SYMPOSIUM GG

Plasmonics–Nanoscale Optics and Photonics Based on Metals

November 27 - 30, 2005

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* Invited paper

TUTORIAL

FTGG: Nanoscale Manipulation and Mega-Enhancement of Light-Utilizing Surface Plasmons
Sunday, November 27, 2005
1:30 PM - 5:00 PM
Room 209 (Hynes)

The tutorial will cover the two main topics of plasmonics that currently undergo the most intensive development in research laboratories, opening new avenues in nanoscience and nanotechnology. The tutorial will begin with a general overview of plasmonics, which is concerned with electromagnetic excitations (plasmons) in metal nanostructures, introducing extended (propagating) and localized surface plasmons (SPs). Propagating SPs will then be considered in detail followed by a presentation of various configurations used for guiding and manipulating of radiation in SP-based photonic components and circuits. Examples of SP-based components, in which radiation is guided along and controlled with the same metal circuitry will be presented.

The latter half of the tutorial will be focused on the theory and applications of localized SP-based nano-imaging and nanospectroscopy, which has been known as tip-enhanced near-field scanning optical microscopy (NSOM). The mechanism for obtaining super resolution beyond the diffraction limit, giant signal enhancement, spectral shift in Raman spectra, and some other interesting effects due to the localized SPs and their interaction with molecules will be discussed. Recent results of Raman/CARS nano-imaging/analysis of advanced nanomaterials including carbon nanotubes and DNA networks will be also shown.

The intention of this tutorial is to give the participants a fundamental background and working knowledge of the main physical ideas used in plasmonics, as well as an overview of modern trends in research and applications.

Instructors:
Sergey I. Bozhevolnyi
Aalborg University, Denmark

Satoshi Kawata
Osaka University, Japan

SESSION GG1: Metallic Nanostructures: Synthesis and Optical Properties
Chairs: Mark Brongersma and Igor Smolyaninov
Monday Morning, November 28, 2005
Room 308 (Hynes)

8:30 AM *GG1.1
Optical Properties of Mixed Monolayer Protected Metal Nanoparticles: from Chains of Chromophores to Chains of Nanoparticles. Francesco Stellacci, Materials Science and Engineering, MIT, Cambridge, Massachusetts.

Monolayer Protected Metal Nanoparticles (MPMNs) are supramolecular assemblies of organic thiolated molecules (ligands) around a small metallic core, 3 to 50 nm in diameter. The ligand shell is a densely-packed, highly-ordered self-assembled monolayer (SAM) whose morphology can be quite different from similar SAMs on flat surfaces. In particular, we have recently found that, when the ligand shell is composed of two types of different molecules, the SAM phase-separates into ring-shaped concentric domains of alternating composition. Here, we will show how this phenomenon can be used to tune and enhance the optical properties of the molecular ligands. In fact, when, in the ligand shell, one of the two types of molecules is optically active, confined domains with controlled inter-molecular spacing and intra-molecular conformation can be formed. Both these parameters can be varied by changing the concentration of the second optically inactive molecule. The optical properties of these MPMNs as a function of molecular morphology will be described. We will also show that the two polar singularities that form where the ring-shaped domains degenerate into points can be chemically modified with specific molecules. By using these 'polar' molecules as chemical handles nanoparticles chains can be easily synthesized. Their plasmonic properties will be discussed. .

9:00 AM GG1.2
Optical and Structural Properties of Photochromic Silver Nanoparticles. Alexander N. Sprafke[1], Christian Dahmen[1], Martina Luysberg[2], Henning Dieker[1], Matthias Wuttig[1] and Gero v. Plessen[1]; [1]I. Physikalisches Institut (IA), RWTH Aachen, 52056 Aachen, Germany; [2]Institut fuer Festkoerperforschung (IFF)/Forschungszentrum Juelich, 52428 Juelich, Germany.

Photochromic materials reversibly change their color under light illumination. In 2003, Ohko *etal.* presented a nanocomposite system with multicolor photochromic properties, consisting of Ag nanoparticles embedded in TiO_2 and fabricated by sol-gel processing [1]. The explanation for the photochromism is spectral hole burning in the particle-plasmon band which is caused by photoemission of electrons from the Ag nanoparticles. This new system has been suggested for applications such as optical data storage and displays [2]. We have recently reported that multicolor photochromic Ag/TiO_2 films can be fabricated by magnetron sputtering and subsequent thermal treatment, an approach that is easily scalable to large-area applications [3]. In the present work, we examine the spectral hole burning process in sputtered Ag/TiO_2 films. By optimizing the fabrication procedure, films with a narrow particle-plasmon line are obtained, which makes optical changes more easily discernible. Transmission spectra measured after laser irradiation of the films show a significant reduction in plasmonic oscillator strength with increased irradiation time. This reduction is accompagnied by a blue-shift of the plasmon band. The intensity dependence of the reduction indicates that the photoemission is a one-photon process. At large irradiation times and high intensities, the plasmon resonance almost disappears, which renders the Ag/TiO_2 films transparent. The bleaching of the plasmon line means that the photoemission significantly reduces the number of conduction electrons in the nanoparticles. For electrostatic reasons, electron depletion will not be uniform throughout the nanoparticle, but is expected to result in the formation of an Ag^+ shell and a neutral Ag core. This scenario is confirmed by X-ray diffraction scans of the photochromic films after various laser-irradiation times which show that the crystalline Ag cores shrink in size with increased irradiation time. Furthermore, chemical experiments indicate that Ag^+ ions are created during irradiation. These results allow one to understand the photochromic bleaching of the particle-plasmon line. In contrast, the irradiation-induced blue shift of the plasmon band is presently not fully understood. Conceivably it is caused by the dielectric properties of the Ag^+ shell, which differ from those of the TiO_2 matrix. [1] Y. Ohko *etal.*, Nature Mat. 2, 29 (2003). [2] K. Naoi *etal.*, J. Am. Chem. Soc. 126, 3664 (2004). [3] J. Okumu *etal.*, J. Appl. Phys. 97, 94305 (2005).

9:15 AM GG1.3
Ag(I)-Assisted Anisotropic Growth of Au Plasmonic Nanostructures. Mingzhao Liu and Philippe Guyot-Sionnest; James Frank Institute, The University of Chicago, Chicago, Illinois.

The mechanisms underlying colloidal nanorods formation from fcc Au are still rather mysterious.[1-5] Murphy et al. first introduced the seed-mediated approach. The seed and growth solution used leads to 5-fold twinned Au nanorods.[3] Subsequently, El-Sayed et al introduced Ag(I) in the growth solution which was shown to improve the yield of the Au nanorods.[1] This study investigates the essential roles of the seed's crystalline structure and of Ag(I) in promoting the formation of nanorods and novel diamond shaped structures. Gold nanorods with aspect ratio between 3 to 5 were synthesized by a Ag(I)-assisted seed-mediated method with a high yield. As a modification of the approach reported by El-Sayed et al,[1] the gold salt was reduced at a lower pH value. Using small single crystalline gold nanocrystals as the seeds (2-3 nm), the nanorods are single crystals, in contrast to the rods grown by Murphy et al. The growth direction is [001], and the side facets are dominated by {110}, which is similar to the gold nanorods prepared electrochemically.[4, 5] Varying the seed concentration in the range of 0.6-15 μM (in gold atoms) indicates that each nanorod is grown from a single seed. Dramatically different results are obtained with slightly larger gold nanocrystals seeds ($\sim$ 4 nm) which are predominantly twinned. With the same growth solution, these seeds lead to predominantly diamond-shaped gold nanocrystals which are twinned and a smaller fraction of single crystal rods. The tip angle of the diamond nanocrystals is around 30°. Optical spectra of the solution of these nanostructures of geometrically defined shape show a strikingly narrow ensemble plasmon resonance (peak at 750-900 nm, width $\sim$ 150 meV). TEM showed that the diamond-shaped nanocrystals have a similar twinned structure as the 5-fold twinned gold nanorods.[3] Their facets are tilted from the {100} plane, instead of the {100} facets in the twinned nanorods. Both the single-crystalline gold nanorods and the diamond-shaped gold nanocrystals have higher energy surface structures, i.e., the {110} surfaces and the steps on {100} surfaces, respectively. It has been suggested that the selective adsorption of Br- on the {110} faces slows down their growth rate and maintain the one-dimensional growth, leaving the role of Ag(I) unclear.[1,2] We propose that Ag(I) has an earlier underpotential deposition onset on these higher energy surface structures. Ag(0) therefore acts as a surfactant slowing down the growth rate of these more open faces which become dominant. (1) Nikoobakht, B.; El-Sayed M. A. Chem. Mater. 2003, 15, 1957 (2) Sau, T. K.; Murphy,

C. J. Langmuir 2004, 20, 6414 (3) Johnson, C. J.; Dujardin, E.; Davis, S. A.; Murphy, C. J.; Mann, S. J. Mater. Chem. 2002, 12, 1765 (4) Wang, Z. L.; Mohamed, M. B.; Link, S.; El-Sayed, M. A. Surf. Sci. 1999, 440, L809 (5) Wang, Z. L.; Gao, R. P.; Nikoobakht, B.; El-Sayed, M. A. J. Phys. Chem. B. 2000, 104, 5417

9:30 AM GG1.4
Monodisperse silver and nickel nanowires for surface enhanced Raman spectroscopy. Paul-Tiberiu Miclea[1], Guido Sauer[2], Georg Brehm[2], Siegfried Schneider[2], Kornelius Nielsch[3], Jinsub Choi[3], Petra Goering[3], Urilich Goesele[3] and Ralf Boris Wehrspohn[1]; [1]Physics, University Paderborn, Paderborn, Germany; [2]Physical and theoretical chemistry, University Erlangen-Nuernberg, Erlangen, Germany; [3]Max Planck Institute of microstructure physics, Halle (Saale), Germany.

Beside fundamental studies of the surface plasmons polaritons, in the last years metal nanoparticles find a direct application in detection and sensing of ultralow concentrations of molecules using surface enhanced Raman scattering (SERS). Metal nanoparticles by self assembling or by electron beam lithography are already as SERS templates. We propose in this paper a new approach, by using metal nanowires prepared from porous templates. 2D arrays of hexagonally arranged, monodisperse silver and nickel nanowires embedded in alumina template were prepared. The degree of template filling is near 100% using improved electrochemical deposition technique. When light is propagating along the long axis of the nanowire, plasmons enhanced absorption and light guidance of the nanowire were observed by optical micro-spectroscopy and scanning near-field optical spectroscopy. The results were compared with generalized Lorenz-Mie theory and with the approximation of infinite long cylinders. In-situ and ex-situ SERS of organic dyes by selectively dissolving of matrix materials were investigated in case of silver nanowires. We find a linear proportionality of the SERS signal intensity with the free surface of the metal nanowire in contact with the dye. Nickel, which is typical SERS-inactive material, shows in in-situ measurements a strong SERS signal. However, this signal can be observed only during the formation of quasi- hot spots formed during the nanowire releasing from the matrix material. The results are compared with the theoretical prediction for SERS enhancement by coupled metal cylinders and with enhancement as a function of dye molecule alignment to the long axis of the nanowire.

9:45 AM GG1.5
Electrical Detection of Plasmon Absorption in Individual Gold Nanorods. Christopher Nicholas LaFratta, Daniel Lim, Juliet Znovena, Linjie Li and John T. Fourkas; Chemistry, University of Maryland College Park, College Park, Maryland.

Electrical detection of Au plasmon absorbance has been accomplished in single nanorods wired using multiphoton absorption polymerization (MAP). Gold nanorods 50 nm in diameter and 5 um long have been individually addressed by fabricating polymer lines over them using MAP. After attaching polymer lines to the rods, the lines can be coated in metal by modifying the polymer surface and using electroless deposition techniques, resulting in an electrically addressable single Au nanorod. The resistance of the rod can be measured in a four probe scheme. Excitation of the nanorod with pulsed or CW radiation in the near infrared leads to a polarization-dependent change in resistance due to excitation of surface plasmons. The resistance has been found to change linearly with excitation power, and its polarization dependence is in good agreement with theory. The ability to electrically detect plasmon absorbance from a single rod may find applications in intra-beam polarization detection, biosensing, and other applications.

10:30 AM *GG1.6
Nanostructures of Noble Metals: Tailoring Their Surface Plasmonic Properties Through Shape Control. Younan Xia, Chemistry, University of Washington, Seattle, Washington.

This talk will discuss how chemical methods can be used to synthesize nanostructures of noble metals with well-defined shapes and hence controlled surface plasmonic properties. I will take three metals (silver, gold, and palaldium) as typical examples to disucss a number of chemical strategies that include polyol reduction, oxidative etching, use of capping agents, template direction. I will specifically focus on the nucleation and growth mechanisms involved in these synthetic routes, as well as their potential extensions to different systems. I will also demonstrate that shape control does provide a simple and powerful means for tailoring the surface plasmonic properties of these nanostructures for various applications.

11:00 AM GG1.7
Coupled-mode theory analysis of lossy plasmonic resonances in periodic metallic nanoparticle arrays. Zheng Wang[1], Shanhui Fan[2], Maozi Liu[3], Kazi Sultana[3], Edmond Chow[3], Danielle Chamberlin[3] and Annette Grot[3]; [1]Applied Physics, Stanford University, Stanford, California; [2]Electrical Engineering, Stanford University, Stanford, California; [3]Agilent Laboratories, Agilent Technologies Inc., Palo Alto, California.

During the past decade, micro-fabrication techniques have been remarkably improved to the extent that metallic nanostructures can be patterned and positioned reliably at a scale of a few nanometers. Taking advantage of the large field discontinuity in highly localized surface plasmon resonances at visible wavelengths, previous research has demonstrated Raman scattering enhanced by up to fourteen orders of magnitude,[1] as well as optical sensors capable of detecting index changes with zeptomole sensitivity.[2] In this paper, based on a temporal coupled-mode theory,[3] we develop general formalism for localized surface plasmon resonances in highly dispersive metallic materials, where frequency-dependent radiative and non-radiative decay rates determine the spectral lineshapes of the energy transfer. In addition, this theory relates the amplitude of the excitation, reflection and transmission to the amplitude of the resonances, thereby quantitatively connecting the scattering property to the resonance enhancement factors. To treat the frequency-dependent dielectric response of noble metals, we employ a set of parametric coupled-mode equations with built-in absorption losses. Since sub-wavelength lattice constants are used in typical nanoparticle arrays, the zero-th order diffractions in normal directions form the two ports in such optical resonator systems. Using this theory, we studied the resonance enhancement in square and triangular lattices of metallic nanoparticles arrays. The interplay between the plasmon-free-space impedance matching and the coherent scattering from a periodic array creates an optimal periodicity that maximizes the overall field enhancement throughout the structure, important for designing surface-enhanced Raman scattering substrates for quantitative analysis. Preliminary finite-difference time-domain (FDTD) calculation shows that a square lattice of silver particles resonating near 488nm has its field enhancement per unit area peaked at a particle spacing 3.5 times greater than the particle diameter, which agrees qualitatively with the analytical coupled-mode theory. The analytical coupled-mode theory predictions will be compared with experimental measurement on lithographically defined silver particles as well as the numerical FDTD calculations. [1] S. M. Nie and S. R. Emory, Science vol. 275, no. 5303, pp. 1102-6, 1997. [2] A. D. McFarland and R. P. Van Duyne, Nano Letters vol. 3, no. 8, pp. 1057-1062, 2003. [3] H. A. Haus and W. P. Huang, Proceedings of the IEEE vol. 79, no. 10, pp. 1505-1518, 1991

11:15 AM GG1.8
Magneto-Optical Kerr Effects of Magnetic Garnet Thin Films Incorporating Gold Nanoparticles: A Possible Coupling between Localized Surface Plasmons and Magneto-Optical Effects. Satoshi Tomita[1], Takeshi Kato[2], Shigeru Tsunashima[2], Satoshi Iwata[3], Minoru Fujii[4] and Shinji Hayashi[4]; [1]PRESTO, Japan Science and Technology Agency, Saitama, Japan; [2]Department of Electronics, Nagoya University, Nagoya, Japan; [3]CCRAST, Nagoya University, Nagoya, Japan; [4]Department of Electrical and Electronics Engineering, Kobe University, Kobe, Japan.

Localized surface plasmons (LSPs) are collective oscillations of free electrons in noble-metal nanoparticles, which are accompanied by an optical near-field in the vicinity of the nanoparticles. An energy transfer [1,2] and local field enhancement [3] via the plasmons in nanostructured materials have been of great interest in the growing field of plasmonics. When the noble-metal nanoparticles are embedded in a magnetized medium, it is expected that the LSPs of the nanoparticles are coupled to magneto-optical (MO) effects of the medium [4], leading to a modification and enhancement of the MO effects [5]. Nevertheless, little is known about an MO effect coupled with the LSPs of noble-metal nanoparticles. In this contribution, an experimental study for MO Kerr effects of magnetic yttrium iron garnet (YIG) thin films incorporating Au nanoparticles is reported. Transmission electron microscopic images and x-ray diffraction profiles indicated that metallic Au nanoparticles about 10 nm in diameter are precipitated in the boundary region of YIG nanocrystals several tens of nanometers in diameter. Ultraviolet-visible transmission spectroscopy showed that YIG without Au is almost transparent above 500 nm [6]. However, by incorporating the Au nanoparticles, a strong absorption peak due to the LSPs of the Au nanoparticles appeared around 600 nm. The polar MO Kerr spectra obtained for the films with Au volume fraction of 10.9% showed that the Kerr rotation angle becomes negative values in the region, where the LSPs of the Au nanoparticles are excited. The anomalous Kerr rotation indicated a possible coupling between the MO Kerr effects of YIG and the LSPs of the Au nanoparticles as a sensitizer. A mechanism for the coupling will be discussed. [1] S. A. Maier et al., Nature Material 2, 229 (2003).[2] P. Andrew and W. L. Barnes, Science 306, 1002 (2004).[3] S. Nie and S. R. Emory, Science 275, 1102 (1997).[4] V. A. Kosobukin, SPIE Proceeding 2535, 9 (1995).[5] V. I. Safarov et al., Phys. Rev. Lett. 73, 3584 (1994).[6] F. J. Kahn et al., Phys. Rev. 186, 891 (1969).

11:30 AM GG1.9
Giant Third-Harmonic Generation in Silver Nanoparticles: New Type of Hyper-Rayleigh Scattering. Oleg A. Aktsipetrov, Tatyana V. Murzina and Evgenia M. Kim; Department of Physics, Moscow State University, Moscow, Russian Federation.

Observation of surface-enhanced nonlinear optical effects in silver nanoparticles traces back to the early1980's when surface-enhanced optical second-harmonic generation (SHG) was observed in the silver island films. The enhancement of the SHG intensity up to three orders of magnitude was attributed to the resonant excitation of local surface plasmon in silver nanoparticles. In this paper, giant incoherent optical third-harmonic generation (THG) is observed in silver island films. The THG intensity from two-dimensional random array of silver nanoparticles is enhanced by two orders of magnitude. Silver island films were prepared by thermal evaporating onto substrate of silicon Si(001) wafers. Two types of samples are used in the nonlinear optical experiments: the silver island film samples with mass thickness of approximately 1 nm and thick homogeneous silver films with mass thickness of 40 nm. The choice of mass thickness of the silver island film samples is crucially important for observation of surface-enhanced THG as mass thickness of 1 nm corresponds to the UV resonant wavelength of local surface plasmons. Thick homogenous silver films are used as the reference samples for measurement of enhancement. The azimuthal dependencies of the THG intensity are measured from (i) silicon wafer (001), (ii) silver island film and (iii) thick flat reference silver film. Comparing the isotropic component of the THG signal from thick flat silver with the isotropic component of the THG signal from silicon (001) one can obtain the enhancement of THG in silver nanoparticles. For correct measurements of the surface enhancement of the THG intensity from island films we must know the diffuseness of THG. Scattering pattern is measured for the third-order HRS. The resulting angular scattering pattern demonstrates the appearance of a diffuse scattering component localized in the angular interval from 40 deg. to 50 deg. For quantitative measurement of enhancement one has to take into account both integration of the THG signal over scattering pattern and over the thickness of silver layer where nonlinear sources are localized in island films and reference thick film. Correct integrations gives the enhancement in nanoparticle silver films of more than two orders of magnitude. Spectral dependence of the THG intensity from silver nanoparticle films as a function of fundamental wavelength was measured with tunable output of OPO laser system. THG intensity demonstrates the sharp spectral peak centered in the vicinity of 810 nm for fundamental wavelength. This spectral peak can be attributed with excitation of local surface plasmons in the array of silver islands. This allows us to associate the mechanism of giant THG with resonant enhancement of local optical fields in random ensample of metal nanoparticles. In conclusion, surface-enhanced optical third-harmonic generation and third-order hyper-Rayleigh scattering is observed in silver island films.

11:45 AM GG1.10
Assembly of Nano-Plasmonic Structures for Robust Nanoscale Devices. Sudhaprasanna Kumar Padigi[1], Jorge Quijano[1] and Shalini Prasad[1,2]; [1]Electrical and Computer Engineering, Portland State University, Portland, Oregon; [2]Bio-Medical Engineering, Oregon Graduate Institute, Hillsboro, Oregon.

A Plasmon based technique for improving the Signal-to-noise ratio for micro and nano-scale devices used for sensing applications has been developed. It consists of dielectric substrate (glass) coated with metallic nanostructures. This results in the generation of Surface Plasmonic Waves (SPW, leading to signal amplification. With the focus on achieving better signal-to-noise ratio from miniaturized devices, there is a need for achieve reliable signals from these devices . The output voltages associated with these devices are in the order of a few tens of micro volts. This results in the interference of noise with the desired signal,lowering the signal-to-noise ratio (S/N). This prevents the signals from being analyzed for valuable data. We overcome this problem through surface plasmonics (SP). The principle of operation is as follows: when an electro magnetic wave is excited at the interface of a metallic nano-structure and a dielectric, Surface Plasmonic Waves (SPW) are generated. The SP waves amplify the electrical field component of the input light wave between 102-104 times. This leads to enhanced output voltage levels, leading to better S/N ratio, making the signals suitable for data analysis. Hence this technology has potential applications in building robust sub-micro size, ultra-sensitive sensors.

SESSION GG2: Structured Metallic Films
Chairs: Shanhui Fan and Stefan Maier
Monday Afternoon, November 28, 2005
Room 308 (Hynes)

1:30 PM *GG2.1
A Revised View of Enhanced Optical Transmission through Subwavelength Hole Arrays. Henri Joseph Lezec, [1]Applied Physics, California Institute of Technology, Pasadena, California; [2]CNRS, Toulouse, France.

Interesting optical transmission properties arise when subwavelength apertures are patterned in wavelength-scale proximity on a metallic surface. For example, when circular holes of subwavelength diameter are arranged to form a periodic array, the transmission spectrum develops a series of peaks. Extraordinary transmission efficiencies at these peak positions have been claimed and associated with discrete grating-coupling conditions that excite surface-plasmon polaritons (SPPs). In this talk, we re-evaluate the magnitude and origin of the effect by proper normalization of the as-collected transmission spectrum of the array to that of the corresponding isolated hole. The normalized spectrum then reveals a sequence of both enhancements and suppressions of similar and modest magnitude (less than a factor of ten). This bi-directional modulation is inconsistent with the SPP-resonance model commonly proposed to date. Instead, we propose a more general mechanism for the observed transmission modulation which is based on interference. A subwavelength aperture must couple inefficiently to a specific surface mode such as an SPP because it diffracts light into a continuum of evanescent surface waves with a large distribution of in-plane k-vectors. We show that these components sum to form an effective surface wave which is coherent over a short range and presents a characteristic phase-shift with respect to the source. We confirm the presence of this composite diffracted evanescent wave (CDEW) via interference experiments involving a single surface-wave launcher (narrow groove) placed at incremental distances from a single collector (slit or hole). We propose a new model for the transmission of hole arrays in which enhancement and suppression result from the interference of light directly incident upon (or emerging from) a given hole with CDEWs originating from neighboring holes. In particular, this simple model fully accounts for the transmission behavior of hole arrays as a function of illumination angle. In addition, it accounts for the salient optical properties of single apertures surrounded by surface corrugations, such as efficient, low-divergence beaming. CDEWs arise generically from in-plane scattering from subwavelength objects, and do not specifically require a metallic-dielectric interface to mediate their propagation (such as for SPPs). Indeed, we show that spectral modulation effects identical to those observed for hole arrays in metal films are also observed for hole arrays in non-metallic films, as well as in systems where no interface is present (a planar array of flat subwavelength-diameter disks suspended in a dielectric medium). Finally, we note that the CDEW contains a wealth of spatial frequency information about its source (as opposed to an SPP which is characterized by one spatial frequency only). Based on this insight, we discuss potential applications of the composite wave to subwavelength microscopy and lithography.

2:00 PM GG2.2
Theoretical and Experimental Study of Enhanced Transmission Through Subwavelength Linear Apertures Flanked by Periodic Corrugations. I. C. Schick[1], J. M. Yarbrough[1], C. G. Allen[1], R. T. Collins[1], R. E. Hollingsworth[2] and G. Nuebel[2]; [1]Physics, Colorado School of Mines, Golden, Colorado; [2]ITN Energy Systems, Inc., Littleton, Colorado.

We present a study of optical transmission through subwavelength apertures in Au films on glass. Samples consisting of 100-500 nm wide, 50 μm long, linear apertures flanked by periodic corrugations were prepared using electron-beam lithography with subsequent broad-beam ion milling. Transmission through these structures was studied experimentally and modeled numerically. Geometric parameters were varied in the numerical simulation which used a 2D Green's function approach and a frequency dependent Au dielectric function. Transmission with flanking corrugations was significantly enhanced related to an isolated aperture at resonant wavelengths for p-polarized incident light in agreement with the literature. Periodic corrugation also affected the spatial dependence of the transmitted field. Transmission characteristics of these structures were measured experimentally in the near- and far-field. Spectrally resolved far-field measurements where obtained for normal incidence with a spectrometer-coupled optical microscope configured for transmission measurements. Transmission spectra were consistent with modeled results as well as with the literature. A near-field scanning optical microscope (NSOM) was invoked to spatially and spectrally map the polarization dependent transmission through these structures. Enhanced transmission and additional structural features were observed in the NSOM images for p-polarization relative to s-polarization. The field intensity distributions were consistent with those observed in the simulations. The NSOM was also used to map the evolution of the transmitted field as a function of distance from the sample surface through the near- to far-field transition. This material is based on work supported by the National Science Foundation under Grant No. DMI-0340259.

2:15 PM GG2.3
Surface electromagnetic wave evolution and the photon-plasmon interaction: Optimizing light in/out-coupling and propagation efficiencies in plasmonic structures.
Jennifer A. Dionne[1], Henri Lezec[1,2], Domenico Pacifici[1], Luke A. Sweatlock[1] and Harry A. Atwater[1]; [1]Thomas J. Watson Laboratory of Applied Physics, California Institute of Technology, Pasadena, California; [2]CNRS, Paris, France.

On subwavelength scales, photon-matter interactions are limited by diffraction. While the diffraction limit restricts the size of device components for photonic confining and guiding, it offers a unique mechanism for coupling into surface electromagnetic waves (SEWs), including surface plasmons (SPs). Here, we investigate the nature of the photon-plasmon interaction and experimentally determine the design parameters which optimize photon-plasmon in- and out-coupling efficiencies. As light couples to a patterned metallic surface, wavelength-scale interactions include scattering, diffraction, and SP excitation. To investigate the nature of these interactions, we patterned in-coupling slits and gratings (launchers) and output slits and gratings (collectors) on 400nm thick Ag on SiO_2 using focused ion beam (FIB) fabrication. The launchers were defined as apertures through the Ag film while the collectors were defined to a depth of 30nm on the top surface of the Ag. Input grating periods ranged over p=550-650nm, and distances from the launchers to the collector varied over d=0, 5, 10, ..., 70μm. Using both broadband and monochromatic light sources (300-1500nm), the samples were illuminated through the SiO_2 substrate, with light in-coupling occurring on the Ag/air interface only in the location of the launchers. Light emitted from the collector was imaged at various detection angles by far-field microscopy and spectrally analyzed using a monochromator coupled to a CCD detector (sensitivity range 200-1100nm) or a Ge detector (sensitivity range 900-1700nm). Propagation distances for grating in-coupling ranged from 24-50μm over free-space wavelengths of 500-800nm. Using grating in-coupling, results for out-coupled intensity as a function of propagation distance suggest that extinction is characterized by a dependence consistent with material-related absorption at large distances and scattered wave evolution at short distances. However, using slit in-coupling, propagation distances are substantially shorter and exhibit a non-exponential distance dependence, suggesting that length scales of SEW spatial evolution are comparable to the launcher-collector distance. For both slit and grating in-coupling, the spectral distribution reveals two transmission maxima around 580 and 750nm, a convolution of the propagating SEW and the output grating response. The study allows us to propose first responses to the questions: Can SPs be discerned in the far-field with a signature distinct from diffracted orders of the subwavelength structure? By which mechanisms are input and output photon-plasmon coupling efficiencies optimized? What happens to input wavevectors exceeding SP momentum? Is the SP a preferred channel for SEWs generated by diffraction? Results highlight the distinction between surface plasmons and general SEWs and indicate the utility of both phenomena in design of efficient in/out-coupling structures between free space and SPP modes.

3:30 PM *GG2.4
Electromagnetic Surface States in Structured Perfect-Conductor Surfaces.
Javier Garcia de Abajo and Juanjo Saenz; CSIC and DIPC, San Sebastian, Spain.

Surface-bound modes in metamaterials forged by drilling periodic hole arrays in perfect-conductor surfaces are investigated by means of both analytical techniques and rigorous numerical solution of Maxwell's equations. It is shown that these metamaterials cannot be described in general by local, frequency-dependent permittivities and permeabilities for small periods compared to the wavelength, except in certain limiting cases that are discussed in detail. New related metamaterials are shown to exhibit exciting optical properties that are elucidated in the light of our simple analytical approach.

4:00 PM GG2.5
Diffractive Coupling in One and Two Dimensional Arrays of Nanoparticles and Nanoholes.
Erin McLellan Hicks, Xiaoyu Zhang, Shengli Zou, Jing Zhao, Kenneth G Spears, George Schatz and Richard P Van Duyne; Chemistry, Northwestern University, Evanston, Illinois.

The improvement of nanofabrication is one of the driving forces behind advancements in the fields of electronics, photonics and sensors. Precise control over nanoscale architecture is an essential aspect in relating new size-dependent material properties. Both direct write methods and natural lithography's offer a unique opportunity to fabricate "user-defined" writing of nanostructures in a wide range of materials. Electron Beam Lithography (EBL) and Nanosphere Lithography (NSL) provide the opportunity to fabricate precise nanostructures on a wide variety of substrates with a large range of materials. Using electrodynamics calculations, Schatz and coworkers have discovered one and two dimensional array structures that produce remarkably narrow plasmon resonance spectra upon irradiation with light that is polarized perpendicularly to the array axis. In order to investigate these interactions, precise control of nanoparticle orientation, size, shape and spacing is necessary. If the overall structures have excessive defects then the effect may not be seen. For the one dimensional arrays, to have the best control over array fabrication and to look at these interactions experimentally, EBL was used to construct lines of circular cylinders of varying interparticle spacings. Dark field microscopy was used to look at overall sample homogeneity and collect the single particle plasmon resonance spectrum. Additionally, a UV-visible spectrometer with a variable angle stage was used to look at the bulk line properties. In investigating of the two dimensional arrays, NSL was used in combinations with reactive ion etching to produce a film over nanowell substrate. Optical testing was done utilizing reflectance spectroscopy and scanning electron microscopy and atomic force microscopy were used to verify overall sample quality. In both cases new theoretical modeling was done to verify the results. This work provides the opportunity to further understand nanoparticle/nanohole interactions to make designed substrates for use in a multitude of fields.

4:15 PM GG2.6
Extraordinary Optical Transmittance through SiC - Ion Beam Processing for Surface Plasmonics.
Herman Hoegstroem[1], Sima Valizadeh[1], Francisco J. Garcia-Vidal[2], Louis Martin-Moreno[3] and Carl G. Ribbing[1]; [1]Solid State Physics, Dept. of Engineering Sciences, Uppsala, Sweden; [2]Dept. de Fisica Teorica de la Materia Condensada, Universidad Autonoma de Madrid, Madrid, Spain; [3]Dept. de Fisica de la Materia Condensada, ICA-CSIC, Universidad de Zaragoza, Zaragoza, Spain.

A number of reports on extraordinary optical transmittance (EOT) through thin metal films have been published.1 A proposed mechanism behind the high collimated transmittance through the subwavelength holes is the excitation of surface plasmons in the film-air interface channeling through the holes and subsequently radiation from the exit side.2 This explanation has been disputed3 and more basic work is required to explore this route to develop subwavelength optical components. A few studies of optical effects of structured thin ceramic samples have been reported.4 In contrast to metals, these compounds are electrically insulating, which is of interest for the selection of mechanisms behind EOT. It may also be useful for applications in which metals cannot be used. The measurements of reflectance and emissivity in ref. 4 have been complemented with transmittance calculations.5 The conclusion was that surface phonon polariton excitation occured, but it is not a neccessary condition for EOT. In a recent study the sensitivity of surface excitations to defect implantation by a focused Be2+-ion beam was demonstrated using near-field optical microscopy.6 In this contribution we report calculations and IR-microscope transmittance measurements for SiC-samples that have been patterned with holes using Ga-ions in a Focused Ion Beam (FIB) instrument starting with a 25 micron thick wafer. The calculations were based on excitation of surface phonon polaritons that channel the electromagnetic energy through the subwavelength holes.2 The calculations indicated that a 10 micron SiC-wafer with a square array of r=2.5 micron circular holes 11 micron apart, would exhibit close to 20 % resonant transmittance at the wavelength12.8 micron. We report on the FIB-preparation of SiC-samples with square hole patterns and their IR transmittance spectra. References 1. H. J. Lezec, A. Degiron, E. Devaux, R. A. Linke, L. Martin-Moreno, F. J. Garcia- Vidal, T. W. Ebbesen Science 297, 820 (2002). 2. L. Martin-Moreno, F. J. Garcia-Vidal, H. J. Lezec, K. M. Pellerin, T. Thio, J. B. Pendry, T. W. Ebbesen Phys. Rev. Lett. 86, 1114 (2001). 3. Q. Cao, P. Lalanne Phys. Rev. Lett. 88, 057403 (2002). 4. L. Gall, M. Olivier, J.-J. Greffet Phys. Rev. B55, 10105 (1997). 5. F. Marquier, K. Joulain, J.-J. Greffet Opt. Lett. 29, 2178 (2004). 6. N. Ocelic, R. Hillenbrand Nature Materials 3, 606 (2004).

4:30 PM *GG2.7
Extraordinary Optical Properties of Structured Metals.
Francisco J. J. Garcia-Vidal[1] and Luis Martin-Moreno[2]; [1]Departamento de Fisica Teorica de la Materia Condensada, Universidad Autonoma de Madrid, Madrid, Spain; [2]Fisica de la Materia Condensada, Universidad de Zaragoza, Zaragoza, Spain.

In this talk we address from a theoretical point of view two physical phenomena that are related to the excitation of surface electromagnetic (EM) resonances (surface plasmons) at structured metal surfaces: i) extraordinary optical transmission through single and periodically structured subwavelength apertures and ii) beaming of light through single apertures surrounded by periodic corrugations. First we analyze the pioneering experiment of Ebbesen et al. [1] on 2D hole arrays in the optical regime by presenting a fully three-dimensional numerical treatment of the structure [2]. Moreover, we will show how subwavelength holes (2D arrays or 1D linear chains)

in a perfect conductor also gives rise to similar extraordinary transmission [3] even though the free surface of an unperforated perfect conductor has no surface modes. We will see that there are not two separate mechanisms: the holes will spoof surface plasmons [4] which play the same resonant role as the real ones on silver or gold in the optical regime. Then, we will discuss the enhanced optical transmission phenomenon and beaming effects both observed in single subwavelength apertures (holes or slits) flanked by periodic corrugations, as reported in [5]. We will present the theoretical foundation for these phenomena and discuss the various dependences of the transmission [6] and emission profiles [7] on the geometrical parameters defining the system, providing guidelines for the optimisation of these profiles for different possible applications, like lensing [8] . Finally, we will explore how these results found for metal surfaces can be exported to dielectric photonic crystals (PCs). We will show that, through appropriate corrugation of the PC surface, it is possible to obtain both enhanced transmission through slits in PC slabs, and strong beaming of light coming out of a PC waveguide [9].
References [1] T.W. Ebbesen, H.J. Lezec, H.F. Ghaemi, T. Thio, and P.A. Wolff, Nature 391, 667 (1998). [2] L. Martin-Moreno, F.J. Garcia-Vidal, H.J. Lezec, K.M. Pellerin, T. Thio, J.B. Pendry and T.W. Ebbesen, Phys. Rev. Lett. 86, 1114 (2001). [3] M. Beruete, M. Sorolla, I. Campillo, J.S. Dolado, L. Martin-Moreno, J. Bravo-Abad, and F.J. Garcia-Vidal, Opt. Lett. 29, 2500 (2004); J. Bravo-Abad, F.J. Garcia-Vidal and L. Martin-Moreno, Phys. Rev. Lett. 93, 227401 (2004). [4] J.B. Pendry, L. Martin-Moreno and F.J. Garcia-Vidal, Science 305, 847 (2004). [5] H.J. Lezec, A. Degiron, E. Devaux, R.A. Linke, L. Martin-Moreno, F.J. Garcia-Vidal and T.W. Ebbesen, Science 297, 820 (2002). [6] F.J. Garcia-Vidal, H.J. Lezec, T.W. Ebbesen and L. Martin-Moreno, Phys. Rev. Lett. 90, 213901 (2003). [7] L.Martin-Moreno, F.J. Garcia-Vidal, H.J. Lezec, A. Degiron and T.W. Ebbesen, Phys. Rev. Lett. 90, 17401 (2003). [8] F.J. Garcia-Vidal, L. Martin-Moreno, H.J. Lezec and T.W. Ebbesen, Appl. Phys. Lett. 83, 4500 (2003). [9] E. Moreno, F.J. Garcia-Vidal and L. Martin-Moreno, Phys. Rev. B 69, 121402(RC) (2004).

SESSION GG3: Poster Session I
Chair: Mark Brongersma
Monday Evening, November 28, 2005
8:00 PM
Exhibition Hall D (Hynes)

GG3.1

The Role of Polaritons in Scattering Near-field Optical Microscopy.
Stefan Grafstrom, Jan Renger, Susanne Schneider and Lukas M. Eng; Institute of Applied Photophysics, Dresden University of Technology, D-01062 Dresden, Germany.

In scattering-type scanning near-field optical microscopy (s-SNOM) a laser beam is focussed on the apex of a sharp tip in close proximity to the sample surface and the light scattered off the tip is detected in the far field. The optical interaction between the tip and the sample can be described in a dipole model [1] in which the incident light induces a dipole in the tip whose field polarizes the sample thereby creating an image dipole, which acts back on the tip and modifies its dipole moment. The field radiated by the two interacting dipoles constitutes the scattered wave. The scattering cross section is strongly enhanced if a polariton is excited in the tip-sample system. The polariton may originate either from the free conduction electrons of a metal (plasmon polariton) or from an optical-phonon resonance (phonon polariton). This effect can be used to improve the sensitivity and resolution in s-SNOM. Depending on the dielectric constants of both tip and sample, either a particle polariton in the probe or a surface polariton on the sample may be excited. For the latter resonance to be excited the presence of the tip is crucial as it breaks the translational symmetry, thereby lifting the requirement of k vector matching. Once the tip-sample separation is reduced, the coupling between tip and sample leads to an increasingly pronounced spectral shift and splitting of the polariton resonances [2]. We use the dipole model for an approximate description of the dependence of the polariton resonances on the tip-sample separation and the optical constants of the materials involved. The scatterer is treated as a sphere or more generally as an ellipsoid. We allow for optical anisotropy of the sample reducing the symmetry of the tip-sample system [3]. The analytical results of the dipole model are complemented by numerical calculations based on the multiple-multipole method [2,4]. They show that the overall behavior is correctly described by the dipole model. Additionally, they provide detailed information on the exact electric-field distribution associated with polariton resonances.
[1] B. Knoll and F. Keilmann, *Opt.Commun.* **182**, 321 (2000). [2] J. Renger, S. Grafstrom, L.M. Eng, and R. Hillenbrand, *Phys.Rev.B* **71**, 115418 (2005). [3] S.C. Schneider, S. Grafstrom, and L.M. Eng, *Phys.Rev.B* **71**, 115418 (2005). [4] J. Renger, S. Grafstrom, L.M. Eng, and V. Deckert, *J.Opt.Soc.Am.A* **21**, 1362 (2004).

GG3.2

Tunable Plasma Resonance of Ag Nanorod Arrays Prepared by Dynamic Oblique Deposition.
Motofumi Suzuki[1], Wataru Maekita[1], Kaoru Nakajima[1], Kenji Kimura[1], Takao Fukuoka[2] and Yasushige Mori[3]; [1]Department of Micro Engineering, Kyoto University, Kyoto, Kyoto, Japan; [2]Kyoto CREATE, JST, Seika, Kyoto, Japan; [3]Department of Chemical Engineering and Materials Science, Doshisha University, Kyotanabe, Kyoto, Japan.

We have succeeded in fabricating arrays of Ag nanorods by a dynamic oblique deposition (DOD) during which the deposition angle and the inplane direction of the substrate are changed. Discontinuous Ag islands are deposited obliquely on the template layer of SiO_2, which has a strong anisotropic surface morphology created by serial bideposition (SBD), a kind of DOD. [1] Due to shadowing effect of the surface corrugation of the template, Ag grows into the rod-like shape, and these Ag nanorods are aligned in such a way that the major axis are quasi parallel to each other. The aspect ratio of the Ag nanorods has been varied from 1 to 6 depending not only on their own deposition conditions (thickness and deposition angle of Ag) but also on those of template (number of SBD and deposition angle of SiO_2). These nanorod arrays show the strong anisotropy in the optical absorption due to the shape-sensitive plasma resonance of the free electrons in the Ag nanorods. The relationship between the resonance energies and the aspect ratio agrees well with the semi-empirical equations proposed by Kuwata et al. [2] so that the resonance energies are tunable by controlling the aspect ratio of the nanorods. Since the method presented here is based on the physical self-organization processes, it can be applied, in principle, to any combination of materials. Moreover, it requires no etching, heating or deformation processes other than vacuum evaporation. Therefore, our thin films with the aligned nanorods are useful for various plasmonic applications such as thin film polarizers and surface enhanced Raman scattering substrates.
[1] M. Suzuki, W. Maekita, K. Kishimoto, S. Teramura, K. Nakajima, K. Kimura and Yasunori Taga, Jpn. J. Appl. Phys., 44 (2005) L193. [2] H. Kuwata et al., Appl. Phys. Lett., 83 (2993) 4625.

GG3.3

Femtosecond Photoluminescence from Gold Nanospheres and Nanorods.
Oleg Varnavski[1], Goodson III Theodore[1], Mohamed Mona[2] and El-Sayed Mostafa[2]; [1]Chemistry, University of Michigan, Ann Arbor, Michigan; [2]Laser Dynamics Laboratory, School of Chemistry and Biochemistry, Georgia Institute of Technology, Atlanta, Georgia.

The dynamics of optical excitations in metal nanoparticles have attracted a wide degree of interest due to possible applications in nanotechnology. In this contribution we report the femtosecond visible photoluminescence from gold nanoparticles using time-resolved fluorescence upconversion spectroscopy. We directly compared this fast luminescence from gold nanospheres (Φ25nm) with that obtained from nanorods (Φ5x40 nm,Φ15x27nm) under Vis (3.02eV) and UV(4.65eV) excitation. The decay of the photoluminescence was found to be very fast, <50fs for both nanorods and nanospheres. The spectrum and efficiency of this emission were the same at different excitation wavelengths for each of the investigated geometries (nanospheres, nanorods of two different aspect ratios). The photoluminescence emerged instantly with excitation (delay<50fs) and it was depolarized. Degenerate femtosecond pump-probe experiments in the low excitation intensity regime demonstrated much slower electron thermalization/equilibration dynamics on the time scale of few hundred femtoseconds (‘540fs). These features strongly indicate a d-hole - conduction electron recombination process as the origin of this photoluminescence. A direct comparison of the fast emission spectra from nanorods and nanospheres is used to discuss the emission enhancement mechanism.
References 1. O. Varnavski, R.G. Ispasoiu, L. Balogh, D. Tomalia, and T. Goodson III, J. Chem. Phys. 114, 1962 (2001). 2. O. Varnavski, M. Mohamed, M. El-Sayed, , T. Goodson III, J. Phys. Chem. B 107, 3101 (2003). 3. T. Goodson III, O. Varnavski, Y. Wang, Int. Rev. Phys. Chem. 23, 109 (2004). 4. O. Varnavski, M. Mohamed, M. El-Sayed, T. Goodson III, Phys. Rev. B, 2005, submitted.

GG3.4

Photoresponsivity and Nanopatterning of Azobenzene-Functionalized Polymer Systems Induced by Surface Plasmons from Periodic Metallodielectric Arrays.
Jae-Chul Hong, Jeong-Ho Park, Chaemin Chun and Dong-Yu Kim; Dept. of Materials Science and Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju, South Korea.

Recently a great deal of interest has been focused on the development of periodic metallic structure and arrays for the propagation of optical waves as surface modes called surface plasmons (SPs) that associated with the excitation of a collective oscillation of electrons on a nano-scaled metal-dielectric interface. Excitation of SP modes on

metallic structures have introduced interesting optical properties to photonic materials including the extraordinary transmission of light through subwavelength hole arrays, plasmonic waveguides, surface enhanced Raman scattering and surface plasmon resonance sensing. In this presentation, we examine the effect of the azobenzene-based polymer media on SP propagation on extended 1, 2-dimensional metallodielectric array structures. When azobenzene-functionalized organic materials are exposed to the polarized light, azobenzene chromophores perform photoisomerization and orientational redistribution. Azobenzene chromophores were reoriented and mass-flowed by the interference pattern of the polarized light. It could result optical fabrication of surface relief grating on azo-based polymer film. We demonstrate the fabrication of the metallodielectric arrays of the silver and the photoresponsive behavior of the azobenzene molecules by the localized SP resonance on the Ag metallodielectric arrays. For the preparation of metallodielectric structures, microcontact printing and electroless plating method were used. This maskless fabrication method could be useful for large-area, periodic metal array structures with submicron features that exhibit efficient SP excitation and propagation characteristics. Epoxy-based azobenzene-functionalized polymer was spin-coated on the Ag arrays, or the other way. The polarized light was irradiated on the azopolymer films with periodic metallic arrays for the excitation of the azobenzene moiety and the surface structure modification of the azopolymer film. We investigated the dynamics of azobenzene molecules in the microscopic point and the derived patterns in the azopolymer-metallic arrays in the macroscopic point.

GG3.5
Second Harmonic Generation by Scattering of Surface Plasmon Polaritons by Metallic Nanodefects. Lina Cao[1], Nicolae C. Panoiu[2], Shuang Zhang[3], Wenjun Fan[3], K. J. Malloy[3], S. R. J. Brueck[3] and Richard M. Osgood[2]; [1]Department of Chemistry, Columbia University, New York, New York; [2]Department of Applied Physics and Applied Mathematics, Columbia University, New York, New York; [3]Center for High Technology Materials, University of New Mexico, Albuquerque, New Mexico.

Surface plasmon-polaritons (SPP) can greatly enhance the electrical field near a metal/dielectric interface. In particular, metal nanoparticles or surface defects at a metal/dielectric interface have been shown to exhibit strong SPP field enhancement to increase the efficiency of nonlinear optical effects, such as Raman scattering, second-harmonic generation (SHG), and third-order nonlinear processes. Among these effects, SHG plays a central role in surface science, as it is used to study surface phenomena such as molecular adsorption, aggregation and orientation, or to study the surface structure of buried interfaces. In addition, since at the surface of a medium the inversion symmetry is broken, the SHG can provide a valuable tool to investigate the surface structure of a centrosymmetric material. In this paper, we analyze the process of the second harmonic generated upon the scattering of a SPP, propagating on a flat metal/dielectric interface, by one-dimensional metallic nanodefects, such as linear stripes (protuberances) or grooves (indentations). For this, using the formalism based on the reduced Rayleigh equations, we first determine the total electromagnetic field at the fundamental frequency, which consists of the incoming SPP wave, the transmitted and reflected SPP waves, and the electromagnetic field scattered into photon states (radiative modes). Subsequently, we compute the induced surface polarization density at the second harmonic and the corresponding electric dipole, magnetic dipole, and electric quadrupole. Finally, these multipoles are used to calculate the electromagnetic field distribution at the second harmonic. We conclude by discussing our results, namely the dependence of the radiated power at the second harmonic on the shape and size of the metallic nanodefects, the SPP wavelength, as well as the components of the surface second order susceptibility tensor (its normal and tangent components, assumed to be isotropic).

GG3.6
Surface Plasmonic Properties of Pd Nanostructures. Yujie Xiong, Jingyi Chen, Benjamin Wiley and Younan Xia; Chemistry, University of Washington, Seattle, Washington.

The surface plasmon resonance (SPR) peaks of Pd nanostructures could be tuned from UV to visible region by controlling their size, shape, or structure (hollow vs. solid). We have recently realized these opportunities by modifying the polyol synthesis with three different approaches: (i) to increase the size of Pd nanocubes from 8 and 50 nm and thus shift the SPR peak from 225 to 390 nm by controlling the number of seeds formed in the nucleation stage via oxidative etching; (ii) to transform solid nanocubes of Pd into hollow nanocages and thus shift of SPR peak from 410 nm to 520 nm via a corrosive pitting process; and (iii) to grow twinned Pd nanoparticles into nanorods, and thus shift their SPR peak towards the visible region. All the measured spectra matched well with the results calculated using discrete dipole approximation (DDA). It is worth noting that the SPR properties of Pd nanostructures remain largely unexplored because Pd nanoparticles below 10 nm in size only have a SPR peak in the UV region (around 225 nm). It is expected that the present work will enable us to shift the SPR peaks of Pd nanostructures towards longer wavelengths and open the door to new applications in the visible and near infrared regions.

GG3.7
Radiation Damping of Plasmons in Metal Nanoparticle Pairs. Christian Dahmen and Gero v. Plessen; I. Physikalisches Institut (IA), RWTH Aachen, 52056 Aachen, Germany.

The optical properties of electromagnetically coupled metal nanoparticles have recently received much interest. One of the reasons for this interest is the large local-field enhancement at the surface of the coupled nanoparticles, which leads to surface-enhanced Raman scattering (SERS) [1]. Another point of interest are the spectral properties of the particle-plasmon resonances, which show large frequency shifts when the particles experience near-field coupling at small particle spacings [2]. Less attention has been devoted to studying the radiation damping in small aggregates of metal nanoparticles. In the present work, we calculate the radiation damping rates of plasmon resonances in metal nanoparticle pairs. Generalized Mie theory is employed to calculate the scattering cross sections of pairs of gold nanospheres, taking retardation effects into full account. The radiation damping rate is extracted from the particle-plasmon linewidth of the scattering-cross-section spectra. The radiation damping of the coupled particle-plasmon alternates between superradiative and subradiative behavior when the interparticle distance is varied. The damping thus depends on the relative phase of the dipole of one particle with respect to the field scattered by the other particle. This behavior is similar to that of the spontaneous emission rate of coupled ions whose ion-ion distance is varied [3], and is linked to the line narrowing recently calculated for nanoparticle chains [4]. For certain polarization conditions, we find electromagnetic coupling between nanoparticles over remarkably large distances, exceeding 20 times the particle diameter. At small particle spacings where near-field coupling sets in, the radiation damping rate lies far below that of an isolated particle. This finding is a consequence of the large plasmon red shift induced by the near-field coupling of the particles. This red shift reduces the coupling to the far field. The reduction of radiation damping implies a reduced dephasing of the coupled plasmon mode and thus tends to increase the local field enhancement between the particles. [1] H. Xu *etal.*, Phys. Rev. E 62, 4318 (2000). [2] P. Nordlander *etal.*, Nano Lett. 4, 899 (2004). [3] R. G. DeVoe *etal.*, Phys. Rev. Lett. 76, 2049 (1996). [4] S. Zou *etal.*, J. Chem. Phys. 120, 10871 (2004).

GG3.8
Iodization of RF Sputtered Ag Thin Films Reveals Surface Plasmon-Exciton 'Transition.'. C. S. Sunandana and D. Bharathi Mohan; School of Physics, University of Hyderabad, HYDERABAD, Andhra Pradesh, India.

Ag thin films 5-25 nm thick were produced by RF magnetron sputtering onto glass substrates. They were subjected to controlled iodization in an hour-glass type chamber for periods ranging between 5 min-60 hr. Apart from structural/microstructural characterization by XRD and AFM they were monitored by optical transimission spectra in the range 300-700 nm. All the films were X-ray amorphous while AFM showed particles in the range 25-75 nm. Thin films of intermediate thickness (10 and 20 nm) show extremely interesting quasiparticle formation and /transition /as a function of irradiation time. plasmons (peak wavelength ∼500 nm) formed for 5 nm irradiation /decay /upon further iodization to reduce (irreversibly) excitons at 423 nm in the manner of a metal-semiconductor or dielectric phase transition. At low levels of iodization (<20 min) the thin film is actually a AgI-Ag nanocomposite with an overall metallic response. For higher levels of iodization (>20 min) the response is semiconductor like with the plasmons completely /decaying /to produce excitons characteristic of nanosized gamma AgI with zincblende structure and a band gap of 2.95eV which is 0.1 eV wider than the thermally evaporated Ag thin films. The favourable kinetics of the formation and decay of these quasiparticles is mainly due to the unique nanostructure stabilized by RF sputtering. Blue shift in the λmax observed for surface plasmon resonance and excitons of AgI for shorter and longer iodization respectively with decreasing film thickness implies the formation of γ-AgI nanoparticles on the Ag nanostructure matrix. As the iodization proceeds from 5 min to 60 hrs, the intensity of the exciton peak increases progressively and for longer iodization time the number of excitons increases with increasing film thickness. Ag thin films produced by thermal evaporation do not apparently produce plasmons upon iodization and thus no plasmon-exciton /transition / occurs in these latter films. A rigorous quantitative monitoring of the entire process is expected to lead to applicable nanostructured devices in the area of microelectronics, optoelectronics, sensor technology, battery cathodes

and excitonic photoluminescence.

GG3.9
Z-scan Studies of the Third-order Optical Nonlinearity of Sol-gel Derived Gold Nanocrystal/Silica Films. Kai Yang[1], Hongyou Fan[2,3], Li Wang[1], Olivier J. Chalus[4], Weiliang Chen[1], Ravi Jain[1], Kevin J. Malloy[1], Jeffrey Brinker[2,3], Jean-claude Diels[4] and Thomas W. Sigmon[1]; [1]Center for High Tech Materials, University of New Mexico, Albuquerque, New Mexico; [2]Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico; [3]Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico; [4]Department of Physics and Astronomy, University of New Mexico, Albuquerque, New Mexico.

Nonlinear optical properties of metal nanoclusters are being investigated by a number of research groups owing to their potential use in nonlinear optical devices. The absorption spectra of these materials show a characteristic surface plasmon resonance (SPR) band, a result of the collective oscillations of the conduction electrons in the metal nanoparticles. Gold nanoparticle composite materials are known to display large optical nonlinearities and rapid time response in the SPR absorption region. These properties have important applications in ultrafast communications, optical data storage, optical limiters and all-optical switching devices. Third-order susceptibilities on the order of 10^{-7} esu have been obtained for silica glasses containing dispersed gold at the SPR frequency using both the degenerate four wave mixing (DFWM) and Z-scan measurement techniques. The main advantages of using the z-scan measurement over the DFWM method is a result of its ability to obtain the imaginary and real parts of $\chi^{(3)}$ and, additionally, to determine the algebraic signs of the two components. When compared to other methods (for example, metal ion implantation), the sol-gel process allows a convenient and cost effective way for producing metal nanoparticle thin film samples. In this work, third-order nonlinear optical properties of sol-gel derived gold nanocrystal(NC)/silica thin films are studied both on and off the SPR wavelength by the z-scan technique using short pulse lasers. The gold nanoparticle mean size diameter is ∼3 nm (variation less than 7%) and the gold volume fraction in the film is about ∼5.6 %. The peak of the SPR spectrum for our gold NC/silica films occurs at ∼520 nm. We have determined the third order nonlinear optical susceptibility for these films using the z-scan method at both 820 nm (mode-locked Ti:Sapphire laser (off SPR)) and 532 nm (Q-switched double frequency Nd:YAG laser (on SPR)). For the 820 nm measurements, the values of the nonlinear absorption coefficient β and nonlinear refractive index coefficient γ determined using open- and closed-aperture Z-scan measurements are $\beta = 3.5 \times 10^{-7}$ m/W and γ = -4.3 $\times 10^{-14}$ m^2/W, respectively. For 532 nm, the corresponding values for β and γ are β = -2.3 $\times 10^{-5}$ m/W and γ = -4.5 $\times 10^{-13}$ m^2/W, respectively. Both the real and the imaginary part of $\chi^{(3)}$ were found to be negative at 532 nm. This corresponds to the absolute value ($-\chi^{(3)}-$) of ∼1.1 $\times 10^{-7}$ esu. Thermal effects and the enhancement of optical nonlinearities resulting from the surface plasmon resonance in these films are discussed in detail.

GG3.10
Surface Palimon Resonance of Golden Nanoparticles in 2D-Layer Characterization at the Interface with ds(ss)DNA-Fullerene C60 and DNA-Au Nanoparticles Supramolecules. Evgenia Buzaneva, Radiophysical Faculty, National Taras Shevshenko University of Kyiv, Kiev, Ukraine.

The kinetic changes of Surface Plasmon Resonance (SPR) response for golden nanoparticles in 2D-layer at the interface with adsorbed molecules from colloidal solutions of C60, C60 oxygen and hydroxyl group derivatives, DNA molecules, and with their mixtures, were recorded [1]. For the understanding of a behavior of SPR signal at time were studied the properties of adsorbed layers on golden nanoparticles layer surface by vibrational mode and UV-visible spectroscopy, and nano-, micro-images. The assembling and bonds at the interface were considered in the models of kinetic changes of SPR response for golden nanoparticles 2D-layer. [1] E. Buzaneva, A. Gorchinskiy, P. Scharff, K. Risch, A. Nassiopoulou, C. Tsamis, Yu. Prilutskyy, O. Ivanuta, A. Zhugayevych, D. Kolomiyets, A. Veligura, DNA, DNA/Metal Nanoparticles, DNA/Nanocarbon and Macrocyclic Metal Complex/Fullerene Molecular Building Blocks for Nanosystems: Electronics and Sensing, in E. Buzaneva and P. Scharff (Eds), Frontiers of multifunctional integrated nanosystems, NATO Science Series, II-Mathematics, Physics and Chemistry, Vol 152, Kluwer Akademic Publishers, Dordrecht, 2004, p. 256-272.

GG3.11
Quantitative Tuning of Silver Nanoparticles Confined on Indium Tin Oxide Surface by Spectroelectrochemistry. Xiaoyu Zhang, Erin M. Hicks, Jing Zhao and Richard P. Van Duyne; Chemistry, Northwestern University, Evanston, Illinois.

An electrochemical method is developed to quantitatively modify and spectroscopically monitor the size and shape of Ag nanotriangles fabricated by nanosphere lithography (NSL) on an indium tin oxide (ITO) electrode surface. AFM and SEM results demonstrate that the preferential order of electrochemical oxidation for a nanotriangle is, surprisingly, bottom edges first, then triangular tips, then out-of-plane height.

SESSION GG4: Microscopy and Imaging
Chairs: Francesco Stellacci and Younan Xia
Tuesday Morning, November 29, 2005
Room 308 (Hynes)

8:30 AM *GG4.1
Immersion Microscopy Based on Plasmonic Crystal Materials. Igor I. Smolyaninov, ECE Department, University of Maryland, College Park, Maryland.

Theoretical model of the enhanced optical resolution of the surface plasmon immersion microscope is developed, which is based on the optics of surface plasmon Bloch waves in the tightly bound approximation. It is shown that a similar resolution enhancement may occur in a more general case of an immersion microscope based on photonic crystal materials with either positive or negative effective refractive index. Both signs of the effective refractive index have been observed in our experiments with surface plasmon immersion microscope, which is also shown to be capable of individual virus imaging.

9:00 AM GG4.2
Direct imaging of the propagation and damping of infrared surface plasmons. Ewold Verhagen and Albert Polman; Center for Nanophotonics, FOM-Institute AMOLF, Amsterdam, Netherlands.

We present a novel method to image infrared surface plasmon polaritons (SPPs) propagating along a Ag/SiO2 interface. The SPPs, excited at 1.49 μm using a semiconductor laser, are coupled to optically active erbium ions implanted in the SiO2 at close proximity to the Ag interface. We study the propagation and damping of the SPP modes in the far-field by two-dimensional imaging of the Er photoluminescence intensity at 1.53 μm using a confocal microscope. The SPP modes are found to remain confined to stripe waveguides, and exponential decay of the mode intensity is observed with characteristic propagation lengths of 100 μm, in agreement with calculations. In these experiments, SPPs at the Ag/SiO2 interface are excited by illumination from the air side of the Ag film using two types of microstructures. First, gratings in ∼50 nm thick Ag films are used to excite SPPs at the Ag/air interface. These gratings are designed with a pitch of 3.3 μm to allow cross coupling to SPPs propagating along the Ag/SiO2 interface. Second, hole arrays in optically thick films, optimized for the excitation of modes at the Ag/SiO2 interface, are used to launch SPPs into waveguides. This imaging technique is a useful tool to investigate the propagation of bound SPPs and long-range surface plasmon polaritons (LR-SPPs) in waveguides and optical components. Although numerous techniques exist to image (leaky) surface plasmons propagating at a metal/air interface, direct observation of bound modes in both symmetric and asymmetric dielectric environments is less straightforward. By taking advantage of cooperative upconversion effects, this technique can also be used to determine the local surface plasmon field intensity.

9:15 AM GG4.3
Optical Microscopy Techniques for Surface Plasmons. David Nicholas Barsic, Benjamin Reddy and Mark L. Brongersma; Stanford University, Stanford, California.

We are investigating a number of optical microscopy techniques useful for studying surface plasmon polariton (SPP) propagation in metal films. The goal of this research is to study the uses and limitations of a few relatively simple microscopy techniques. First, techniques for launching surface plasmons through grating couplers patterned in the metal films or through oil immersion condenser lenses will be considered. Next, a method using a layer of fluorescent molecules or particles to map out SPP fields in a fluorescent microscope are shown. Finally, a technique which collects and images light emitted into a high index substrate by leaky SPP modes is presented. All of these techniques can be easily implemented with relatively simple modifications to an optical microscope, and can provide some insight to the behavior of SPP waves through the observation and interpretation of far-field images.

9:30 AM *GG4.4
Metallic Photon Reservoirs for Nano-imaging and Lasing. Satoshi Kawata, [1]Applied Physics, Osaka University, Suita, Osaka, Japan; [2]RIKEN, Saitama, Japan.

When photons are incident on a metal nanostructure, the photon energy is transferred to surface plasmon polariton (SPP) energy. The SPP energy is accumulated on the surface of metal, resulting in the amplification of the local field of light. Here, the metal surface works as a photon reservoir. We will talk about two types of metallic nanostructures as photon reservoirs: nano-cone and periodic corrugation. A nano-cone has been used as a near field scanning optical microscope probe, in particular, for Raman imaging and spectroscopy of nano-materials. This configuration is quite similar to that of surface enhanced Raman scattering spectroscopy, except that the force is exerted between the metal tip and the molecule in contact-mode AFM operation. We will show some recent results on DNA imaging and carbon nanotube analysis with metallic nano-cone tip [1, 2]. A periodically corrugated surface of metal also reserves photons in metal as SPPs at the edge of certain energy band, in which surface plasmon propagation is prohibited in all lateral directions. By careful design of the periodic structure topography on both sides of the metal film, SPPs can be made to accumulate at the band gap edge frequency, and can then exhibit a lasing effect. We will show the fabrication, configuration, and experimental results of plasmonic band gap laser [3]. The design of the corrugation profile of both sides of a metal film and the implications of film thickness for minimal loss and maximal amplification [4] will be discussed. [1] H. Watanabe, et. al, Phys. Rev. B. 69, 155418, 2004 [2] T. Ichimura, et. al. Phys. Rev. Lett. 92, 220801, 2004 [3] T. Okamoto et. al, Appl. Phys. Lett. 85, 3968, 2004. [4] F. H'Dhili, T. Okamoto, and S. Kawata, submitted.

10:30 AM *GG4.5
Optical Antennas for Scanning Near-field Optical Microscopy.
Kenneth Crozier and Eric Kort; DEAS, Harvard University, Cambridge, MA, Massachusetts.

Currently, a significant obstacle to the experimental realization of photonic devices such as photonic crystals and plasmonic nanostructures is the absence of a tool for the reliable imaging of the optical fields in these devices on subwavelength scales. Because of this, the photonic device becomes a 'black box', whose output is an inexplicable function of its input. Scanning near-field optical microscopy offers a means for the direct measurement of fields in photonic devices, as well as a method for overcoming the classical diffraction limit. The technique has never become widespread, however, mainly due to the disadvantages of traditional near-field probes based on tapered optical fibers. These include poor optical efficiency, a resolution limit of approximately twice the skin depth of the aluminum coating, fragile probes, and poor reproducibility. Silicon microfabrication offers a means for producing high efficiency and robust probes in a batch fashion. In addition, thse probes present the opportunity for high resolution imaging, through the incorporation of a metallic nanostructure, known as an 'optical antenna', at the end of the tip. We have previously demonstrated that microfabricated silicon nitride cantilevers with integrated microlenses ('solid immersion lenses') allow focusing of light into the tip to a spot size of ∼130nm [1]. We have previously demonstrated optical antennas operating at mid-infrared wavelengths [2]. Here, we will present optical antennas fabricated at the ends of atomic force microscope tips, designed for operation at near infrared wavelengths. References [1] K.B. Crozier, D.A. Fletcher, G.S. Kino and C.F. Quate, 'Micromachined silicon nitride solid immersion lens,' Journal of Microelectromechanical Systems 11, (5), pp. 470-478 (2002) [2] K.B. Crozier, A. Sundaramurthy, G.S. Kino, and C.F. Quate, 'Optical antennas: resonators for local field enhancement,' Journal of Applied Physics 94 (7), pp. 4632-4642 (2003)

11:00 AM GG4.6
High-resolution cathodoluminescence imaging of short-range near-resonance thin-film surface plasmon polaritons.
Timon van Wijngaarden and Albert Polman; Center for Nanophotonics, FOM-Institute AMOLF, Amsterdam, Netherlands.

Surface plasmon polaritons at frequencies close to the plasmon resonance possess wave vectors much larger than their far-field photonic counterpart. As a consequence, they may enable nanophotonic integration at length scales below the free-space wavelength. A major challenge in analyzing these near-resonance plasmons is to image their spatial distribution, propagation and damping, as this requires a measurement resolution well below the optical wavelength. Here, we present the use of cathodoluminescence imaging spectroscopy to image the propagation of surface plasmon polaritons in thin Ag and Au films at nanometer resolution. A 30 keV electron beam in a high-resolution scanning electron microscope is used to generate surface plasmons by normal incidence on the metal film at a well-defined distance from a grating that is integrated in the metal film. The grating is designed to couple the plasmon out into far-field radiation that is then collected by a parabolic mirror and imaged on a CCD array detector. By fabricating samples with varying grating pitch, clearly distinct emission spectra are observed. Taking into account the grating vector and plasmon dispersion the spectrum of the electron-beam generated surface plasmon polaritons is derived. By measuring the light intensity emitted from the grating while varying the distance between the excitation spot and the grating, the characteristic surface plasmon propagation distance can be determined as a function of wavelength. Pure exponential decays are observed, from which the characteristic propagation length is derived. For a 75 nm thick Au films we find, at a free-space wavelength of λ=550 nm (i.e. close to resonance), a propagation length of 750 nm, confirming the high spatial resolution of this technique. Further away from resonance, at λ=700 nm a longer propagation of 20 μm is found. This increased trend with wavelength is in agreement with calculations for a semi-infinite film using experimentally determined optical constants. Measurements on Ag films and on thin films ($<$ 30 nm) with coupled plasmon modes, will also be presented. Given the extremely high spatial resolution of cathodoluminescence imaging spectroscopy (beam spot diameter $<$ 5 nm), this technique may prove ideal for the characterization of a wide range of plasmonic nanostructures.

11:15 AM GG4.7
Plasmonic probe of Cu interconnect quality.
Gary Daniel Knight, c/o electronics, OCIP (Ottawa-Carleton Inst. of Physics), Ottawa, Ontario, Canada.

At submicron device scales photonic and electronic interconnects are challenged. In subwavelength dimensions dielectric cores are unable to confine radiant energy, while submicron electrical interconnects and devices have nonlinear size-effects and RC delays. We show that metallized lines and stripes retain promise in plasmonics, through surface plasmon polaritons (SPPs [5]) able to characterize lines of about 100 nm width or thickness in relation both to optical and to electronic function. As technology strives for metal surfaces specular to electrons and granular purity supportive of high current density, a hurdle is the roughness of boundaries. This misnomer encompasses anything that scatters electrons non-specularly: impurities and defects embedded at metal grain boundaries, and includes random orientations of microfacets with scatter locally specular but unpredictable from the surface plane. This roughness creates transmission losses for surface-plasmon polaritons (SPPs) on the metal surface, and we propose re-radiated infrared light as a probe of the surface quality for the metal line. Undesirable roughness is random, while engineered surface textures – periodic terraces, vicinal structures, and nano-embossings are correlated, making texture useful directly, in momentum transport, or in resistence and photonic loss characterization [1,2]. SPP waveguiding (or re-radiation) offers an attractive probe of in-situ metal surface quality that is directly related to current transport. DC resistivity is also a probe, but its scattering factors are not separable (even across operating temperatures). Through local size-effects on the negative permittivity in metal, we show SPP probing to be a well poised complement by injecting p-polarized IR light into a metal-core test structure patterned on-chip. We address the topic of nanoscale metal line as dual-purpose conduit of electronic {and} photonic pulse-trains, with added possibilities of modulating one of these momentum transports with the other, or through mutual interaction with gate fields. REFERENCES: 1 G.D. Knight in: Chapter 4 {Materials for Information Technologies}, {and references therein} (Springer, 2005) 2 G.D. Knight, T.J. Smy in: ECS special proceedings on ULSI, {ECS 207, Quebec} and references therein (Claeys Cor, Ed) (2005) 3 G.D. Knight, T.J. Smy: Microelect Eng {64} {417-28} (2002) 4 G.D. Knight, T.J. Smy: ECS Proceedings (M. Yang, Ed) {ISTC 2002} {295-304} (2002) 5 H. Raether: {Surface Plasmons on smooth and rough surfaces and on gratings} Tract Mod Phys {111}(Springer, 1988)

11:30 AM GG4.8
Microscopic near-field optics of metallic nanostructures.
Yongqiang Xue, College of Nanoscale Science and Engineering, University at Albany-SUNY, Albany, New York.

The further miniaturization of integrated optical devices requires investigating optical elements with dimensions on the nano-scale. Methods are therefore needed for detecting and guiding light on a scale much smaller than the wavelength of light. The study of near-field optical phenomena thus provides a unifying theme for the further miniaturization of photonic-band-gap materials into the nano/atom- regime and for the continuing advancement of optical information gathering and processing through scanning near field optical microscopy techniques. It is clear that to investigate light-matter interaction in a spatial extension much less than the optical wavelength, one can not in general have confidence in the macroscopic electrodynamics so far popular in near-field optics and photonic band-structures. Instead a microscopic approach is highly desirable that describes the electromagnetic field in the real space, includes microscopic model of the light-nanostructure interaction from the beginning and facilitates the quantization of the electromagnetic field in nanostructured media. In this work we present a microscopic theory of near-field optical effect in single and coupled metallic

nanostructures. Our theory is based on the rigorous Lagrangian and Hamiltonian formulation of local-field electrodynamics, where the nanostructure optical response is treated quantum-mechanically, while the electromagnetic field is treated either classically or quantum-electrodynamically within a unified space-time picture. We demonstrate the power and discuss the insights obtained from such microscopic analysis for understanding light confinement in sub-wavelength structures and near-field mediated electromagnetic energy transport phenomena through applications to metal nanoparticles. Both single and coupled metal nanoparticles are studied, where the electromagnetic wave propagation is mediated by resonant excitation of surface plasmon polariton modes.

11:45 AM GG4.9
Two Photon Photoemission Microscopy of Ag Nanoislands and Wires. Frank Joachim Meyer zu Heringdorf, Liviu Ionut Chelaru, Dagmar Thien and Michael Horn-von Hoegen; Fachbereich Physik, Universitat Duisburg-Essen, Essen, Germany.

Although the Mie-Plasmon in Ag islands and particles is well known, it is still a challenge to analyze the plasmonic behaviour of single Ag particles and nanowires on surfaces. Photoemission Microscopy offers a way to directly observe particle plasmons by means of two photon photoemission. Here, the plasmon as an intermediate state completely dominates the photoemission signal: if an intense 400nm laser pulse is focused on the surface, a first photon is used to excite a particle plasmon before a second photon is used to trigger plasmon-assisted photoemission. We will show, how the particle plasmon evolves during growth of Ag islands and wires on 4 degree vicinal Si(001) and demonstrate that depending on the size of the particle only distinct plasmon modes can be excited. The strong nonlinear nature of the two-photon photoemission process also allows to directly image the fringe field around the particle on the surface and to study the interaction of the fringe field with the plasmonic excitation.

SESSION GG5: Plasmonic Waveguides and Devices
Chairs: Mark Brongersma and Vladimir Shalaev
Tuesday Afternoon, November 29, 2005
Room 308 (Hynes)

1:30 PM *GG5.1
Channel plasmon polariton guiding by subwavelength metal grooves. Sergey I Bozhevolnyi, Department of Physics and Nanotechnology, Aalborg University, Aalborg East, Denmark.

Surface plasmon polaritons (SPPs) are tightly bound to the metal surface penetrating on $\sim$ 100 nm in dielectric and $\sim$ 10 nm in metal, a feature that implies the possibility of using SPPs for miniature photonic circuits and optical interconnects attracting a great deal of attention to SPPs [1]. The main issue in this context is to strongly confine the SPP field in the cross section perpendicular to the SPP propagation direction (smaller cross sections ensure smaller bend losses and higher densities of components), while keeping relatively low propagation losses. Several configurations have been proposed and investigated for this purpose. Thus, nanometer-sized metal rods and linear chains of metal nano-spheres were shown to support extremely confined SPP modes propagating though only over submicron distances. SPP propagation along metal stripes and channels in periodically corrugated regions has also exhibited propagation losses increasing drastically with the decrease in the stripe/channel width. It should be noted that, in general, the SPP confinement is achieved primarily by decreasing the SPP spatial extent into dielectric, thereby increasing the portion of SPP power being absorbed by metal, so that the choice of optimum guiding configuration is subject to trade-off with many intricate issues yet to be elucidated. Here, the results of our investigations of SPP guiding by channels cut into metal are presented, demonstrating the realization of channel plasmon polariton (CPP) propagation along a subwavelength metal groove. Using imaging with a near-field microscope and end-fire coupling with a tapered fiber, it is shown that a fundamental CPP mode can be both confined within subwavelength gold grooves and guided over tens of microns at telecom wavelengths [2]. A simple model based on the effective-index method that accounts for the main features of CPP guiding and provides a clear physical picture of this phenomenon is also reported. References: [1] W. L. Barnes, A. Dereux, and T. W. Ebbesen, Nature 424, 824 (2003). [2] S. I. Bozhevolnyi, V. S. Volkov, E. Devaux, and T. W. Ebbesen, Phys. Rev. Lett., in press.

2:00 PM GG5.2
Optical non-linearities in subwavelength particle plasmon structures. Luke A. Sweatlock, Jennifer Dionne, Henri Lezec, Sungjee Kim, Domenico Pacifici and Harry A. Atwater; Applied Physics, Caltech, Pasadena, California.

The advancement of integrated nanophotonics requires development of novel materials to facilitate the interaction of light and matter at the nanoscale. A promising avenue is the integration of semiconductor quantum dots such as CdSe, which have strong optical nonlinearity near bandgap, with arrays of small metal particles. The third order susceptibilities of quantum dot structures near bandgap have been reported in the range of 0.01 esu, corresponding to a nonlinear index on the order of 1E-5 cm$\hat{2}$/W. Therefore the possibility exists for unity change in optical index with modest input power density, given a light-harvesting mechanism to enhance the effective interaction cross section of the quantum dots. Plasmonic nanostructures are suited to perform this role as nano-concentrators. Finite difference simulations indicate that a 5000-fold local field enhancement is achievable in a system consisting of 10 nm diameter metal particles in glass with 1 nm interparticle seperation. The frequency of the collective plasmon resonance and the degree of field enhancement depend strongly on the geometrical parameters of the array. We fabricate via electron beam lithography linear arrays of 2 to 20 Au nanoparticles with various diameters from 50 to 200 nm, and various interparticle spacings of 20% to 100% of the diameter. Confocal spectroscopy is utilized to determine the power-dependent optical properties, both in a linear dielectric matrix and in the presence of semiconductor quantum dots. Simulations are used to connect macroscopic observations to the local nanoscale optical response. Applications to development of active nonlinear plasmonic device technologies are explored.

2:15 PM GG5.3
Tunnel Junction Surface Plasmon Source. Anu Chandran and Mark L. Brongersma; Geballe Laboratory for Advanced Materials, Stanford University, Stanford, California.

Surface plasmons in metallic nanostructures allow propagation and confinement of light on sub wavelength length scales. Currently, most experimental device studies employ an optical route for exciting surface plasmons. However, from a device stand point it is attractive to have an electrically excitable surface plasmon source. Broad band light emission from metal insulator metal (MIM) tunnel junctions under bias has been studied extensively since the first observation in 1976 [1]. This light emission was attributed to the excitation of surface plasmons by tunneling electrons which get scattered out as free space radiation by the roughness in the junctions. In spite of the inherent processing advantages and wide range tunability, these devices are not attractive as light sources due to their low power efficiency. The best reported efficiencies are in the range of one photon per 104 tunneling electrons[2]. Theory of surface plasmon excitation by tunneling electrons suggest that the efficiency of excitation of modes with fields concentrated at the tunneling barrier may be high, but the low efficiency of scattering these modes into free space radiation lowers the overall light emission efficiency[3]. This makes these junctions attractive as an electrically pumped surface plasmon source. We have fabricated MIM junctions with Al and Au electrodes deposited by electron beam evaporation, with film thicknesses between 20nm to 60nm. The AlOx barrier (2 to 5nm thickness) was formed by thermal as well as plasma oxidation. Room temperature light emission was observed from these junctions. Here we study Al-AlOx-Au junctions to create a narrow line width, efficient surface plasmon source. Design and fabrication of tunnel junction structures optimized for surface plasmon generation will be outlined. Experimental and numerical studies aimed at reducing the emission line width and increasing the efficiency of surface plasmon excitation will be presented. References [1] Lambe J. and McCarthy S.L.; Phys. Rev. Lett. 32, 545 (1976) [2] Shu Q.Q., Wen W. J. and Xu S.J.; J. Appl. Phys., 65, 373 (1989) [3]Davis L.C; Phys. Rev. B 16, 2482 (1977)

3:30 PM *GG5.4
Integrated optics based on long-ranging surface plasmon-polaritons. Pierre Berini, [1]School of Information Technology and Engineering, University of Ottawa, Ottawa, Ontario, Canada; [2]Spectalis Corporation, Ottawa, Ontario, Canada.

Recent theoretical and experimental progress on the development of a new integrated optics technology based on the propagation of long-ranging surface plasmon-polariton waves will be reviewed. The foundation waveguide is a thin metal film of finite width embedded in an optically infinite homogeneous background dielectric, and the mode utilized is the ssb0 mode, which is the fundamental long-ranging mode in this structure [1-3]. The metal film in this waveguide can be dimensioned such that: (i) the ssb0 mode propagates with low loss, (ii) the ssb0 mode couples efficiently to the TM mode propagating in a single mode dielectric waveguide butt-coupled to the input, (iii) all long-ranging higher-order modes are cut off, and (iv) any remaining high-loss modes are unexcited, or at best, excited with very low efficiency by the dielectric waveguide input means. These characteristics are all desirable in a waveguide for integrated optics. Progress on defining the foundation passive elements required for integrated optics will be reviewed. In particular, structures such as straight waveguides, bends, Y-junctions, couplers, Mach-Zehnder interferometers and Bragg gratings will be discussed. Robust and accurate theoretical models for all of these elements will be presented

along with the experimental validation of the models. Active devices can be designed to exploit the benefits of: (i) having a metal in the optical path, and/or (ii) operating in a surface wave. These particularities lead to novel thermo-optic, electro-optic or charge carrier based devices in polymeric, electro-optic and semiconductor claddings, respectively. Active devices based on a metal (or metal silicide) buried in polymer, lithium niobate or silicon claddings will be presented along with theoretical and experimental results achieved to date. Fabrication requirements, challenges and solutions will be briefly discussed for the principal metal/dielectric combinations of interest. [1] Berini, P., "Plasmon-Polariton Modes Guided by a Metal Film of Finite Width", Optics Letters, Vol. 24, No. 15, pp. 1011-1013, August 1999 [2] Berini, P., "Plasmon-Polariton Waves Guided by Thin Lossy Metal Films of Finite Width: Bound Modes of Symmetric Structures", Physical Review B, Vol. 61, No. 15, pp. 10484-10503, April 2000 [3] Charbonneau, R., Berini, P., Berolo, E., Lisicka-Skrzek, E., "Experimental Observation of Plasmon-Polariton Waves Supported by a Thin Metal Film of Finite Width", Optics Letters, Vol. 25, No. 11, pp.844-846, June 2000

4:00 PM GG5.5
Compact Bragg Gratings for Integrated Optics Utilizing Long-Range Surface Plasmon Polaritons. Alexandra Boltasseva[1] and Sergey I. Bozhevolnyi[2]; [1]Research Center COM, Technical University of Denmark, Kgs. Lyngby, Denmark; [2]Department of Physics and Nanotechnology, Aalborg University, Aalborg, Denmark.

In recent years, surface-bound electromagnetic excitations propagating along metal-dielectric interface, so-called surface plasmon polaritons (SPPs), have attracted much interest in the context of highly integrated photonic devices and nano-optics. Field enhancement at the interface and exponential decay into both of the neighboring media make SPPs very sensitive to surface features allowing one to efficiently control the SPP propagation in the surface plane by using different types of surface metal nanostructures. However, SPPs suffer from rather strong damping due to absorption in metals so that their propagation length is limited to micrometer distances. The way of increasing the propagation length is to consider the symmetric configuration of two very close metal/dielectric interfaces. Such a structure of a thin metal film embedded in dielectric supports propagation of the field-symmetric modal solution, so-called a long-range SPP (LR-SPP), formed by the coupled SPPs at two metal-dielectric interfaces. Such coupling results in a very small field concentration within the metal film and, hence, low propagation loss for the LR-SPP. Due to low propagation and coupling losses, LR-SPP stripe waveguides constitute a new alternative for integrated optical (IO) circuits with one unique feature - the possibility of using the same metal-stripe circuitry for both guiding optical radiation and transmitting electrical signals that control its guidance. Different LR-SPP-based passive and active IO components including straight and bent waveguides, Y-splitters, directional couplers, various modulators and switches have been recently experimentally demonstrated. Here, we demonstrate Bragg gratings based on LR-SPP-supporting configurations, having in mind great interest in Bragg gratings for broad range of applications in distributed-feedback (DFB) and distributed Bragg reflection (DBR) lasers, Bragg filters, (de)multiplexers, etc. By introducing periodic thickness-modulation of thin metal stripes embedded in a dielectric, we realize compact and efficient Bragg gratings for LR-SPPs operating around 1550 nm. We measure reflection and transmission spectra of the gratings having different lengths (from 20 to 160 μm), heights (tens of nm) and widths of the metal ridges forming the grating, and demonstrate the reflectivity of up to 60% and bandwidths ranging from 5 to 40 nm. By using a simple lossless-uniform-grating description, we estimate the effective refractive index modulation in LR-SPP gratings to be of the order of 10^{-2}. We show that the use of such LR-SPP Bragg gratings gives an advantage of choosing many grating parameters (for example, grating size and bandwidth) in a very broad range combined with the robust and reproducible fabrication procedure. The LR-SPP loss incurred in the gratings is also discussed as well as the possibilities for their future applications.

4:15 PM GG5.6
Photonic Band Gap Structures for Surface Plasmon Waveguiding: Influence of the Filling Factor.
Anne-Laure Baudrion, Jean-Claude Weeber and Alain Dereux; Physics Laboratory of the University of Burgundy, Dijon, France.

In analogy to the band gaps occurring in the electronic energy spectrum due to the periodic arrangement of atoms, a periodic modulation of the dielectric constant can inhibit the propagation of electromagnetic waves in a certain range of wavelengths creating a so-called photonic band gap (PBG). In recent years, this effect has been extended to surface plasmons (SP), which are quasi-two-dimensional electromagnetic waves that propagate along a dielectric-metal interface with amplitudes that decay exponentially in the perpendicular direction of the interface. Within this context, a periodically structured metal surface can create a surface plasmon photonic band-gap (SPPBG) effect in such a way that a channel of defects opened in the structure acts as a SP waveguide providing a good confinement of the SP propagation. SP waveguiding is of fundamental interest in the development of integrated optical devices. Thus, a deep understanding of the wave propagation properties in SPPBG structures, also called plasmonic crystals, as well as of the behavior of channels of defects opened in them is required in order to obtain an optimum design of devices based upon this kind of structures. This work reports on the optical properties of plasmonic crystals as a function of the filling factor, obtained by means of near-field optical characterization.

4:30 PM GG5.7
A Diffraction Limit for Surface Plasmon Waveguides: Guided Polariton Propagation and Diffraction. Rashid Zia and Mark L. Brongersma; Geballe Laboratory for Advanced Materials, Stanford University, Stanford, California.

To date, theoretical and experimental works on surface plasmon waveguides have highlighted their differences from conventional, diffraction-limited dielectric waveguides. It has been suggested in the literature that guided plasmon-polaritons are not diffraction-limited, and furthermore, that the modes of surface plasmon waveguides are inconsistent with a ray-optics interpretation of guided wave phenomena. However, we have recently derived solutions for the leaky plasmon modes [1] which suggest an interpretation that is consistent with conventional waveguide theory, [2] and we have expanded upon this analogy to propose a dielectric waveguide model for guided polariton optics. [3] This model, based upon a ray-optics representation of surface polaritons, accurately predicts both the real and imaginary propagation constants for the modes supported by single and coupled interfaces of finite width. Here, we discuss an effective diffraction limit for surface polaritons formulated with the aid of this model. Specifically, we present combined numerical, analytical, and experimental studies supporting the dielectric waveguide model for guided polariton propagation and diffraction. We have developed a numerical method to solve for the leaky and bounds modes of arbitrary geometry polariton waveguides. With this method, we demonstrate that published experimental results for the field profiles and propagation lengths of metal stripe waveguides are anticipated by their leaky modal solutions. Although surface polaritons supported by stripes guide electromagnetic energy in three dimensions, we establish that such modes can be modeled by the solutions of two-dimensional dielectric slab waveguides. Leveraging this physical model, we have designed and fabricated a variety of plasmonic devices, and we present experimental characterization by photon scanning tunneling microscopy (PSTM) to support the model's validity. [4] References [1] Leaky modes are those excited in the Kretschman configuration, and hence those most relevant for many experimental studies. [2] R. Zia, M.D. Selker, and M.L. Brongersma, Phys. Rev. B 71 (2005), 165431. [3] R. Zia, A. Chandran, and M.L. Brongersma, Opt. Lett. 30 (2005), 1473-1475. [4] R. Zia and M.L. Brongersma, "Near-Field Observation of Guided Polariton Propagation and Diffraction" (in preparation).

4:45 PM GG5.8
Integratable Metallic Slot Waveguide on SOI Platform for Strong Confinement. Long Chen and Michal Lipson; Cornell University, Ithaca, New York.

There is a growing interest in nanophotonics based on noble metals like silver and gold. The key feature of metals at optical and infrared frequencies is the large negative permittivity which supports plasmas around the surface. Strong confinement and considerably enhanced field intensity could be achieved due to localized plasma resonance or propagating surface waves, for applications in nanoscale devices, single molecule detection and nonlinear switching etc. Metallic components as surface plasma waveguides have been demonstrated both theoretically and experimentally. However, there is a fundamental trade-off between higher confinement and loss: metal strips buried in homogeneous dielectric have been shown to either support loosely confined long range surface plasma of micron-scale wide propagating up to several millimeters, or, using metallic nano-wires, to present tight confinement with very high losses, with propagation length of only several microns. Therefore, in order to become practical such high confinement plasmonic waveguides would need to be integrated with low-loss dielectric photonics Such coupling has been shown to be difficult: The end-fire coupling efficiency between dielectric waveguides and metallic waveguides is poor due to the significant mode mismatch (while the dielectric waveguides have the field concentrated within the high index core region, the field of a metallic waveguide is mainly around the surface of the metal core). We investigate a silicon wire waveguide embedded in a metal region for seemless integration between surface plasma and dielectric waveguide. The waveguide is embedded in the metal region laterally and in silicon dioxide vertically. The confinement is achieved through both the dielectric index contrast and the strong discontinuity of the

normal E components at the metal-silicon interfaces. This configuration forms a metallic slot waveguide with light tightly concentrated within the silicon slot, in analogy to the dielectric slot waveguides demonstrated recently. We analyzed the dependence of the propagation, attenuation and strong confinement characteristics of such a waveguide on the silicon slot width at 1.55um wavelength while the thickness is fixed at 250nm. For the mode well confined within a slot of 150nm width, the loss is estimated to be around 1dB/um, less than the loss of metal nanowires for the same level of confinement. We also demonstrate a compact taper of 3 um length for coupling to a 100nm metallic slot waveguide from the conventional SOI waveguide of 450nm wide with reflection loss than 5%. Therefore the metallic slot waveguide could be designed as only a small functional segment with the SOI waveguides as the transmission path on chip.

SESSION GG6: Poster Session II
Chair: Stefan Maier
Tuesday Evening, November 29, 2005
8:00 PM
Exhibition Hall D (Hynes)

GG6.1
Optical Properties of Au-ZnO Plasmonic Nanostructures.
Maria Michela Giangregorio, Maria Losurdo, Marianna Luchena, Marianna Ambrico, Pio Capezzuto and Giovanni Bruno; Chemistry, Univ. Bari, IMIP-CNR, Bari, Italy.

Gold (Au) thin films and nanoparticles are extremely interesting because they exhibit plasmon resonance (SPR) in the visible range. Au nanoparticles near the plasmon resonance enable the excitation of surface and resonant modes in nanostructures that can be used for optical and sensing plasmonic devices. These devices require a careful control of gold nanoparticle size, shape, and distribution to result in systems with well tailored functional properties. Furthermore, zinc oxide ZnO, as one of the most important direct band gap (3.37eV) semiconductors with a high exciton binding energy of 60meV, good lasing properties, high resistance to radiation, has recently attracted interest for synthesizing and fabricating various nanostructured devices. ZnO nanostructures, including nanoparticles and nanorods, are also interesting for many optoelectronic applications, since they expected to further lower the lasing threshold because of quantum effects resulting in enhancement of radiative recombination due to carrier confinement, and for sensors by increasing the surface active area. Therein, we focus on coupling the Au SPR with ZnO nanostructures. Specifically, in this contribution, we present data on the growth and characterization of (i) Au nanoparticles and of (ii) Au/ZnO nanostructures on different substrates, including Si(100), SiC (0001) and sapphire (0001), by r.f. (13.56 MHz) plasma enhanced chemical vapor deposition (PECVD). R.f. sputtering of an Au target by Ar plasmas is used for deposition of Au nanoparticles; successively, ZnO nanoparticles and nanorods are grown using the Zn(TPA)2Tmed precursor carried by an Ar flux with or without enhancement by O2 plasmas. We present and discuss data on the dependence of the size and distribution of Au nanoparticles on (i) the surface tension of the substrate; (ii) the substrate surface treatments using various wet-chemicals and dry plasma treatments (N2, Ar, H2) that impact on Au wetting and, hence, Au nanoparticles size; (iii) the effect of the experimental parameters, such as substrate temperature and plasma power. The optical properties of Au nanoparticles at the plasmon resonance and amplitude are investigated by spectroscopic ellipsometry, which also highlights interfacial reactivity between the Au nanoparticles and substrates that can result in an enhancement of the plasmon resonance. Subsequently, ZnO is grown on the Au/substrates taking advantage of the Au catalysed mechanism, which driven the Au nanoparticles ontop the ZnO nanostructure. Data on the dependence of the ZnO nanoparticles and nanorods size on deposition temperature and plasma parameters will also be presented. The emphasis is on the optical monitoring of the Au plasmon resonance frequency and amplitude depending on all the above factors and coupling with ZnO using spectroscopic ellipsometry (SE) in the photon energy range 0.75-6.5 eV. Ellipsometry data are also corroborated by XPS, XRD, AFM and I-V characteristics.

GG6.2
Nonlinear plasmonic microarray of gold nanoparticles above a gold surface with nanogap.
Kazuma Tsuboi[1], Shinya Abe[2] and Kotaro Kajikawa[2,1]; [1]PRESTO, Japan Science and Technology Agency (JST), Saitama, Japan; [2]Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology, Yokohama, Japan.

We have reported that gold nanopartciles located above a gold surface with a gap distance of a few nanometers show considerable second-order nonlinear optical activity. This comes from enhancement of the electric field owing to localized surface plasmon resonance in the system of gold nanoparicles and the gold surface. In order to clarify the mechanism of this enhancement, we have performed spectroscopic measurements of second-harmonic generation (SHG) and have found that the SHG intensity form this system is in good agreement with the reflection absorption spectra in oblique incidence, in which the absorption band due to the plasmon resonance can be observed. Since the absorption band is sensitive to the dielectric constant or adsorption of molecules on the surface of the nanoparticles, the plasmonic microarray of gold nanopartciles will be useful for sensing technology. In this paper we will report the fabrication methods to make such microarray using self-assembled monolayers by means of UV-photolithography. The microarrays were probed by linear and nonlinear (SHG) optical microscope with a cooled CCD camera. Clear SHG microscope images can be observed from the gold nanoparticle arrays and the intensity of the SHG was sensitive to the surrounding dielectrics as well as the linear optical measurements previously reported.

GG6.3
Localized Surface Plasmons on Torus Geometry Theoretical Analysis.
Alexandre Mary[1], Alain Dereux[1] and Thomas L. Ferrell[2]; [1]LPUB - Nanosciences, University of Burgundy, Dijon, France; [2]University of Tennessee, Knoxville, Tennessee.

The importence of surface plasmon effects on the optical properties of small metal particles is known since pionneering work of Ritchie[1] and Otto[2]. However, it has received a renewed attention in the last years due to their promising technological applications, ranging from non-linear phenomena to photonic devices [3]. As the properties of surface plasmon depend very strongly on the material and shape of the particles, many geometrical forms have been studied. In the literature, the plasmon dispersion relations associated to geometric coordinates systems allowing variable separation in Laplace's equation (11 systems possible) i.e. for particles size for which the non-retarded approximation is acceptable, can been found. However, the toroidal geometry [4] has not been extensively studied because of the relative complexity of its solutions [5] as well as the difficulty up to now of practically fabricating the torus. Nevertheless, this last impediment has been recently overcome since fabrication of nanorings is possible by microfabrication methods [6], in such a way that knowing the properties of torus surface plasmons could have a practical interest. In this study, we propose a procedure that allows to establish the surface plasmon dispersion relations in the toroidal coordinates system. The plasmon mode frequencies, associated potential and electric field can be numerically calculated. We have demonstrated that localized surface plasmon associated to a gold nanotorus have peculiar properties. From the electric field distributions of modes, we show the existence of even surface plasmon eigenmode, which carry an electric dipole moment and a vanishing magnetic dipole moment, and odd surface plasmon eigenmode, where this feature is reversed [7]. According to the dispersion relation, these even and odd modes are expected to be degenerated. In general, coupling among particles and betweeen particles and the environment leads to energy shifts and splitting the degenerate modes, where short and long-range couplings drive the properties of the plasmon resonances. On this basis, perspectives about designing metal nanostructures featuring an effective magnetic dipole moment at optical frequencies can be discussed. [1] R. H. Ritchie, Phys. Rev. 106, 874 (1957). [2] A. Otto, Zeit. Fur Physik, 216, 398 (1968). [3] W. Barnes, A. Dereux and T. W. Ebbesen, Nature, 424, 824 (2003). [4] J. Segura and A. Gil, Computer Phys Com., 124 (200) 104-122. [5] P. M. Morse and H. Feshbach, Methods of Theorical Physics, MacGraw-Hill, NY, 1953. [6] J. Aizpura, P. Hanarp, D.S. Sutherland, M. Kall, Garnett W. Bryant and F.J. Garcia de Abajo, Phys. Rev. Lett. 90, 057401 (2003). [7] A. Mary, A. Dereux and T. L. Ferrel, Phys. Rev. B, to be published.

GG6.4
Abstract Withdrawn

GG6.5
Au and Ag Nanoparticle Chains Prepared via Wet Chemistry.
James Sioss and Christine Keating; Penn State University, University Park, Pennsylvania.

We have developed a simple process to fabricate linear chains of metal nanoparticles by incorporating sacrificial layers of metal into striped nanowires. Striped nanowires are electrodeposited in the pores of aluminum oxide and the composition, sizes, and spacing can be controlled. Coating the nanowires with silica and etching the sacrificial metals leaves nanoparticle chains. The size of the porous aluminum oxide determines the diameter, and the electrodeposition time controls the size and spacing of the nanoparticle chains. We have used Ni as a sacrificial layer to produce Au, Au/Ag, and Ag nanoparticles chains with diameters down to 30 nm and spacing greater than, equal to, and smaller than the nanoparticle sizes. These nanoparticle chains were characterized with transmission electron microscopy, energy dispersive x-ray spectroscopy and optical spectra.

This method may be useful for producing plasmonic devices.

GG6.6
Abstract Withdrawn

GG6.7
Controllable Plasmon Resonanses Of Gold Nanoparticles Higher-Order Aggregates. Evgenia Buzaneva, [1]Radiophysical Faculty, National Taras Shevshenko University of Kyiv, Kiev, Ukraine; [2]Radiophysics and Chemical Faculties, Kyiv National Taras Shevchenko University, Kiev, Ukraine.

The methods of strong-controllable changes of the plasmon band of gold nanoparticles in colloidal biosolutions were developed. The methods are based on the fact that the reason for the plasmon band shift is that this band is very sensitive to the interparticle distance, as well as to the aggregate size. In contrast to known method (S.A.Mirkin, 1999), using these DNA molecule as bridge between nanoparticles we selected ssDNA tails for separated negative charged citrate-capped Au nanoparticles (8.6, 10.6 and 16.5 nm in diameters) and biosolutions, for example with ascorbic acid. The UV-VIS spectroscopy of dispersed nanoshells fabricated with 80 nm diameter core and a 8.6, 10.6, 16.5 nm gold and nanoshells aggregates confirmed that the extinction of aggregates in the presence of biomolecules is at 740 nm in contrast to the extinction of separated gold nanoparticles. The aggregation of nanoshells by SEM, AFM images of their adsorbed layer on a substrates has been confirmed. All this has extended our knowledge of metalnanoparticles and may lead to the design and construction of new artificial nanophotonic systems. The methods of strong-controllable changes of the plasmon band of gold nanoparticles in colloidal biosolutions were developed. The methods are based on the fact that the reason for the plasmon band shift is that this band is very sensitive to the interparticle distance, as well as to the aggregate size. In contrast to known method, using these DNA molecule as bridge between nanoparticles we selected ssDNA tails for separated negative charged citrate-capped Au nanoparticles (8.6, 10.6 and 16.5 nm in diameters) and biosolutions, for example with ascorbic acid. Then UV-VIS spectroscopy of dispersed nanoshells fabricated with 80 nm diameter core and a 8.6, 10.6, 16.5 nm gold and nanoshells aggregates confirmed that the extinction of aggregates in the presence of biomolecules is at 740 nm in contrast to the extinction of separated gold nanoparticles. All this has extended our knowledge of metalnanoparticles and may lead to the design and construction of new artificial nanophotonic systems.

GG6.8
A New Type of Inverse Opal: Drude-Like Gold Spheres in Titania Matrix. Poorna Praveen Rajendran and Chekesha Liddell; Materials Science and Engineering, Cornell University, Ithaca, New York.

This work examines a new type of inverted opal structure based on metallodiectric photonic crystals near the plasma frequency of the metal. A macroporous titania (anatase) inverse opal was backfilled with Au using electroless deposition methods. Au exhibits Drude-like behavior, with its frequency dependent dielectric function approaching zero near the plasma frequency. This allows the potential to obtain dramatic improvements in refractive index contrast, i.e., at 650nm, $n_c = 12.6 = n_{titania}(2.4)/n_{Au}(0.19)$. The configuration of Au at FCC lattice sites with titania interstitial filling leads to an "effectively inverted" structure where the low index region is provided by low-dispersive metal spheres rather than by air voids. Similar theoretical work on noble metal spheres in an air matrix predicts up to four complete band gaps with a maximum relative gap-width of 10%. At 650nm, an index contrast of $5.2 = n_{air}(1)/n_{Au}(0.19)$ would be expected for such a structure. The same configuration for dielectric spheres in air does not exhibit a bandgap regardless of high refractive index contrast. The present work investigates the existence of a bandgap near the Au plasma frequency using reflectance/transmission spectra measurements. By altering the size of the sphere templates, the midgap frequency of the metallodiectric photonic crystal can be tuned to overlap with the non-absorptive frequency window near the plasma frequency in otherwise highly absorptive Au materials. Scanning electron microscopy images confirmed an "effectively inverted" structure. Studies using a General Area Detector Diffraction System (GADDS) confirmed the crystalline anatase phase of titania and the presence of gold nanocrystals.

GG6.9
Localized Surface Plasmon Resonance Au Nanobar Array Sensors Fabricated Via Nanoimprint Lithography. Brandon D. Lucas[1], Jin Sung Kim[2] and L. Jay Guo[2,1]; [1]Applied Physics, University of Michigan, Ann Arbor, Michigan; [2]Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, Michigan.

The desire to create metallic nanostructures has grown tremendously due to their ability to act as miniature chemical and biological sensors based on their localized surface plasmon resonance (LSPR). LSPR is highly sensitive to the size, shape, interparticle distance and local dielectric environment of the nanoparticles. In order to exploit LSPR for sensor applications there must be an efficient, reliable and cost-effective means to create and immobilize these nanoparticles on optical substrates for measurement using well-established spectroscopic systems. Two commonly used fabrication methods are chemical synthesis and nanosphere lithography (NSL). Chemical synthesis requires the use of reaction parameters such as time, relative concentrations of reactants and temperature to control the size and shape of the resulting nanostructures. This method is severely limited by its difficulty to produce homogenous populations of nanoparticles. Further, this approach requires the generation of novel surface chemistries for nanostructure immobilization on an optical substrate and minimization of nanoparticle aggregation. NSL employs the use of a single or double layer of polystyrene nanospheres to create a mask for subsequent nanoparticle formation through metal deposition. This technique has proven to be an effective means for producing periodic arrays of nanostructures on optical substrates, however it has demonstrated a limitation in surface coverage. We have recently utilized nanoimprint lithography (NIL) to fabricate an array of Au nanobars on a rectangular lattice with a period of 700nm and 200nm in the respective long and short dimensions on glass substrate. Using NIL as the fabrication technique offers the advantages of creating large ($>cm^2$) low-defect sample areas with an inherent control of the relative placement and orientation of the nanobars. Previous studies have demonstrated the presence of longitudinal and transverse plasmon modes related to the long and short dimensions of these structures. The high-degree of control of nanobar orientation capable with NIL will allow the full exploitation of these modes for sensing purposes. Based on our initial theoretical calculations using the discrete dipole approximation (DDA), the extinction efficiency maxima corresponding to the two plasmon modes exhibit strong polarization dependence. Characterization and theoretical modeling of these structures with different aspect ratios, including their ability to detect changes in the dielectric environment, is ongoing and results will be presented. Optimization of the structures produced by NIL is imperative for the development of these nanostructures as biological and environmental sensors.

GG6.10
Active manipulation of surface plasmons in metal-molecule-metal devices. Ragip Pala and Mark Brongersma; Stanford University, Stanford, California.

Molecular electro-optical switches are one of the major milestones in the development of optoelectronic memory elements as well as a long-standing goal of highly integrated photonic circuits. We are investigating the use of photochromic compounds to realize such structures. We have fabricated gold and aluminum metal-molecule-metal junctions with photochromic molecules such as spiropyrans and azobenzenes as the active switching element. The switching of these photochromic molecules results in a change in the real and imaginary part of the refractive index that was detected in a home-built surface plasmon resonance sensing system. The experimental results and modeling of these switching events will be presented. After characterizing the switching behavior, we have also investigated the possibility to manipulate the propagation of surface plasmon waves with external stimuli. In these experiments, surface plasmons polaritons were excited locally using a nanoscale grating and directed towards an area with photochromic molecules. Depending on the state of the molecules, the surface plasmon waves were allowed to propagate or strongly attenuated. These structures act as prototypes for the future plasmonic switches.

GG6.11
Omni-directional emission via surface plasmons in a metal-insulator-metal structure. John Liu and Mark Brongersma; Stanford University, Stanford, California.

Light extraction from light emitting devices is an important design problem. Planar metallic microcavities have been used in the past to allow facile electrical excitation and obtain resonantly enhanced emission. In general this emission is only enhanced in a narrow angle range, while for some applications a wide emission angle is desirable. We will demonstrate that by using a properly designed, planar, metallic microcavity it is possible to enhance the free space emission via low momentum surface plasmons that lie above the light line. Additionally, for an optimized cavity thickness this enhanced emission can be observed at a well-defined frequency for all angles due to a nearly flat surface plasmon dispersion relation, hence the term omni-directional emission. This effect is predicted through simulations of dipole emission.

SESSION GG7: Photonic Crystals and Cavity Effects

Chair: Shanhui Fan
Wednesday Morning, November 30, 2005
Room 308 (Hynes)

8:30 AM *GG7.1
Nanofabricated metallic structures for optical observation of biomolecular processes. Harold Craighead, Applied Physics, Cornell University, Ithaca, New York.

We have used several methods to create nanometer-scale metallic devices for localization of optical excitation. High resolution lithography and non-lithographically fabricated templates have been used to create metallic structures used to confine or concentrate optical excitation and observation to dimensions on the order of ten nanometers. These devices have been used to observe single molecule binding events or observe the motion of individual fluorescent molecules in solution or to observe the motion of molecules confined in lipid membranes.

9:00 AM GG7.2
Effects of SiO2 Cover Layers on Surface-Enhanced Raman-Scattering Molecular Sensors Utilizing Silver Oxide Thin Films. Makoto Fujimaki[1], Yasuhiko Iwanabe[2,1], Koichi Awazu[1] and Junji Tominaga[1]; [1]Center for Applied Near-Field Optics Research, National Institute of Advanced Industrial Science and Technology, Tsukuba, Ibaraki, Japan; [2]Graduate School of Engineering, Tokyo Denki University, Chiyoda, Tokyo, Japan.

High sensitivity molecular detection systems have been expected in many fields. Our group has developed a molecular sensor that utilizes silver nanoparticles formed from silver oxide thin films, where surface enhanced Raman scattering (SERS) by the silver nanoparticles is used for detection of small amount of molecules. The formation of silver nanoparticles is easily triggered by visible laser irradiation. The laser is also used as a light source to induce the SERS. Therefore, the system is quite simple and is expected to be an effective molecular sensing tool. In the present research, we have succeeded in the increment of the sensitivity of the sensor by applying a SiO2 thin film layer on a silver oxide thin film. The SiO2 thin film shifts a localized-plasmon-resonance wavelength, resulting in higher efficiency of SERS. Furthermore, the SiO2 thin film prevents molecules to be detected from being oxidized by the silver oxide thin film. Accordingly, stable and high sensitivity molecular detection has been realized.

9:15 AM GG7.3
Stimulated Emission of Surface Plasmons at the Interface Between a Silver Film and an Optically Pumped Dye Solution. Stefan Grafstrom, Jan Seidel and Lukas M. Eng; Institute of Applied Photophysics, Dresden University of Technology, D-01062 Dresden, Germany.

Surface plasmons (SPs) are surface-bound electromagnetic waves supported by metals, offering the possibility of strong spatial confinement of electromagnetic fields on the micrometer and nanometer scale. They suffer, however, from strong damping caused by absorption and radiation losses. Here, we demonstrate amplification of SPs by stimulated emission [1] which marks a possible solution to this problem. Amplification of SPs can be considered analogous to photon amplification in a laser, thereby suggesting novel approaches in the field of nanooptics [2]. The surface plasmon to be amplified is excited by a probe laser beam in an attenuated-total-reflection setup (Kretschmann-Raether configuration) at the interface between a flat continuous silver film and a liquid containing organic dye molecules. Optical pumping by a second laser creates a population inversion in the dye molecules, which then act to deliver energy to the plasmon field by stimulated emission, thereby reducing the damping of the plasmon. To detect this effect, we measure the reflectance of the probe laser beam as a function of the incidence angle. The width and depth of the reflectance dip associated with the excitation of SPs depend on the degree of damping that the SPs experience. A reduction of the damping caused by the presence of an amplifying medium at the film surface (in our case an ethanolic solution of either rhodamine 101 or cresyl violet) will therefore result in a characteristic modification of the reflectance curve providing a signature of stimulated SP emission. We find an excellent agreement of the experimental observations with a theoretical analysis for both dyes in use and for various values of the metal film thickness. [1] J. Seidel, S. Grafstrom, and L.M. Eng, *Phys.Rev.Lett.* **94**, 177401 (2005). [2] D.J. Bergman and M.I. Stockman, *Phys.Rev.Lett.* **90**, 027402 (2003).

9:30 AM GG7.4
Enhancement of 1.54 μm emission from Er and Yb co-doped Al_2O_3 films by Au thin film. Toshihiro Nakamura, Minoru Fujii and Shinji Hayashi; Electrical and Electronics Engineearing, Kobe University, Kobe, Hyogo, Japan.

It is well known that emission properties of an emitter can be modified by placing it near a metal surface. This modification is caused by an interference of the emitter's field with the field reflected by the metal surface. Depending on the phase of the reflected field, the spontaneous emission rate of the emitter is either enhanced or decreased. The phenomena can also be described by the change of local photonic mode density (PMD), which results in the modification of the rate. Recently, it is demonstrated that the modification of the PMD can affect the rate of F ö rster energy transfer. The control of the energy transfer by changing PMD is of great importance for a lot of fields where sensitized excitation of optically forbidden transition of an accepter by the energy transfer from a donor is required. In this work, we attempt to enhance the energy transfer from Yb ions to Er ions by altering PMD. We prepared Al_2O_3 films co-doped with Er and Yb covered by a Au thin film, and measured photoluminescence excitation (PLE) spectra of 1.54 μm emission of Er ions and time transients of the 980 nm PL of Yb ions. By placing the Au layer on top of the sample, the 1.54 μm PL is strongly enhanced in the excitation wavelength range between 850 and 980 nm, where the Er ions are excited by energy transfer from Yb ions, while no significant change is observed on the spectra when the Er ions are excited directly. Furthermore, from the time transient PL of Yb ions, enhancement of energy transfer rate in the presence of the Au layer is concluded. These results suggest that enhancement of 1.54 μm PL of Er is due to the enhancement of energy transfer rate by the modification of the PMD. Another possible mechanism of the enhanced luminescence is the local field enhancement of incident light due to the excitation of surface plasmons in the Au layer, i.e., the small roughness allows the direct coupling of the surface plamons with the incident light.

9:45 AM GG7.5
Surface plasmon enhanced high-speed spontaneous emission of InGaN/GaN. Koichi Okamoto[1], Isamu Niki[1,2], Yukio Narukawa[2], Takashi Mukai[2], Yoichi Kawakami[3] and Axel Scherer[1]; [1]Physics, California Institute of Technology, Pasadena, California; [2]Nitride Semiconductor Research Laboratory, Nichia Corporation, Anan, Tokushima, Japan; [3]Electronic Science and Engineering, Kyoto University, Kyoto, Kyoto, Japan.

Currently, InGaN/GaN quantum well (QW) based light-emitting diodes (LEDs) have been developed and expected to eventually replace more traditional fluorescent tubes as illumination sources. Recently, we have reported a method for enhancing the light emission efficiency from InGaN QWs by controlling the energy transfer between QW emitters and surface plasmons (SPs) [1]. This technique has potential to enhance spontaneous emission rate dramatically. Here, we investigate the direct observation of SP coupled spontaneous emission rate by using the time-resolved photoluminescence (PL) measurements. InGaN/GaN QW wafers were grown by MOCVD. The structures consist of a GaN (4000 nm), an InGaN single QW (3 nm) followed by a GaN cap layer (10 nm). A 50 nm thick silver layer was then evaporated on top of the wafer surface. To perform time-resolved PL measurements, the frequency doubled output from a mode-locked Ti: Al2O3 laser (1.5 ps, 400 nm, 80 MHz) and a streak camera were used for excitation and detection. The PL intensity of an Ag-coated sample was found to be about 12 times stronger than that of an uncoated sample. The shape of PL peaks from Ag-coated samples were not symmetric, but broaden at shorter wavelengths compared with peaks from uncoated sample. The streak camera output profile of each sample was quite different and the decay rates of Ag-coated samples were faster than those of uncoated samples. The decay profiles of the Ag-coated sample strongly depend on the wavelength and become faster at shorter wavelengths, whereas PL lifetimes of uncoated samples were almost constant at 9 ns. The fastest emission lifetime of Ag-coated sample is about 200 ps at 440 nm. This is about 32 times faster than that of an uncoated sample. We attribute the increases in both emission intensities and decay rates from Ag-coated samples to the efficient energy transfer from electron-hole pair recombination in the QW to electron vibrations of SPs at the silver/GaN interface. The SP energy of silver/GaN interface is estimated to about 2.8 eV (440 nm). SP coupling should therefore be more effective at shorter wavelengths because the wavelength is closer to the SP energy. That is why enhancements of both emission intensities and decay rates from Ag-coated sample were most remarkable at 440 nm. We also measured the temperature (T) dependency of the time-resolved PL measurements. We find that SP coupling rates become larger with increasing with T, similar to the behaviour of nonradiative recombination rates. This suggests that the coupling mechanism from exciton to phonon modes may be similar to that from exciton to SP mode. This new understanding would help to optimise the QW-SP coupling to develop super bright and high-speed LEDs that offer realistic alternatives to conventional fluorescent tubes and light bulbs. [1] Okamoto et al. Nature Material, 3, 601 (2004).

10:30 AM GG7.6
Effective mode volume in plasmonic nanoresonators: towards a common description of dielectric and metallic cavities.

Stefan Maier[1] and Oskar Painter[2]; [1]Dep. of Physics, University of Bath, Bath, United Kingdom; [2]California Institute of Technology, Pasadena, California.

The localization of light using surface plasmon-polaritons allows the creation of cavities and waveguides with mode profiles below the diffraction limit of light, thus enabling the creation of a subwavelength photonic infrastructure. However, the generation of light volumes below the diffraction limit in the dielectric space surrounding a metallic nanostructure does not imply that the effective electromagnetic mode volume V_{eff} is smaller than the diffraction limit, due to the fact that upon resonance a significant amount of the electromagnetic energy resides inside the metal. Thus, it is not a priori clear under which circumstances plasmonic cavities are preferable to the use of dielectric cavities, e.g. for sensing purposes. While for the latter a description of their ability to both spectrally and spatially localize electromagnetic energy is well established in the form of quality factor Q and V_{eff}, metallic cavities have to the best of our knowledge up to this point not been described in these terms. As a canonical example, we analyze the electromagnetic confinement for a metal-insulator-metal heterostructure. For small separation of the metallic half-spaces, such a structure can indeed support modes with V_{eff} far below the diffraction limit, taking proper account of both dispersion and the energy inside the metallic regions. We show that metallic nanoresonators can show Q/V_{eff} surpassing those of their dielectric counterparts for infrared frequencies. Additionally, we present a description of surface enhanced Raman scattering in terms of V_{eff}.

10:45 AM GG7.7
Three Dimensional Metallodielectric Photonic Crystal Based on Self-Assembled Gold Nanoshells. Jin-hyoung Lee and Wounjhang Park; Department of Electrical and Computer Engineering, University of Colorado at Boulder, Boulder, Colorado.

This report describes the synthesis and self-assembly of gold nanoshells and the optical properties of the self-assembled nanoshell opal structures. Our theoretical modeling studies found that metal nanoshell opal should exhibit a complete 3D photonic bandgap up to 5 times greater than the purely dielectric system such as the Si inverse opal. Self-assembled gold nanoshell opals are therefore an excellent platform for highly integrated 3D nanophotonic systems. Gold nanoshells were synthesized by first synthesizing monodispersed silica spheres, salinizing their surfaces for gold attachment and subsequently growing thin gold layers. The scanning electron micrographs clearly showed the formation of continuous and uniform gold nanoshells and the optical extinction spectra exhibited pronounced and well-defined surface plasmon resonances. The position and lineshape of the surface plasmon peaks were in excellent agreement with the prediction of the Mie scattering theory. We then used the sedimentation method for the self-assembly of nanoshells. In this process, the nanoshell solution was spread inside a circular ring so that a concave meniscus is formed. By the lateral capillary force, ordered structure begins to form at the center and grows to the radial direction. The reflection spectrum of the self-assembled nanoshell exhibited strong Bragg reflection peaks, indicating the formation of fcc close-packed structure. More specifically, the opal structure made of nanoshells with core diameter of 420 nm and shell thickness of 20 nm exhibited two strong reflection peaks at 1230 nm and 630 nm. The photonic band calculation suggested that the former corresponds to the pseudo-gap along (111) direction and the latter to the complete 3D photonic bandgap. We are currently undertaking a systematic study on the dependence of photonic bandgap properties on the structural parameters such as core size and shell thickness. The experimental results will be complemented and verified by extensive theoretical studies using the finite-difference time-domain method.

11:00 AM GG7.8
Simulations of plasmon resonances in sub-wavelength metallo-dielectric photonic crystals. Rana Biswas[1,3], Changgeng Ding[1], Irina Puscasu[2], Martin Pralle[2], Michael McNeal[2], James Daly[2], Anton Greenwald[2] and Ed Johnson[2]; [1]Dept. of Physics, Iowa State University, Ames, Iowa; [2]Ion Optics, Waltham, Massachusetts; [3]MRC, ECpE, Ames Lab, Iowa Stae Univ, Ames, Iowa.

A rigorous S-matrix (scattering matrix) theory is developed to simulate and understand the optical properties of a sub-wavelength metallo-dielectric photonic crystal structure. The metallo-dielectric photonic crystal has been fabricated by Ion-Optics [1] for infrared sensing applications. The device structure consists of a sub-wavelength array of holes in a metal layer residing on a photonic crystal. The S-matrix approach simulates the reflection and transmission through this structure by solution of eigenmodes within each layer. Realistic material absorption and dispersion is utilized. We find in the simulations sharp resonant absorption and corresponding resonant emission modes, at wavelengths close to the lattice spacing, in agreement with measurement. There is an enormous enhancement of the fields within the sub-wavelength apertures with maxima near the edges of the holes and minima in the interior of the apertures, for the resonant modes. Analysis of the dominant eigenmodes connects the resonances to surface plasmon modes at the metal-dielectric interfaces. The thin metal layer with a sub-wavelength array of holes for a range of hole sizes, exhibits the well-known extraordinary transmission peaks for with our simulation approach. Simulations find that photonic crystals (without metal coatings) have transmission resonances for out-of-plane propagation, which correlate well with measurements. These effects combine to generate the unusual surface-plasmon driven absorption of the metallo-dielectric photonic crystal. The role of the photonic crystal, lattice symmetry, aperture shapes and material parameters in supporting such resonant plasmon modes will be discussed. [1] I. Puscasu, J. Applied Physics (2005). Supported by the NSF under grant DMR-0346508.

11:15 AM GG7.9
Spectroscopical Properties of Artificial Opals Infiltrated with Gold Nanoparticles. Davide Comoretto[1], Valentina Morandi[2], Franco Marabelli[2], Vincenzo Amendola[3] and Moreno Meneghetti[3]; [1]Dipartimento di Chimica e Chimica Industriale, Universita' di Genova, Genova, Italy; [2]Dipartimento di Fisica "A. Volta", Universita' di Pavia, Pavia, Italy; [3]Dipartimento di Chimica Fisica, Universita' di Padova, Padova, Italy.

We report on the spectroscopical and morphological characterization of polystyrene artificial opal films infiltrated with gold nanoparticles (NpAu) obtained by laser ablation. Opal films are grown with the meniscus technique from water and alcohol mixtures of monodisperse nanosphere and NpAu suspensions having different relative concentrations. Polarized variable incidence angle reflectance and transmittance micro-spectroscopy shows that the bare opal photonic structure is modified upon infiltration of NpAu. As a matter of fact, the photonic crystal stop band is shifted up to 1500 cm-1 below its original position (depending on the NpAu loading degree) and the high energy high incidence angle spectral features due to excitation of photonic modes are modified. Moreover, additional spectral structures are observed. The dispersion of the stop band by changing the incidence angle indicates that a modification of about 10% of the effective refractive index has been achieved. Atomic Force Microscopy shows that NpAu are embedded in the interstices between spheres preserving their nanoparticle nature without creating bulk metal. In fact, by dissolving the infiltrated opal with the proper solvent, a reddish film is obtained whose transmittance spectrum resembles that of original NpAu suspensions. This fact is important since the optical nonlinearities of NpAu inside the photonic crystal are also preserved. The optical switching properties of these films are currently under investigation.

11:30 AM GG7.10
2-D Photonic Crystals Metal on Polymers. Anton C. Greenwald[1], Irina Pucasu[1], Martin U. Pralle[1], Mark P. McNeal[1], James T. Daly[1], Edward A. Johnson[1] and William L. Schaich[2]; [1]Ion Optics, Inc., Waltham, Massachusetts; [2]Dept. of Physcis, Indiana University, Bloomington, Indiana.

Thin patterned metal layers on structured semiconductors have been shown to emit narrow band radiation when heated. [1] This effect is due to plasmon coupling at the semiconductor-metal interface and the metal-air interface. Emission pattern is strongly affected by the metal pattern and metal and semiconductor properties. Computer simulation models matched experimental results. To expand applications to very large areas we constructed photonic crystals of metal on Kapton and recovered strong resonances and emission peaks for heated samples, similar to data from semiconductors. Variations of optical properties with materials and patterns, both the top conductor (gold, aluminum, platinum, titanium-nitride) and support (silicon, Kapton, odies) will be presented. 1 "Angular Roll-Off Dependence Of Spectral Emission From A Metallodielectric Photonic Crystal", Irina Puscasu, et. al. Proc. MRS, paper, **v797**, paper W4.7 (2004)

11:45 AM GG7.11
Plasmons in Very Strongly Coupled Metallic Nanoparticles: The Singular Limit of Touching Particles. Garnett W. Bryant[1], Christopher Kakovitch[1], Isabel Romero[2], Javier Aizpurua[2] and F. J. Garcia de Abajo[2]; [1]Atomic Physics Division, National Institute of Standards and Technology, Gaithersburg, Maryland; [2]Donostia International Physics Center, San Sebastian, Spain.

Understanding coupling between metallic nanoparticles is critical for exploiting plasmon nanooptics in sensors, for nanooptical communication, and in new metamaterials. Coupled nanoparticles exhibit redshifts of the dipolar plasmonic intraparticle response. Experiments [T. Atay, J. H. Song, and A. V. Nurmikko, Nano Lett. 4, 1627 (2004)] show that the plasmonic response of nearly touching, very strongly coupled gold nanoparticles exhibit drastic redshifts. When the nanoparticles overlap, the plasmonic modes are strongly

mixed and blueshift. We use the boundary element method to calculate the classical plasmonic response of coupled metallic nanoparticles in the limit of very strong coupling. We use the results to understand the singular limit when the nanoparticles are nearly touching or just overlapping. The plasmonic response drastically redshifts as the coupled nanoparticles are brought to nearly touching. Drastic charge buildup at the gap occurs. However the dipolar response is suppressed in the limit of nearly touching particles because the gap is too small to sustain a dipolar response. When the nanoparticles overlap, the charge buildup remains at the edges of the interparticle junction. There is strong coupling between the charge at either edge of the junction and strong hybridization of the plasmonic response from each of the nanoparticles. As the overlap at the junction increases, the coupling weakens, the plasmonic response blueshifts and the strength of the dipolar response builds back. These results explain the observed singular response in the limit of nearly touching and just overlapping nanoparticles. To fully understand the limit of nearly touching nanoparticles, we have also developed a density functional theory for the quantum mechanical response of these particles. A jellium model is used for the coupled gold nanoparticles. Initial results are presented to understand when the classically separated nanoparticles with an interparticle gap are shorted out by quantum mechanical electronic interparticle tunneling. This defines a cutoff for the singular limit for nearly touching nanoparticles.