

SYMPOSIUM MM
Ultrafast Lasers for Materials Science

November 30 - December 1, 2004

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* Invited paper

8:30 AM *MM1.1
Phase Change Mechanisms in Laser-Matter Interaction.
Xianfan Xu, Purdue University, West Lafayette, Indiana.

Laser micro-machining is finding many applications in materials processing and manufacturing. At Purdue, various laser techniques are being used to fabricate sensors, micro-fluidic systems, bio-MEMS, and nano-structures. This presentation will mainly deal with the fundamental issues involved in laser-matter interaction. Our studies are focused on laser induced thermal and thermomechanical phenomena and phase change mechanisms that determine the materials removal process during laser machining, as well as the non-linear laser-material interaction in dielectric materials. It is shown that during nanosecond laser heating, explosive phase change could occur, during which the liquid is superheated to close to the thermodynamic critical point, followed by an explosive, homogeneous phase transformation. On the other hand, it is observed in the experiment that the time for nucleation during laser induced phase explosion is on the order of one nanosecond. Thus, when a laser with pulsewidth of one picosecond or less is used, it is likely that the material can be heated above the critical point, and another type of phase change, spinodal decomposition becomes possible. The time scales obtained from the fundamental studies, i.e., the time scale of non-linear optical response in ultrafast laser-dielectrics interactions, non-equilibrium energy transfer between electrons and the lattice, and the time scales of lattice melting and removal provide a good guidance for synthesizing pulse trains with specific temporal waveforms to improve results of ultrafast laser micro-machining. The experimental studies are aided with numerical simulations. We have developed finite difference (FD) and molecular dynamics (MD) simulation techniques. Results of these studies also showed the explosive nature of pulse laser-matter interaction. Our current effort is on large-scale hybrid FD/MD type of numerical studies to develop parallel computational techniques, which may provide a better understanding of the material removal process.

9:00 AM MM1.2
Polaritonic Materials Fabricated and Tested with Ultrashort-Pulse Lasers. David W. Ward, Eric Statz, Thomas Feurer and Keith A. Nelson; Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts.

Using ultrashort-pulse laser machining, we have fabricated guided-wave, photonic bandgap, effective index, and aperiodic structures designed to control propagation of phonon-polaritons in ferroelectric crystals with thicknesses from 10 microns to 500 microns. Broadband and narrowband polaritons were generated with impulsive stimulated Raman scattering (ISRS) using an ultrashort laser pulse. The spatial and temporal evolution of the polaritons were recorded as they propagated through the structures. Functional devices such as near-field THz microscope tips and slabs, tunable THz filters, waveguides, and resonators as well as the design and fabrication procedures for these devices will be demonstrated.

9:15 AM *MM1.3
Ultrafast Laser Processing for Lab-on-Chip Device Manufacture. Koji Sugioka, Ya Cheng and Katsumi Midorikawa; Laser Technology Laboratory, RIKEN - The Institute of Physical and Chemical Research, Wako, Saitama, Japan.

There has been rapid progress in research and development of lab-on-chip device that is used for field-deployable chemical analysis with high-efficiency, high-accuracy, and high-performance. The lab-on-chip device is composed of microfluidic components, such as microcell, and microchannel for infusion of reagents and storage of reactants, micromechanical components, such as microvalve, micromixer, and micropump for control of reagent flow and reaction, and microoptical components, such as micromirror, microlens, micrograting, waveguide and microoptical sensor, for in-situ analysis of the reactants. These components are integrated in a small chip and then infusion, mixture, and reaction of the reagents and analysis of the reactants are successively conducted. Consequently, the lab-on-chip device makes it possible to reduce reaction and analysis time in human gene and protein analysis, medical inspection, new drug development, and environmental monitoring by measuring a trace amount of reagents. For the lab-on-chip device manufacture, development of fabrication technique of three-dimensional (3-D) hollow microstructures is in great demand. Since it is well known that ultrafast laser realizes internal modification of transparent materials, use of such a laser would be a promising and simple approach for embedding 3-D hollow microstructures inside the materials. In fact, the internal modification of glass by ultrafast laser followed by

selective wet etching of the modified regions can fabricate such structures. In the present paper, fabrication of 3-D hollow microstructures in glass is demonstrated for the lab-on-chip device application. We use a commercially available photostructurable glass (Foturan glass) from Schott Glass Corp. The fabrication process includes three steps: (1) direct writing of latent images in the glass by tightly focused fs laser beam (775 nm, 150 fs); (2) baking of the laser irradiated sample in a programmable furnace for formation of modified regions; and (3) etching of the modified sample in a 10% diluted solution of hydrofluoric acid for the selective removal of the modified regions. The modified region is preferentially etched away with a contrast ratio of about 50 in etching rate compared with unmodified regions, resulting in formation of 3-D hollow internal microstructures. By using this technique, we fabricate a variety of microfluidic structures. Mixing of two different aqueous solutions in the embedded channel is demonstrated using the fabricated structure. Then, we embedded a freely movable microplate inside the microfluidics that has a function of microvalve controlling flow direction of fluids. We also apply the present technique to integrate microoptics, such as a micromirror and a micro beam splitter, and micro laser inside the glass for optical analysis of reactants in the lab-on-chip device.

10:15 AM *MM1.4
Generation of New Nanomaterials by Interfering Femtosecond Laser Processing. Yoshiki Nakata, Tatsuo Okada and Mitsuo Maeda; Grad. school of ISEE, Kyushu University, Fukuoka, Japan.

New nanomaterials such as nanobump, nanomesh, nanobelt were generated from thin film processed by interfering femtosecond laser beams. To split and correlate femtosecond laser, a demagnification optical system with a transmission beam splitter was used. Different interference pattern were generated depending upon the number of the beams, magnification factor of the optical system, and the period of the transmission grating. Metallic single- or multi-layered film deposited on a silica substrate was used as a sample. With four interfering femtosecond laser beams, a conical nanobump arrayed in a matrix was generated with single laser shot. The diameter and height became larger and a bead was formed at the top of a bump as the femtosecond laser fluence increased. Moreover, with three or two interfering femtosecond laser beams, ellipsoidal or linear nanobump array was generated. As an application of a conical nanobump array, field emission from the array was demonstrated, and the I-V characteristics were measured. On the other hand, with higher fluence and four interfering femtosecond laser beams, a nanohole array was generated. A nanomesh was also generated from the nanohole array by scratching the film. A grating was also generated with two interfering femtosecond laser beams, and nanobelts were generated from the grating by scratching. Bimetallic nanobelt was also generated from multi-layered thin film. This technique for the generation of nanomaterials has advantages as follows; the structures are unique and controllable, and automatically aligned. Moreover, any raw film can be used, which may be amorphous, crystalline, single- or multi-layer, composite, compound, or even living materials. No special ambient condition such as high vacuum, temperature control, special gas or liquid is required during fabrication. In addition, single process will result in low photon cost. This approach is expected to improve the existing applications and open new opportunities for nanomaterials.

10:45 AM MM1.5
Femtosecond-Laser-Encoded Distributed-Feedback Color Center Laser in Lithium Fluoride Single Crystal.
Ken-ichi Kawamura¹, Toshio Kurobori², Masahiro Hirano¹, Toshio Kamiya³ and Hideo Hosono^{3,1}; ¹Hosono Project, Japan Science and Technology, Yokohama, Kanagawa, Japan; ²Kanazawa University, Kanazawa, Japan; ³Tokyo Institute of Technology, Yokohama, Japan.

Lithium fluoride (LiF) offers good physical and chemical properties including high photothermal stability of laser active color centers, therefore it has been used for room temperature color center lasers. Especially, F² and F³⁺ centers in LiF are very promising candidates for visible laser action. Recently, Montecchi¹ has reported enhancement in the refractive index due to the formation of the color centers and fabricated active channel wave waveguides. However, high-energy deposition techniques such as ion implantation, electron beams and γ -ray irradiation are generally needed for producing the color centers in LiF because of its large optical bandgap (14 eV). To our knowledge, producing the color center in LiF using optical light has not been demonstrated. Here we report the fabrication of three-dimensional active optical devices inside LiF by holographic encoding method using an infrared fs laser². We present gratings, waveguides and a distributed-feedback (DFB) color-center laser. The femtosecond laser pulses were focused on the surface and inside of LiF plates to give a spot size of 50 micrometer in diameter. As a result, formation of F, F², F³⁺ and F²⁺ color centers were observed by optical transmission measurements. Estimated concentration of the F² color center was $2 \times 10^{18} \text{ cm}^{-3}$ and the induced refractive index change was about 1%. Then, we fabricated DFB F² color center laser

using microgratings aligned in a stripe. Microgratings 40 μm in diameter were written continuously in a 5 mm line at a depth of 100 μm^3 . As we aimed at lasing from 2centers, the line-spacing of the grating was designed to be 510 nm, which corresponds to the theoretically expected DFB oscillation wavelength of 710 nm. Much narrowed, intense emission is clearly observed at 707 nm from the grating line. The observed oscillation wavelength is almost consistent with the theoretically expected value 710 nm. The line width is less than the resolution of our measurement system (1 nm). Such narrow emission spectrum was not observed without gratings. 1. M. Montecchi, et al., J. Appl. Phys. **86**, 1 (1999). 2. K. Kawamura, et al., Appl. Phys. Lett. **78**, 1038 (2001); Appl. Phys. Lett. **79**, 1228 (2001); Appl. Phys. Lett. **81**, 1137 (2002); Rev. Sci. Instr. **73**, 1711 (2002). 3. K. Kawamura, et al. Appl. Phys. Lett. **84**, 311 (2004).

11:00 AM MM1.6

Femtosecond Laser Micromachining of Periodical Structures in Si <100>, Mohamed El-Bandrawy and Mool C. Gupta; Applied Research Center, Old Dominion University, Newport News, Virginia.

A femtosecond Ti: sapphire laser from Spectra Physics was frequency doubled to give a wavelength of 400 nm, pulse width of 160 fs, and a repetition rate of 1 kHz. The laser was used with a computer controlled galvo head to write periodic structures on Si <100> samples, which were fixed on a high resolution X-Y stage. Laser pulses of 130 nJ were focused using an objective lens of 0.65 NA. Laser parameters were optimized for efficient submicron ablation, yielding lines 700 nm wide by 600 nm deep. 1-D and 2-D periodic structures of 5 and 5 x 5 micron periods, respectively, were fabricated and studied using optical and atomic force microscopy. Light diffraction characteristics of the periodic 1-D and 2-D structures were examined. Diffraction properties of the 1-D structures were highly dependent on the light polarization orientation with respect to micromachining direction.

11:15 AM MM1.7

WDXRF of Aluminum on Silicon wafers with Si-K α radiation generated by interaction of fs-laser radiation with Silicon wafers. Robert S. Ritschel¹, Christian Kaiser², Torsten Mans³, Peter Russbuehldt², Heinrich Schwenke³, Joachim Knoth³, P. Beaven³, E. W. Kreutz¹ and Reinhart Poprawe^{1,2}; ¹Lehrstuhl fuer Lasertechnik, RWTH, Aachen, Germany; ²Institut fuer Lasertechnik, Fraunhofer Gesellschaft, Aachen, Germany; ³GKSS Forschungszentrum, Geesthacht, Germany.

The miniaturisation of semiconductor structures on silicon requires clean silicon wafers to reduce leakage currents which are mainly caused by Aluminum impurities. For detection of Aluminum on silicon wafers X-ray fluorescence of Aluminum is stimulated by X-ray radiation generated by focussed femtosecond laser radiation. The occurring intensities are smaller than $3*10^{16}$ W/cm² on a thin (20 μm) silicon target wafer which is positioned directly above the silicon wafer under consideration. The generated X-rays with an energy between Aluminum K α edge and Silicon K α edge transmit the target wafer and stimulate X-ray fluorescence of Aluminum on the silicon wafer. The spectrum of the X-ray radiation emitted from the target wafer and the X-ray fluorescence spectrum of the silicon wafer is recorded by a von Hamos spectrograph or by a multilayer mirror spectrograph. The intensity of the measured Aluminum K α line is directly proportional to the concentration of Aluminum atoms on the silicon wafer. A direct diode pumped, compact, high energy efficient Cr:LiSAF / Cr:LiSGaF MOPA laser system is used which generates laser pulses with a pulse energy of 100 μJ , a pulse length of < 100 fs and a repetition rate < 2.5 kHz. The laser pulses are focused by a chromatically corrected aspheric lens to the target wafer down to a spot diameter of 2 μm . Due to the small Rayleigh length (3.5 μm) of the focused laser radiation the vibrations of the target wafer, which is translated at a velocity of 20 mm/s by an X-Y-table, must be compensated by an autofocus system based on an astigmatic method which can compensate oscillations with amplitudes < 10 μm and oscillation frequencies < 100 Hz. For optimized absorption a femtosecond prepulse is focused to the target wafer to establish a plasma above the target wafer. After a delay time which depends on the intensity of the prepulse the mainpulse is focused into the plasma. With this technique the conversion efficiency from laser pulse energy into X-ray pulse energy can be increased by an order of magnitude necessary to reduce the measurement time for X-ray fluorescence analysis.

11:30 AM MM1.8

Femtosecond Laser-Induced Micro-Structuring of Thin a-Si:H Films. Barada K. Nayak and Mool C. Gupta; Applied Research Center, Old Dominion University, Newport News, Virginia.

Improving light trapping is a major challenge in solar cell research. In a-Si:H thin film solar cells, light trapping has been sought as a method to enhance absorption and improve the efficiencies of these devices. Laser assisted surface texturing of silicon surfaces, especially

wafers, has been reported by many research groups as a means of enhancing absorption. Here we present results for surface texturing of 2 micron a-Si:H thin films by femtosecond laser. The texture led to a significant enhancement in optical absorption (80%) in visible through near IR (60%). The textured surface was observed by a scanning electron microscope (SEM), and showed a forest of spikes. The surface was also analyzed using an atomic force microscope (AFM), giving spike heights of 65-75 nm. The samples were completely black after laser treatment.

11:45 AM MM1.9

Femtosecond Laser Interactions with Co/Al Reactive Foils. Yoosuf N. Picard^{1,2}, Kim M. Archuleta³, David P. Adams³ and Steven M. Yalisove^{1,2}; ¹Materials Science & Engineering, Univ. of Michigan-Ann Arbor, Ann Arbor, Michigan; ²Center for Ultrafast Optical Science, Univ. of Michigan-Ann Arbor, Ann Arbor, Michigan; ³Advanced Manufacturing Processes Lab, Sandia National Labs, Albuquerque, New Mexico.

Femtosecond lasers have recently demonstrated controlled laser cutting of energetic materials, including reactive multilayer systems, without igniting a self-propagating reaction or explosion. We continue this work by exploring the potential benefits and limitations of femtosecond laser cutting in Co/Al reactive multilayer material systems. Co/Al reactive foils are made using magnetron sputter deposition on thermally oxidized silicon wafers and released as a free standing foil. Femtosecond laser pulses at 780 nm wavelength and 150 femtoseconds in pulse length are generated from a Ti:sapphire femtosecond laser system and focused at the surface of these Co/Al foils. Studies of the resultant morphology for single and multiple pulses are conducted using scanning electron microscopy and atomic force microscopy. The nature of the material ablation mechanisms are explored as well as thermal propagation and critical fluences for laser ignition.

SESSION MM2:

Chair: E. W. Kreutz

Tuesday Afternoon, November 30, 2004

Room 303 (Hynes)

1:30 PM *MM2.1

Femtosecond Laser Induced Phenomena in Glasses and Photonic Device Applications. Kazuyuki Hirao^{1,2}, Yasuhiko

Shimotsuma¹, Jianrong Qiu² and Kiyotaka Miura²; ¹Graduate school of Engineering, Kyoto University, Kyoto, Japan; ²Photon Craft Project, JST, Seika-cho, Kyoto, Japan.

Femtosecond laser is a perfect laser source for materials processing when high accuracy and small structure size are required. Due to the ultra short interaction time and the high peak power, the process is generally characterized by the absence of heat diffusion and, consequently molten layers. Various induced structures have been observed inside glasses after the femtosecond laser irradiation. Here, we report the refractive index change, space-selective valence state manipulation of active ions, nano-grating and precipitation control of nanoparticles by a femtosecond laser in glasses. The mechanisms of the observed phenomena were also discussed.

2:00 PM MM2.2

Induction of Photosensitivity in Transparent Materials by Pre-Pulse Irradiation and Micrograting Encode by a Single Pulse Holographic Method with Femtosecond Laser.

Ken-ichi Kawamura¹, Takukazu Otsuka², Masahiro Hirano¹, Toshio Kamiya² and Hideo Hosono^{2,1}; ¹Hosono Project, Japan Science and Technology, Yokohama, Kanagawa, Japan; ²Tokyo Institute of Technology, Yokohama, Japan.

We have proposed a "holographic encoding technique by an interfered infrared femtosecond (fs) laser pulse"¹. In this technique, holographic gratings are encoded on the surfaces of various nonphotosensitive materials as a consequence of the interference of two fs pulses split from a single pulse. Furthermore, we have succeeded in encoding refractive index-modulated volume-type gratings deep inside transparent materials if we use a single chirped (500 fs) laser pulse. This technique provides a fast method applicable for encoding gratings and for fabricating optical devices such as distributed-feed-back (DFB) lasers. DFB F2-color center lasers were fabricated using encoded volume-type gratings in LiF single crystals and oscillated at room temperature². These results prove that our fs laser interference technique has good applicability to produce unique devices. However, the technique has still a few points to be improved. In this study, we have tried to control shapes of gratings and to reduce power densities for the encoding by employing a pre-excitation technique. Furthermore, we observed ultrafast electron dynamics related to optical permanent damages occurring during the encoding

of the gratings in femtosecond regime. Generally, laser damages in transparent materials is described in terms of three major processes: (I) the excitation of high-density (e.g. 10^{21} cm^{-3}) free electrons in the conduction band by multiphoton ionization, (II) laser heating of the conduction band electrons by absorbing the residual laser pulse, and (III) transfer of the plasma energy to the lattice³. We separated the contributions of these processes using the pre-pulse irradiation technique. We used pure fused silica glass and MgO single crystal for samples. A mode-locked Ti:Sapphire laser system (800 nm, 100 fs, 10 Hz) was used. One pulse was split into three pulses. A split pulse was used for pre-excitation (pre-pulse) and the other pulses were irradiated as an interfered pulse. The interfered pulse was irradiated following the pre-pulse with time delays varied from 0 to 10 ps. The pulse energies of both the pre-pulse and interfered pulse were always below the threshold energy for the encoding by a single pulse technique without a pre-pulse. We observed that threshold energy was drastically reduced by a pre-pulse irradiation and diffraction efficiency of the grating strongly depended on the delay time. Furthermore, we have succeeded in modifying the shape of a grating by changing beam patterns of the pre-pulse. 1. K. Kawamura, et al., Appl. Phys. Lett. **78**, 1038 (2001); Appl. Phys. Lett. **79**, 1228 (2001); Appl. Phys. Lett. **81**, 1137 (2002); Rev. Sci. Instr. **73**, 1711 (2002). 2. K. Kawamura, et al., Appl. Phys. Lett. **84**, 311 (2004). 3. B. C. Stuart et al., Phys. Rev. Lett. **74**, 2248 (1995).

2:15 PM MM2.3

Direct Femtosecond Inscription of Fibre Bragg Gratings.

Amos Martinez, Michael V. Dubov, Igor Y. Khrushchev and Ian Bennion; Photonics Research Group, Aston University, Birmingham, United Kingdom.

We present a method for inscribing FBG using direct, point by point writing by an infrared femtosecond laser. This method requires neither phase masks nor photosensitised fibres and hence offers a remarkable technological flexibility. Unlike known point-by point methods of making FBGs, it presents a very short process time of less than 60 seconds per grating. High quality, strong gratings were produced using this method in commercial, non-photosensitive fibres. This technique presents distinct practical benefits since the physical mechanism of femtosecond inscription is different to that of UV inscription; such as the possibility to inscribe gratings in undoped standard fibres without photosensitising and producing structures with improved thermal robustness. Gratings were produced in an amplified laser system, operating at a wavelength of 800nm and generating 150fs-long pulses at a repetition rate of 1kHz, with 0.5 μJ pulse energy. The beam was focused into the fibre core by a X100 microscopic objective. No photosensitisation procedure was carried out prior to the exposure. The fibre was placed on a high-precision, two-coordinate translation stage. The stage moved at a constant speed along the focal point of the beam. Each laser pulse produced a grating pitch. The grating period was set by changing the ratio of the translation speed to the pulse repetition rate. Reflection and transmission were monitored in-situ by using two optical spectrum analysers. Gratings were later characterised using a tuneable laser with 1pm resolution and a high-performance analyser with a 5pm resolution. The grating length ranged from 10mm to 55mm. Gratings of first, second and third order were inscribed at speeds of 0.535mm/s, 1.07mm/s and 1.605mm/s respectively for the resonance at a wavelength of 1550nm. Second order grating presented the strongest response. We believe this is a feature of the femtosecond-inscribed FBGs. The refractive index modulation along the fibre after femtosecond inscription is likely to be a sequence of relatively sharp peaks, different to the smooth sinusoidal profile of the UV-inscribed gratings. This is caused by the geometry of femtosecond inscription process and further emphasised by its nonlinear nature. Hence, femto-inscribed FBGs have more pronounced higher order resonances compared to the UV-written ones. On the other hand, a relatively large size of the laser spot in the fibre core causes overlapping of pitches in fundamental order gratings, thus reducing the quality of the first order grating. The FWHM linewidth of the grating resonance typically ranged between 0.1nm and 0.2nm, out of band losses were below 1.5dB. Gratings showed significant birefringence due to the asymmetry induced in the writing process.

2:30 PM MM2.4

Laser Shot Peening and Surface Morphology. **A. A. Bugayev**¹,

Mool C. Gupta¹, S. Levesque², J. Orr² and R. Payne²; ¹Applied Research Center, Old Dominion University, Newport News, Virginia; ²Framatome ANP, Inc., Lynchburg, Virginia.

The results on laser shot peening and its characterization for Inconel 600 are presented. A brief description of the physical processes involved in the pressure generation mechanism is given. Using an X-ray diffraction technique we show that the residual compressive stresses can be successfully induced in Inconel 600 with parameters of laser peening, which are close to that of 316L stainless steel. The on-line monitoring system involving the acoustic pulse measurements and auto focusing of laser beam is described for quality control of

laser shot peening process. The system was designed to provide delivery of controlled laser energy to metal surface for most effective laser peening conditions. We found that sample scanning during laser processing results in a system of column-like microstructure, which is tilted in direction of scanning. The features of optical properties of tilted microstructure are described. We revealed that the base material injected into the confining water due to laser ablation is transformed to spherical nanoparticles with diameter of 60 nm.

3:15 PM *MM2.5

Direct-Write Micro- and Nanostructuring with Femtosecond Lasers. **Boris N. Chichkov**, Laser Zentrum Hannover e.V., Hannover, Germany.

I will report on our recent progress in direct-write femtosecond laser material processing technologies, nanostructuring, fabrication of photonic devices, and biomedical components. Special attention will be given to sub-wavelength microstructuring of metals and two-photon polymerization (2PP) technique. Formation of microbumps and nanojets on thin metal films under single pulse laser irradiation will be discussed. 2PP of different photosensitive materials and resolution limits of this technology will be analyzed. Numerous applications of this technology for the fabrication of 3d structures, waveguides, photonic crystals, etc. will be presented.

3:45 PM *MM2.6

A Bunch of Excellent Properties for Application of Solid-State Laser Radiation. **R. Poprawe**¹, P. Loosen² and D.

Hoffmann³; ¹Lehrstuhl für Lasertechnik der Rheinisch-Westfälischen Technischen Hochschule Aachen, Fraunhofer-Institut für Lasertechnik, Aachen, Germany; ²Lehrstuhl für Technologie optischer Systeme, Fraunhofer-Institut für Lasertechnik, Aachen, Germany; ³Fraunhofer-Institute für Lasertechnik, Aachen.

Diode laser pumped solid-state lasers such as rod, disk, slab or fiber lasers are on the way to govern as new systems developed in the last years the applications of laser radiation for various application in innovative technologies. The laser radiation is used either as a tool for materials processing or as a source for radiation generation beside of applications in life science, environment, medicine, analytics and others. The state-of-the-art of diode laser pumped solid state lasers is described highlighting beam quality, efficiency, mean time between failure and compactness in order to discuss new laser radiation governed processes aimed for rapid manufacturing by direct metal deposition, surface cleaning, surface polishing and generation of EUV radiation in relation to simulation and modeling.

4:15 PM MM2.7

Bimodal Size Distribution of Gold Nanoparticles in Aqueous Solution during Pulsed Laser Irradiation. **Susumu Inasawa** and Yukio Yamaguchi; The University of Tokyo, Tokyo, Japan.

Size distribution of gold nanoparticles in aqueous solution during pulsed laser irradiation is observed by transmission electron microscopy (TEM). Interestingly, our results show that size distribution of gold nanoparticles, which is initially mono-modal distribution, changes into bimodal one with pulsed laser irradiation. Irradiation of Nd:YAG laser pulses of wavelength 355 nm, pulse width 30 ps, to chemically prepared gold nanoparticles in water is carried out. Whole solution of gold nanoparticles in quartz cuvette is irradiated by each laser pulse. The mean length and aspect ratio of initial gold nanoparticles are 37 nm, 1.28 respectively, with standard deviation 5 nm. With the increase of number of pulses, number of larger particles decreases while smaller particles less than 10 nm diameter increases. There is a "valley" in number of particles at about 15 nm in size distribution, which makes size distribution bimodal. This "valley" could be interpreted that gold nanoparticles with diameter of 15 nm are unstable in this system for a certain reason. It is well known that noble metal nanoparticles, such as gold and silver, can be reduced their sizes by the irradiation of intense laser pulses. Two kinetic processes have been proposed to explain laser-induced size reduction phenomenon, one is the vaporization of particles (Takami et al., J. Phys. Chem. B, 1999, 103, 1226) and another is coulomb explosion of particles caused by the electron ejection out of the particles (Kamat et al., J. Phys. Chem. B, 1998, 102, 3123). Both could explain the size reduction phenomenon itself, but neither of them could precisely explain the change of size distribution from mono-modal to bimodal, for lack of information on the formation kinetics of smaller particles. Precise observation of time evolution of size distribution of gold nanoparticles during pulsed laser irradiation tells us the mechanism of smaller particles formation. We are studying size reduction kinetics, formation kinetics of smaller particles and the origin of bimodal size distribution of gold nanoparticles under laser irradiation.

4:30 PM MM2.8

Nanometer Material Processing Using NSOM-delivered

Femtosecond Laser Pulses. Chen-Hsiung Cheng and Ming Li; Panasonic Boston Laboratory, Cambridge, Massachusetts.

Nanometer-scale surface topology modification has been demonstrated using NSOM (near-field scanning optical microscope) delivered femto-second pulses. The ablation laser has a pulse width of 150 femto-second and wavelength of 387-nm. The laser pulses are coupled into the free end of a multimode optical fiber that a nanometer-size NSOM probe was fabricated on the other end with small orifice. The transmitted laser pulses from the probe orifice illuminates and machines the substrate surface when the probe is in contact with the substrate. The produced feature on Silicon surface is as least 200-nm deep with hole diameter around 200-nm. Near-field coupling of the laser has the potential to achieve ablation of feature size less than diffraction limit. Using NSOM delivery method also allows us to take advantage of nanometer metrology in precision surface ablation or other type of surface modification. The ability of monitoring surface topology of substrate in real time enables us to accomplish the in-situ surface processing. We have demonstrated the technique of drilling 200-nm air holes on a pre-formed 600-nm wide wave guide. This method can be used to fabricate one-dimensional photonic crystal on a waveguide in ambient environment. The experiment design and performance evaluation will be discussed. Acknowledgements This work was performed under the Femtosecond Technology Research Association (FESTA), which is supported by the New Energy and Industrial Technology Development Organization (NEDO) in Japan.

4:45 PM MM2.9

Transmission Electron Microscopy Studies of Damage and Modification Induced by Femtosecond Lasers.

Yousuf N. Picard^{1,2}, Qiang Feng¹, Tresa M. Pollock¹ and Steven M. Yalisove^{1,2}; ¹Materials Science & Engineering, Univ. of Michigan–Ann Arbor, Ann Arbor, Michigan; ²Center for Ultrafast Optical Science, Univ. of Michigan–Ann Arbor, Ann Arbor, Michigan.

Recent direct observation of the modifications induced by femtosecond laser irradiation using transmission electron microscopy have yielded greater insight into the extent of amorphization in single crystal materials (silicon, InP, and GaAs). When irradiating at fluences near the critical threshold for ablation, it is not clear if any amorphization by femtosecond lasers is thermally or mechanically induced. We and other researchers have observed evidence for both. To better elucidate the amorphization mechanism, we have conducted near and above ablation threshold machining of pre-thinned single crystal silicon and metal specimens. The 100 nm thin foils are more susceptible to pressure-induced mechanical deformation and localized melting due to the restricted thermal conduction paths. High and low resolution transmission electron microscopy is conducted to assess the extent of amorphization in the materials and the presence and extent of dislocations.

SESSION MM3: Poster Session
Chair: Alberto Pique
Tuesday Evening, November 30, 2004
8:00 PM
Exhibition Hall D (Hynes)

MM3.1

ZnO nanowire synthesis using femtosecond laser. Lei Luo, Yanfeng Zhang, Richard Russo, Liwei Lin and Samuel S. Mao; Lawrence Berkeley National Laboratory and Department of Mechanical Engineering, University of California at Berkeley, Berkeley, California.

One-dimensional semiconductor nanostructures have attracted much recent interests for their potential in realizing nanoscale electronic, optical, and mechanical devices. ZnO nanowires, due to their unique properties such as wide band gap and large piezoelectric coefficient, are under intensive investigations for applications such as lasers, sensors and actuators. While thermal evaporation remains a standard approach for growing ZnO nanowires, no alternative method for making high quality ZnO nanowires has been reported. Using high power femtosecond laser pulses, we successfully synthesized ZnO nanowires based on vapor-liquid-solid process. In our experiments, a ZnO target was ablated using the femtosecond laser pulse, whereas a silicon substrate was placed a few mm away from the target. Gold catalysts, which promote the formation of Zn-Au alloy droplets that help initialize one-dimensional growth, were prepared by thermal evaporation onto the substrate. After nanowire growth, the sample was imaged using a scanning electron microscope. The ZnO nanowires are approximately 100 nm in diameter and several microns in length. In addition to growth and structural characterizations, we will discuss the optical properties of ZnO nanowires. We believe that our demonstration opens up a new route for the application of ultrafast lasers for semiconductor nanomaterial fabrication.

MM3.2

Catalyst-free Growth of Oxide Semiconductor Nanowires by Pulsed Laser Deposition. Yanfeng Zhang, Richard Russo and Samuel S. Mao; Lawrence Berkeley National Laboratory, Berkeley, California.

Owing to their potential in nanoscale device applications as well as in fundamental research, one-dimensional oxide semiconductor nanowires have attracted substantial interest. In this presentation, we report our recent success in synthesizing oxide semiconductor nanowires using femtosecond pulsed laser deposition (PLD) approach. PLD is a widely used method for fabricating oxide thin films; it has also been applied to synthesize different types of nanowires based on vapor-liquid-solid (VLS) one-dimensional growth model. A standard VLS process requires the presence of metallic catalysts such as Au, which promote the formation of alloy droplets that help initialize one-dimensional growth. Nevertheless, our recent oxide nanowire growth experiments suggest that catalyst-based VLS model may not be the only mechanism for nanowire growth. By using femtosecond laser pulses, we successfully synthesized different oxide semiconductor nanowires on silicon substrate without any metallic catalyst. We will discuss the influence of growth parameters, such as the growth temperature, oxygen partial pressure, and laser power on nanowire growth. We will propose an alternative three stage vapor-solid growth model for oxide semiconductor nanowire growth.

MM3.3

Carrier Dynamics in Sr₂RuO₄. Prasenjit Guptasarma¹, Marshall Onellion², Michael Schneider², Steve Sendelbach² and Greg Taft³; ¹Physics, University of Wisconsin- Milwaukee, Milwaukee, Wisconsin; ²Physics, University of Wisconsin- Madison, Madison, Wisconsin; ³Physics, University of Wisconsin- Stevens Point, Stevens Point, Wisconsin.

Using femtosecond pump-probe methods to measure the transient optical reflectivity, we report on the carrier dynamics of Sr₂RuO₄ in the normal state, from 7K to room temperature. At room temperature, there is a single fast thermalization (time constant ~ 70 fs) and a single fast relaxation (~ 120 fs). As the temperature decreases, there are two qualitative changes. First, a second, slower, component to the relaxation develops and becomes dominant. The slower component of the relaxation is as slow as > 500 ps time constant at 7K. Also, the increase in reflectivity, usually ascribed to carrier thermalization, develops two components. The second increase becomes slower (time constant ~ 10 ps at 7K) and larger at low temperatures. The data are in direct conflict with the idea, arising from electrical resistivity measurements, that Sr₂RuO₄ is an anisotropic three dimensional Fermi liquid below ~ 25K. [1,2] Sr₂RuO₄ is also qualitatively different from the cuprates in that both the thermalization and the relaxation develop two components at low temperatures. We discuss implications of the data for modeling carrier-carrier interactions in Sr₂RuO₄. 1. Y. Maeno et al., J. Phys. Soc. Jpn. 66 (1997) 1405. 2. A.W. Tyler et al., Phys. Rev. B 58 (1998) R10107.

MM3.4

Excited States Dynamics of Silicon Clusters and Nanowires in Intense Laser Fields Studied Quantum Mechanically. Anna Maria Mazzone and M. Bianconi; IMM, Sezione di Bologna, CNR, Bologna, Italy.

Dynamical processes at the molecular level occur on ultrafast time scales and are often associated with structural as well as electronic changes. In poly-atomic structures these excitations result in the rapid mixing of vibrational and electronic motions. Though this non-adiabatic coupling is a key step in photochemical and photobiological processes, the notion of distinct and readily observable electronic and vibrational states becomes obscure. Therefore particular attention has been given in theory and experiments to the smallest molecular systems and intense, femtosecond laser irradiation has been adopted as a powerful experimental tool to improve the current understanding of excited ion-electron dynamics by breaking the linear regime usually dealt with by perturbative expansions. The purpose of this work is to extend this study to structures of current interest in nanoscience and it centres on crystalline silicon clusters and atomic wires under femtosecond laser pumping. The study is performed within a real-space, real-time implementation of the Density Functional theory. The results demonstrate that it is possible to obtain detailed insights into ultrafast non-adiabatic processes as well as on their dependence on the irradiated structure.

MM3.5

Transient Photoconductivity of vertically aligned crystalline TiO₂ nanorod arrays. Abul K. Azad², W.-L. Zhang², S.-H. Li¹ and Yiping Zhao¹; ¹Physics and Astronomy, University of Georgia,

Athens, Georgia; ²School of Electrical and Computer Engineering, Oklahoma State University, Stillwater, Oklahoma.

Dye-sensitized solar cells based on nanocrystalline TiO₂ films have attracted enormous interest because of their high photo-current conversion efficiencies [1]. The transient photoconductivity of sintered TiO₂ nanoparticles has been characterized at liquid nitrogen temperature with a relative thick nanoparticle layer (3 μm) [2]. However, the operation of a solar cell usually is at room temperature or higher. Here, using incandescent halogen white light as the photoexcitation source, we have studied the photoconductivity of vertically aligned TiO₂ nanorod arrays at room temperature. The TiO₂ nanorod sample is a 0.42-μm-thick film deposited on 1-mm-thick quartz substrate by glancing angle deposition [3]. The sample has been annealed at 748C for 4 hrs in ambient, and the XRD shows that there is only Anatase phase TiO₂. The TiO₂ sample was sensitized in ethanol solution of Ru-535 in order to broaden its absorption bandwidth. Frequency dependent conductivity, power absorption and optical dispersion are measured by terahertz differential time-domain spectroscopy over the frequency range of 0.4 - 2.5 THz. The measured complex conductivity shows good agreement with the Smith model. Acknowledgement: This work is partially supported by the Oklahoma EPSCoR and NSF. 1. Nazeeruddin, M. K.; Pechy, P.; Renouard, T.; Zakeeruddin, S. M.; Baker, R. H.; Gratzel, M. J. *Am. Chem. Soc.* 2000, 123, 1613. 2. Gordon, T. M.; Beard, M. C.; Schmittenmear, C. A. *J. Phys. Chem. B* 2002, 106, 11716. 3. Zhao, Y. -P.; Ye, D. -X.; Wang, Pei-I.; Wang, G. -C.; Lu, T. -M. *International Journal of Nanoscience* 2002, 1, 87.

SESSION MM4:

Chair: Ming Li

Wednesday Morning, December 1, 2004

Room 303 (Hynes)

8:30 AM MM4.1

Producing ultrashort Terahertz to UV photons at high repetition rates for research into materials. George R. Neil, C. Behre, S. V. Benson, G. Biallas, J. Boyce, D. Douglas, H. F. Dylla, R. Evans, A. Grippo, D. Gruber, J. Gubeli, C. Hernandez-Garcia, K. Jordan, M. J. Kelley, L. Meringa, J. Mammosser, J. Preble, M. Shinn, T. Siggins, R. Walker, G. P. Williams, B. Yunn and S. Zhang; Jefferson Lab, Newport News, Virginia.

A new THz/IR/UV source at Jefferson Lab is the first of a new generation of light sources based on an Energy-Recovered, (superconducting) Linac (ERL). The machine has a 150 MeV electron beam and an average current of 10 mA in high repetition rate bunches that are in the hundred femtosecond regime. These electron bunches pass through a magnetic chicane and therefore emit synchrotron radiation. For wavelengths longer than the electron bunch the electrons radiate coherently a broadband THz half cycle pulse whose average brightness is > 5 orders of magnitude higher than synchrotron IR sources. Previous measurements showed 20 W of average power extracted[1]. The new facility offers simultaneous synchrotron light from the visible through the FIR along with broadband THz production of 100 fs pulses with >200 W of average power (see G. P. Williams, this conference). The FELs also provide record-breaking power [2]: up to 10 kW of average power in the IR from 1 to 14 microns in 400 fs pulses at up to 74.85 MHz repetition rates and similar pulses of 300-1000 nm light at up to 3 kW of average power from the UV FEL. These ultrashort pulses are ideal for maximizing the interaction with material surfaces. The optical beams are Gaussian with nearly perfect beam quality. See www.jlab.org/FEL for details of the operating characteristics; a wide variety of pulse train configurations are feasible from 10 microseconds long at high repetition rates to continuous operation. The THz and IR system has been commissioned. The UV system is to follow in 2005. The light is transported to User laboratories for basic and applied research. Additional lasers synchronized to the FEL are also available. Past activities have included production on carbon nanotubes, studies of vibrational relaxation of interstitial hydrogen in silicon, pulsed laser vapor deposition, nitriding of metals, and energy flow in proteins. This paper will present the status of the system and discuss some of the opportunities provided by this unique light source for modifying and studying materials. [1] Carr, et al., *Nature* 420, 153-156 (2002). [2] Neil, et al., *Phys. Rev. Lett.* 84, 662-665 (2000). This work was supported by the Office of Naval Research, the Joint Technology Office, the Commonwealth of Virginia, the Air Force Research Laboratory, and by DOE Contract DE-AC05-84ER40150.

8:45 AM *MM4.2

Resonant Infrared Free-Electron Laser Deposition of Crystalline Poly(tetrafluoroethylene). Michael R. Papantonakis^{1,2}, Michelle L. Baltz-Knorr^{1,2}, Stephen L. Johnson^{1,2}, Kenneth E. Schriver^{1,2} and Richard F. Haglund^{1,2}; ¹Physics and

Astronomy, Vanderbilt University, Nashville, Tennessee; ²W. M. Keck Foundation Free Electron Laser Center, Vanderbilt University, Nashville, Tennessee.

Since the laser ablation and deposition of intact poly(ethylene glycol) (molecular weight 1.5 kDa) was first reported in 2001, it has been demonstrated that a number of other polymers can be ejected substantially intact into the gas phase by resonant infrared excitation of bulk substrates using a picosecond micropulse free-electron laser (FEL). In most of this work, the ablated polymers were deposited for subsequent analysis on NaCl or Si substrates, then subjected to analysis by Fourier-transform infrared spectrometry, size-selective chromatography and mass spectrometry. Preservation of local electronic structure and of the polymer mass distribution were noted in most, though not all, cases of resonant excitation. Recent experiments with poly(tetrafluoroethylene) (PTFE) have demonstrated the even more interesting result that crystalline PTFE can be deposited by the same technique at substantially lower temperatures than can be achieved by either conventional ultraviolet pulsed laser deposition or femtosecond near-infrared (800 nm) laser ablation. The crystallinity was confirmed by X-ray photoelectron spectroscopy and X-ray diffraction; the smoothness of the films, observed by atomic-force microscopy and electron microscopy, was excellent. Ablation yields and thresholds were compared at wavelengths resonant with PTFE vibrational modes (4.2 and 8.26 μm) and off-resonance (7.1 μm). PTFE is a particularly interesting case because of its technological importance, and the fact that its insolubility means that other thin-film growth techniques, such as spin-coating, are not feasible. In order to more fully understand how picosecond resonant vibrational excitation can lead to intermolecular bond-breaking and vaporization of relatively large polymer units, we have characterized the FEL ablation plumes on and off resonance by pulsed laser shadowgraphy. The results show that the ejection of the polymer target material by off-resonance irradiation has a substantially longer incubation time before the formation of a plume compared to on-resonance irradiation (6-8 μs vs 4 μs). Given the high photon flux density characteristic of the FEL micropulses, this probably reflects the efficient vibrational pumping of the anharmonic resonant modes of the polymers in resonant excitation. Photographs of the PTFE ablation target show clear evidence of damage (carbonization) in off-resonance ablation, damage that is absent in the case of resonant FEL excitation. Taken together with the shadowgraphy data, this suggests that the resonant excitation has a non-thermal component, while the off-resonance laser irradiation proceeds thermally. This research was supported by grant FA9550-04-1-0045 from the DOD MFEL Program.

9:15 AM *MM4.3

High Power THz Generation from Sub-ps Bunches of Relativistic Electrons. Steve Benson, Dave Douglas, Joe Gubeli, Kevin Jordan, George Neil, Michelle Shinn, Shukui Zhang and Gwyn P. Williams; Jefferson Lab, Newport News, Virginia.

We describe a > 100 Watt broadband THz source that takes advantage of the relativistic enhancement of the radiation from accelerating electrons according to the formula assigned the name of Sir Joseph Larmor[1,2]. This is in contrast to the typical 1 milliwatt sources available in a laboratory. Specifically, for relativistic electrons the emission is enhanced by the fourth power of the increase in mass. Thus for 100 MeV electrons, for which the mass increases by a factor of 200, the enhancement is > 109. The experiments use a new generation of light source called an energy recovery linac (ERL) [3], in which bunches of electrons circulate once, but in which their energy is recovered. In such a machine the electron bunches can be very much shorter than those, say, in storage rings or synchrotrons. The Jefferson Lab facility operates in new limits of emission from relativistic particles involving both multiparticle coherence and near-field emission in which the velocity (Coulomb) term in the classical electro-dynamical theory becomes as important as the acceleration term (synchrotron radiation). The sub-picosecond pulses of light offer unique capabilities in 2 specific areas, namely time-resolved dynamics, and imaging. High resolution THz spectroscopy has recently revealed sharp vibrational modes for many materials including malignant tissue, proteins, DNA, pharmaceuticals and explosive materials. Energetically the THz range embraces superconducting bandgaps, and regions of intense interest in the understanding of systems in which correlated motions of electrons are important, such as colossal magneto-resistive and high-Tc materials. The very high power levels of the new source will allow non-linear effects to be observed as well as the creation of novel states of materials, including electric-field driven localization[4]. We will give examples of existing work in these areas and present opportunities afforded by the new source. 1. G.L. Carr, M.C. Martin, W.R. McKinney, K. Jordan, G.R. Neil and G.P. Williams "High Power THz Radiation from Relativistic Electrons", *Nature* 420 153-156 (2002). 2. G.P. Williams, "High Power THz Synchrotron Sources", *Phil Trans. R. Soc. Lond. A* 362 403 (2004). 3. G.R. Neil, et al. "Sustained Kilowatt Lasing in a Free-Electron Laser

with Same Cell Energy Recovery". Phys. Rev. Lett. 84, 662-665 (2000). 4. D.K. Campbell, S. Flach and Y.S. Kivshar, Physics Today, January 2004, p 43. Acknowledgements: This work at Jefferson Lab was supported the U.S. DoE and DoD. We are grateful to our colleagues at JLab, Berkeley and Brookhaven for critical support without which these experiments would not have been possible.

10:15 AM *MM4.4
Modifications and Color Markings in Glass by UV Laser Radiation. M. Talkenberg¹, G. Falkenberg² and M. Krauss³;

¹Lehrstuhl für Lasertechnik der Rheinisch-Westfälischen Technischen Hochschule Aachen, Aachen, Germany; ²Hamburger Synchrotronstrahlungslabor at Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany; ³Fraunhofer-Institut Silicaforschung, Würzburg, Germany.

Rare earth-, transition metal-, and metal doped soda lime-, BK7-, and photosensitive lithiumalumosilicate glass are irradiated by pulsed UV laser radiation (wavelength $\gamma_L=248$ to 355 nm, pulse length $t_p=10$ to 80 ns) as a function of repetition rate, pulse energy and overlap between successive pulses. The solid state laser radiation is focused by an optics into the volume and the excimer laser radiation is projected by a circular mask onto the surface with fluences below the removal-threshold. Depending on the chemical composition, the laser parameters and the processing parameters the laser radiation is absorbed by optically- and photochemically active polyvalent ions with conversion of the optical energy into fluorescence stimulation and ionisation as well as color center formation resulting in modifications and/or color markings in the UV-VIS. The absorption and concentration changes of the polyvalent ions are probed by optical spectroscopy, transient absorption spectroscopy, electron paramagnetic resonance and X-ray absorption near edge spectroscopy. The photochemical and -physical processes such as oxidation and reduction of polyvalent ions and/or color center generation underlying the modifications and color markings are discussed.

10:45 AM MM4.5
Three-Dimensional Micro- and Nano-Fabrication with Multiphoton Absorption. John T. Fourkas, Chemistry, Boston College, Chestnut Hill, Massachusetts.

Because the probability of multiphoton absorption depends nonlinearly upon the intensity of the laser employed, it is possible to localize photochemical and photophysical events within the focal volume of an ultrafast laser beam that passes through a microscope objective. By controlling the position of the focus in three dimensions it is possible to sculpt arbitrarily complex 3-D structures with resolution that is significantly better than would be expected from diffractive considerations. I will discuss some of the recent advances made by our team in the fabrication and replication of functional polymeric, metallic and hybrid structures.

11:00 AM MM4.6
Photonic Crystal Templates Obtained by Two-Photon Laser Lithography in Photoresist SU-8. Vyngantas Mizeikis, Kock Khuen Seet, Saulius Juodkazis, Vyngandas Jarutis and Hiroaki Misawa; Research Institute for Electronic Science, Hokkaido University, Sapporo, Japan.

The evolution of modern photonic technologies depends on the possibilities to obtain large-scale, three-dimensional photonic crystals cheaply and efficiently. Since existing planar semiconductor fabrication techniques can not fully satisfy these requirements, two-photon laser lithography of photoresists emerges as a strong alternative candidate. We report laser fabrication of 3D photonic crystals in photoresist SU-8 by two-photon lithography. Extended structures having woodpile and square spiral geometries were fabricated with submicrometric resolution; they exhibit negligible structural deformations due to shrinkage, show clear signatures of photonic stop-gaps at mid-infrared wavelengths (2.0-6.0 micrometer), and may be used as templates for infiltration by materials with a higher refractive index.

11:15 AM *MM4.7
Ultrafast Lasers and Nanowires: Growth, Mechanisms, and Properties. Samuel Mao, Lawrence Berkeley National Lab, Berkeley, California.

In this presentation, I will give an overview of recent progress on the applications of ultrafast lasers for the development of one-dimensional (1D) nanostructures - crystalline nanowires. Since the first report of nanowire synthesis using pulsed lasers approximately eight years ago, a variety of materials in the form of nanoscale wire structures have been realized. Two representative 1D nanostructured materials that will be covered in this presentation include Si and ZnO nanowires. While Si nanowires were among the first nanomaterials synthesized using pulsed laser-assisted process, it is only very recently that crystalline ZnO nanowires were grown using femtosecond laser pulses.

The underlying growth mechanisms for the formation of crystalline nanowire structures will be discussed, along with systematic measurements of physical properties of nanowires. The presentation will conclude by showing the potential of ultrafast laser synthesized nanowires as the building blocks for future nanoscale electronic and optical devices.

11:45 AM MM4.8
Waveguide Structures Written with FS-Laser Pulses above the Critical Self-Focusing Threshold in SF57 Glass.

Victor Diez¹, Jan Siegel¹, Fidel Vega², Jesus Armengol² and Javier Solis¹; ¹Instituto de Optica, CSIC, Madrid, Spain; ²Depto. Optica y Optometria, UPC, Terrasa, Spain.

Waveguide writing with fs-laser pulses in glasses can be hampered by the appearance of uncontrollable non-linear propagation phenomena. This occurs when the pulse power exceeds a certain threshold, P_{cr} , above which critical self-focusing takes place and leads to catastrophic damage. In a first approach, P_{cr} is inversely proportional to the linear and non-linear refractive indices, n_0 and n_2 , which makes the processing of materials with high n_0 and n_2 values to become a challenge. The aim of the present work is to analyze the behavior of one of these materials, Schott SF57 glass, when it is processed with 800 and 1260 nm, 100 fs laser pulses to produce waveguide structures. SF57 is a commercial glass essentially formed by SiO_2 and PbO_2 having a 40-50% cationic content of Pb and that shows large linear ($n_0=1.85$) and non-linear ($n_2\approx 10^{-18} m^2/W$) refractive indices. Two types of structures have been produced inside this glass, transversal (t-) and longitudinal (l-) ones, depending on whether the sample translation was perpendicular or parallel to the writing laser beam. In the t-structures, extremely elongated damaged zones are induced due to critical self-focusing. Off-center light guiding is observed nearby the damaged filaments indicating the formation of a refractive index increased region in the vicinity of this damaged zone. In the l-structures, the damaged zone shows a circular shape, corresponding to the focal volume of the critical self-focused writing beam. In this case, off-center light guiding is again observed nearby the damaged region. We attribute the formation of these off-center guiding regions, in both the l- and t- structures, to the high local pressure generated by the ultrafast temperature increase at constant volume that takes place in the focal volume of the critical self-focused beam. This pressure can be high enough to compress the material nearby giving rise to the observed permanent refractive index increase in the guiding region ($\Delta n > 5 \times 10^{-4}$). For both l- and t- structures, critical self focusing effects appear for pulse energies just above the structural modification threshold, and the size of the increased refractive index region is conditioned by the pulse energy and wavelength used; for instance, when writing at 1260 nm, the size of this latter region is not large enough to support a guided mode at 633 nm. As an alternative means to produce efficient waveguides with a controllable guiding zone size, the use of multiple structures has been investigated. The basic idea relies on writing several l-channels that do not individually support a guided mode. However, when they are written close enough to each other the resulting multiple structure effectively supports a guided mode with a gaussian profile, as it will be shown for a writing wavelength of 1260 nm.

SESSION MM5:

Chair: Alberto Pique

Wednesday Afternoon, December 1, 2004

Room 303 (Hynes)

1:30 PM MM5.1
Investigation of Femtosecond Laser Irradiation on Fused Silica HF Etching Selectivity. Yves Bellouard¹, Ali A. Said², Mark Dugan² and Philippe Bado²; ¹CAT, Rensselaer Polytechnic Institute, Troy, New York; ²Translume, Ann Arbor, Michigan.

Femtosecond laser irradiation has various noticeable effects on fused silica. It can locally increase the index of refraction or modify the material chemical selectivity. Regions that have been exposed to the laser are etched several times faster than unexposed regions. Various observations reported in the literature seem to show that those effects are possibly related to a combination of structural changes and the presence of internal stress. However, a detailed analysis of the contribution of both effects is still lacking. In this paper, we present systematic SEM-based investigations performed on fused silica substrate (a-SiO₂). Line-patterns were first scanned on the substrate using a femtosecond laser (Ti-Sapphire, repetition rate 250kHz, pulse width 100fs) and then etched in a low-concentration HF solution. The effects of various laser parameters such that the power and the scanning speed are analyzed and we show further evidence of an interface between two different etching regimes.

1:45 PM *MM5.2

Diagnosis and Modelling of Nonlinear Dynamics in Laser Cutting, Welding and Drilling. Wolfgang Schulz, Fraunhofer Institut Lasertechnik, Aachen, Germany.

Laser cutting and welding are well established industrial applications. To maintain productivity and to guarantee product quality the industry tries to introduce monitoring and control systems. The long term goal is the autonomous laser machine. Signal assessment is advancing by monitoring and simulation of the dynamical processes. Applying the advanced results about diagnosis and modelling broadens the potentials to cope with productivity and quality features in drilling, trepanning and fine cutting. As result, in cutting two mechanisms for the formation of adherent dross are revealed theoretically, identified by the monitoring system and can be avoided by modulation of the laser beam power. In welding, the dynamic model predicts the formation of pores sets in or is suspended depending on modulation frequency for the laser power. In drilling the mechanisms governing the maximum depth of the drilled hole – still showing efficient melt removal – are identified experimentally and can be related to the processing parameters theoretically.

2:15 PM *MM5.3

Micro- and Nano-Structured Optical Fibers - Artificial Media for Amplification of Light. Andreas Liem¹, A. Tunnermann^{1,2} and H. Zellmer¹; ¹Friedrich Schiller University Jena, Institute of Applied Physics, Jena, Germany; ²Fraunhofer Institute for Applied Optics and Precision Engineering, Jena, Germany.

Rare-earth-doped fibers have established themselves as an attractive and power scalable solid-state laser concept. In continuous-wave operation output powers approaching the kW-regime with diffraction-limited beam quality have been demonstrated. Ytterbium-doped fibers provide also several key advantages regarding the amplification of short optical pulses. The gain bandwidth supports pulses as short as ~ 30 fs, the saturation fluence allows for the generation of high energy pulses and high optical pumping efficiencies (up to 80%) make an ytterbium-doped fiber amplifier to an outstanding gain medium. However, power and energy scaling of single-mode fiber amplifiers is restricted due to nonlinear pulse distortions. Novel fiber geometries based on micro- and nano-structured fiber-cores allow for a significant scaling of power in continuous and pulse operation. The optical properties of these artificial media are discussed, scaling laws are presented.

3:15 PM MM5.4

Ultrafast generation and detection of shear and longitudinal acoustic phonons in GaAs. David Howard Hurley¹, Osamu

Matsuda², Oliver B. Wright² and Vitali E. Gusev³; ¹Physics Department, INEEL, Idaho Falls, Idaho; ²Department of Applied Physics, Hokkaido University, Sapporo, Japan; ³Universite du Maine, Le Mans, France.

Gaining a better understanding of the way in which electrons interact with the vibrational modes of a lattice (phonons) is of fundamental importance for controlling quantum processes in semiconductors. For example, devices that rely on the coherent control of excitons are limited by decay, which is mediated by electron-phonon coupling. Coupling between electrons and phonons is described in terms of deformation potentials that relate shifts in the energy of band extrema to strain. For semiconductors that lack inversion symmetry, electron-phonon coupling can also arise through changes in electric polarization caused by electron motion, that in turn give rise to strain via the piezoelectric effect. Ultrafast optical techniques provide an opportune means to probe directly the interaction of photoexcited electrons and phonons. In this study we make use of ultrashort optical pulses to generate and detect acoustic phonon pulses in (411) GaAs. Unlike previous studies involving ultrafast phonon generation in semiconductor surfaces cut in high-symmetry orientations, the reduced symmetry of the (411) GaAs surface allows shear as well as longitudinal phonon generation. The role of anisotropy in relation to phonon generation through thermoelasticity, deformation potential coupling, and piezoelectric effects caused by screening of built-in electric fields by photoexcited carriers is considered in detail. In addition, the pump power dependences of the various generation mechanisms are discussed.

3:30 PM *MM5.5

Fundamental Mechanisms and Applications of Ultrafast Laser Ablation of Solids. Michel Meunier¹, Patrick Lorazo¹, Laurent J. Lewis², A. V. Kabashin¹, J.-P. Sylvestre¹ and E. Sacher¹; ¹Laser Processing Lab, Department of Engineering Physics, Ecole Polytechnique de Montreal, Montreal, Quebec, Canada; ²Department of Physics, University of Montreal, Montreal, Quebec, Canada.

Recent works on the fundamental mechanisms of ultrafast laser interaction with solids and applications of fs laser ablation towards

the fabrication of nanomaterials will be reviewed. In the first subject, we will address the physical pathways to laser-induced matter removal from surfaces by studying the routes to ablation of silicon under UV pulses with duration ranging from 500 fs to 100 ps. The detailed electronic and atomic dynamics are described by the Monte Carlo and molecular-dynamics methods, respectively, and the density, temperature, and pressure of the system are computed during heating, cooling, and expansion. By mapping these results onto the phase diagram of silicon, the complete thermodynamic evolution of the system can be obtained. Under ultrashort pulses ($= 1$ ps), isochoric heating and rapid adiabatic cooling of the system below the binodal line provide a natural pathway to *phase explosion* while under "long" ($= 10$ ps) pulses, matter removal is, rather, driven by a *fragmentation* process within the supercritically expanding matter [Phys. Rev. Lett. **91**, 225502 (2003)]. In the second subject, we consider the fs laser ablation of metal targets in aqueous solutions and the resultant formation of colloidal metal nanoparticles. The distribution of ablated particles give evidence for two different mechanisms of material ablation in liquid environments. The first mechanism, associated with thermal-free fs ablation, leads to very small (2.5 -10 nm), almost monodisperse, gold colloids. The second, attributed to the subsequent plasma-induced heating and ablation of the target, gives rise to much larger particles, with broad size distributions [J. Appl. Phys. **94**, 7941, (2003)]. Chemically active products (e.g. cyclodextrins, etc.) are also used to surround and stabilize the particles on formation, permitting the fabrication of contamination-free, almost monodisperse, 2-3 nm colloidal particles, whose surfaces could be modified for specific biochemical interactions [J. Phys. Chem. B, **107**, 4527 (2003)].