Progress reports of groups

- Information (Storage) Group
- Information (Quantum Information) Group
- Energy Group
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- Supramolecules and Self-Assembly Group
- Nanomaterials and Nanodevices Group
- Quantum Theory and Computation Group

Information Storage Group

Katsuji Nakagawa*, Akiyoshi Itoh , Arata Tsukamoto

1. Overview of the research plan in 2010

1) Preparation to measure the surface plasmon response to femto-second laser pulse
2) Simulation method for the analysis of the surface plasmon response
3) Surface plasmon antenna to create circularly polarized light
4) Preparation to measure the magnetic and optical response of material within sub-picosecond time resolution by femto-second laser pulse
5) Excitation of laser induced magnetization reversal phenomena in TbFeCo thin film
6) Preparation of a thin nano-porous SiO_2 film and application as etching musk of metallic films
7) Introduction of vacuumed etching chamber for fabrication of nano structured structure

2. Advances and achievements in 2010

1) Femto-second laser and the optics have been designed and constructed to analyze the surface plasmon response.
2) Electro-magnetic transit analysis method for nano-scale dispersion materials has been developed by the cooperation with Associated Professor Ohnuki.
3) We have studied some surface plasmon antennas to find a antenna, which can create circularly polarized resonance mode. It was revealed that some antennas had a resonance mode for circularly polarized light.
4) All-optical time resolved measurement system was constructed with femto-second pulse laser. Magnetic and optical response from material was monitored by the magneto optical effect and the change of reflectivity caused by heating of electrons, respectively. Ultrafast demagnetization phenomena were observed which must be caused by heating of electron system.
5) Perpendicular magnetized TbFeCo thin film consisted from rare earth and transition metal alloy was prepared. We clarified that the magnetization direction of TbFeCo can be switched by irradiation of circularly polarized ultra short pulsed laser without magnetic field.
6) We have succeeded preliminarily to prepare thin SiO_2 layer on metallic films which have two dimensional closed packed nano-pores prepared by the polymer micelles technique. As another type of nano structured template having a conjugate relation with nano-pore structure, nano convex pattern was prepared by using self-assembled spherical silica particles followed by inductively coupled plasma reactive ion etching.
7) We prepared test batches of Si and metallic film by Ar ion etching and confirmed the etching rates satisfied our study.

3. Collaborations and activities in 2010 as the group

We had one or two meetings every month with Associated Professor Ohnuki to find a method to calculate a surface plasmon response. The results were reported at IEEE International Symposium on Antennas and Propagation, July 11-17, 2010, Toronto, Ontario, Canada. We also invented a new antenna structure to confine a circularly polarized light, and submitted a patent. We also reported this result at 55th Annual Conference on Magnetism & Magnetic Materials, 14-18 November 2010, Atlanta, Georgia, USA.

Information (Quantum Information) Group

Shuichiro Inoue,* Takeshi Kuwamoto, Hideomi Hashiba

1. Overview of the research plan in 2010

We will construct a quantum key distribution (QKD) system based on BBM92 protocol using high-speed single-photon detectors and the cross-polarization-entangled photon pair source we developed last year. Using the system, we will demonstrate 200 km QKD. And also, we will demonstrate the entanglement swapping using the detectors and two cross-polarization-entangled photon-pair sources. Moreover, we will try to store a cross-polarization-entangled photon pair resonant with a transition in rubidium atoms using electrically induced transparency. On the other hand, we will fabricate Au nano-stripe waveguides and 50/50 couplers to investigate physical properties of surface plasmon polaritons.

2. Advances and achievements in 2010

We demonstrated the distribution of a cross-polarization-entangled photon pair which is generated using a Type-II PPLN waveguide. To achieve a high-rate distribution of entangled photon pairs, we used the high-speed and efficient single photon detector based on the sinusoidally gated InGaAs/InP avalanche photodiodes (SG-APDs). The measured two photon interference visibility was 97 %. Moreover, the entanglement distribution rate exceeded 2.8 kilobit per second. This value is the highest distribution rate in the entanglement distribution experiment at telecommunication wavelength to date.

Surface Plasmon polaritons have attracted much interest in the research area of quantum information technologies because of their enhanced electric field and efficient coupling with nanoscale electronic systems. We fabricated low loss surface plasmon polariton straight waveguides using ZPU12 polymer. The coupling loss and propagation loss of the waveguide were 0.81 dB and 1.7 dB / mm, respectively. Moreover, we fabricated a 50:50 coupler using the low loss surface plasmon polariton waveguides. The insertion loss of the coupler was approximately 25 dB.

Photon-number resolving detectors are indispensable requirements for many quantum information applications. We have developed titanium-based transition edge sensors (Ti-TESs) to improve the response speed of the sensor. We fabricated Ti-TESs with the mult-layerd absorption structures, which consist of a 20-layer dielectric mirror and a 7-layer AR coating. The size and the thickness of the Ti film are 10 μm×10 μm and 22 nm, respectively. The detection efficiency at 844 nm was 98.4 % which is the highest detection efficiency reported for an optical photon detector.

A superconducting nanowire single photon detector (SNSPD) is one of the promising candidates for a single-photon detector at 1550 nm because of their low timing jitter and dark counts. The Nb film with 3 nm thin alumina has a critical temperature at 6.9 K and its critical current density is 4.4 MA/cm^2, which are much higher than those of previous results obtained by uncoated Nb films. We have fabricated a SNSPD using the Nb film. Limiting the dark count to 100 Hz, the detection efficiency of the SNSPD at 850 nm and 1550 nm are 3 % and 3×10^{-3} %, respectively.

We constructed the fluorescence microscope system for the spin-coated colloidal quantum dots. Using the total internal reflection setup and the high sensitive CCD camera, we achieved the imaging of the fluorescence from a single CdSe/ZnS quantum dot. We are now concentrating on the Hanbry-Brown Twiss experiment in which we can investigate the photon statistics of the fluorescence.

3. Collaborations and activities in 2010 as the group

We have collaborated with Otsuki Laboratory on single-photon generation from a CdSe/ZnS quantum dot. We also have collaborated with Ohnuki Laboratory on the designing of superconducting nanowire single-photon detector. We had three group meetings in which we reported individual research activities and discussed the next direction we should take.

Energy Group

Takuya Hashimoto*, Yasuo Asada, Joe Otsuki, Nobuyuki Nishimiya, Sachiko Matsushita

1. Overview of the research plan in 2010

1) In order to improve energy conversion efficiency of solar cell, preparation of a dye-sensitized photonic-crystal electrode with a full-photonic band gap is continuously examined using an electron-beam lithography. Development of new dyes is also examined. Pore-size dependence of the photovoltaic characters and electrochemical impedances of dye-sensitized TiO_2 inverse opals electrodes is also elucidated.
2) For innovation of technique to generate H_2 from H_2O with photo energy, improvement of efficiency of supra-molecule catalyst is challenged.
3) In order to use H_2 as a clean energy source, generation of H_2 employing biotechnology, preparation of materials for H_2 storage and generation, or solid oxide fuel cells are tried. The aim of operation temperature of solid oxide fuel cells is 600 ºC, at which enough energy conversion efficiency can be obtained and long term endurance is realized.

2. Advances and achievements in 2010

1) Three kinds of TiO_2 inverse opals using different-size polystyrene particles were prepared and the electrochemical characteristics of the dye-sensitized inverse-opal electrodes have been measured. The large diffusion resistance of the electrolyte suggests that the improvement of the photoelectric conversion efficiency was not caused by nano/mesoscopic effect because nano/mesoscopic effect should be appeared as small diffusion resistance.

As a new material for dye sensitized solar cells, three kinds of new dye have been prepared and solar cell employing prepared dye is fabricated. Although efficiency is as low as 0.5-1.9 %, it can be improved by optimization of photo absorption, electrical conduction property.
2) The development of new homogeneous photo catalyst for generation of H_2 from H_2O which can be worked in aqueous solution has been succeeded. At present, number of turn-over has reached to 1300 for photo irradiation of 2 hours.
3) It has been discovered that H_2 generation by biotechnology can be improved by coexistence of H_2 absorption alloys. H_2 generation from $LiBH_4$ can be controlled by irradiation of ultraviolet light with addition of WO_3 as a photo catalyst and CuO.

The apparatus for measurement of mechanical strength at high temperature under various gas atmospheres has been developed. *In-situ* measurements of mechanical strength of materials for solid oxide fuel cells will be performed.

For development of new proton conducting materials, preparation of $Ba_{1-x}Sr_xZrO_3$ via liquid phase has been examined. It has been revealed that specimen with x=0.4 has superior property with high sintering property, conduction property and no structural phase transition. As a new electrode material, we have clarified that $LaNi_{0.6}Fe_{0.4}O_3$ has excellent property from the viewpoints of electrical conduction, thermodynamic stability and sintering property. Fabrication of new solid oxide fuel cells employing thus discovered materials are now in progress.

3. Collaborations and activities in 2010 as the group

Pore-size dependence of the photovoltaic characters and electrochemical impedances of dye-sensitized TiO_2 inverse opals electrodes is a collaboration work of Dr. Matsushita and Dr. Hashimoto, College of Humanities and Sciences (one journal paper, submitted). This research was also awarded a poster prize. Improvement of H_2 generation from biotechnology using H_2 storage alloys is collaboration work of Dr. Asada and Dr. Nishimiya.

Medical Group

Noboru Fukuda*, Katsumi Abe, Satoru Takahashi, Tsugumichi Koshinaga, Kyoko Fujiwara, Koichiro Kano, Noriaki Matsumoto, Hiroki Nagase

1. Overview of the research plan in 2010

We made 3 research plannings. (1) Developing probes for Her2, molecular marker to diagnose cancer early, and MYCN, then labeling those probes with luminescent chemicals or radioisotopes to generate probes for cancer diagnostic imaging. (2) Drug discovery of pyrrole-imidazole (PI) polyamides based on chemical genomics. (3) Developing new radio sensitizer by labeling porphyrin compound with iodine

2. Advances and achievements in 2010

(1) We have been trying to down regulate MYCN gene by using PI polyamide. PI polyamide molecules are composed with aromatic amino acides *N*-methylpyrrole (Py) and *N*-methylimidazole (Im), can bind to DNA in sequence specific manner. By inhibiting the binding of transcription factor, PI polyamide can change expression level of specific genes. We developed PI polyamides, which can bind to binding site of E2F and SP1 on the promoter region of MYCN, and found that those polyamides could down regulate MYCN expression and reduce the growth rate of NB cell line CHP134. Those polyamides also showed growth inhibiting effect on the xenograft made by hypodermic injection of CHP134 to mice.

(2) 1) PI polyamide conjugated with Histone deacetylase (HDAC) inhibitor has been developed also. Since aberrant methylaion and/or aberrant histone acetylation around the promoter region of tumor suppressor genes have been observed in cancer tissues frequently, we combined HDAC inhibitor SAHA with p16 polyamide, which can bind to p16 tumor suppressor gene, to recover expression level of p16 in cancer cells. It was confirmed that the conjugated molecule still kept SAHA activity and could induce acetylation of histone H3 around the target sequence. This molecule was also shown to be able to reduce the growth rate of human cervical cancer cell line. 2) Effects of PI polyamide targeting TGF- β1 on EPS. To develop PI polyamide targeting TGF-β1 promoter (Polyamide) as a new therapeutic medicine for encapsulating peritoneal sclerosis (EPS). To create an animal model of peritoneal sclerosis, rats were given a daily intraperitoneal injection of chlorhexidine gluconate and ethanol (CHX) for 14 days. One mg of Polyamide was ip injected once or 3 times. Polyamide significantly suppressed thickness of peritoneum in the injection-time dependent manner. Lead optimization of PI polyamides targeting human TGF-β1. 3) We synthesized seven PI polyamides targeting human TGF-β1 on sis-elements of human TGF-β1 promoter. We chose three lead optima such as GB1101, GB1105, GB1106 by inhibition of expression of TGF-β1 mRNA. 4) Induction of human iPS cells with PI polyamide targeting human TGF-β1. We started a collaborating project to induce human iPS cells by PI polyamide targeting human TGF-β1 with Florida University and Nihon Univ. Coll. Bioresource Sci.

(3) The porphyrin compound HPPH (3-(1`-hexylocyethyl)-3-devinylpyropheophorbide-a), which is photosensitizer and accumulated in cancer tissues preferentially rather than in normal tissues, has been used in photodynamic therapy (PDT). We conjugated this molecule with iodine, which is used as a contrast agent in X-ray imaging, to develop radio sensitizer compound 531 and 717. Human bladder cancer cell line T24 treated with 531 or 717, followed by irradiation of 33.17KeV parametric x-ray, showed significantly lower viability compared with the cells irradiated without preceding 531, 717 treatment. Now we are trying to confirm those results in vivo using mouse xenograft model.

3. Collaborations and activities in 2010 as the group

We have had a joint meeting once a month to share the results and discuss in School of Medicine. All of the members have been interacting very well, sharing information, equipment and reagents etc. Fukuda became Medical leader of N. Project from Sept. 2010 instead of Nagase. The collaborating project to induce human iPS cells by PI polyamide targeting human TGF-β1 with Florida University and Nihon Univ. Coll. Bioresource Sci.

Supramolecules and Self-Assembly Group

Hiroki Ikake* , Akiyoshi Itoh, Joe Otsuki, Arata Tsukamoto, and Sachiko Matsushita

The goal of the supramolecules and self-assembly group is to develop advanced technologies on nanomaterials and nanostructures and to supply these technologies to the application-oriented groups, *i.e.*, the information, energy, and medical groups, thus strongly promoting networking among these groups on diverse fields. As follows, each groups theme in 2010.

· Itoh & Tsukamoto Group

1. Overview of the research plan in 2010

Preparation of a thin nano-porous SiO_2 film and application as etching musk of metallic films

2. Advances and achievements in 2010

We have succeeded preliminarily to prepare thin SiO_2 layer on metallic films which have two dimensional closed packed nano-pores prepared by the polymer micelles technique. As another type of nano structured template having a conjugate relation with nano-pore structure, nano convex pattern was prepared by using self-assembled spherical silica particles followed by inductively coupled plasma reactive ion etching.

· Otsuki Group

1. Structures and dynamic behaviors of molecular self-assemblies at the molecular level
2. Light-driven hydrogen evolution from water with self-assembled complexes
3. Preparation of compounds for the X-ray based photodynamic therapy

· Matsushita Group

1. Overview of the research plan in 2010

The applications as a metamaterial and as a template for single-cell analysis were examined using self-assembled particle structures. The activation of the spontaneous movement by chlorobenzene addition occurred at nitrobenzene/water interface was elucidated by interfacial tension measurement and solubility measurement.

2. Advances and achievements in 2010

Recently, the application of a self-assembled spherical-particle structure to hydrogels is actively studied. The application is mainly as sensors, for example, as a glucose sensor and temperature sensor. In this research, with the viewpoints of the applications as a cell culture substratum, a micro-pocket gel array was prepared by templating of a two-dimensional self-assembled structure composed of cell size particles.

3. Collaborations and activities in 2010 as the group.

This work is a part of collaboration with Prof. Kano, College of bioresource science.

· Ikake Group

1. Development of Poly(lactic acid) Films with Exhibiting the Piezoelectricity
2. Preparation of Poly(lactic acid)/Silica, and /Carbon Nanotube Hybrid Materials
3. Preparation of Stereo-complex type Poly(lactic acid) Films

Nanomaterials and Nanodevices Group

Kaoru Suzuki*, Yoshiki Takano, Tomohiko Asai, Nobuyuki Iwata, Hideomi Hashiba, Shigeru Chaen and Shosuke Mochizuki

1. Overview of the research plan in 2010

This group aims at fabrication of nanomaterials and nanodevices for high functional applications such as 1) Quantum dot single-photon terahertz detector by Fe-based superconductor films, 2) Magnetic probe of Scanning probe microscope by Ni, etc. encapsulated carbon nanotubes and semiconducting single-walled carbon nanotubes by controlling specific chirality for field effect transistor, 3) Photocatalytic La,Sr,Ca:TiO2 films for hydrogen generation, storage and oxygen (hydrogen) storage and release with metal oxides (metal hydrides) nanoparticles, 4) New photo-memory by highly-photoluminescent material (NiO-ZrO_2 solid solutions) and in-vitro single molecule imaging of these proteins by membrane receptors.

2. Advances and achievements in 2010

Prof. K. Suzuki approached synthesis of ferromagnetic metal (Ni, Fe, and Co) encapsulated carbon nanotubes for probe of magnetic force microscope. The diameter and length of the metal core is in the range of 10 – 80 nm and 100 – 800 nm, respectively. The walls consist of cylindrical graphene sheets with 3 -50 layer. The hydrogen yield on water decomposition with the TiO:La,Sr,Ca films by photo catalytic reaction which was measured to yield about 16 m*l*/h・cm^2 hydrogen gas under visible light irradiation.

Prof. Y. Takano has mainly studied the Fe-based superconductors. He has firstly prepared $Sr_{1-x}Nd_xFFeAs$ and obtained T_c=49 K for x=0.5. This value is close to the highest value in $Sr_{1-x}R_xFFeAs$ (R= rare earth ions). He has also prepared $SrFFe_{1-x}Ni_xAs$ where Fe sites are directly substituted by Ni ions. Li_xFeAs itself is a superconductor with T_c=18 K. He has prepared $Li_xFe_{1-y}Co_yAs$ where Fe sites are directly substituted by Co ions.

Asso. Prof. T. Asai has developed a magnetized coaxial plasma gun for rapid generation of TiFe Series thin-film. Preliminary experiments have been performed with stainless steel electrode and deposited metal thin-film has been confirmed by Prof. Nishimiya. He has also been developed an electrode with composite metallic material for generation of TiZrFeMn film.

Asso. Prof. N. Iwata has studied the selective growth of single-walled carbon nanotubes (SWNTs) with specific chirality controlled by irradiating the FEL. The semiconducting SWNTs grew only the area between electrodes. Expected superlattice structure was [7units CFO / 7units LFO]×14. From the results of low angle x-ray reflection from the surface, superlattice structures were [CFO(1.16nm) / BFO(1.30nm)]×14 and [SFO(1.36nm) / BFO(1.00nm)]×14.

Asso. Prof. H. Hashiba has studied the "Single-electron transistor". His sensitive THz range single photon detector is assembled from a GaAs/AlGaAs quantum dot, electron reservoir and superconducting single-electron transistor. The quantum dot is isolated from the surrounding electron reservoir in such a way that when the excited plasma wave decays, an electron could tunnel off the dot to the reservoir. The detector shows high noise equivalent power of ~10^{-19} $WHz^{-1/2}$.

Prof. S. Mochizuki has studied the photoluminescence properties of various kinds of metal oxides under oxygen gas, vacuum and CO_2 gas atmospheres. It is found for the first time that the photoluminescence intensity of metal oxides is enhanced by UV-laser light irradiation under CO_2 gas.

Prof. S. Chaen and Aso. Prof. T. Tojo has studied the "Single-Molecule Imaging of Bio-nanomachines". He report studies on the biomolecular motor using the ordinary fluorescent imaging and the receptor protein on the biomembrane using the single-moecule fluorescent imaging technique.

3. Collaborations and activities in 2010 as the group

Each crew has team meetings and offered the research sample, respectively. We obtained the several collaborations results such as superconductor films, nanotube device, hydrogen generation/storage/release nano-particles/films and single-moecule fluorescent imaging.

Quantum Theory and Computation Group

Hiroshi Ishida, Sinichiro Ohnuki, Tokuei Sako*, Kazuo Fujikawa, Tsuneki Yamasaki

1. Overview of the research plan in 2010

The principal aim of this group is to understand the interaction between lights and nano materials. For grasping fundamental insight into this subject the members of this group have conducted their researches, focusing particularly on (i) propagation of electromagnetic fields, (ii) accurate electromagnetic responses of small systems, (iii) electronic properties of nano materials, and (iv) reexamination of fundamental concepts in quantum theory. In this year they have planned to study **(i-1)** designing nano-scale antennas for locally exciting magnetic materials by circularly polarized lights (by Ohnuki), **(i-2)** developing a high-speed and accurate method for electromagnetic simulation (by Ohnuki and Yamasaki) and studying propagation characteristics of photonic nano-waveguides (by Yamasaki), **(ii)** elucidating the origin of Hund's multiplicity rule in atoms (by Sako), **(iii)** understanding the electronic structure of heterointerface of strongly correlated systems by dynamic mean-field theory (by Ishida), **(iv)** detailed investigation of the uncertainty relation in quantum theory (by Fujikawa). They have also launched a new project involving all members that aims at solving Maxwell-Schrödinger equations for simulating simultaneous propagation of electromagnetic fields and quantum systems (Project **(v)**).

2. Advances and achievements in 2010

For the first project **(i-1)**, optimum structure for a nano-scale antenna generating circularly-polarized local fields has been examined in cooperation with Nakagawa, Ito, and Tsukamoto, and prospective results have been obtained. The results were presented in an international conference and a paper out of the results will appear in J. Appl. Phys. A patent has been also applied based on the results. Regarding the project **(i-2)** a new theoretical method for analyzing transmission of electromagnetic fields has been developed in cooperation with Nakagawa, which is applicable to nano-scale dispersive materials with numerical accuracy controlled.

Project **(ii)**: The origin of Hund's multiplicity rule accounting for the relation between the electron spin and the energy-level ordering has been clarified for the helium atom by visualizing the internal part of the wave functions. The existence of a *conjugate Fermi hole* in the lower spin states has been also clarified for the first time. A paper based on the results has been submitted to Phys. Rev. A.

Project **(iii)**: The electronic structure of a strongly-correlated single-atom-layer membrane adsorbed on a metal surface has been studied by the dynamic mean-field theory. The adsorbed atomic layer has shown to become metallic owing to orbital mixing with the grounding surface even when it is a Mott-insulator in an isolated condition. On the other hand, its electronic structure is not that of a simple Fermi metal but shows a similar phase transition as it does in an isolated condition.

Project **(iv)**: A detailed investigation into the uncertainty relation in quantum theory has shown that the uncertainty relation is not a principle but a consequence of quantum theory that is governed by Copenhagen interpretation. A resultant paper has been accepted in Prog. of Theor. Phys. Mathematical formulas for the uncertainty relation under periodic boundary conditions have been also successfully derived for the first time and the results have been published in Prog. of Theor. Phys.

Project **(v)**: A theoretical formulation has been derived to solve Maxwell-Schrödinger equations and coding based on the symplectic integrator method has been started.

3. Collaborations and activities in 2010 as the group

Study meetings were regularly organized in order to launch the new project involving all members (project (v)) and keep it on track. For other inter-group researches, there have been started collaborative projects between Ohnuki and Inoue, "Electromagnetic simulation for plasmon wave guides", and between Ohnuki and Otsuki, "Particle simulation for molecular motor".