US-Japan TeraNano Workshop on Nanophotonics & Nanoelectronics

08:30 – 17:30, Friday May 11th 2012
Agrusa Auditorium, Davis Hall,
University at Buffalo

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08:45:	*Opening Remarks* Kenneth M. Tramposch, Ph.D., Associate Vice President for Research
09:00	*Dynamic Terahertz Emission Microscope* Prof. Masahisa Tonouchi, Osaka University
09:35	*The Many Applications of THz light: Plasmons, Proteins & Topological Insulators* Prof. Andrea Markelz, University at Buffalo
10.10	Coffee Break
10.30	*Structured Light in Nanostructured Media* Prof. Natasha Litchinitser, University at Buffalo
11.05	*Cooperative phenomena in electron-hole and electron-hole-photon systems* Prof. Tetsuo Ogawa, Osaka University
11.40	*Analysis and Applications of Pulsed-Laser Plasmon-assisted Nanoscale Photothermal Energy Transfer in Fluid* Prof. Ed Furlani, University at Buffalo
12.15	*Lunch*
14.00	*Nonlinear Terahertz Spectroscopy in Semiconductors* Prof. Koichiro Tanaka, Kyoto University
14.35	*Rectifying Terahertz Nanosensors* Prof. Jonathan Bird, University at Buffalo
15.10	*Compact and Integrable THz Polarization Converter* Prof. Qiaoqiang Gan, University at Buffalo
15.45	Coffee Break
16.00	*Spectroscopic study on ultrafast carrier dynamics and terahertz amplified stimulated emission in optically pumped graphene* Prof. Taiichi Otsuji, Tohoku University
16:35	*Nonequilibrium carriers in graphene* Prof. Vladimir Mitin, University at Buffalo
17.10	*Closing remarks*

Workshop co-sponsored by the Dean's Office of the School of Engineering and Applied Sciences, and the Department of Electrical Engineering, of the University at Buffalo, and the JSPS core-to-core program.

Dynamic Terahertz Emission Microscope

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One can observe terahertz (THz) radiation from various kinds of materials [1], when excited with a femtosecond laser, owing to ultrafast current modulation. THz waves reflect various kinds of properties such as local electric field, particularly ultrafast transient phenomena, in their waveforms. The observation of the THz waveforms enables us to explore ultrafast nature of electronic materials and devices as a THz emission spectroscopy. When one excites the THz emission from a certain substance with the femtosecond optical pulses and visualizes the emission image by scanning the laser beam on it, the resolution of the image is limited by the laser beam diameter rather than THz wavelength. Thus construction of a laser-THz emission microscope (LTEM) would provide a new tool for material science and application [2,3].

Here we propose and demonstrate new type of LTEM combining with pump and probe THz emission system, which is referred as Dynamic Terahertz Emission Microscope (DTEM). The pump and probe emission study enable us to disclose ultrafast carrier dynamics with a time resolution close to optical pulse width [4]. Thus the combination would extend the performance of LTEM greatly in terms of ultrafast spatial transient carrier behavior.

DTEM is the pump-probe LTEM. LTEM system is explained in Ref. 5. Here we examined ultrafast dynamic behavior of photoconductive THz emission antennas made of LT-GaAs and SI-GaAs. Dynamic response of the switches are successfully visualized and explained by dynamic local screening and resultant enhancement of internal electric field at unscreened location.

This work is partially supported by Grant-in-Aid A22246043, Core to Core, JSPS, and Industry-Academia Collaborative R&D, JST.

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Applications of Terahertz Light: Plasmons, Proteins and Topological Insulators

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Several recent results using the unique strength of terahertz radiation to probe material properties will be discussed: detection of colossal Kerr response from topological insulators; harmonic enhancement of plasmonic response in a two-dimensional electron gas and detection of underdamped structural motions in proteins.

Topological insulators (TI) are a distinct class of materials as their states do not originate from local parameters such as broken symmetries but from topologically protected properties encoded in their wavefunctions. THz measurements have revealed colossal Kerr rotation for Bi2Se3 [1]. We will discuss these measurements and a new broad band technique for rapidly measuring the complex Faraday and Kerr rotations in a single measurement, combining the distinct advantages of THz time domain spectroscopy and polarization modulation techniques [2].

Plasmonic devices in the terahertz range can be challenging as they require submicron features over millimeter areas. Here we demonstrate large area plasmonic materials fabricated using self -assembly and find that the response at higher harmonics is surprisingly enhanced relative to the fundamental.

Finally we will discuss THz applied to biomolecular dynamics. Correlated motions in proteins have long been predicted to lay in the terahertz frequency range [3]. Unfortunately the measurement of these modes has been problematic due to overlap with the broadband response of biological water and librational motions of surface side chains. Polarization difference spectroscopy is a method that can be used to suppress a homogeneous background from orientation sensitive resonances, however the size for typical protein crystals is below the diffraction limit at terahertz frequencies. Recently great advances in THz near field microscopy have been made[4]. Using THz near field microscopy we find strong orientation sensitive features from hen egg white lysozyme tetragonal protein crystals and suggest that these resonances are related to the intra molecular vibrations.

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Structured Light in Nanostructured Media

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In this talk we discuss our recent studies of fundamental optical phenomena at the interface of two emerging fields of modern optical physics – singular optics and optical metamaterials (MMs). Singular optics is a fascinating emerging area of modern optics that considers spin and orbital angular momentum (OAM) properties of light, and brings a new dimension to the science of light and physics in general. Indeed, light captures many of the basic features of relativistic spinning particles. These similarities result from the wave-particle dual nature of quantum particles. Optics facilitates the realization of many effects that are predicted in other physical systems where direct experimental observations are challenging or impossible. Recent developments in the field of MMs and transformation optics have enabled unprecedented control over light propagation and made possible the "engineering" of space for light propagation. These developments have opened a new paradigm in spin and OAM related phenomena in optical physics. In this project, we conduct basic theoretical studies, extensive numerical modeling and design, fabrication, and characterization of nanostructures to understand spin and OAM interaction effects that appear when "structured light" interacts with linear and nonlinear nanostructured MMs. We will discuss our recent initial results on the generation of radially polarized optical vortices and their interactions in novel materials, including magnetic and nonlinear MMs. We exploit the fact that both the electric and magnetic field components of a light beam are involved in such interactions, and we consider the possibilities of dispersion engineering in MMs for manipulating beams carrying OAM, as well as the role of backward phase-matching in the processes of second harmonic generation and parametric down-conversion with such beams. Finally, we outline the potential of structured light interactions in MMs for quantum communications and optical imaging.

Cooperative Phenomena in Electron-Hole and Electron-Hole-Photon Systems

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We have been focusing in recent years on interacting electron-hole-photon (ehp) systems realized in semiconductor quantum wells in microcavities, which show two different types of coherent light emission: lasing and polariton Bose-Einstein condensation (BEC). The main interests are in their many-body aspects, arising from the strong light-matter coupling and Coulomb interaction, which can play a role both in the nonequilibrium (with large cavity loss) [1] and quasi-thermal-equilibrium (under fast thermalization) [2] situations. Here we report our recent progress in the theory for the ehp systems. A many-body feature in semiconductor lasers is recently found when the BCS-type (Bardeen-Cooper-Schrieffer-type) electron-hole (eh) pairing instability occurs during low-temperature operation. In this situation, the Fano resonance is found in the gain spectra due to the interference of the optical transitions via an eh Cooper pair (discrete level) and unbound eh pairs (continuum). Since their interference occurs only if the dephasing of eh pairs is present, we understood the Fano-resonance gain is one of nonequilibrium characteristics found near the BCS phase transition. As a result of the Fano-resonance gain, the single-mode laser operation is strongly modified relative to the conventional laser operation with the Lorentzian-type eh plasma gain. We have studied the stationary state of lasing using the Maxwell-semiconductor-Bloch equations (MSBE), and found a strong reduction in the carrier density at the lasing threshold and an enhancement in the stability of the continuous-wave operation. Macroscopic numbers of the cavity polaritons in ehp systems can be condensed into a single energy level and exhibit polariton BEC in a quasi-thermal equilibrium situation. Here, the stationary state of the system is determined by the minimization of Free energy [2]. Whereas the polariton BEC is a coherent light source similar to semiconductor lasers, the condition for the stationary states to be sustained in quasi-thermal equilibrium and nonequilibrium situations are quite different. Thus, their description had been treated with different theories (BCS-like theory for polariton BEC, and MSBE for semiconductor lasers). Recently, we successfully unify these two theories for the Coulomb-correlated ehp systems [3] based on the Keldysh Green's functions in full range of temperature and nonequilibrium parameters. We found the crossover of the stationary states from those of the polariton BEC to those of lasers. By using the unified theory, it becomes now possible to describe the lasing from the nonequilibrium BCS phase, which will be discussed with some numerical results.

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Analysis and Applications of Pulsed-Laser Plasmon-Assisted Nanoscale Photothermal Energy Transfer in Fluid

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Rapid progress in the development and applications of nanostructured materials has spawned a growing interest in understanding and controlling thermal energy generation and transport at the nanoscale [1]. While various approaches have been proposed to address this need, one of the most promising involves photothermal energy conversion wherein a laser is used to heat noble metal nanostructures at their plasmon resonant frequency [2]. This enables efficient and highly localized heating, with considerable tuning potential, which makes the process suitable for a wide range of applications. Such applications include nanoparticle synthesis (transformation of particle size, shape, and phase) [3], optothermal imaging [4] and targeted photothermal medical therapies [5-6]. In this presentation we demonstrate results on the analysis of photothermal effects associated with nanosecond-pulsed laser-illuminated subwavelength metallic nanoparticles in aqueous solutions. We use combined computational electromagnetic and fluid analysis to model fundamental aspects of the photothermal process. More specifically, we study energy conversion within the nanoparticle at plasmon resonance, heat transfer to the fluid, homogeneous bubble nucleation and the dynamic behavior of the bubble and surrounding fluid. We demonstrate the theory via application to various nanoparticle geometries including spheres, nanorods and toroids. We show that process parameters such as laser intensity, pulse duration, wavelength, polarization and particle geometry can be tuned to control the size and behavior of nucleated bubbles. We also present results on multi-particle systems, and demonstrate that cooperative heating enables controllable bubble nucleation using substantially reduced laser energy compared to single-particle systems. We discuss details of the modeling approach and specific applications including recent advances in photothermal tissue therapy at the cellular level.

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Terahertz Spectroscopy in Solids with Single-Cycle Terahertz Pulses

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Recent development of ultra-short pulse technologies allows us to drive large amplitude motion of electron and ion coherently. The intense terahertz (THz) pulse resonant with the vibration frequency is promising to drive vibrations more directly and in coherent manner. In the case of semiconductors, one may coherently control the electronic system in the sub-level structures of quantum structures with intense THz waves. We recently succeeded to generate intense terahertz pulses by Cherenkov scheme with tilted wave-front technique in the LiNbO3 crystal [1, 2]. The maximum electric field is now larger than 1 MV/cm, which ponderomotive energy is as large as 10 eV. The ponderomotive energy is strong enough to ionize bound electronic states in solids such as donors and accepters and easy to induce nonlinear optical effects in solids [3].

In this talk, we would like to review first the state of the art of the high-power THz-wave generation with femtosecond lasers including Cherenkov-scheme in non-linear crystals and air-plasma generation. Then we will focus recent results on THz nonlinear optical phenomena induced by THz electric field larger than 100 kV/cm in molecular crystals [4], semiconductors [5] and ferroelectric materials [6].

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Nanoscale Terahertz Rectifiers

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Recently, there have been many attempts to utilize semiconductor nanostructures for the detection of terahertz (THz) radiation, motivated by the compact size of these structures, and by the fact that their characteristic energy scales are comparable to the THz-photon energy. Among the mechanisms that allow for detection, rectification of THz signals due to non-linear electrical characteristics are extremely attractive. Potentially, this essentially classical mechanism can allow for broadband THz detection, independent of the blackbody background. In this talk, we review our recent efforts [1-3] to develop nanoscale terahertz (THz) rectifiers that are capable of broadband THz detection. These devices are implemented in high-mobility two-dimensional electron systems that are confined either electrostatically, or through chemical etching, to nanoscale channels. We discuss how these efforts provide a path towards our ultimate goal of real-time (video-rate) THz imaging at room temperature.

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Compact and Integrable THz Polarization Converter

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The terahertz (THz) frequency range is one of the least developed spectral regions in which various important applications have been recognized, including security screening, nondestructive evaluation, communication and biological and medical sciences. However, due to the relatively immature material and device achievements, radiation sources, detectors, transmission technologies and passive components are limited in this spectral region. Particularly, compact, coherent and continuous solid-state sources with desirable polarization states are of high interest [1] depending on special application requirements. Conventionally, the polarization state of THz light was usually manipulated by external bulky and expensive optical components, which limited the dimension of the entire light source system. In recent years, various compact THz metamaterials and plasmonic structures were proposed to manipulate the polarization state of free space light. However, as a passive component, it is critical to achieve broadband and high efficiency simultaneously, which is challenging to achieve based on these resonant mechanisms. The polarization management can only be realized at individual resonant peaks in most reported THz metamaterial and plasmonic structures, which, on the other hand, is more attractive to be implemented in compact laser systems. Semiconductor-based quantum-cascade lasers (QCL) with integrated optical components are highly desired for applications where the compact dimension of the device/system is critical. For example, one dimensional metallic gratings were employed on the front facet of QCLs to control the polarization state of the output laser radiation [2]. However, due to the tiny area of the subwavelength slit, the output power from the plasmonic-manipulated QCL is weak [2], which may constrain its usefulness for practical applications. To address this low output limitation, we propose single-layered and double-layered L-shaped metallic structures to achieve the high efficient polarization management, i.e. to realize high transmission efficiency and tunable polarization control simultaneously.

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Spectroscopic Study on Ultrafast Carrier Dynamics and Terahertz Amplified Stimulated Emission in Optically Pumped Graphene

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Graphene is a one-atom-thick planar sheet of a sp2-bonded honeycomb carbon crystal [1]. Its gapless, linear and symmetric energy spectra of electrons and holes lead to nontrivial features such as giant carrier mobility, ultra-broadband flat optical response [2], as well as negative dynamic conductivity in the terahertz spectral range [3]. Carrier-phonon interactions via intravalley, intervalley, intraband, and interband processes take the dominant roll to cool the elevated temperatures of photoelectrons and photoholes[4], which leads to extraordinary nonequilibrium energy relaxation processes in graphene. In case of room temperature environment and/or strong pumping, collective excitations due to the carrier-carrier (CC) scattering, e.g., intraband plasmons perform ultrafast carrier redistribution along the energy [5]. Then optical phonons (OPs) are emitted by carriers on the high-energy tail of the carrier distributions. This energy relaxation process accumulates the nonequilibrium carriers around the Dirac points. Due to a fast intraband relaxation (ps or less) and relatively slow interband recombination of photoelectrons/holes, one can obtain the population inversion under a sufficiently high pumping intensity [6]. The authors have first analytically found the possibility of terahertz gain in such a system under a cryogenic condition [5] and have recently numerically verified the occurrence of the terahertz negative dynamic conductivity even at high temperatures [6]. We have measured the carrier relaxation and recombination dynamics in optically pumped graphene using terahertz time-domain spectroscopy based on an optical pump/terahertz-and-optical-probe technique [7]. The observed results clearly show amplification of the terahertz probe pulse with a threshold behavior against the pumping intensity. Time-dependent gain-spectral profiles well agree with the simulated ones. We conclude that the terahertz emission from graphene is stimulated by the coherent terahertz probe radiation and that the terahertz emission is amplified via electron-hole recombination in the range of the negative dynamic conductivity.

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Nonequilibrium Carriers in Graphene: Diffusion, Recombination, and Transient Population Inversion

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Spatio-temporal evolution of carriers after ultrafast photoexcitation is determined by the energy relaxation, recombination, and diffusion processes. [1] As reported recently [2,3], these processes govern the transient THz emission. We are presenting our results on recombination and diffusion of electron-hole pairs in graphene as well as on transient stimulated emission of radiation under initial population inversion of carrier distribution. Generation-recombination interband transitions via acoustic phonons are allowed in a disordered graphene because the energy-momentum conservation requirements become softer. The generation-recombination dynamics of carrier concentration is analyzed based on the quasiequilibrium approach for the scattering by a short-range disorder and for the deformation interaction of carriers with in-plane acoustic modes. [4] The transient evolution of nonequilibrium carriers is described by the exponential fit which depends on doping conditions and disorder level. The characteristic carrier recombination time is estimated to be about □rec~150 ns at room temperature for a sample with the maximal sheet resistance ~5 k□. The spatial diffusion of carriers excited by a micrometer-size laser spot is determined by an effective elastic scattering and peculiarities of energy distribution which is formed after the cascade emission of optical phonons. [5] The diffusion coefficient is found to be of the order of 104 cm2/s, in agreement with the experimental data. [1] The process of transient stimulated emission during time intervals t <<□□rec is described based on the population equation for mid-IR mode propagating along THz resonator. The stimulated emission time is about 10 ps and the mid-IR gain ~ 20 cm-1 at initial concentrations above ~5.1011 cm-2 if one uses the resonator of length larger than 0.5 mm. This gain is comparable to the gain of quantum cascade lasers, so our estimates demonstrate that a multi-layer graphene can be used for mid-IR lasers with the same efficiency as quantum cascade lasers.

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