical structures for advanced nanoscale sensing: We develop a new technology for high sensitivity nanoscale transport measurements and THz spectroscopy using MEMS/NEMS technology.

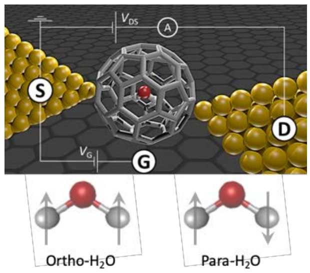

Fig. 1 Schematic illustration of a single molecule transistor structure with a $H_2O@C_{60}$ molecule as a quantum dot.

【Expected Research Achievements and Scientific Significance】

Physics and applications of nm-scale systems are just in a phase of rapid rise and this project will be a major stepping stone. Furthermore, the miniaturization of silicon technology represented by CMOS is approaching its limit, and there is an urgent need to search for new device principles. This research provides a major scientific basis for creating nanodevices with novel functions such as quantum information processing by combining the unique physical properties of nanogap electrodes and molecules.

【Publications Relevant to the Project】

· S. Du, K. Yoshida, Y. Zhang, I. Hamada, and K. Hirakawa: “Terahertz dynamics of electron–vibron coupling in single molecules with tunable electrostatic potential”, Nature Photonics, vol.12, pp. 608-612 (2018).
· K. Yoshida, K. Shibata, and K. Hirakawa: “Terahertz field enhancement and photon-assisted tunneling in single-molecule transistors”, Physical Review Letters, vol. 115, pp. 138302-1~5 (2015).

【Term of Project】 FY2020-2024

【Budget Allocation】 146,800Thousand Yen

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Title of Project : Universal quantum media conversion in diamond quantum storage

KOSAKA Hideo
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Research Project Number: 20H05661 Researcher Number : 20361199
Keyword : Quantum information, Spintronics

【Purpose and Background of the Research】

In recent years, competition for the development of quantum computers using superconducting qubits has become worldwide research trend, but on the other hand, the development of the quantum internet, which enables encrypted communication with physically guaranteed security, has also begun. Furthermore, with the spread of DNA banks and Bitcoins, the need for quantum storage for the safe storage of personal information that cannot be leaked is rapidly increasing. By constructing a quantum computer network, it will be possible to perform distributed quantum computing, blind quantum computing, and quantum internet (Fig. 1). To realize these, it is indispensable to develop a large-scale quantum storage and its quantum interface, which are highly consistent with the superconducting qubit, which is the heart of the quantum computer, and can hold the quantum state for a long time.

The purpose of this study is to establish an universal quantum media conversion technology in quantum storage composed of carbon isotopes (^{13}C) distributed in a cloud shape around a single nitrogen-vacancy (NV) center in diamond. The development of 1M-bit scale quantum storage that is highly consistent with superconducting qubits and operates in the absence of a magnetic field paves the way for distributed quantum computation and blind quantum computation by quantum computer networks connected by the quantum internet, and brings dramatic evolution to the highly computerized society.

【Research Methods】

In previous research, we have developed a technology to convert the quantum state of photons into surface carbon based on quantum teleportation principle, using the surface carbons near an NV center as a quantum memory that operates under a zero magnetic field. On the other hand, in this study, the deep carbons distant from NV is used as quantum storage, and individual quantum entanglement of deep carbons by geometric decoupling, selective quantum media conversion from single photon to a single deep carbon, quantum entanglement measurement between arbitrary deep carbons, fault tolerantization by quantum coding, and large-scale quantum storage by extension to NV ensemble are realized.

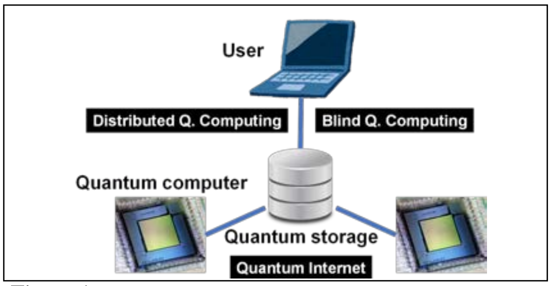

Figure 1 Concept of a quantum computer network quantum-mechanically connected by a quantum storage.

At the center of the NV, there are electrons captured by the defect and a nitrogen impurity adjacent to them, and two types of carbon isotopes (^{13}C) are distributed in layers according to the distance from the electrons. The feature of this research is to use these as a quantum processor, quantum buffer, quantum memory, and quantum storage, and to increase the scale by using an ensemble (Fig. 2).

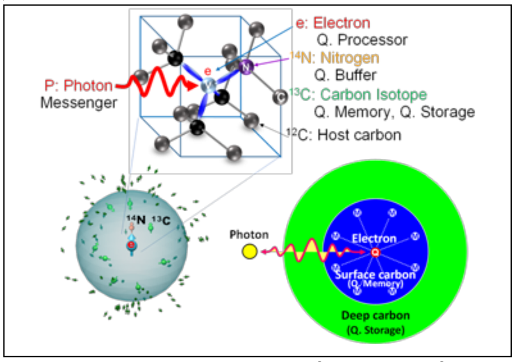

Figure 2 A layered structure consisting of electrons-surface carbons-deep carbons associated with a single nitrogen vacancy (NV) center in diamond.

【Expected Research Achievements and Scientific Significance】

Quantum storage that operates under a completely zero magnetic field is highly consistent with superconducting qubits, paving the way for distributed and blind quantum computations using quantum computer networks connected via the quantum internet

【Publications Relevant to the Project】

· Kodai Nagata, Hideo Kosaka*, et.al., “Universal holonomic quantum gates over geometric spin qubits with polarised microwaves”, Nature Communications, 9, 3227 (2018).
· Kazuya Tsurumoto, Hideo Kosaka*, et.al., "Quantum teleportation-based state transfer of photon polarization into a carbon spin in diamond", Communications Physics (Nature publishing), 2, 74 (2019).

【Term of Project】 FY2020-2024

【Budget Allocation】 150,800 Thousand Yen

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Title of Project： Development of Phase-Controlled Near Field Spectroscopy with Extremely High Spatiotemporal Resolution

TAKEDA Jun

(Yokohama National University, Faculty of Engineering Science, Professor)

Research Project Number: 20H05662 Researcher Number：60202165

Keyword：terahertz, scanning tunneling microscopy, near field, luminescence, mid-infrared pulse

【Purpose and Background of the Research】

Manipulating the structure and function in matters by controlling their electronic and charged states at arbitrary time and space is one of the most important issues in the fields of materials science and nanoscience. In terms of the time resolution, attosecond control of electrons has been achieved in elaborate nanostructures by modulating a carrier-envelope phase of near infrared optical pulses. However, it is difficult to apply this technique to a variety of materials. In terms of the spatial resolution, on the other hand, scanning tunneling microscopy (STM) has been widely utilized as an analytical tool with atomic-scale resolution yet with low time resolution, making it difficult to reveal energy conversion and dissipation in quantum systems.

In this study, we develop terahertz-field-driven scanning tunneling luminescence (THz-STL) spectroscopy by combining the THz-STM and STL techniques to capture the energy dynamics triggered by ultrafast electron tunneling. In addition, we also utilize single-cycle mid-infrared (MIR) near fields to control vibrational states in functional and biological materials. Our final goal is to hybridize these techniques that can manipulate a variety of physical properties of materials in THz to MIR frequency regions (Fig. 1).

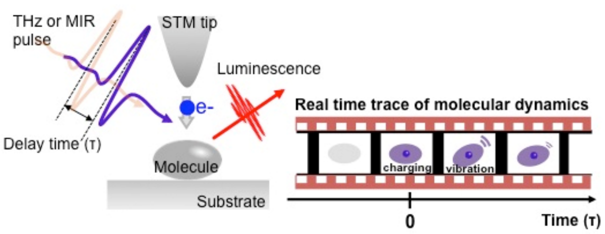

Figure 1 Schematic of phase-controlled terahertz (mid-infrared) field-driven near-field spectroscopy.

【Research Methods】

Conventional STL spectroscopy can measure photons converted from the tunneling electrons of an STM. In our new THz-STL spectroscopy, by injecting a charge using phase-controlled THz pulses that are generated using a $LiNbO_3$ prism with tilted pulse-front configuration, we can trace time-resolved excited-state dynamics in a single molecule. The phase of THz pulses can be tuned by a THz phase shifter originally developed. In addition, we produce phase-stable MIR pulses via optical rectification of 10 fs near-infrared laser pulses compressed by either a hollow fiber or a chirped mirror. After a combination of these field-driven nanoscopic techniques, we try to explore radiative decay processes of a single molecule and trace local structural dynamics of hydrogen-bonded network.

【Expected Research Achievements and Scientific Significance】

Our goal is to establish a new spectroscopic tool with unprecedented spatiotemporal resolutions to capture energy dissipations and conversions among various quanta, which offers prospects for sensing and controlling quantum systems, providing novel insights and advances in nanoscale science and technology.

Our challenge will also provide information on how smaller and how faster we can access the quantum nature of materials, which is the supreme proposition for the development of next-generation nanoelectronics and plasmonic devices with high performances.

【Publications Relevant to the Project】

- K. Yoshioka, I. Katayama, Y. Minami, M. Kitajima, S. Yoshida, H. Shigekawa, and J. Takeda, "Real-space coherent manipulation of electrons in a single tunnel junction by single-cycle terahertz electric fields", **Nature Photon. 10**, pp. 762-765 (2016).
- K. Yoshioka, I. Katayama, Y. Arashida, A. Ban, Y. Kawada, K. Konishi, H. Takahashi, and J. Takeda, "Tailoring single-cycle near-field in a tunnel junction with carrier-envelope phase-controlled terahertz electric fields", **Nano Lett. 18**, pp. 5198-5204 (2018).
- H. Mashiko Y. Chisuga, K. Oguri, H. Masuda, I. Katayama, J. Takeda and H. Gotoh, "Multi-petahertz electron interference in Cr:Al_2O_3 solid-state material", **Nature Commun. 9**, 1468 (2018).

【Term of Project】 FY2020-2024

【Budget Allocation】 146,600 Thousand Yen

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project: **Analysis, Design, and Construction of Highly Concentrated Electrolytes for Innovative Electrodeposition Technologie**

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(Kyoto University, Graduate School of Engineering, Professor)

Research Project Number: 20H05663 Researcher Number: 30283633
Keyword: Electrodeposition, Concentrated solutions, Solution chemistry, Metal complexes, Metallographic structure

【Purpose and Background of the Research】

Electrodeposition (ED) that uses electrochemical reduction of metal ion species in electrolytes to form metals and alloys is an elemental technology for various industrial processes. We recently developed novel ED technologies that utilize a series of highly concentrated electrolytes (HCEs) for, for example, environmentally friendly trivalent chromium plating, ED of aluminum for battery anodes, and formation of Cu_2O semiconductor layers for photovoltaics. These HCEs provide environments depleted of free solvent molecules, and we believe that metal complexes that are stable only in such special environments control the ED behaviors and the resulting deposits. However, their details are still unclear.

In this study, we will look deeper into the metal complexes formed specifically in HCEs and, at the same time, investigate the metallography and physical properties of the electrodeposits to get comprehensive understanding of the new ED processes (Fig. 1).

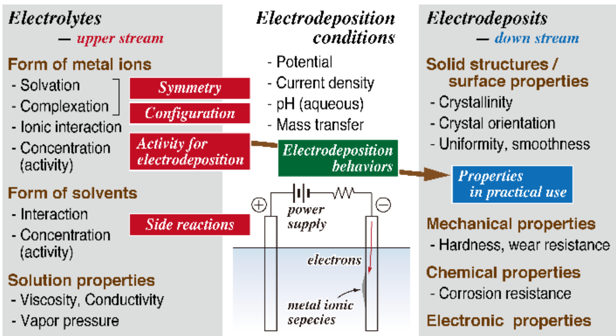

Figure 1 Possible governing factors in ED process

【Research Methods】

This study includes three subjects: (*i*) environmentally friendly electroplating using hydrate melts, (*ii*) ED of Al using glyme-based deep eutectic solvents, and (*iii*) ED of Cu_2O layers using aqueous electrolytes of concentrated α-hydroxyl acids. Co-investigators are Prof. UMEBAYASHI Yasuhiro (Niigata University) and Prof. NAKANO Hiroaki (Kyushu University). The analysis of dissolved species in the electrolytes is performed using vibrational spectroscopy (Raman, FT-IR), UV-visible spectroscopy, nuclear magnetic resonance, and mass spectrometry. Computational chemistry, *e.g.* MO and MD calculations, is employed in addition to the multivariate analysis of the spectra. Crystallographic features of the deposits are analyzed by a variety of electron microscopy techniques (SEM, TEM, EBSD). The data on mechanical, chemical, and electronic properties will also be collected.

【Expected Research Achievements and Scientific Significance】

The HCEs give a new class of solution system different from general aqueous solutions or ionic liquids. The data on the complexes therein, *i.e.* chemical structure (Fig. 2) and their thermodynamics, and on the correlation among the complexes, ED behaviors, and resulting deposits will help us to design HCEs to obtain more sophisticated materials in terms of practical use.

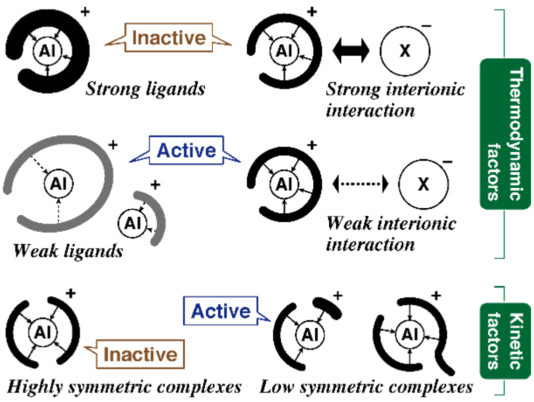

Figure 2 Possible relationship between Al complex in the electrolyte and the resulting ED behavior.

【Publications Relevant to the Project】

- K. Adachi, A. Kitada, K. Fukami, K. Murase, Crystalline Chromium Electroplating with High Current Efficiency Using Chloride Hydrate Melt-based Trivalent Chromium Baths, *Electrochim. Acta*, **338**, 135873/1-8 (2020).
- A. Kitada, K. Nakamura, K. Fukami, K. Murase, Electrochemically Active Species in Aluminum Electrodeposition Baths of $AlCl_3$/Glyme Solutions, *Electrochim. Acta*, **211**, 561-567 (2016).

【Term of Project】 FY2020-2024

【Budget Allocation】 153,000 Thousand Yen

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Title of Project : Development of valley-spin quantum optics in atomically thin artificial hetero-structures

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(Kyoto University, Institute of Advanced Energy, Professor)

Research Project Number: 20H05664 Researcher Number : 40311435
Keyword : Atomically thin material, Photonics, Quantum Optics

【Purpose and Background of the Research】

Here we will tackle to create new research field of "valley-spin quantum optics" leading to novel optical quantum information devices, which is based on the scientific insights combined with atomically thin material science and quantum optics. There is a coupling of degree of freedom between valley in the momentum space and spin in the atomically thin materials (MX_2:M=Mo, W, X=S, Se, Te), called as valley-spin due to breaking of Kramers degeneracy. In our previous studies, the detail physical understanding of valley-spin degree of freedom and continuous control of valley-spin polarization by external-field were successfully realized as important milestones toward the valleytronics. We found the new strategy to control the valley-spin degree of freedom as quantum states. Thus, the research field of "valley-spin quantum optics" based on the control of quantum states will be newly opened.

In this research project, we will construct the scientific framework of "valley-spin quantum optics" with overcoming the conventional quantum optics in the ultimate zero-dimensional (0D) quantum dots by atomically thin hetero-structures. Moreover, we will develop toward the application of "valley-spin quantum optics" as "valley-spin quantum photonics".

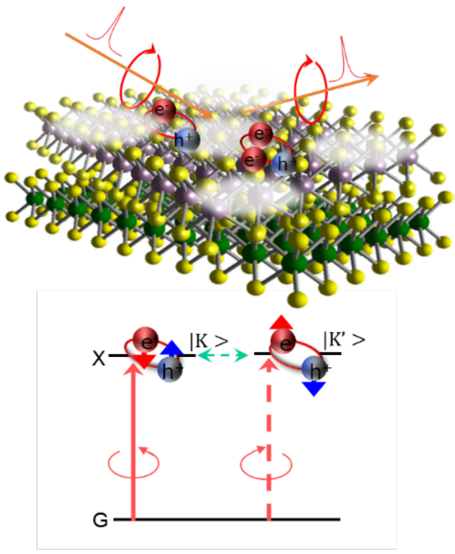

Figure 1 Schematics of atomically thin artificial hetero-structure and valley-spin quantum system

【Research Methods】

According to our studies, the 0D (quantum two-level system) will be realized in the atomically thin hetero-structures by introduction of quantum confined moiré potential. The proposed valley-spin quantum optics are studied as follows, 1) development of fabrication technique of atomically thin artificial hetero-structures and their devices, 2) exploring quantum optical phenomena in the atomically thin artificial hetero-structures, 3) quantum control in the atomically thin quantum dots toward valley-spin quantum optics, 4) application of quantum information devices based on valley-spin control.

【Expected Research Achievements and Scientific Significance】

New routes for the application of optical quantum devices (quantum bit and a single photon source) are expected through the realization of long-term retention of valley-spin quantum coherence and quantum state control. We also expect the novel quantum systems with external interface and controllability of interactions between the quantum bits in the atomically thin hetero-structures, which is much different from other quantum systems. Thus, this project is important not only in the viewpoint of fundamental science but also in the application.

【Publications Relevant to the Project】

· Y. Miyauchi, S. Konabe, F. Wang, W. Zhang, A. Hwang, Y. Hasegawa, L. Zhou, S. Mouri, M. Toh, G. Eda, and K. Matsuda: Evidence for line width and carrier screening effects on excitonic valley relaxation in 2D semiconductors, Nat. Commun. **9**, (2018) 2598.
· K. Shinokita, X. Wang, Y. Miyauchi, K. Watanabe, T. Taniguchi, and K. Matsuda: Continuous control and enhancement of excitonic valley polarization in monolayer WSe_2 by electrostatic doping, Adv. Func. Mater. **29**, (2019) 1900260.
·

【Term of Project】 FY2020-2024

【Budget Allocation】 151,000 Thousand Yen

【Homepage Address and Other Contact Information】

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project： Ferrimagnetic spintronics and device application

ONO Teruo
(Kyoto University, Institute for Chemical Research, Professor)

Research Project Number: 20H05665 Researcher Number：90296749
Keyword：spintronics

【Purpose and Background of the Research】

Spintronics is an attractive field of research where the discovery of fundamental phenomena directly leads to innovation, such as the giant magnetoresistance effect being used for the hard disk read head and the nonvolatile magnetic memory using the tunnel magnetoresistance effect being developed.

Until now, spintronics has been developed by controlling the magnetization of ferromagnetic materials, but the leakage field and the resonant frequency of gigahertz, which are characteristics of ferromagnetic materials, are obstacles to the further development of spintronics. The leakage magnetic field becomes a problem for magnetic memory, and the device operating speed is limited to be up to nanoseconds due to the resonance frequency of gigahertz. To solve these bottlenecks, an antiferromagnet with no leakage magnetic field and resonance frequency in the terahertz region has attracted attention. However, the response of antiferromagnet to the magnetic field is extremely small, leading to the limitation of research methods and applications of antiferromagnets.

Ferrimagnet is a material having a net magnetization while two kinds of magnetic moments are coupled antiparallelly. The magnitude of the magnetization can be adjusted by tuning the composition and/or temperature, and it is possible to realize a situation where total magnetization is completely zero.

We have recently found that the ferrimagnetic GdFeCo alloy behaves as an antiferromagnet with magnetization. The purpose of this study is to construct a new field called "ferrimagnetic spintronics" by clarifying the universality and diversity of the behavior of ferrimagnet as "antiferromagnet with magnetization", and to seek the possibility of device applications.

【Research Methods】

In order to achieve the objectives, we executes the following research items.
(1) Universality and diversity of behavior of ferrimagnet as antiferromagnet with magnetization
[1-1] Confirmation of the increase in the domain wall propagation speed at the angular momentum compensation temperature
[1-2] Universality of relation between angular momentum compensation temperature, magnetization compensation temperature, and Curie temperature.
[1-3] Spin damping of ferrimagnet
[1-4] Dzyaloshinskii-Moriya interaction induced by composition modulation
(2) Development of device application utilizing the characteristics of antiferromagnet with magnetization
[2-1] Deployment to the skyrmion device
[2-2] Development of terahertz spintronics
[2-3] Polarization control of antiferromagnetic spin wave

【Expected Research Achievements and Scientific Significance】

It is expected that the operation of the device which utilizes the excellent features of the antiferromagnet proposed so far can be demonstrated by using the ferrimagnet. Furthermore, it is possible to deploy the novel devices that cannot be realized by the antiferromagnet by utilizing the feature of ferrimagnets that spin dynamics can be easily excited by a magnetic field.

【Publications Relevant to the Project】

- Takaya Okuno et al., “Spin-transfer torques for domain wall motion in antiferromagnetically coupled ferrimagnets”, Nature Electronics 2, 389 (2019).
- Duck-Ho Kim et al., “Bulk Dzyaloshinskii–Moriya interaction in amorphous ferrimagnetic alloys”, Nature Materials 18, 685 (2019).
- Yuushou Hirata et al., “Vanishing skyrmion Hall effect at the angular momentum compensation temperature of a ferrimagnet”, Nature Nanotechnology14, 232 (2019).
- K.-J. Kim et al., “Fast Domain Wall Motion in the Vicinity of the Angular Momentum Compensation Temperature of Ferrimagnets”, Nature Materials 16, 1187 (2017).

【Term of Project】 FY2020-2024

【Budget Allocation】 153,200 Thousand Yen

【Homepage Address and Other Contact Information】

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project：　Spintronics based on the Information thermodynamics

SUZUKI Yoshishige
(Osaka University, Graduate school of Engineering science, Professor)

Research Project Number:　20H05666　　Researcher Number：50344437
Keyword：Magnetic skyrmion, Information thermodynamics, Spintronics

【Purpose and Background of the Research】

In this research, the concept of "information flow (to be exact, called transfer entropy)" will be introduced into the field of spintronics, which has been focusing on the generation, control, and conversion of "spin currents". By that way, a new field of the spintronics that is based on the information thermodynamics will be established. The new science will be a base to design highly intelligent and energy saving spintronic information devices and systems. (see Fig. 1).

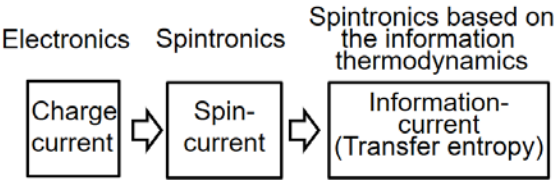

Fig. 1 Paradigm shift from spin current to spintronics that handles information flow (transfer entropy).

【Research Methods】

Specifically, we will realize an information heat engine that uses the thermal motion of the magnetic skyrmion (see Fig. 2), clarify the performance of spintronics elements from an information thermodynamic point of view, and reduce the energy consumption required for the information heat engine. We will pursue ultra-low energy sensing technology. Furthermore, as applications, we will demonstrate the operation of an ultra-low energy consumption spintronics computer. (See Figure 3)

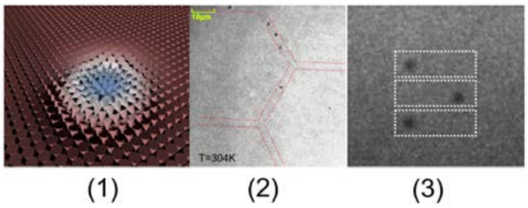

Figure 2 (1) Schematic image of the magnetic structure of the magnetic skyrmion. The vertices of the small cones indicate the direction of magnetization in the thin film (direction of N-pole). (2) Skyrmion channel created using local potential control. Skyrmions smoothly pass through the three-way junctions by Brownian motion without being trapped. (3) Interaction between skyrmions confined in rectangular wells. Skyrmions oscillate between equivalent energy states.

【Expected Research Achievements and Scientific Significance】

Our goal is to elucidate a possibility to realize IT equipment that has the same energy efficiency as biological systems and can process information as much as the human brain by expanding and developing the theory and technology of the spintronics.

In this research, there is engineering significance in creating elements and systems that approach the thermodynamic limits, using information thermodynamics as a guiding principle, rather than simply applying information thermodynamics to the spintronics.

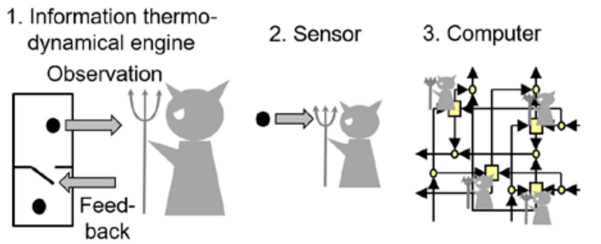

Figure 3　Three R & D subjects of the spintronics based on the information thermodynamics.

【Publications Relevant to the Project】

[1] “Skyrmion Brownian circuit implemented in continuous ferromagnetic thin film”, Yuma Jibiki, Minori Goto, Eiiti Tamura, Jaehun Cho, Soma Miki, Ryo Ishikawa, Hikaru Nomura, Titiksha Srivastava, Willy Lim, Stephane Auffret, Claire Baraduc, Helene Bea, and Yoshishige Suzuki, Applied Physics Letters, 117, 082402 (2020).

[2] “Theory of Skyrmionic Diffusion: Hidden Diffusion Coefficients and Breathing Diffusion”, E. Tamura, Y. Suzuki, arXiv：1907.06926.

【Term of Project】 FY2020-2024

【Budget Allocation】 147,400 Thousand Yen

【Homepage Address and Other Contact Information】

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Title of Project：Whole gamma imaging to break through the physical limitation of positron emission tomography

YAMAYA Taiga
(National Institutes for Quantum and Radiological Science and Technology, National Institute of Radiological Sciences, Group Leader)

Research Project Number: 20H05667 Researcher Number：40392245
Keyword：PET, Compton camera, SPECT, nuclear medicine, scintillator

【Purpose and Background of the Research】

Positron emission tomography (PET), which uses a diagnostic drug labelled by a trace amount of weak radioactivity, has become a standard method for cancer diagnosis (figure 1), but PET does not make full use of detectable gamma-rays for imaging. Therefore, we aimed at replacing PET with whole gamma imaging (WGI), which is our original idea to utilize all detectable gamma-rays, for earlier diagnosis of intractable cancers such as multiple myeloma.

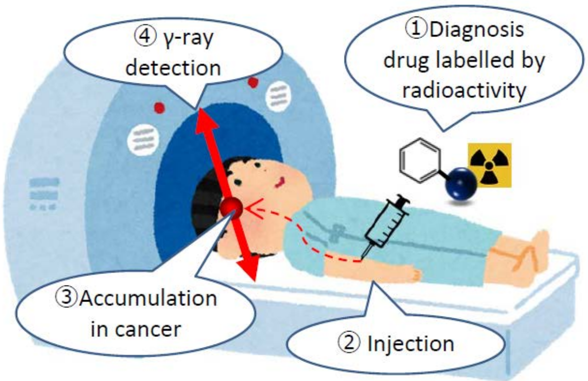

Figure 1 Schematic of the nuclear medicine imaging concept.

【Research Methods】

In WGI, an additional detector ring is inserted in a conventional PET ring so as to add a Compton camera function to a PET system. In addition to the coincidence detection of a pair of 511 keV photons, all other gamma-rays can be detected by means of the Compton camera, which localizes a radioisotope on a surface of a cone. Key methods to be studied in this project are (1) replacing a typical PET radioisotope such as ^{18}F (110 min half-life) in a radiopharmaceutical by an unusual radioisotope such as ^{89}Zr (3.3 d half-life), which emits a 909 keV gamma-ray in addition to a positron; (2) hybrid imaging of PET and Compton imaging; and (3) changing the principle of cancer diagnosis from glucose metabolism to an antigen-antibody reaction (figure 2).

【Expected Research Achievements and Scientific Significance】

There are three step-by-step goals in this project. First, a WGI system in which 909 keV Compton imaging shows better spatial resolution than PET will be developed. For this goal, a new scintillator which has better energy resolution than conventional PET scintillators will be developed. Second, a new imaging algorithm to combine both PET data and Compton imaging data will be developed so as to improve image quality. Third, diagnosis of multiple myeloma will be investigated as a clinical output of this project. Imaging demonstration of model mice will be done with the developed WGI system.

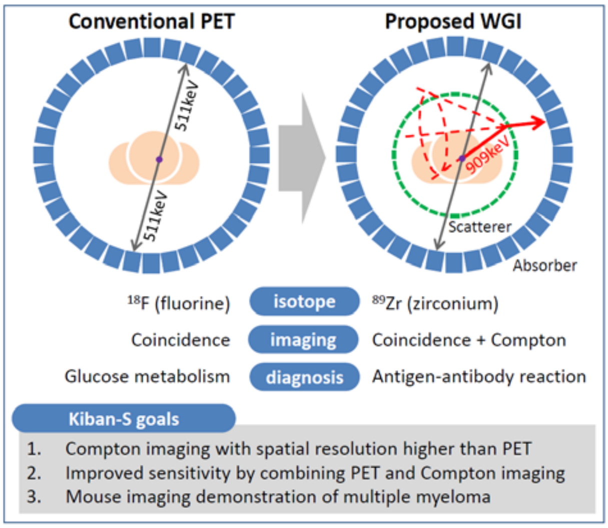

Figure 2 Schematic of whole gamma imaging (WGI) concept and the study.

【Publications Relevant to the Project】

・T. Yamaya, E. Yoshida, H. Tashima, et al., “Whole gamma imaging (WGI) concept: simulation study of triple-gamma imaging,” J. Nucl. Med., vol. 58, no. supplement 1, 152, 2017.
・E. Yoshida, H. Tashima, K. Nagatsu, et al., "Whole gamma imaging: a new concept of PET combined with Compton imaging," Phys. Med. Biol., 65, 125013, 2020.

【Term of Project】 FY2020-2024

【Budget Allocation】 151,900 Thousand Yen

【Homepage Address and Other Contact Information】

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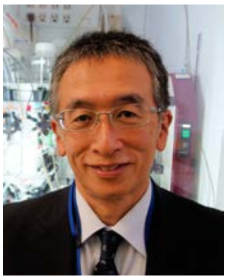
【Grant-in-Aid for Scientific Research (S)】

Broad Section D



Title of Project：Creation and application of perfect structure carbon nanotubes

KATAURA Hiromichi
(National Institute of Advanced Industrial Science and Technology, Department of Materials and Chemistry, Visiting Scientist)

Research Project Number: 20H05668　Researcher Number：30194757
Keyword：Carbon nanotube, Structure sorting, Healing

【Purpose and Background of the Research】

Carbon nanotubes (CNTs) were discovered in Japan in 1991 as multi-walled and in 1993 as single-walled. Single-wall CNTs are tubular structures with a diameter of about one nanometer made of a single atomic layer carbon. Due to their excellent physical and electrical properties, it is expected to be applied in various fields including electronic devices. In order to achieve this, our previous work has enabled the automatic sorting of 20 different structure single-wall CNTs from as grown mixture. By combining with the mass CNT synthesis method, it has become possible to easily obtain single-wall CNTs with controlled structures, but enough performance has not yet been obtained for practical use. In recent years, it has become clear that one of the causes is "defects". Single-walled CNTs are composed of a network (network) in which carbon atoms are covalently bonded and have a structure in which all atoms are located on the surface. It is not easy to build this network perfectly, and the single-wall CNTs currently available contain many defects. The purpose of this research project is to create CNTs that do not contain "defects" and to bring out the original excellent physical properties of CNTs.

【Research Methods】

In this research, we will utilize a new technology that can separate low-defect CNTs simply by pouring them into a column (see Fig. 1). With this technology, it is possible to sort low-defect CNTs and quantitatively investigate the defect density distribution of raw CNTs. By thermally and chemically treating the raw material CNT, it is possible to precisely investigate how much defect repair has progressed. Using this high-sensitivity defect detection method, we will try to heal CNT defects, which was difficult until now. We aim to realize perfect structure CNTs by selecting those with low defects from them.

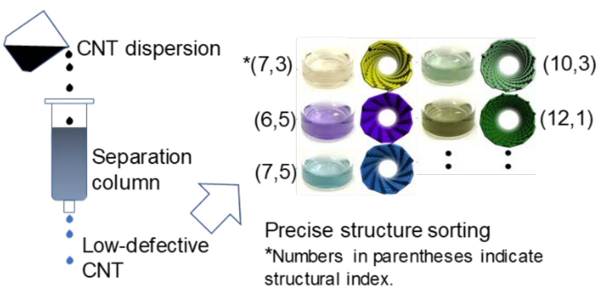


Figure 1　Separation of CNTs

【Expected Research Achievements and Scientific Significance】

The research results of CNTs obtained so far have been for CNTs containing many defects. By eliminating defects, it will be possible to bring out the amazing physical properties of CNTs, and rapid application development such as electronic devices is expected. As a high luminous efficiency fluorescent material in the near infrared region with high bio permeability, it can be expected to be applied to pathological research by bioimaging.

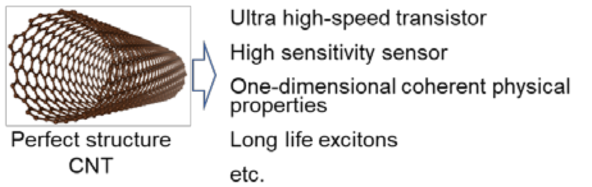


Figure 2　Research and application of perfect CNT

【Publications Relevant to the Project】

- Y. Yomogida *et al.* “Industrial-scale separation of high-purity single-chirality single-wall carbon nanotubes for biological imaging”, Nat. Commun. **7**, 12056 (2016).
- X. Wei *et al.* “Experimental determination of excitonic band structures of single-walled carbon nanotubes using circular dichroism spectra”, Nat. Commun. **7**, 12899 (2016).
- H. Liu *et al*. “Large-scale single-chirality separation of single-wall carbon nanotubes by simple gel chromatography”, Nat. Commun. **2**, 309 (2011).

【Term of Project】 FY2020-2024

【Budget Allocation】 151,300 Thousand Yen

【Homepage Address and Other Contact Information】

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【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project： Creation of Neurophotonics and Elucidation of Brain Funcitons

NEMOTO Tomomi

(National Institute of Natural Sciences, Exploratory Research Center on Life and Living Systems (ExCELLS), Professor)

Research Project Number: 20H05669 Researcher Number：50291084

Keyword：Bioimaging, neuroscience, laser

【Purpose and Background of the Research】

The question "what kinds of neural cells' collective activity realize our mental activity" always fascinates many people as an eternal object. To understand the emergent and the operating principles of brain function, functions of local neural circuits and its true nature -propagation and synchronization of neural activity between cells - are critically required to be clarified under a truly intact condition (*in vivo*). The collective action with network synchronization is deterministically crucial for realizing the function of local neural circuits. However, information transmission at neural synapses occurs stochastically at the molecular and cellular levels. To integrally understand brain function beyond this divergence, a cutting-edge method must directly visualize the population activity and quantitatively analyze the transmission process as it is.

【Research Methods】

The principal investigator has promoted *in vivo* two-photon excitation microscopy to measure the living brain and nervous system worldwide. Based on this, we will utilize optical technologies such as high-power compact laser light sources with variable wavelength, adaptive optics, and second harmonic generation. We are going to finally realize the world's first high-speed super-resolution optical imaging that visualizes biomolecule activities within deeper layers in biological tissues and morphological changes of living cells less-invasively.

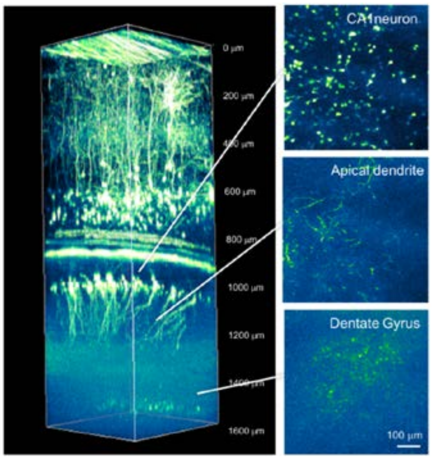

Fig.1: *in vivo* observation of mouse living brain

Using this innovative microscope, we will accurately visualize and analyze synchronous neuronal population activity and neurotransmitter exocytosis in the deep part of the mouse living brain in the "as is" state. Furthermore, we will track three-dimensional morphological changes of nerve cells and the dynamics of exocytosis in real-time, which will lead to an understanding of the signal transmission mechanism by the interaction between nerve cells and glial cells and the emergent principle of brain function.

【Expected Research Achievements and Scientific Significance】

This project will develop and improve novel microscopy, leading us to understand the essence of information transmission in the brain by analyzing multiple neural cell responses and synchronous changes in collective activity. The microscopy enables super-resolution imaging for revealing the nature of neurotransmission, incredibly deep in living organs. It will visualize biomolecular dynamics during the exocytotic process from the accumulation of exocytotic molecules at presynaptic terminals, neurotransmitter release at synapses to response in the posterior region, without damaging the neural circuits in the living brain. Besides, by combining it with localized photoactivation and/or administration of drugs, it will lead to developing a route leading to the elucidation of the molecular basis and treating diseases such as mental illness and diabetes.

As described above, the new "neurophotonics" promoted by this research project will contribute to life science innovations such as control of physiological functions by light and photocell therapy by advancing deep-body imaging.

【Publications Relevant to the Project】

・ M. Inoue, *et al.*, “Rational engineering of XCaMPs, a multicolor GECI suite for in vivo imaging of complex brain circuit dynamics”, ***Cell***, **177**:1346-1360.e24 (2019)
・ K. Yamaguchi, *et al.*, “In vivo two-photon microscopic observation and ablation in deeper brain regions realized by modifications of excitation beam diameter and immersion liquid”, ***PLoS ONE***, (2020)

【Term of Project】 FY2020-2024

【Budget Allocation】 153,800 Thousand Yen

【Homepage Address and Other Contact Information】

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Title of Project : Petaherz-scale solid state physics exploring by attosecond high-harmonic-based ultrafast spectroscopy

OGURI Katsuya
(NTT Basic Research Laboratories, Quantum Science and Technology Laboratory, Executive manager)

Research Project Number: 20H05670 Researcher Number : 10374068
Keyword : Petahertz electronics, Attosecond physics, Lightwave-driven phenomena, 2D materials

【Purpose and Background of the Research】

The three innovative optical technologies, which were invented in the early 21st century, attosecond pulse generation technology, optical clock technology, and carrier-envelope phase stabilization technology have allowed light to be measured in the time domain on a 10^{-18} second scale and to be stabilized in the frequency domain with a 10^{-18} fractional uncertainty. The technological progress has currently enabled us to utilize a light wave as a precisely controllable electromagnetic wave with an extremely high frequency approaching 1 petahertz (PHz: 10^{15} Hz) or "PHz wave".

In this project, we will explore ultrafast dynamics of the electronic coherent responses and nonequilibrium relaxation induced by an interaction between a lightwave electric field and internal quantum degrees of freedom of electrons such as polarization, spin, and valley on a time scale from 100 as to 10 fs. We call this time scale, which corresponds to one cycle of light wave from visible to infrared, as “PHz scale”. By investigating with both experimental and theoretical approaches, we will pioneer a new framework of “PHz-scale solid state physics”.

【Research Methods】

In this project, we investigate an ultrafast lightwave-electron interaction in various 2D materials including semiconductors, ferromagnets, and topological insulators, which show characteristic band structures, spin and magnetic properties, and topological orders. In order to proceed this research comprehensively, we will combine the following three approaches of (i) development of new extreme attosecond spectroscopic systems, (ii) high quality 2D-materials growth and characterization, and (iii) the first-principles computation and real-time quantum simulation for solid state materials.

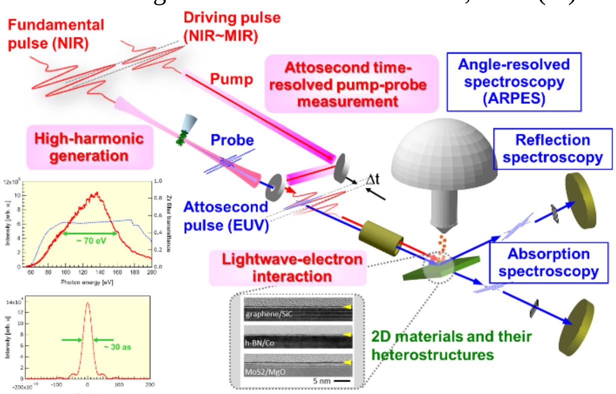

Figure 1. Schematic illustration of an extreme attosecond spectroscopic system for measuring solid state materials.

For the new attosecond spectroscopic techniques, we will develop a high-repetition-rate system with a frequency of from 0.1 to 1 MHz to improve the quality of data by increasing the data acquisition. The high-repetition attosecond time-resolved system will be combined with not only reflection and absorption spectroscopy but also angle-resolved photoelectron spectroscopy (ATTO-ARPES), and a magneto-optical Kerr effect (ATTO-MOKE). By applying these techniques to various 2D materials and their layered structures and heterostructures, we explore various ultrafast interaction between lightwave and electronic polarization, spin, and valley. In addition, we seek a better understanding of these experimental observations by theoretical approach such as the time-dependent density-matrix (TD-DM) method.

【Expected Research Achievements and Scientific Significance】

This research opens a new scientific field of “PHz-scale solid state physics”, which provide a framework beyond the usual approximations such as the envelope approximation and the rotating wave approximation assumed in the conventional optical solid state physics. The PHz-scale solid state physics will pave the way for the breakthrough in the novel ultrafast electronic functionalities with unprecedented operational speed controlled by light-wave field.

【Publications Relevant to the Project】

· K. Oguri, H. Mashiko, T. Ogawa, Y. Hanada, H. Nakano, and H. Gotoh, “Sub-50-as isolated extreme ultraviolet continua generated by 1.6-cycle near-infrared pulse combined with double optical gating scheme,” Appl. Phys. Lett. 112, 181105 (2018).
· H. Mashiko, K. Oguri, T. Yamaguchi, A. Suda, and H. Gotoh “Petaheltz optical drive with wide-bandgap semiconductor,” Nature Physics 12, 741 (2016).

【Term of Project】 FY2020-2024

【Budget Allocation】 154,900 Thousand Yen

【Homepage Address and Other Contact Information】

http://www.brl.ntt.co.jp/J/group_010/group_010.html