

## Progress reports of groups

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- 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 2011

- 1) A method to analyze fast plasmon response to femto-second laser will be established and reschedule our plan.
- 2) Local surface plasmon antenna structure will be studied for circularly polarized light.
- 3) The system to measure fast optical response will be revised and rebuilt to analyze the magnetization mechanism.
- 4) Metal and/or magnetic material on nano-meter structured substrate will be studied.
- 5) Magnetizing process for nano-meter structured magnetic material will be analyzed by computer simulation.

### 2. Advances and achievements in 2011

1) We had trials to write magnetic domains on a magnetic film by femto-second laser. The magnetic film was coated with surface plasmon antennas which were made by E-beam lithography. The shapes of the antennas were designed by electro-magneto simulation as well as thermal diffusion simulation. From the observation of film structure dependency on optical/magnetic response during tens of ps after laser irradiation, it was found that conductive metallic layer play an important role in ultrafast energy transfer process.

We also set up the system applying a fine adjustment manipulator. Laser pumping and probing system can be easily adjusted by the new system.

- 2) We found some specific aperture shape which can confine a circularly polarized light. We also studied another structure for antennas which can create a localized circularly polarized light with a liner polarized light.
- 3) A new magnetic bias coil was prepared for applying up to 1 kOe. The bias field can be synchronized to the laser system.
- 4) We started to fabricate nano-meter structure using etching method. Nano-size silica particles were placed over a metal film before etching.
- 5) We found that the magnetization in a small one magnetic particle can be reversed by applying surface plasmon antenna by simulation.

### 3. Collaborations and activities in 2011 as the group

We have had regular meetings once or twice a month meetings every month with Associated Professor Ohnuki, belonged to Quantum Theory & Computation Group, to find a method to calculate a surface plasmon response. The results were reported at IEEE International Symposium on Antennas and Propagation, July 5-6, 2011, Spokane, Washington, USA. We had two invited talks about magnetic nano-particle media and dynamics of ultra fast magnetic reversal at Moscow International Symposium on Magnetism, Aug. 21-25, 2011, Moscow, Russia. We also reported about thermally assisted recording, ultra-high speed magnetization, and circular polarized light generation with a plasmon antenna at Magnetic and Optical Research International Symposium, June 21-24, 2011, Nijmegen, The Netherlands.

## Quantum information Group

Shuichiro Inoue\*, Takeshi Kuwamoto, Hideomi Hashiba

### 1. Overview of the research plan in 2011

- 1) Development of the entanglement swapping system
- 2) Fabrication of superconducting nano-wire single-photon detectors and the evaluation of their performances
- 3) Fabrication of Au nano-stripe directional couplers and demonstration of quantum interference of single surface plasmon-polaritons
- 4) Storage of polarization entangled photon pairs in Rb atoms using electromagnetically induced transparency
- 5) Development of simple and reliable etching technique of Si on ICP
- 6) Study of anatase formation on sputtered TiO<sub>2</sub> nano-structures with different baking temperature

### 2. Advances and achievements in 2011

- 1) We made two polarization entangled photon-pair sources, and investigated the indistinguishability of the photons from each source using Hong-Ou-Mandel interferometer. We are going to demonstrate the entanglement swapping by performing Bell-state measurements.
- 2) We have investigated the coupling of surface plasmon-polaritons with a superconducting nano-wire by computer simulations collaborating with Ohnuki laboratory.
- 3) We have fabricated Au nano-stripe directional couplers using long-range SPP straight waveguides and S-bends. The insertion loss of the fabricated directional coupler was approximately 30 dB and the output ratio was 64:36. Moreover, we have demonstrated quantum interference of single surface plasmon-polaritons using the directional coupler, a correlated photon-pair source, and the superconducting photon-number resolving detectors. It turned out that the quantum interference of single surface plasmon-polaritons output two photons together in one port or the other, which is the evidence that the single surface plasmon-polariton is a bosonic quantum particle.
- 4) The 397.5-nm pump light, which is used for generating polarization entangled photon pairs at 795 nm through a spontaneous parametric down conversion process, was enhanced by developing a bow-tie type cavity for the second harmonic generation (SHG) with a type I PPKTP crystal, which led to about a three-fold increase of the number of orthogonally polarized photon pairs. And also we have improved the efficiency of light storage in Rb atoms using electrically induced transparency by developing a high-speed injection-current controller of the laser and a hot-water temperature control system for Rb atoms to reduce magnetic field around the cell, which led to a two-fold increase of the light storage efficiency.
- 5) CF<sub>4</sub> plasma of high pressure and low power on ICP has been tested for etching of metal-masked Si samples and calibrated suitable etching ratio of few hundred nano-meter wide patterns.
- 6) We studied transformation of TiO<sub>2</sub> nano-structures fabricated from standard resist mask technique to anatase.

### 3. Collaborations and activities in 2011 as the group

We have proposed a colloidal quantum dot array as a new device for the quantum repeaters and collaborated with Otsuki laboratory to fabricate the device. 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 2011

- 1) In order to improve energy conversion efficiency of dye-sensitized solar cell, preparation of a photonic-crystal electrode with a full-photonic band gap composed of TiO<sub>2</sub> and liquid electrolyte is continuously examined using electron-beam lithography. Also tried is life-time measurement of luminescence to clarify the origin of energy conversion efficiency increase of solar cells by employing self-assembled photonic crystals. Development of new dyes is also examined for improvement of energy conversion efficiency of solar cells.
- 2) The analysis of the structure and reaction mechanism of the supra-molecule catalyst to generate H<sub>2</sub> from H<sub>2</sub>O with photo energy, which has been discovered in 2010 by this project, is challenged.
- 3) In order to use H<sub>2</sub> as a clean energy source, establishments of H<sub>2</sub> generation and storage methods using enzyme prepared with biotechnology and H<sub>2</sub> storage materials is tried. The development of new methods of photo-controlled H<sub>2</sub> storage is also examined. As an electrical power generation system using thus obtained H<sub>2</sub>, development of solid oxide fuel cells operating at about 600 °C, at which enough energy conversion efficiency can be obtained and long term endurance is realized, is tried by development of new materials.

### 2. Advances and achievements in 2011

- 1) Photo-electrical conversion efficiency of dye-sensitized solar cells has been measured employing monochromatic light source. The dependence of conversion efficiency on grain size of inverse-opal electrode with a parameter of wavelength has been clarified, which is inevitable information on analysis of mechanism of grain size effect on conversion efficiency.

As a new material for dye sensitized solar cells, some derivatives of peryleneimide have been prepared. Solar cells employing the prepared dye were fabricated and efficiency reached 3.1 % at the highest.

- 2) The H<sub>2</sub> generation from triethylamine using photocatalyst has been discovered in spite of dark conditions. The mechanism of H<sub>2</sub> generation is under investigation.
- 3) H<sub>2</sub> generation using cyano-bacteria under conditions with inert gas and light irradiation has been examined. Quantity of H<sub>2</sub> gas has increased by coexistence of H<sub>2</sub> absorption alloys. It has been also revealed that not only H<sub>2</sub> absorption alloys but also carbon nanotube and BCN cluster increase the quantity of H<sub>2</sub> gas production.

It has also been found that stability of absorbed H<sub>2</sub> gas increases by irradiation of ultraviolet light, resulting in increase of H<sub>2</sub> desorption temperature. This suggests probability of charge of second type battery as Ni-H<sub>2</sub> one using photo irradiation.

The apparatus for measurement of solid oxide fuel cell (SOFC) property has been developed. At present, fundamental property of Ba<sub>0.6</sub>Sr<sub>0.4</sub>Zr<sub>0.9</sub>Y<sub>0.1</sub>O<sub>3</sub>, LaNi<sub>0.6</sub>Fe<sub>0.4</sub>O<sub>3</sub> and Sr<sub>2</sub>FeWO<sub>6</sub> has been minutely investigated, showing that they are promising as new materials for SOFC. At present, SOFC composed of the above materials are now preparing.

### 3. Collaborations and activities in 2011 as the group

Evaluation of efficiency using monochromatic light source was carried out under collaboration of Dr. Matsushita and Dr. Hashimoto. The achievement has been reported as a paper published in *J. New Mat. Electr. Sys.* Production of H<sub>2</sub> using bacteria and H<sub>2</sub> absorption alloys has been achieved as collaboration of Dr. Asada and Dr. Nishimiya. The patent has been applied.

## Medical Group

### Medical Group Members

ARISH: Fukuda N, Cancer Genetics: Saito K, Igarashi J, Fujiwara K, Soma M, Radiology: Abe O, Ishibashi N, Urology Takahashi S, Pediatric Surgery: Koshinaga S, Uekusa S, Science and Technology: Watanabe T, Bioresource Science: Masuhiro Y, Kano K, Pharmacy: Matsumoto Y, Aoyama T, Chiba Cancer: Nagase H

### Progress and Production by Whole Medical Group in 2011

- 1. Applied chemical, physical biology: Strategy to cure cancer patients** (Fujiwara, Takahashi, Abe, Ishibashi, Soma): We invented the radiation-sensitizing chemical of the porphyrin derivatives, which can be used for PDT and may also induce photon activation therapy (PAT), provoking the emission of Auger electrons after inducing a photoelectric effect. X-ray radiation allows for the treatment of cancers that are deep inside the human body. We observed an induced cancer cell death after irradiation following administration of the porphyrin derivative.
- 2. Development of antitumor PI polyamides for pediatric cancer** (Uekusa, Koshinaga): PI polyamides (h-CCAAT1, h-CCAAT3) designed on the CAAT box in promoter region of LIT1 gene efficiently suppressed expression of p57<sup>KIP2</sup> gene.
- 3. Determination of a lead compound targeting human TGF- $\beta$ 1** (Igarashi, Fukuda): Read polyamides were selected and examined on expression of TGF- $\beta$ 1 mRNA in human cells. GB1105 and GB1106 strongly inhibited expression of TGF- $\beta$ 1 mRNA in a dose-dependent manner. Effect of GB1105 was strongest to inhibit the expression of TGF- $\beta$ 1 mRNA in human cultured fibroblast.
- 4. Development of the Nihon University original methodology inducing iPS cells using the PI polyamide targeting human TGF- $\beta$ 1** (Saito, Fukuda, Masuhiro) : We examined the iPS-producing method establishment using proteolysis resistant cell-permeable proteins and the initiation factor, TGF- $\beta$ 1 inhibitor, PI polyamide targeting human TGF- $\beta$ 1, Apigenin, TGF- $\beta$ 1 antagonist and Apigenin, and TGF- $\beta$ 1 and PI polyamide targeting human TGF- $\beta$ 1.
- 5. Establishment of a breast cancer-inducing mouse model by the transplantation of DFAT** (Kano): We tried the creation of a breast cancer-inducing mouse model by the transplantation of DFAT-GFP transfected oncogene, which is transformed to epithelial cells.
- 6. Histone acetylation of specific genomic region induced by PI polyamide-SAHA conjugate** (Nagase) : We made considerable advances in coupling of existing drug SAHA that is being used as an HDAC inhibitor, to PI polyamides for targeting specific subsets of genes for reactivation in cancers (e.g. Cdkn2a / p16).
- 7. Solid phase synthesis of cyclic PI polyamide** (Watanabe) : the method of simple synthesis with solid phase synthesis method using glutamic acid which is the usual amino acid. Six ring cyclic PI polyamide was synthesized.
- 8. Pharmacokinetic/Pharmacodynamic Analysis of tumor-localizing photosensitizing compounds** (Matsumoto, Aoyama): To describe the relationships between effects following photodynamic

therapy, light dose, and plasma compound concentration, they developed a high-performance liquid chromatography (HPLC) method for the determination of plasma concentration and investigate the pharmacokinetics of novel compound CT101019a.

#### **Activities of Medical Team in 2011**

- 1) Meeting of N. Research team of Cancer Genetics every week.
- 2) Aug 3<sup>rd</sup>: Presentation “Innovation by PI polyamide based on the chemical biology” at Colledge of Engineering.
- 3) Sept 22<sup>nd</sup>: N. Research project meeting of School of Medicine about the research progress and planning in 2011.
- 4) Dec 17<sup>th</sup>: Report for the research progress of the medical team in the 7<sup>th</sup> Nihon University Advanced Bioforum.
- 5) Made contract with the Drug Preparation Room in Nihon University School of Medicine Itabashi Hospital.
- 6) Made contract with Central Institute for Experimental Animals as prace for the practical experiments.

## Supramolecules and Self-Assembly Group

**Hiroki Ikake<sup>1</sup>, Akiyoshi Itoh<sup>2</sup>, Joe Otsuki<sup>1</sup>, Arata Tsukamoto<sup>2</sup>, and Sachiko Matsushita<sup>3</sup>**

1 College of Science and Technology, Department of Materials and Applied Chemistry

2 College of Science and Technology, Department of Electronics and Computer Science

3 Tokyo Institute of Technology, Department of Metallurgy and Ceramic Science

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 2011.

### · **Itoh & Tsukamoto Group**

Preparation of a thin nano-porous SiO<sub>2</sub> film and application as etching mask of metallic films  
Self-assembly prepared SiO<sub>2</sub> nano-dent-template (Dent pitch ~8 nm and ordering domain size of ~1 μ m<sup>2</sup>) was formed by (1) utilizing triblock copolymer of adequate molecular weight and (2) development of the preparation procedure of precursor solution.

### · **Otsuki Group**

Self-assembly of appropriately designed molecules will afford a bottom-up method for producing nanostructures. This work aims at developing new molecular self-assembling systems, revealing self-assembled structures and dynamic behaviors at the molecular level, and searching for applications of self-assembly to energy, medical, and information technologies through the collaboration with researchers of the *N.* research project.

- 1. Structures and dynamic behaviors of molecular self-assemblies at the molecular level**
- 2. Piperidine-Substituted Perylene Sensitizer for Dye-Sensitized Solar Cells**
- 3. Preparation of compounds for the X-ray based photodynamic therapy**

### · **Matsushita Group**

Overview of the research plan in 2011

Six-rayed star-like nanostructures in prospective plasmonic devices

Advances and achievements in 2011

- **Preparation of two-dimensional arrangement of SiO<sub>2</sub> particulate by self- integrated**
- **Crosslinks were formed between arranged particles with heat-treatment**
- **Preparation of microstructure with protruded structure applied HFaq etching for arrangement particles**
- **Absorption peak shift for surface plasmon resonance was confirmed with structural change**

### · **Ikake Group**

In our group, the aim of development of poly(lactic acid) (PLA) films as biopolymer with the high thermal- and mechanical- resistance. And then, the improved PLA was submitted to new material field.

- 1. Development of Poly(lactic acid) Films with Exhibiting the Piezoelectricity**
- 2. Preparation of High Crystallinity and High Orientation Poly(L-lactic acid) Films under Electric Field**
- 3. Morphological change of Poly(L-lactic acid) Films with Magnetic Irradiation**

## 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 2011

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:TiO<sub>2</sub> 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<sub>2</sub> solid solutions) and in-vitro single molecule imaging of these proteins by membrane receptors.

### 2. Advances and achievements in 2011

Prof. K. Suzuki approached synthesis of ferromagnetic metal (Ni, Fe, and Cu) 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 deposited on thin polymer films by laser induced forward transfer method.

Prof. Y. Takano has mainly studied the Fe-based superconductors. He has firstly prepared Sr<sub>1-x</sub>Nd<sub>x</sub>FFeAs and obtained  $T_c=49$  K for  $x=0.5$ . This value is close to the highest value in Sr<sub>1-x</sub>R<sub>x</sub>FFeAs (R= rare earth ions). He has also prepared SrFFe<sub>1-x</sub>Ni<sub>x</sub>As where Fe sites are directly substituted by Ni ions. Li<sub>x</sub>FeAs itself is a superconductor with  $T_c=18$  K. He has prepared Li<sub>x</sub>Fe<sub>1-y</sub>Co<sub>y</sub>As 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 with an electrode made of composite metallic materials. He has also proposed innovative technique for EUV (~13-14nm) light source by plasmoid collision heating which can be utilized for the next generation lithography. Prototype of the light source and measurement apparatus for performance evaluation have been developed.

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 in THz range“. THz range single photon detectors are assembled from a GaAs/AlGaAs quantum dot coupled with a metallic single electron transistor which senses appearance of charge state of the QD. Plasma excitations of the QD arises with a formation of confinement potential barrier from the reservoir having resistances more than resistance quanta, and we revealed that appropriate shape of the barriers lowers dark counts by suppression of flow of hot electrons from the reservoir. We also show that the QD works as a heat bath from THz plasma excitations. This will promise high temperature operation of the THz detection.

Prof. S. Mochizuki and Mr. K. Yoshida have studied the photoluminescence properties of various kinds of metal oxides under oxygen gas, hydrogen gas, vacuum and CO<sub>2</sub> 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<sub>2</sub> gas.

Prof. S. Chaen and Prof. T. Tojo have succeeded in estimating ADP release rate from the displacement



of fluorescent nucleotides bound to myosin heads in the *in vitro* motility assay system by flash photolysis of caged ATP. And they have developed a new wet cell system of Scanning Electron Microscopy to observe a living cell in aqueous solution with at nanometer resolution.

Asso. Prof. K. Judai has studied the “Self-assembled nano helix”. He found silver tolyl-acetylide molecules self-assemble into nano helical morphology. This may open a new technique to combine nano-materials with bio-molecules.

### **3. Collaborations and activities in 2011 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-molecule fluorescent imaging.

## Quantum Theory and Computation Group

**Hiroshi Ishida, Shinichiro Ohnuki, Tokuei Sako\*, Kazuo Fujikawa, Tsuneki Yamasaki**

### 1. Overview of the research plan in 2011

- 1) Fundamentals of entanglement in quantum information theory
- 2) Investigation of electronic properties of nanosized structures by many-body techniques such as dynamical mean-field theory
- 3) Origin of Hund's rule in atoms by conjugate-Fermi-hole analysis
- 4) Design of plasmonic antennas for all-optical magnetic recording
- 5) Development of high-speed and high-precision methods for electromagnetic simulation
- 6) Photon-energy storing in photonic crystals with defect structures
- 7) Development of coupled solver for Maxwell and Schrödinger equations

### 2. Advances and achievements in 2011

1) A currently hot topic of "systems having quantum-mechanical features but not entangled" has been investigated and the results have been summarized in the following two papers:

1. K. Fujikawa, "Quantum discord and noncontextual hidden variables models", submitted.

2. K. Fujikawa, "Does CHSH inequality test the models of local hidden variables in quantum mechanics?", submitted.

2) (i) 1. Transport properties of the Mott insulating layer adsorbed on a metal substrate have been studied by using a cluster extension of dynamical mean-field theory. The Mott insulating layer has shown to become metallic when it is coupled with a substrate metal. Moreover, the in-plane electrical resistivity of the overlayer has shown to increase with decreasing temperature. These results will appear in the following accepted paper:

H. Ishida, A. Liebsch, "First-order metal-to-metal phase transition and non-Fermi liquid behavior in a two-dimensional Mott insulating layer adsorbed on a metal substrate", *Physical Review B* 2012, Vol.85 (in press) 1-14.

(ii) The electron conductance of monoatomic chains suspended between two metal electrodes has been studied by considering the on-site Coulomb repulsion energy in the atomic chain. It is shown that the calculated conductance becomes much smaller than that of the non-interacting chain.

3) The internal structure of the many-body electronic wave functions for He and He-like atomic ions has been investigated by focusing on the recently proposed "conjugate Fermi holes". The results have shown that the wave function of the singlet state extends much broader than does the triplet wave function due to the conjugate Fermi holes, resulting in a larger increase in the total energy. This rationalizes the mechanism of how Hund's rule operates in these systems.

4) The arrangement of particulate media under the nano-scale plasmonic cross antenna has been investigated. The results show that the structure of a plasmonic antenna enhances the intensity ratios of the center particle to the adjacent particles. When the linear polarized light impinges, the localized circular polarized light is produced only in the center particle and the electric field intensity becomes double compared with adjacent particles.

5) (i) Time domain solvers based on integral equation methods

We have developed fast and accurate solvers based on integral equation methods with fast inverse Laplace transform for time domain electromagnetic problems. Applying the multilevel fast multipole method, the computational time becomes about five times faster. Our method is suitable for parallel computing and efficiency keeps almost 100 % for increasing the number of nodes. We investigate nano-aperture antennas and charge distribution on a nano-particle due to an electric dipole source.

(ii) High-precision analysis of electromagnetic scattering problems

A point matching method can analyze electromagnetic scattering problems with a high degree of accuracy. We extend our previous idea to 3D problems and investigate computational accuracy of the electromagnetic scattering from homogeneous dielectric spheres. Compared with the analytical solutions, our prediction method for 3D problems is verified.

6) We have proposed fabrication of defect structures filled with dielectric materials in the follow wave-guides of photonic crystals in order to store larger electromagnetic energy. By analyzing the distribution of electromagnetic fields in the wave guides, an optimal structure of the dielectric materials has been investigated. It is shown that a diamond is the best structure for storing electromagnetic energy. The results have been published in the following paper:

R. Ozaki and T. Yamasaki, "Propagation Characteristics of Dielectric Waveguides with Arbitrary Inhomogeneous Media along the Middle Layer", *IEICE TRANSACTIONS on Electronics*, Vol.E95-C, No.1, pp.53-62 (2012).

7) A novel algorithm is proposed for solving coupled Maxwell and Schrödinger equations relying on the use of a length gauge form of the coupling between an electromagnetic field and electrons. The proposed algorithm can reduce computational time almost by half as compared with the conventional method.

### **3. Collaborations and activities in 2011 as the group**

We have organized a meeting every month at Prof. Ohnuki's laboratory with Prof. Nakagawa and Dr. Ashizawa of Information Storage Group to study theoretical methods for solving coupled Maxwell and Schrödinger equations.