Degenerate and Nondegenerate Nonlinear Spectroscopy: Eric Van Stryland, David Hagan, Scott Webster, Dmitry Fishman, Zhigang Hu, Trevor Easley and Markus Seidel; CREOL, University of Central Florida, Orlando, Florida.

We will describe methods for rapidly characterizing the spectrum of nonlinear optical absorption as well as the dispersion of the nonlinear refractive index in materials containing semiconductors and organic dyes. A weak White-Light Continuum (WLC) can be used as the probe in pump-probe experiments and thus yield the spectrum of the nondegenerate nonlinear absorption. Kramers-Kronig integrals can then be used to calculate the dispersion of the nonlinear refractive index. Temporally delaying the probe gives information on the temporal dynamics of the nonlinear material. Alternatively, the WLC E-scan relies on a spectrally intense femtosecond WLC which we have recently developed and made available over more than an octave bandwidth using up to 1 fs pulses at 800 nm weakly focused into a 2-μm tube filled with krypton gas at -2.5 atm. By seeding the WLC with a very weak (-10^-6 of the pump beam) visible pulse we can increase the total energy in the continuum by 100 to obtain a few nJ/nm spectral energy density. Thus, WLC-E-scan experiments can simultaneously yield the nonlinear absorption spectrum and index dispersion for frequency degenerate nonlinearities. Such experiments have provided a wealth of data on the third-order nonlinear response of materials. Recently we demonstrated orders of magnitude enhancement in the two-photon absorption (TPA) coefficient of direct bandgap semiconductors when going to extremely nondegenerate photon pair states (i.e., energy ratios of -10^5) which is in excellent agreement with a simple 2-parabolic band model. This has allowed gated detection of subpico-second, IR detection, using wide-gap semiconductors. Further applications, such as all-optical switching, should be possible. In addition, the inverse process of 2-photon gain should be significantly enhanced, making the likelihood of 2-photon semiconductor laser realistic. Analogous processes in organic materials are under study. Measurements of nonlinearities as a function of pulsewidth can further help determine the dynamics of the nonlinear response. Separating the bound electronic and nuclear contributions to the nonlinear refractive index, n2, is of interest to fully understand the nonlinear mechanisms in many materials but particularly for organic dyes. A detailed study of C60 shows that measurements of n2 using pulses 10 fs are dominated by the bound-bound electronic response, while as the pulsewidth is increased nuclear contributions become dominant. A simple model of the kinetics shows the magnitude and temporal response of such nonlinear contribution to n2. Extending these models and measurements to organic dyes is underway.

8:30 AM KL.2
Chromophores for Nonlinear Absorption at Telecommunications Wavelengths: Chantal Andreux, UMR CNRS-UCBL 5182, Ecole Normale Superieure de Lyon, Lyon, Ceden, France.

We will present two different families of chromophores (heptamethine cyanines and aza-bodipy) for two-photon absorption (TPA) based optical power limiting in the IR (particularly at telecommunications wavelengths). Spectroscopic properties of these chromophores will be discussed in this talk. Optical power limiting will be presented on the basis of TPA and excited state absorption (ESA) properties. P.A. Bouit, G. Wietzel, G. Berginc, B. Lesueur, L. Tropet, P. Fenayroz, Y. Breitenbicher, K. Kamara, O. Maury, C. Andreux, Chem. Mat. 2007, 19, 2215-2222; P.A. Bouit, R. Westlund, P. Fenayroz, O. Maury, M. Mailloch, E. Malemstrom, C. Andreux, Adv. Mat. 2009, 21, 1153-1156; P.A. Bouit, C. Andreux, L. Tropet, B. La Gruenica, C. Andreux, O. Maury JACS 2010, 132, 4328-4335; F. Bellot, S. Pigeon, C. Andreux, B. La Gruenica, C. Andreux, O. Maury Org. Lett. 2015, 13, 29-35.

9:00 AM KL.3
Principles and Applications of Small Molecule Assemblies for Organic Nonlinear Integrated Optics: Igor Lizzi, Physics, Lehigh University, Bethlehem, Pennsylvania.

For applications in third-order nonlinear integrated optics it is necessary to develop organic materials that combine a large third-order susceptibility, a high optical quality, and the ability to integrate them with existing guided wave technology. This work will discuss how this aim was achieved by developing small molecules and their corresponding dense single-component supramolecular assemblies. While larger molecules can have larger third-order polarisabilities, they are generally more difficult to handle and to assemble into a dense solid state where their nonlinearities is not diluted. Small molecules, on the other hand, can be
chromophores and the need to take electron correlation effects properly into account, a sufficiently accurate analysis of the non-linear response of cyanines is a challenging task for modern computational chemistry. In this talk, I will discuss our recent results for several promising cyanines using a combined approach of density-functional theory and configuration interaction methods. In particular, I will discuss the influence of solvents and counterions as well as the impact of aggregation on the ground- and excited-state electronic properties of cyanine molecules.

11:30 AM K4.7
Nanosolvent-Free Hybrid Silica Coatings: A Versatile Tool for Light Manipulation

Ali A. Letallier\(^1,2,3,6\), Cedric Boisserie\(^4\), Francois O. Ribot\(^5\), Clement Sandor\(^6\), Jerome Taisset\(^7\), Etienne Berthe\(^6\), Elie Sonderegger\(^6\), Christophe Costeau\(^6\), Gilles Lecomte\(^6\) and Nicolas Chemin\(^6\),

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4Laboratoire d’Electromagnetisme et d’Optique, Universite de Paris-Sud, Orsay, France.
5Laboratoire d’Electromagnétisme et d’Optique, Universite Paris-Sud, Orsay, France.
6Laboratoire de Chimie de La Matiere Condensee et d’Instrumentation Optique, Universite de Technologie de Troyes, Troyes, France.
7Surface du Verre et Interfaces, Saint-Gobain Recherche, Aubervilliers, France.

In light-emitting devices, a large amount of the light produced by the active layer is trapped inside the device because of internal reflection. Patternning the different interfaces induces an index gradient and light scattering and can therefore lead to an increase of the light output. Among the existing methods, Nanosolvent Lithography (NIL) emerges as a simple route for surface patterning at the sub-micrometer scale over large areas. To avoid multiple step processing and the poor stability of polymers resists, imprint of functional materials is required. Hybrid sol-gel materials form an innovative class of resists for NIL. They are based on the solution processing of organic precursors to obtain metal oxide. For instance we previously demonstrated the replication of patterns with sub-100 nm lateral size and aspect ratio greater than 1 into hybrid sol-gel silica. (1) A combination of low viscosity and high reactivity enables fast and conformal imprint over several tens of cm. (2) Furthermore, we demonstrate how the elaboration of the chemicals can be modified to adjust the process. (3) Due to their low dielectric constant, nanopatterned silica coatings are very suitable for direct applications to photonic and integration in displays. Finally, thanks to the versatility of the sol-gel processing, other functionalities have been added to obtain multifunctional coatings. For instance, it was possible to graft NIL chromophores such as 2,4-dinitrophenylsilane into the silica network and to successfully imprint the resulting layer. In the context of displays and lighting, we developed the introduction of colloidal quantum dots inside the patterned silica layer. (4) This last system is well suited for light conversion and extraction. As it absorbs around 400 nm and emits light in the visible range, such a layer can set as a conversion layer in LED devices. Moreover, the light output in the patterned area increased 60% compared to the flat surfaces. References: (1) Penn, C., Claessens, V., Berthe, E., Sonderegger, E., Advanced materials 2009, 21, 355-361; Letallier, A., Taisset, J., Chemia, N., Barbi, E., Sondergard, E., Chem. Mater. 2010, 22, 3343; (2) A. Letallier, F. Ribot, C. Boisserie, C. Boisserie, J. Taisset, E. Berthe, N. Chemin, J. Chem. Soc., submitted; (3) A. A. Letallier, T. Richard, C. Boisserie, C. Boisserie, C. Costeau, G. Lecomte, E. Berthe, S. Sonderegger, N. Chemin, and Francois Ribot, Advanced materials, submitted.

11:45 AM K4.8
Hybrid Silicon Nitride and Colloidal Nanocrystal Waveguides and Microlenses: A Highly Versatile Test Bed for Visible to Near-Infrared Active Integrated Photonics

Bren De Graaf\(^1,2,3,8\), Katayana Komorowska\(^1,9\), Zeger Hem\(^8\) and Dries Van Thourhout\(^1\), Photonic Research Group, DTGEC, Ghent University IMEC, Gent, Belgium. 2Physics and Chemistry of Nanostructures, Inorganic and Physical Chemistry Department, Ghent University, Gent, Belgium. 3Center for Nano- and Biophotonics (NB-Photonics), Ghent University, Gent, Belgium.

Recent advances in colloidal nanocrystal synthesis have extended the material’s library greatly to more complex colloidal heterostructures. These new heterostructures are engineered to solve intrinsic problems of colloidal quantum materials, such as ultraviolet multiexciton Auger recombination and overall material stability. These new properties, together with their ease of production and processing, and the tunability of their optical properties, make them excellent candidates to render passive integrated photonic devices on silicon-on-insulator, silica and silicon nitride, active. In this work, we present an extensive study of hybrid silicon nitride and colloidal nanocrystal heterostructures. An extensive review of the field is presented in reference 1.


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SESSION K1: Photonics IV
Chair: Steven Herr

Thursday Afternoon, November 24, 2011
Independence E (Dawes)

1:30 PM K4.5
Strong Multimode Photonic Microresonator and Nanoparticle Interactions

Xueyi Zhu\(^1\) and Patrick Pignatello\(^2\), NYU and CUNY, New York City, New York. 1Massachusetts Institute of Technology, Cambridge, Massachusetts.

We have demonstrated strong multimode photonic microresonator and nanoparticle interactions by using an integrated microdisk resonator from through port of the laser coupling bus waveguide. In addition to the fundamental resonance mode, disk resonator has higher order resonance modes. The excited higher order mode has a node at the position where the electromagnetic energy of the fundamental mode is close to a maximum. Here we report that a self-referencing mechanism can be achieved by simultaneous excitation of both fundamental and second order micro disk optical resonance modes. Additionally, we are able to measure the area around the maximum of the fundamental resonance mode and the node of the higher order mode, which have overlap in the disk. In this work, we used chip disk microresonator as the example, as a variety of types of optical microresonators have been investigated; we used nanoparticle to interact with the two optical resonance modes excited by the coupling bus waveguide, where the nanoparticle can be either electronic materials or metallic materials. The strong photonic microresonator and nanoparticle interactions has variety of applications for optical switches, waveguide and detection. The self-referencing characteristics of the two optical resonance modes have potential to achieve photonics functions independent of external perturbation, such as temperature change.

1:45 PM K4.6
Highly Efficient Double-Doped Solid-State White Light-Emitting Electrochemical Cells

Chih-Tung Lai\(^1\), Yu-Chun Shen\(^1\), Hsi-An Pan Chien\(^2\), Hai-Ching Su\(^3\) and Ken-Tsung Wong\(^4\), Institute of Lighting and Energy Photonics, National Chiao Tung University, Taiwan, Taiwan. 1Department of Chemistry, National Taiwan University, Taipei, Taiwan.

While organic light-emitting diodes (OLEDs) based on polymers and small-molecule materials have attracted intense attention due to their potential applications in flat-panel displays and solid-state lighting. Compared with conventional white OLEDs, solid-state white light-emitting electrochemical cells (WLECs) possess several promising advantages. WLECs generally require only a single emissive layer, which can be easily processed from solutions, and can therefore conveniently use air-stable electrodes. The emissive layer of WLECs contains mobile ions, which can drift towards electrodes under an applied bias. The spatially separated ions induce electrochemical doping (oxidation and reduction) of the emissive materials near the electrodes, i.e. p-type doping near the anode and n-type doping near the cathode. The doped regions induce ionic contacts with the electrodes and consequently facilitate the injection of both holes and electrons, which recombine at the junction between p- and n-type regions. As a result, a single-layered LED device can be operated at very low voltages (close to E_g/2, where E_g is the energy gap of the emissive material) and is elementary charge with balanced carrier injection, giving high power efficiencies. Furthermore, air-stable metals, e.g. Ag and Au, can be used since carrier injection in WLECs is relatively insensitive to work functions of electrodes. In this work, we highly efficient solid-state white LEDs based on a double-doped strategy, which judiciously introduces an orange-emitting quest, [(pyppy)(dip)], (1)PFP (where pyppy is 2-phenylpyridine and dip is 2,5-diphenyl-l,4-pyrazine), which can be directly integrated in a single-doped emissive layer comprising of an efficient blue-green emitting host, [tr(dipppy)(dib-hpy)], (2)PFP (where dipppy is 4,5-diphenyl-1,2,4-triazoline and dib-hpy is 1,4,5-dicyanobenzene-2,2'-bipyridine) and a red-emitting quest, [tr(ppy)2](1)PFP. http://www.nrvi.org/111-abstracts/index.html

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