SYMPOSIUM Q

Femtosecond Materials Science and Technology

April 16 – 18, 2001

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* Invited paper

TUTORIAL

ST Q: FEMTOSECOND TECHNIQUES FOR MATERIALS SCIENTISTS
Monday, April 16, 2001
1:00 p.m. - 5:00 p.m.
Salon 14/15 (Marriott)

- Linear and nonlinear propagation of light
  - Propagation of electromagnetic waves in dense media
  - Dielectric function
  - Lorentz equations, Drude model
  - Pulse dispersion
  - Nonlinear response
  - Second harmonic generation and inversion symmetry†
  - Self phase modulation and self- focusing
  - Continuum generation
- Femtosecond measurements
  - Pump-probe technique
  - Dispersion compensation techniques†
  - Representation of pulses; Wigner representation
  - Temporal characterization of pulses
  - Joint time-frequency measurements†
  - Frequency-resolved optical gating
  - Limits of frequency and time resolution†

†Work-sheet format

Instructor:
Eric Mazur, Harvard University, Cambridge, MA

SESSION Q1: MATERIAL INTERACTIONS, MECHANISMS, AND MODELING
Chairs: Eric Mazur and S. K. Sundaram
Tuesday Morning, April 17, 2001
Franciscan III (Argent)

Professor Nicolaas Bloembergen, our opening speaker, was awarded the 1981 Nobel Laureate in Physics for his contribution to the development of laser spectroscopy.

8:30 AM *Q1.1
FEMTOSECOND SCIENCE AND MATERIAL INTERACTIONS. N. Bloembergen, Optical Sciences Ctr, Univ of Arizona, Tucson, AZ.

The development of femtosecond pulse generation during the past quarter century is briefly reviewed. With strongly focused short pulses of only nanojoule energy content it is possible to create sub-micron damage spots in transparent materials by creating a plasma strongly localized in space and time. Heat damage to the surrounding material is minimized. With this technique optical wave guides, junctions and Bragg reflectors can be constructed, In medical applications the reduction of thermal damage to surrounding tissue is also essential. The technique of chirped pulse amplification can yield femtosceond pulses with energy content from one millijoule to one joule. When such pulses are focused, power flux densities exceeding watts/cm are obtainable where the light field amplitudes exceeds the Coulomb field responsible for binding of valence electrons by several orders of magnitude. The physics of very dense relativistic plasmas with large temporal and spatial gradients can be studied experimentally.

9:00 AM Q1.2
ELECTRON STRUCTURE AND ELECTRON DYNAMICS AT III-V SEMICONDUCTOR SURFACES. Michael A. Grishin, Henrik S. Karlsson, Martin Mansson, Ulf O. Karlsson, Dept of Materials Physics, Royal Institute of Technology, Stockholm, SWEDEN.

With the evolution of high-speed electronics, the understanding of electron and hole dynamics and scattering at surfaces and at interfaces has become of prime importance. These phenomena can be investigated by pump-and-probe photoemission based upon ultra-short pulsed laser system. Utilizing a regenaratively amplified titanium:sapphire laser system, a time- and angle-resolved photoelectron spectrometer has been developed. The system is tunable between 720 and 950 nm and produces 600 mJ pulses at a rate of 1 kHz and the high peak power is used to create UV light at 9.32 eV by cascaded frequency doubling and tripling [1]. In addition a new multi channel detector has been developed which significantly increases the counting rate. This detector consists of a 32-channel detector mounted on a 8"-ConFlat flange with a chevron-mounted pair of 40-mm-diameter micro-channel plates in front of a pixel array. Pump-and-probe photoemission spectra from different III-V semiconductors and surfaces will be presented. By changing the polarization of the photoemitting probe pulse, different states in the conduction band with widely different life-times can be observed. It is also shown that the surface electron dynamic depends strongly on both the surface orientation and the material. The results from laser-based photoemission will be compared to results obtained with conventional photoelectron spectroscopy and scanning tunneling microscopy.
[1] H.S. Karlsson, G. Chiaia and U.O. Karlsson, Rev. Sci. Instrum. 67, 3610 (1997).

9:15 AM Q1.3
THE ROLE OF ELECTRONIC EXCITATIONS IN INITIATION OF CHEMISTRY IN MOLECULAR SOLIDS. Maija M. Kuklja, Department of Electrical and Computer Engineering, Michigan Technological University, Houghton, MI.

We have studied initiation of chemistry process in high explosive crystals from a solid-state physics viewpoint. In particular, we were looking for the relationship between the defect-induced deformation of the electronic structure of solids, electronic excitations, and fast chemical reactions under shock conditions. The combined theoretical and experimental study is performed. Band structure calculations by means of Hartree-Fock method with correlation corrections were done to model an effect of the strong compression induced by a shock/impact wave on crystals with and without edge dislocations. Based on the obtained results, an excitonic mechanism of the earliest stages for initiation of high explosive solids is suggested with an application to RDX [$C_3H_6N_6O_6$]. Experimental tests of this mechanism for AgN_3 decomposition controlled by the dislocation density were worked out. The use of pulse radiolysis techniques allows us to observe pre-explosion modifications in properties and behavior of the solids. Measurements of the pre-explosion conductivity and pre-explosion luminescence lead us to the model of heavy metal azides decomposition chain reaction. We found that electronic excitations promoted by the lattice defects is a candidate mechanism for contributing to initiation and detonation phenomena in practical explosive solids. This fact brings essentially new possibilities to apply the large solid state physics experience to the femtosecond technology and physics of high explosives also for a discovery of a very new perspective for the development of the modern initiation and detonation theory. Several important applications of the suggested mechanisms are discussed. The use of the methods governing the properties of the materials by the controlled creation of lattice defects and deformations, which are well studied in solid state physics, seems to be a very perspective tool for successful solution of safety, aging, and handling of explosives issues.

9:30 AM *Q1.4
SILICON NANOSTRUCTURES VIA INTENSE ULTRAFAST ELECTRONIC EXCITATION. Alex Hamza, Mike Newman, Thomas Schenkel, Howard Lee, Joe McDonald, Dieter Schneider, University of California, Lawrence Livermore National Laboratory, Livermore, CA.

Due to the indirect nature of its band gap, bulk silicon is typically a poor photon emitter upon external excitation. However, as the crystal size approaches nanometer scales, the band gap widens due to quantum confinement and may become direct allowing for more efficient photon emission. Phase transformations induced by intense, ultrafast electronic excitation from slow, highly charged ions have produced nanometer-sized structures in silicon. Beams of highly charged ions of various charge state from 20 to 69 and various kinetic energies from 5 to 14 keV times charge have been utilized to induce this phase transformation in clean, silicon surfaces. The new phase is characterized by ex situ photoluminescence from the irradiated area after excitation with laser wavelengths from 379 - 514 nm. Photoluminescence spectra from the exposed areas show emission centered at ~540 nm. This is consistent with emission observed from 1-2 nm silicon nanocrystals. A series of sharp lines at 565, 555, and 548 nm are present in the photoluminescence spectrum from areas exposed to Xe44 which are characteristic of an excitonic series in nanometer-sized material.

10:15 AM *Q1.5
C.V. Shank, Lawrence Berkeley National Lab, Berkeley, CA.

(ABSTRACT NOT AVAILABLE)

10:45 AM Q1.6
ULTRAFAST OPTICAL STUDIES OF NONEQUILIBRIUM THERMOELASTICITY IN METALS. James B. Spicer, Johns Hopkins University, Department of MS&E, Baltimore, MD; Christopher J.K. Richardson, University of Maryland, Laboratory for Physical Sciences, College Park, MD.

A theoretical description is presented for the thermoelastic transients in a solid metal following the absorption of a sub-picosecond duration laser pulse. This description is shown to agree with experimental observations on time scales ranging from a few picoseconds to a few hundred picoseconds using material properties that are consistent with accepted values. In various studies, material property values are adjusted for ultrafast measurements to produce agreement with experimental observations. Often, these values deviate significantly from bulk values and the structure of the surface material is cited as being the cause for the observed deviation. In this work, the foundation of the material description is a non-linear, two-temperature thermal diffusion theory that models the electrons and phonons as coupled systems. Absorption of the optical energy occurs directly via interactions with electrons that transfer the energy to the lattice through electron-phonon collisions. Experimental results from an electrochemically polished aluminum single-crystal are compared to models based-on the nonlinear thermoelastic theory presented here and are also compared to conventional models that only consider changes in the temperature of the sample. By accounting for the thermoelastic response of the material, results are found to be more consistent with accepted values for material properties. Direct comparisons between experimental and calculated results illustrate the improvement in the agreement for both the short-duration and long-duration transients when the thermoelastic description is used. The improved modeling of experimental observations provides confidence in the measurement and in the determination of thermal and elastic properties of thin films for picosecond duration transients as well as for material properties that govern equilibrium or long-time duration events.

11:00 AM *Q1.7
PHYSICS OF ULTRASHORT LASER PULSE INTERACTION WITH DIELECTRICS AND METALS. M.D. Feit, A.M. Komashko, A.M. Rubenchik, Lawrence Livermore National Laboratory, Livermore, CA.

Intense laser pulses of duration shorter than the electron-phonon inelastic scattering time lead to high deposited energy density in a thin surface layer in both dielectric and metallic materials before significant thermal conduction or hydrodynamic motion can occur. This layer can be ablated without significant energy loss to nonablated material, thus minimizing collateral damage for material processing. In biomedical applications, the definition of collateral damage is more subtle and the effect of short duration shock waves must be determined.
In wide bandgap dielectrics, high order multiphoton absorption provides free charge carriers for subsequent dielectric breakdown. Plasma mirrors can be created by such breakdown. Metals have conducting electrons from the start, but a careful treatment of the change of laser coupling during even ultrashort pulses is necessary for quantitative understanding. Theoretical descriptions of these processes and modeling of applications will be described.
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

11:30 AM Q1.8
MOLECULAR DYNAMICS STUDIES OF THE KINETICS OF PHASE CHANGES IN NANOPARTICLES: CRYSTAL NUCLEATION OF (RbCl)108 PARTICLE AT 600K, 550K, AND 500K. Haishan Deng, Department of Chemistry, Nanjing Normal University, Nanjing, PR CHINA; Jinfan Huang, Department of Chemistry, University of Michigan, Ann Arbor, MI.

Molecular dynamics computer simulation based on the Born-Mayer-Huggins potential function have been employed to study the crystal nucleation of (RbCl)108 particles. The melting point of the (RbCl)108 particle was obtained to be ~880K. When a completely melted particle was cooled down nucleation and crystal growth into the NaCl type structure crystal was observed at ~600K. Quenching a melted (RbCl)108 particle from 900K to a bath with temperatures from 600K to 500K and keeping in the bath for a time period up to ~720 ps both nucleation and crystal growth have been observed. Nucleation rate of crystallization at 600K, 550K, and 500K was estimated. The interfacial free energy of the liquid-solid boundary was derived from the nucleation rate using the classical nucleation theory. Based on the solid-liquid interfacial free energy data the Laplace pressure on small RbCl particles was estimated and the possiblity of the solid-solid transition from NaCl type to CsCl type structure under such a high Laplace pressure is discussed.

11:45 AM Q1.9
NON-THERMAL MECHANISMS OF FEMTOSECOND LASER ABLATION AND BREAKDOWN OF TRANSPARENT MATERIALS. Vitali E. Gruzdev, Anastasia S. Gruzdeva, State Research Center 'S.I. Vavilov State Optical Institute', St. Petersburg, RUSSIA.

The paper is devoted to analysis of experimental data and theoretical consideration of possible mechanisms of single-shot femtosecond damage and ablation of wide band-gap materials. We distinguish three phases of femtosecond action - initial phase (non-thermal energy transfer), intermediate phase (non-equilibrium processes) and final phase (after-heating). Non-thermal effects are shown to dominate at the first stage of single-shot action. They are connected with nonlinear electrodynamical processes (higher harmonic generation, action of gradient forces and straight action of electric field on crystal lattice) as they are the least inertial. The processes must provide significant modification of transparent material without heating during single-pulse action. Shock electromagnetic wave (SHEW) are considered as the most suitable candidate for that. SHEW is shown to appear at laser fluence characteristic of femtosecond interactions with transparent materials. Among its properties the most important are generation of higher harmonics and formation of abrupt SHEW front near which gradient of laser intensity can be very large. The harmonics are shown to produce ionization 100 - 1000 times more effectively than traditional ionization mechanisms (multiphoton, impact and tunneling ionization). On the other hand, gradient of laser intensity near SHEW front is so large that laser pressure can reach 1000 atmospheres and electric field can even move atoms and ions away from crystal lattice. It is presented description of interaction between SEW and transparent material within approach of classical mechanics and electrodynamics. Obtained results and predictions of theoretical model are shown to be capable of explaining many observed peculiarities of single-shot femtosecond laser interaction with transparent media.

SESSION Q2: PROCESSING AND THIN FILMS I
Chairs: Francois Y. Génin and David G. Cahill
Tuesday Afternoon, April 17, 2001
Franciscan III (Argent)

1:30 PM *Q2.1
LASER CHEMICAL PROCESSING WITH SHORT HIGH ENERGY PULSES. M. Stuke, M. Koch, K. Mueller, G. Padeletti*, Max-Planck-Institut f. Biophys. Chemie, Goettingen, GERMANY. *CNR Monterotondo.

Short pulses deliver energy faster than relaxation into heat can occur, and easily do force transparent [2] materials to absorb whatever necessary. Any material can be processed. For more than a decade [1], laser processing with short pulses has been studied in detail in many groups. In this talk, the special advantages of short pulses will be presented (a) for removal of material by laser ablation, (b) for deposition of species by laser photo-CVD using surface adsorbed precursor molecules, and (c) for time-resolved diagnostics including laser ionization time-of-flight detection. A comparison will be made to other processing schemes such us VUV laser and focussed ion beam (FIB) processing, in particular with respect to the achievable precision and efficiency, revealing the bright future of short pulse laser processing for many applications.
[1] S. Kuper, M. Stuke, Appl. Phys. B 44 (1987)199-204 "Femtosecond UV Excimer Laser Ablation".
[2] S. Kuper, M. Stuke, Appl. Phys. Lett. 54 (1989) 4-6 "Ablation of Teflon with Femtosecond UV Laser Pulses".

2:00 PM Q2.2
A TRANSMISSION ELECTRON MICROSCOPY (TEM) STUDY OF FEMTO-SECOND LASER INDUCED MODIFICATIONS IN QUARTZ. T. Gorelik, Technical Institute, FSU, Jena, GERMANY; M. Will, A. Tuennermann, Institute of Applied Physics, FSU, Jena, GERMANY; U. Glatzel, Technical Institute, FSU, Jena, GERMANY.

Sub-micrometer structures produced inside a transparent material by tightly focused ultrashort laser pulses result in a complex distribution of the refraction index. If these pulses are so close together that the damage regions overlap a continuous line can be written inside the matrix. It has been shown that such structures realised in monocrystaline quartz have waveguide properties. To understand the

mechanism of the waveguide formation experiments with varying laser parameters such as pulse duration and energy, polarisation and repetition rate have been carried out. TEM samples of different views were prepared from laser treated crystals. From bright and dark field TEM images the morphology of the damaged region has been determined. The matrix nearby is distorted by a high density of point defects. A strong strain field surrounding the beam-damaged area is seen. Finite Element calculations have been performed using the observed shape as input. The calculated strain field can be transferred to a refraction index distribution. The local maximums of this refraction index profile correspond to the areas where waveguiding is observed.

2:15 PM Q2.3
EXPANSION DYNAMICS OF THE PLASMA PRODUCED BY FEMTOSECOND LASER ABLATION. O. Albert, J. Etchepare, J.C. Loulergue*, A. Dos Santos LOA - ENSTA/Ecole Polytechnique, FRANCE. *also at LMOPS, Université de Metz et Supelec, France J. Perriére, E. Millon* GPS, Universités Paris VI et VII, FRANCE. *also at LSMCL, Université de Metz , FRANCE; C. Boulmer-Leborgne, E. Le Menn GREMI, Université d'Orléans, FRANCE.

The expansion dynamic of the plasma produced by femtosecond laser ablation of different materials has been studied with a time resolved CCD camera in the UV-visible range. About 1 mJ energy of a laser operating at 620 nm issued from an amplified femtosecond CPM dye laser is focused (focusing diameter : 0.5 mm) onto a rotating target. The laser pulse width can be tuned from femtosecond to picosecond and nanosecond duration. In the femtosecond pulse regime the plume behavior is linked to physical properties of the target : metals (Ti, Al), semiconductor (GaAs), large gap semiconductors (ZnO, GaN) and insulators ($BaTiO_3$, BN). Time resolved analysis of the plasma plume from 10 nanoseconds to several milliseconds, indicates that several populations of species are ejected from the target. A first one with high velocity component is emitted a few tens of nanoseconds after the laser pulse while a slower and dense component expends in the vacuum during a few microseconds. Some specific behavior is observed with ZnO and GaN targets for which the femtosecond ablation process is found likely similar to the nanosecond ablation regime. Indeed, the study of expansion dynamic is an important issue to determine the optimal growth conditions for pulsed laser deposition (PLD) of thin films. Thus, the surface morphology, the chemical composition and the crystalline features (epitaxial growth, oriented layers...) of the deposited films are strongly influenced by the nature and kinetic energy of the species reaching the substrate. This issue will be highlighted in the precise case of the femtosecond PLD of $BaTiO_3$.

3:00 PM *Q2.4
ULTRASHORT-PULSE LASER MATERIALS PROCESSING. B.C. Stuart, Lawrence Livermore National Laboratory, Livermore, CA.

The use of ultrashort-pulse lasers allows materials processing of any material (e.g., hardened steel, ceramics, diamond, high-explosives) with extremely high precision and minimal collateral damage. Advantages over conventional laser machining (using pulses longer than a few tens of picoseconds) are realized by depositing the laser energy into the electrons of the material on a time scale short compared to the transfer time of this energy to the bulk of the material (either electron-phonon coupling or thermal diffusion). Using ultrashort laser pulses, material is removed by ionizing the material. The ionized plasma expands away from the surface too quickly for significant energy transfer to the remaining material, resulting in negligible shock or thermal stress. This distinct mechanism produces extremely precise and clean-edged holes without melting or degrading the remaining material. Since only a very small amount of material (¡micron) is removed per laser pulse, extremely precise machining can be achieved. High machining speed is achieved by operating the lasers at repetition rates of 10 kHz or higher. The plasma plume generated by the short-pulse laser ablation process can also be utilized in two different ways. First, the spectroscopic character of the plasma enables determination of the material being machined. This can be accomplished on a pulse-to-pulse basis. This feature allows the machining of multilayer materials, metal on metal or metal on ceramic where one material can be machined without damaging the next. This process also allows for micron-scale 3-D compositional analysis of complex structures. Second, the plasma plume can be used as a clean and well-controlled source for thin-film deposition. Since any material can be ablated, and the ablation is not sensitive to the melting temperatures of differing materials, a great variety of film types may be produced by this method. We will present several materials-processing examples that demonstrate the benefits of, and in many cases are enabled by using the technology of ultrashort-pulse lasers.

3:30 PM Q2.5
ULTRAFAST OPTICAL NON-LINEARITIES IN METAL NANOCOMPOSITE FILMS PREPARED BY PULSED LASER DEPOSITION. R. del Coso, R. de Nalda, J. Olivares, J. Requejo-Isidro, A. Suarez-Garcia, J. Gonzalo, J. Solis, Instituto de Optica, CSIC, Madrid, SPAIN.

Nanocomposite films formed by semiconductor or metal nanocrystals (NCs) embedded in a transparent host have large third-order non-linear susceptibilities with ultrafast response times (few picoseconds or less). We have used pulsed laser deposition to prepare metal nanocomposite films formed by Cu NCs embedded in an amorphous Al_2O_3 host. The films are prepared by alternate deposition of the host and the metal in a sequence which allows to independently control the dimensions and in-depth distribution of the nanocrystals. Films having 3-6 nm sized Cu NCs separated in depth 10 nm are routinely produced and characterised with both linear and non-linear optical techniques. Spectroscopy absorption measurements are performed in order to determine the linear optical properties and the position of the surface plasmon resonance (SPR). Degenerate four wave mixing measurements are used to determine the different components of the third-order susceptibility tensor of the composite using both ps and fs laser pulses at a wavelengths in the vicinity of the SPR. The laser beam used is generated by a cavity-dumped, synchronously pumped, mode-locked Rhodamine 6G laser tuneable in the 580-620 nm range providing 15 ps laser pulses at a repetition rate variable from 400 Hz to 4 MHz. The pulse can be alternatively injected in a fibre-grating compressor to lead to a pulse duration in the range of hundreds of fs. The results obtained from the four-wave mixing experiments using either ps or fs laser pulses will be presented and discussed in terms of the various contributions to the third order non-linear optical susceptibility which operate in different time scales.

3:45 PM Q2.6
FEMTOSECOND RESOLVED DIELECTRIC FUNCTION MEASUREMENTS OF ULTRAFAST PHASE CHANGES IN GeSb FILMS. J. Solis, J. Siegel, C.N Afonso, Instituto de Optica (CSIC), Madrid, SPAIN; J.C.G. de Sande, Departamento de Ingeniería de Circuitos, E.U.I.T.T, UPM, Madrid, SPAIN; J.P. Callan, A.M.-T. Kim, C.A.D. Roeser, E. Mazur, Harvard Univ., Dept. of Physics, Cambridge, MA.

The interaction of ultrashort laser pulses with non-stoichiometric, Sb-rich, Sb_xGe_{1-x} films has recently received considerable attention. This material has been shown to be a potential candidate for the development of ultrashort laser pulse driven rewriteable optical memories. Also it has been recently shown that upon irradiation with fs pulses above a threshold fluence, the amorphous state of this material undergoes an ultrafast transition to a non-equilibrium transient phase within 300 fs. Based on results obtained from time-resolved reflectivity measurements at a single wavelength this phase transition has been considered as a possible candidate for an electronic excitation induced disorder-to-order ultrafast phase transition. The present work reports very recent measurements of the time evolution of the dielectric function of 50 nm-thick amorphous $Ge_{0.06}Sb_{0.94}$ material over the spectral interval from 1.5 to 3.5 eV upon irradiation with 50 fs laser pulses. The results obtained show that the highly excited phase differs from that of the crystalline material and is close, but different, to that of the thermal liquid, indicating the occurrence of an ultrafast disorder-to-disorder phase transition from the amorphous phase to a glassy (liquid-like disordered) state.

4:00 PM Q2.7
PULSED LASER DEPOSITED FERROELECTRIC $Pb(Zr,Ti)O_3$ THIN-FILM OXYGEN SENSORS. M.A. McCormick, M. McElfresh, B. Stuart, A. Wynne and François Génin, University of California, Lawrence Livermore National Laboratory, Livermore, CA.

Ferroelectric Pb(Zr,Ti)O3 (PZT) thin films are deposited by pulsed laser deposition using both 150 fs and 0.5 ns pulses. The films are studied to understand the role of a few processing parameters (pulse length and laser fluence) on their microstructure and properties. First, the characteristics of the ablation plume (mass, charge, and velocity of the ablated species) are measured with a time-of-flight mass spectrometer. The microstructure and the surface morphology of the films are then characterized by SEM, AFM, and TEM. Finally, ferroelectric polarization is measured as a function of applied electric field for different oxygen partial pressures in a controlled-atmosphere probe station. The presentation will discuss the correlation between processing parameters and film properties.
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

4:15 PM Q2.8
FEMTO-SECOND LASER DEPOSITION OF CARBIDES FOR EUV REFLECTANCE. Peter Blake[1], François Génin[2], Ritva Keski-Kuha[1], Alexis Wynne[2], Brent Stuart[2]. [1]Optics Branch, Goddard Space Flight

Center, Greenbelt, MD. [2]Lawrence Livermore National Laboratory, Livermore CA.

Progress is reported in the use of femto-second laser deposition to produce thin stoichiometric films of silicon carbide and titanium carbide. The application is thin-film optical coatings for astronomic optics in the EUV region (between 600$\AA$ and 1000$\AA$). The goal is to produce dense, smooth, stoichiometric films that will have a high and stable reflectance in the EUV.
Although polished SiC produced by chemical vapor deposition (CVD has the highest reflectivity in the EUV of any currently used optical material (>40%), the high temperature required for the CVD process limits its suitability for coating optical components. Instead, space flight programs have turned to room-temperature ion-beam sputtering of SiC for reflective coatings on mirrors and gratings. These ion-beam-sputtered SiC films suffer an irreversible asymptotic loss in refectance over time, stabilizing at less than 30% reflectance at 920$\AA$, with surface oxidation as the likely mechanism.
Femto-second laser deposition presents an attractive opportunity to produce SiC films of higher density, and with a lower proportion of dangling silicon bonds, than is available with ion-beam sputtering. Such films are expected to be more resistant to passive surface oxidation. Current progress is reported, along with measurements of mechanical, physical and thermal properties, chemical make-up, bond characterization, and reflectance over time.
Acknowledgments:
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48 and by NASA RTOP grant # 344-01-23 for Space Astrophysics Research and Analysis.

4:30 PM Q2.9
TRIBOLOGICAL COATINGS VIA SUBPICOSECOND-REGIME PULSED LASER DEPOSITION. I. EFFECTS OF PULSE WIDTH IN THE NANOSECOND AND SUBPICOSECOND REGIMES.
Lowell R. Matthews, Michael D. Moon, and Kenneth H. Church, CMS Technetronics, Inc., Stillwater, OK; Yin-Ming Wang and Ranga Komanduri, Oklahoma State University, Department of Mechanical & Aerospace Engineering, Stillwater, OK; Kai Dou and Edward T. Knobbe, Oklahoma State University, Department of Chemistry, Stillwater, OK.

The CMS-OSU project team has constructed a number of tribological films from boron carbide (B_4C) and layered composites of B_4C with other materials, primarily the solid lubricant molybdenum disulfide (MoS_2), via pulsed-laser deposition (PLD) in two time regimes. The construction of an automated chamber enabled the deposition of composite multilayer structures with potentially large improvements in physical properties over single-component films. The team paid particular attention to the influence of such processing parameters as laser wavelength (λ), fluence (Φ), pulse count (N_p), pulse width or duration (τ_p), and repetition rate (R_p); substrate temperature (T_s); chamber pressure (P_c); and substrate-target separation (D_{st}). Subpicosecond-regime (SPR) PLD offers significant improvements over traditional nanosecond-regime (NR) PLD in terms of film quality and ablation rate, especially for the ultrahard materials like B_4C. The SPR laser (248 nm) produced noticeably stronger ablation plumes of B_4C than did conventional NR excimer lasers operating at both shorter and longer wavelengths (193 and 308 nm). This observation supports the hypothesis that, for B_4C, ablation phenomena are driven by multiple-photon absorption events, for which conditions are far more favorable in the compressed time frame of the SPR pulse. The resultant SPR-PLD B_4C films were much smoother than the NR-PLD films, indicating a greater degree of particle dissociation within the ablation plume; the occurrence of "splashes" or large particulates is lower. In general, SPR-PLD films also demonstrated greater adhesion than NR-PLD films.

4:45 PM Q2.10
TRIBOLOGICAL COATINGS VIA SUBPICOSECOND-REGIME PULSED LASER DEPOSITION. II. COMPARISON WITH UNBALANCED MAGNETRON SPUTTERING DEPOSITION.
Lowell R. Matthews, Frank M. Kustas and Kenneth H. Church, CMS Technetronics, Inc., Stillwater, OK; Kai Dou and Edward T. Knobbe, Oklahoma State University, Department of Chemistry, Stillwater, OK.

The CMS-OSU project team has constructed a number of tribological films from such ultrahard materials as boron carbide (B_4C) and composites with softer materials, including metals and solid lubricants like molybdenum disulfide (MoS_2). The films were constructed via the competitive techniques of subpicosecond-regime pulsed-laser deposition (SPR-PLD) and unbalanced magnetron sputtering deposition (UBM-SD). These techniques offer complementary abilities with regard to the deposition of composite multilayer structures with potentially large improvements in physical properties over single-component films. SPR-PLD offers superior utility in the construction of graded films or those composed of alternating layers, while UBM-SD offers advantages in terms of throughput for single-component or codeposited materials. Both approaches can lead to superior tribological coatings with practical military, space, and tool-industry applications. The work presented herein focuses on coatings deposited onto industrial aluminum, steel, and titanium alloys.

SESSION Q3: PHASE TRANSFORMATION AND MATERIALS MODIFICATION
Chair: Francois Y. Génin
Wednesday Morning, April 18, 2001
Franciscan III (Argent)

8:30 AM *Q3.1
MATERIALS RESEARCH WITH FEMTOSECOND LASERS.
Gerard Mourou, Director, Center for Ultrafast Optical Sciences; A.D. Moore, Distinguished University Professor of Electrical Engineering and Computer Science and Professor of Physics, Ann Arbor, MI.

The application of femtosecond lasers to materials research covers a broad area. In this talk we will discuss those areas that have been under investigation at CUOS. These include micromachining, thin film deposition, isotope enrichment, and time resolved electron diffraction. Basic studies have been carried out on the fundamental aspects of ultrafast laser pulse interactions with materials with emphasis made on understanding the deterministic aspects of laser induced breakdown in transparent and absorbing materials. Thin film deposition has been investigated by taking advantage of the thin clean plasmas produced with ultrafast pulses and the very low thresholds needed to induce ablation plasma formation. Elimination of particulates in ablation plumes is investigated through adjustment of pulse duration, repetition rate, wavelength, and fluence. Extreme versatility in formation of superlattices, heterostructures, and thin film devices is possible through this technology, with high quality epitaxial tin dioxide films having been demonstrated for gas sensor applications. Other areas of research involve the direct enrichment of isotopes in ablation plumes through use of laser-plasma driven magnetic field effects and time resolved electron diffraction for use in studying sub-picosecond lattice heating, melting, and other surface science phenomena.

9:00 AM Q3.2
CONTROLLED LASER-INDUCED DAMAGE STUDIES IN FUSED SILICA.
M.D. Shirk, M.D. Feit, M.J. Fluss, L.W. Hrubesh, B.C. Stuart, Lawrence Livermore National Lab, Livermore, CA.

Abstract: Short-pulse Ti:Sapphire lasers have been used to create precise, repeatable damage spots in fused silica as a means to study laser damage evolution in high-power laser optics. The deterministic nature of short-pulsed laser damage thresholds allows for repeatable structures to be ablated on fused silica. A range of pulse durations, energies, and spot sizes are used to create damage spots with various characteristics.
We show that damage initiation sites can be created precisely with good repeatability for further studies on the mechanisms of growth and failure of optical defects. The relationship between the laser parameters and the morphology of these structures will be presented.
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

9:15 AM Q3.3
ABOUT POSSIBLE ELECTRODYNAMICAL EFFECTS OF FEMTOSECOND LASER ACTION ON TRANSPARENT MATERIALS.
Vitali E. Gruzdev, Anastasia S. Gruzdeva, State Research Center, S.I. Vavilov State Optical Institute, St. Petersburg, RUSSIA.

The paper is devoted to theoretical consideration of possible nonlinear electrodynamical effect accompanying single-shot femtosecond damage and ablation of wide band-gap materials. The effect is formation of shock electromagnetic wave on optical cycle due to generation of higher harmonics. Main features of that process are studied on the basis of the simplest model of plane-wave propagation in isotropic dielectric with nonlinear optical response of general type. There are considered influence of absorption and dispersion on formation of SHEW. Correct estimations show that threshold of SHEW formation can be obtained without taking into account dispersion. Bearing that in mind, we obtain necessary conditions for SHEW formation, in particular, threshold amplitude is estimated. The latter does not depend on laser wavelength and pulse duration and is determined by laser intensity and focusing geometry. Comparison of threshold estimation with experimental data shows that SHEW can be generated in experiments with femtosecond laser pulses where light intensity can reach 1 - 100 TW/cm^2. Computational modeling of dynamics of SHEW formation is based on the model taking into

account dispersion of both linear and nonlinear parts of refractive index. It is shown that laser-induced dispersion can play important role during SEW formation together with material dispersion and can compensate normal material dispersion if laser intensity is close to SHEW threshold. Obtained theoretical estimations are confirmed by results of computational modeling of nonlinear wave propagation in transparent dispersive nonlinear materials. Special attention is paid to study of higher harmonics generated at SHEW front and influence of absorption on SHEW dynamics. It is considered possible method for detection of SHEW based on frequency shift of low-intensity probe signal resulting from reflection of the signal at SHEW front.

10:00 AM *Q3.4
PHOTOSTIMULATED DESORPTION FROM CRYSTALLINE SOLIDS: PRODUCT STATE CONTROL AND TIME-RESOLVED STUDIES. Wayne Hess, Alan Joly, Ken Beck, Pacific Northwest National Laboratory, Richland, WA; Daniel Gerrity, Reed College, Portland, OR; Petr Sushko, Alexander Shluger, University College, UNITED KINGDOM.

Laser excitation of KBr crystal leads to desorption of fast and slow neutral bromine atoms. The source of the fast bromine atom emission is attributed to decay of a surface exciton. The slow component is derived from excitation within the crystal. We have for the first time produced separately the fast and slow components of desorbed neutral halogen atoms from an alkali halide. Resonant excitation of the surface exciton produces almost exclusively fast Br ground state atoms. Surprisingly, changing the laser wavelength to photon energies below the surface exciton resonance produces excited state Br* atoms. We explore the mechanism of excited state atomic desorption in experiments using multiple laser pulses. We have also measured desorption of positive ions as a function of time-delay between two ultraviolet femtosecond laser pulses. We find the ion yield, from 265 nm irradiated MgO, KBr, and LiF, depends critically on the time delay between the laser pulses. For example, the Mg ion desorption yield persists for laser delays of over 100 ps. In contrast, the Li ion desorption yield from LiF decays much more rapidly on the order of 3 ps. The femtosecond two-pulse technique allows direct observation of solid state and surface dynamics on a femtosecond timescale. Preliminary results suggest a possible route to laser control of solid-state reactions.

10:30 AM Q3.5
HOLOGRAPHIC ENCODING OF MICRO-GRATING STRUCTURES IN TRANSPARENT DIELECTRICS BY INTERFERENCE OF A SINGLE PULSE FROM ML-FEMTOSECOND LASER. Hideo Hosono, Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama, JAPAN; Ken-ichi Kawamura, Nobihiko Sarukura, Masahiro Hirano, Hosono Transparent ElectroActive Materials, ERARO, JST, Kawasaki, JAPAN.

A mode-locked Ti: sapphire laser has two unique features, ultrashort pulses and Fourier-limited pulse. The latter means that a pulse has high coherency. Although the former has been utilized successfully, the latter has not intentionally applied for micro machining. Here we report that non-erasable micro-grating structures may be holographically encoded on / in transparent dielectrics by colliding a pair of pulses split from a single high peak power fs laser pulse. Substrates used are single crystal of Al_2O_3, SiC, $LiNbO_3$, ZrO_2, ZnO, CaF_2, CdF_2, MgO, TiO_2, glasses, plastics, and metals (Pt, Au, WC). Near infrared light (800nm) pulses from a ML-Ti: sapphire laser were amplified by a regenerative amplifying system using an Nd: YAG laser operated at 10Hz, separated into 2 paths, and finally these two focused pulses were crossed on the top surface or beneath the surface of the substrate. Beam diameter on the substrate surface is 70 micron. The beam crossing angle was varied in the range 10 to 160 degree and the time delay between the two pulses were adjusted in the range of 0.2$\sim$2 ps by changing the optical path. The pulse duration was $\sim$ 100 fs and the laser power at the surface was varied in the range of 50mJ$\sim$3mJ. Only when a pair of pulses overlap temporarily and spatially, grating structure was encoded on / in all materials examined. Surface relief-type gratings were encoded for high laser power via ablation mechanism. Embedded gratings were encoded by shifting beam focus position into the inside of substrates. Index modulation via laser-induced structural alternation (ex. crystalline $\rightarrow$ amorphous) is the primary origin of the formation of embedded-type gratings.

10:45 AM *Q3.6
PZT THIN FILMS FOR BIOMEMS APPLICATIONS. Dennis Polla, François Génin, Peter Krulevitch and Brent Stuart, Lawrence Livermore National Laboratory, Livermore, CA.

Biomedical microsensors and microactuators based on the piezoelectric effect in PZT have been prepared by several deposition methods including sputtering, metal-organic decomposition, and femtosecond pulsed laser deposition. The BioMEMS devices realized have applications to surgery, laboratory medicine, and autonomous therapy. Several case examples including human clinical trials will be presented to demonstrate the challenges of materials, devices, and systems technology integration issues in meeting future health care needs. The application of the three PZT thin film deposition methods above have specific process integration challenges in 1) overall process flow, 2) thermal constraints, 3) deposition on micromechanical structures, 4) integration with other non-standard materials, 5) integration with electronics, and 6) packaging and reliability constraints. These challenges will be reviewed in both successful and unsuccessful biomedical systems realization. New results based on the application on femtosecond pulsed laser deposition of PZT on silicon and non-silicon MEMS structures will be presented.
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

11:15 AM Q3.7
MICROFABRICATION BY USING FEMTOSECOND IRRADIATION. Hiroaki Misawa, Andrius Marcinkevicius, Saulius Juodkazis, Mitsuru Watanabe, Vygantas Mizeikis, Shigeki Matsuo, Tokushima Univ., JAPAN; Jyunji Nishii, Osaka National Research Inst., JAPAN.

Three-dimensional (3D) microstructuring of transparent materials by femtosecond pulses is a promising technique for fabrication of 3D optical memory, photonic crystals, interconnected capillary networks in biochips, etc. In this work we investigate fs microfabrication in several glass-like materials such as PET, PMMA, silica, and in crystalline sapphire. Direct writing by a point-like photomodification inside transparent materials by a tightly focused Gaussian pulses was used in these applications in a single or multishot irradiation mode. Furthermore, we demonstrate microfabrication by "non-diffracting" Bessel-Gauss beams, which allow to achieve photomodification of the material along the line of pulse propagation over macroscopic distances without translating the sample, as would be required for Gaussian beams. The Gauss-Bessel beams are obtained by shaping conventional Gauss beams (λ = 800 nm, τ = 140 fs at the target) with glass axicon (cone angle 10°). The beam was projected into the sample by a telescope consisting of two positive lenses. This allows us to vary the focusing cone angle from 5° to 19°, as measured outside the sample. By this, the maximum distance of non-diffracting propagation of the Bessel beam was tuned within the range of 0.1$\sim$1 cm. Linear photomodification filament of the same length, and having a width as small as λ (theoretical limit is $4\lambda/3$), were fabricated along the line of fs pulse propagation in silica, sapphire and transparent plastics. Temperature control over the light induced damage threshold of silica is demonstrated. Advantages and current shortcomings of fs microfabrication in transparent solid materials are discussed.

11:30 AM Q3.8
FEMTOSECOND RESPONSE OF SQUARYLIUM DYE J-AGGREGATES FILMS AND THE APPLICATION TO ULTRAFAST OPTICAL SWITCHES. Lyong Sun Pu, Makoto Furuki, Min-quan Tian, Yasuhiro Sato and Izumi Iwasa, Corporate Research Center, Advanced Research Lab., Fuji Xerox Co., Sakai, Nakai-machi, Ashigarakami, Kanagawa, JAPAN; Satoshi Tatsuura, Osamu Wada, The Femtosecond Technology Research Association, Tsukuba, JAPAN.

Observation of femtosecond response of squarylium(SQ) dye J-aggregates films and the application of ultrafast optical switches are reported. We have investigated various SQ dyes with different molecular structures and been succeeded in making uniform J-aggregates films of SQ dyes with specific molecular structures by simple spin-coated technique. So far, femtosecond optical dynamics of organic films has been investigated in phthalocyanines and pseudoisocyanine bromide. However, for the application to ultrafast optical switches with these films, further improvements should be made in response time, nonlinear optical coefficient and thermal stability. We have studied SQ dye J-aggregates as an ultrafast optical switching material, as SQ dye has a potential to solve these problems. With pump-probe measurements by femtosecond lasers, we have shown that SQ dye J-aggregates films exhibit ultrafast recovery of the bleached and induced absorption and that they are suitable for the application of ultrafast optical switches. Femtosecond optical measurements revealed that this SQ dye J-aggregates film exhibited decay time of less than 100 fs and pump energy of 80 fJ/μm^2, which are one of the best values compared with other materials including semiconductor devices. With this ultrafast SQ optical film, multi-output demultiplex operations for T bps pulses were performed. A serial 4 femtosecond pulses of 1 ps interval (corresponding to 1 T bps signals) were converted to parallel signals at one step with a femtosecond gate pulses on this SQ film and four clear demultiplexed signals were observed at different area on photodetector.
This work was supported by New Energy and Industrial Technology

Development Organization (NEDO) within the framework of the Femtosecond Technology Project.

SESSION Q4: PROCESSING AND THIN FILMS II
Chairs: Francois Y. Génin and S. K. Sundaram
Wednesday Afternoon, April 18, 2001
Franciscan III (Argent)

1:30 PM *Q4.1
WAVELENGTH-SELECTIVE ULTRAFAST-LASER MATERIALS PROCESSING IN THE MID-INFRARED. Richard F. Haglund, Jr., David R. Ermer, Michael R. Papantonakis and Michelle Baltz-Knorr, Vanderbilt University, Nashville, TN.

At mid-infrared wavelengths, one can efficiently ablate glassy and crystalline dielectric materials by laser excitation of vibrational degrees of freedom, provided that the pulse duration is short compared to vibrational relaxation times. In this paper, we describe the use of a tunable, mid-infrared, subpicosecond free-electron laser to study fundamental processes of desorption, ionization and ablation in silica, calcite, sodium nitrate and polymers. We observe that ablation efficiency depends strongly on the relative magnitudes of the thermal diffusion length and optical absorption depth. The absorption depth can be shortened by orders of magnitude compared to non-resonant excitation, depending on wavelength and absorption mechanism, and further decreased by nonlinear absorption processes. In fused silica, for example, where absorption increases over five decades from 4 to 9 μm wavelength, a range of behaviors from fracture to explosive vaporization is observed. In crystalline calcite or its isoelectronic cousin sodium nitrate, efficient desorption, ionization and ablation occur at the fundamental ν_2-ν_4 resonance and also at the first overtone frequency. Measured energy distributions of ablated ions and neutrals show energies in the eV range, hinting at the possibility of multiphoton excitation processes, as do relaxation times measured in a reflection pump-probe geometry. Calculations based on a quantum anharmonic oscillator model indicate that there is a substantial probability for multiquantum excitation leading to ion desorption. Thermal models do not accurately describe the melt depths or particle yields for either desorption or ablation. A simple explosive vaporization model is more nearly consistent with the experimental results in ablation. The potential for making use of these results in thin-film deposition of organic materials will be discussed.
Research supported by the Office of Naval Research, Medical Free-Electron Laser Program and by the Office of Science, U.S. Department of Energy.

2:00 PM Q4.2
MICRO-EXPLOSIONS IN A TELLURITE GLASS BY FEMTOSECOND LASER. S.K. Sundaram, Pacific Northwest National Laboratory, Richland, WA; Chris B. Schaffer and E. Mazur, Division of Applied Sciences and Department of Physics, Harvard University, Cambridge, MA.

We present internal microstructuring in a tellurite glass by initiating micro-explosions inside the material. Tellurite glass ($0.1Na_2O{\cdot}0.9TeO_2$) is prepared by mixing the starting materials followed by melting and cooling per specific schedule. We use tightly focused 100 fs laser pulses to write an array of voxels about 100 μm below the surface. The internal cross-section of resulting structures is examined using optical microscopy, electron diffraction, and atomic force microscopy. We present our preliminary results.

2:15 PM Q4.3
STRUCTURAL MODIFICATION OF OPTICAL GLASSES ASSOCIATED WITH WAVEGUIDE WRITING BY FEMTOSECOND LASER PULSES. James W. Chan[1,2], Thomas Huser[1], Joseph Hayden[4], Subhash H. Risbud[2], Denise M. Krol[1,3]. [1]Lawrence Livermore National Laboratory, Livermore, CA. [2]University of California at Davis, Dept. of Chemical Engineering and Materials Science, Davis, CA. [3]University of California at Davis, Dept of Applied Science, Davis, CA. [4]Schott Glass Technolgies, Inc., Duryea, PA.

Recently, it has been shown that high intensity focused femtosecond pulses can be used to induce local index changes inside bulk glasses for fabrication of waveguides and other optical devices. The exact mechanism responsible for the change in the glass is not clear. We have studied the spectroscopic changes that occur in various glasses after modification by near-infrared focused femtosecond pulses. Pulses from a regeneratively amplified Ti:Sapphire laser (800 nm, 125 fs, 1kHz) are focused inside bulk glasses using a microscope objective. Waveguides are written by translating the sample parallel to the beam at a speed of 20 μm/s. Raman and fluorescence spectroscopy is performed on the modified glasses using a confocal microscopy setup. In vitreous silica, we have observed clear changes in the Raman spectra under conditions used for waveguide writing. The relative intensities of the Raman features at 490 and 605 cm^{-1}, which are associated with ring structures in the silica network, increase by a factor of roughly 2 after exposure to the femtosecond laser. The Raman spectrum of the modified glass corresponds to that of a glass with increased fictive temperature.[1] For vitreous silica, it is well known that a density and refractive index increase is associated with this higher fictive temperature.[2] In addition to the Raman features, we observe a broad fluorescence band centered at roughly 600 nm. The details of the observed spectroscopic behavior as a function of femtosecond laser power, intensity, and exposure time will be discussed for vitreous silica as well as for various phosphate glasses.
This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory, through the Institute for Laser Science and Applications, under contract No. W-7405-Eng-48.
References:
1. A.E. Geissberger and F.L. Galeener, Phys. Rev. B, 28, 3266 (1983).
2. R. Bruckner, J. Non-Crystalline Solids, 5, 123 (1970).

3:00 PM *Q4.4
INTERNAL MODIFICATION OF GLASS MATERIALS BY FEMTOSECOND LASER IRRADIATION. NEW APPLICATION TECHNIQUES FOR PHOTONIC DEVICES. K. Hirao, Kyoto University, Kyoto, JAPAN.

Since femtosecond laser beam has high strength of electric field, focusing a femtosecond pulse makes it possible to produce electric field of 108~109V/cm (light intensities of about 100TW/cm^2). This field strength is high enough to cause various optical nonlinearities. From this view point, a femtosecond laser is not only a light source for studying optical nonlinearities in an ultrashort time but also becoming an instrument for creating induced structure within materials. Focused irradiation of femtosecond pulses causes permanent refractive-index change within glasses and optical waveguide can be formed by inducing continuous refractive-index change. The presentation will be done about inside-modification of glass materials (selective crystallization and photo-bleaching) by focused irradiation of femtosecond pulses at visible wavelengths.

3:30 PM *Q4.5
DIRECT WRITING OF OPTICAL WAVEGUIDES IN BULK GLASS USING A FEMTOSECOND LASER OSCILLATOR. Chris B. Schaffer, Jose F. Garcia, Alan Jamison, Jonathan B. Ashcom and Eric Mazur, Harvard University, Department of Physics and Division of Engineering and Applied Sciences, Cambridge, MA.

In recent years, femtosecond lasers have proven to be extremely useful for micromachining the surface and bulk of transparent materials. When a femtosecond laser pulse is focused into a transparent material, the intensity in the focal volume can become high enough to cause absorption through nonlinear processes, leading to optical breakdown and permanent structural change to the material. Because the absorption is nonlinear, this structural change can be localized in the bulk of the sample, allowing a three-dimensional structure to be micromachined.
In this paper, we show that by focusing a femtosecond laser pulse very tightly, one can micromachine bulk glass using an unamplified laser. A 1.4 numerical aperture microscope objective is used to focus 5 nJ, sub-100-fs pulses from a 25-MHz Ti:Sapphire laser oscillator to a 0.5-μm diameter spot inside bulk glass. The intensity at the focus reaches 3 X 1017 W/m^2, causing optical breakdown and structural change in the material.
Micromachining with a high repetition-rate, unamplified laser enables a new kind of multiple-shot, thermal effect to be utilized for waveguide fabrication. Successive laser pulses deposit a small amount of heat (< 1 nJ) at a rate faster than energy can diffuse out of the focal volume. Over thousands of pulses, a micrometer-sized volume of the glass melts. By scanning the sample perpendicular to the incident direction of the laser beam, single-mode optical waveguides with a near-gaussian output profile are formed inside the glass. From the divergence of the waveguide output we determined the refractive index change in the core to be about 5 X 10-4.
The ability to directly write waveguides in three dimensions into bulk optical materials enables the fabrication of a wide variety of passive and active optical devices for the telecommunications industry. Here we have shown that this task can be achieved using only a femtosecond laser oscillator.

4:00 PM Q4.6
SURFACE PROCESSING OF ALUMINUM ALLOY 2024-T3 VIA FEMTOSECOND PULSED LASER IRRADIATION. Kai Dou, Edward T. Knobbe, The Environmental Institute and The University Center for Laser and Photonics Research, Oklahoma State University, Stillwater, OK; Robert L. Parkhill, CMS Technetronics, Inc., Stillwater, OK.

Advances in the development of ultrashort laser pulses have created many new aspects in laser processing of materials. Significant works in laser material processing, such as thin film deposition, ablation, microstructuring, laser etching, and surface cleaning, have been demonstrated in recent years. The extremely short pulse-width facilitates the achievement of a very high peak laser intensity, and electrons are driven to a much higher temperature than the ions. The interaction is thus highly nonequilibrium. Subsequent electron-lattice energy exchange takes place on a much longer time than the laser-matter interaction time. In this paper, these features of femtosecond pulses will be demonstrated for laser surface modification on the aluminum substrates, and the physics of the laser-matter interaction leading to materials modification will be discussed. In this work, our earlier studies on aluminum alloy 2024 are extended by using femtosecond and nanosecond pulse laser irradiation at a laser fluence ranging from 0.02 to 10 J/cm^2, and the micrographs of the scanning electron microscopy (SEM) have been characterized as a function of incident laser fluence. Nano- to micro-dimensioned surface texturing has been found under lower irradiation fluence. In addition to the surface texturing, the fluence dependence of ablation of metals has been studied, and two ablation regimes are found in the logarithmic dependence of ablation depth per pulse on the laser fluence. The physics of ultrashort pulse laser-matter interaction leading to material ablation and differences in ablation mechanisms are discussed.

4:15 PM *Q4.7
ULTRASHORT LASER PULSE DRIVEN REWRITEABLE PHASE CHANGE OPTICAL RECORDING IN GeSb FILMS. J. Solis, Instituto de Optica (CSIC), Madrid, SPAIN.

This presentation provides an overview of the work carried out up-to-date on the development of ultrashort laser pulse driven re-writeable phase optical memories. Non-stoichiometric, Sb-rich, amorphous thin films, such as Sb_xGe_{1-x} with x > 0.85 can be crystallized upon irradiation with ps and fs laser pulses and show a large optical contrast upon transformation. This makes these materials very promising for the development of ultrashort laser pulse driven rewriteable phase-change optical memories, since the reversibility of the process has been demonstrated in some cases. An adequate control of the heat flow conditions has allowed to achieve a full transformation time faster than 400 ps for write and erase operations with 30 ps pulses, which could open the possibility of performing static addressing. It has also been shown that the fluence required to induce crystallization decreases upon irradiation with pulses shorter than 800 fs, thus indicating the relevance of electronic excitation processes in the amorphous-to-crystalline phase transition. Using time-resolved optical measurements including femtosecond resolved photography and more recently dielectric function measurements upon irradiation of the amorphous phase with fs laser pulses, the presence of an ultrafast non-thermal phase transition to a non-equilibrium transient state of the material has been shown to occur within 300 fs after the arrival of the laser pulse. Even when the nature of this transient phase is not yet fully elucidated, the results obtained from the latter time-resolved experiments are consistent with a disorder-to-disorder transition from the amorphous phase to a glass-like state. The presence of this transient phase surprisingly conditions the final state induced upon irradiation and thus the feasibility of the studied material as ultrafast rewritable recording media since the full crystallization of the amorphous phase is only induced for fluences above the threshold for the formation of the non-thermal transient phase.

4:45 PM Q4.8
BRAGG REFLECTION POLYMER DISPERSED LIQUID CRYSTAL GRATINGS VIA ULTRAFAST HOLOGRAPHIC TWO-PHOTON INDUCED POLYMERIZATION.
Sean M. Kirkpatrick, SAIC, Dayton, OH; David Pikas TMCI, Dayton, OH; Lalgudi V. Natarajan, Vince Tondiglia SAIC, Dayton, OH; Tim J. Bunning AFRL/MLPJ WPAFB, OH.

Molecular excitation via the simultaneous absorption of two photons can lead to improved three-dimensional control of photochemical or photophysical processes due to the quadratic dependence of the absorption probability on the incident radiation intensity. This has lead to the development of improved three-dimensional fluorescence imaging, optical data storage, and microfabrication. Recently, the first use of ultrafast holographic two-photon induced polymerization (H-TPIP) has been employed to fabricate transmission-type polymer dispersed liquid crystal (PDLC) gratings. Conventional use of CW lasers to holographically induced phase separation of liquid crystal domains in a polymer matrix has been well established. Several formulations and reaction pathways have been optimized for the linear fabrication of such structures. However, it has been found that certain classes of two-photon dyes can be used as self-initiators, eliminating many of the other components of the linear formulations. Specifically, in this work, bulk Bragg reflection PDLC gratings are fabricated using H-TPIP and a class of self-initiating two-photon dyes (bis(diphenylamino) diphenyl hexatriene). A comparison to the analogous linear CW laser fabricated gratings both morphologically and electro-optically is also presented.