# ICPEPA-8
## Collected Abstracts
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UV-Laser interactions with wide bandgap insulators and semiconductors has generated a number of examples of point defect production, surface and bulk modification, etching and re-deposition processes, as well as numerous PLD related applications involving the emitted particles. In this talk we examine these phenomena and modifications in oriented single crystals of the transparent semiconductor ZnO which has a band-gap of ~3.4 eV. This material is of high interest for production of transparent conductors and transistors, solar cells, lasers, sensors, and in catalysis (including potential use in the photo-dissociation of water). We explore exposure of single crystal ZnO to UV laser radiation under ultra high vacuum conditions. We examine the underlying mechanisms for nanoparticle growth on the exposed surface, the emissions of ions, the intense and energetic emissions of neutral Zn, O, and O2, and the extraordinary clean etching of this very brittle but highly important technological material.
In situ measurement of crystallization of oxide thin films during irradiation of pulsed UV laser in chemical solution method

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Photo-induced crystallization of oxide thin films including epitaxial growth can occur at atmospheric pressure and low temperatures less than 500 °C by an excimer laser-assisted metal organic deposition (ELAMOD) process. We previously reported that epitaxial growth of La\textsubscript{1-x}Sr\textsubscript{x}MnO\textsubscript{3} (LSMO) films occurred on SrTiO\textsubscript{3} (STO) single crystals, while polycrystalline growth occurred on LaAlO\textsubscript{3} (LAO) single crystals under the irradiation of a XeCl excimer laser in the ELAMOD process. The selection of crystal growth modes can be attributed to photochemical reaction and photothermal heating effects when the sample surface is irradiated by a pulsed ultraviolet laser. However, their contributions to the growth mechanism are yet to be elucidated quantitatively. In the present paper, we report the development of an in situ measurement system to observe surface state changes when the sample surface was irradiated by an excimer laser. Various single crystals such as STO and LAO were selected as substrates. Amorphous LSMO films were fabricated onto these substrates by spin-coating, followed by prebaking. The obtained films were irradiated by a XeCl excimer laser (the wavelength: 308 nm, the full width at half maximum: 26 ns). InGaAs and Si photo diodes with narrow band pass filters coupled with converging lenses were utilized to collect thermal radiations from the specimens during the irradiation. The validity of the measurement was confirmed by comparing the result with a numerical simulation. Interesting findings are that the intensity of the thermal radiation from the LSMO film on an LAO substrate decayed much faster than that on an STO substrate, suggesting a more rapid cooling on the LAO substrate than on the STO substrate. This difference in cooling behavior can be related to the selection of crystal growth modes in the ELAMOD process.
Epitaxial growth and electrical properties of Sb-doped SnO₂ thin film grown by ArF excimer laser assisted metal organic deposition

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Tin oxide is a promising material for next generation electronic devices because it offers good properties, such as high conductivity, transparency, and chemical stability. It is also an abundantly available natural resource. Epitaxial growth of the oxide material is of considerable practical concern because its electrical and optical properties strongly depend on the orientation of the thin film. Therefore, controlling the orientation of the electrode thin film for functional oxide materials is very important. However, in most cases, the processes for oxide epitaxial growth require both vacuum and high temperature, making device production expensive. To decrease the processing temperature, we developed an excimer laser-assisted metal organic deposition (ELAMOD) method. By using ELAMOD, we prepared the epitaxial Sb-doped SnO₂ film on TiO₂ substrate by using the KrF and XeCl laser [1, 2, 3]. In this paper, we tried to prepare on the Sb- doped SnO₂ film by using ArF laser. Also, the effects of the crystal orientation and Sb concentration on the crystal growth and electrical properties were investigated.

Fig. 1 shows XRD patterns of the Sb-doped SnO₂ film prepared by ELAMOD using ArF laser. As can be seen from figure 1, an epitaxial Sb-doped SnO₂ film was obtained by laser irradiation at a fluence more than 100mJ/cm². In addition, all the films on the (110), (001) and (100) TiO₂ substrates was found to be epitaxially grown whereas the lattice mismatching between film and different crystal orientation of the TiO₂ substrates.

The electrical resistivity and carrier concentration and mobility of the film were measured. The resistivity of the (001) Sb-doped SnO₂ film was found to be lower than that of the (001) and (110) oriented Sb-doped SnO₂ films on TiO₂ substrate as show in Fig .2. In order to control the resistivity, the effect of the concentration of the Sb-doping on the electrical properties was investigated. When the 2% Sb was doped to SnO₂, the lowest resistivity of the (001) Sb-doped SnO₂ films on (001) TiO₂ substrate is 2.80×10⁻³Ωcm. Detail electrical properties such as mobility and carrier concentration will be presented

Laser Interactions in Nanomaterial Synthesis with Real-Time Diagnostics: SWNTs and Graphene

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Abstract

Laser interactions can provide highly nonequilibrium growth conditions to explore a wider range of parameter space for the synthesis of novel nanomaterials. In addition, laser interactions provide remote spectroscopic probes of the growth environment, to investigate the building blocks and kinetics in nanomaterial nucleation and growth, and to serve as real-time diagnostics to control the nanomanufacturing of nanomaterials. Here, progress in the laser-based synthesis and spectroscopic diagnostics of carbon nanomaterial growth will be briefly reviewed with an emphasis on single-wall carbon nanotubes (SWNTs) and graphene.

Laser plasmas provide high-temperature growth environments to rapidly self-assemble pure carbon into a variety of forms including metal catalyst-free synthesis of graphene flakes, single-wall carbon nanohorns, and with catalyst-assistance, SWNTs. Time-resolved, in situ spectroscopic and imaging diagnostics in combination with ex situ atomic resolution images of SWNTs, SWNHs, and graphene reveals that graphene flakes are likely building blocks for the growth of these materials.

Chemical vapor deposition (CVD) methods suitable for mass production of nanomaterials will be examined. Pulsed-CVD and pulsed laser deposition (PLD) are described to investigate the catalyst-assisted growth kinetics of graphene and SWNTs. By varying the flux of acetylene in 0.2-second pulse exposures, the dependence of the nucleation time and growth kinetics were measured. Time-resolved laser reflectivity and Raman spectroscopy studies reveal that autocatalytic kinetics and intermediates appear to govern the synthesis of both graphene and SWNTs on catalytic substrates.

Acknowledgments

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Material micro and nano structures from femtosecond laser condensation of carbon atoms

by

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ABSTRACT

Femtosecond laser ablation of graphite into a roughing pump vacuum atmosphere with subsequent condensation of the plasma plume onto a smooth, amorphous and/or crystalline substrate produces an allotropic carbon form. The solid film condensate has an open, buckled, hillock structure with textured layers of growth planes that have an interplanar spacing of 0.4696 nm. This growth plane is spaced mid-way between the (111) planes of diamond at 0.2059 nm and (0001) basal planes of hexagonal graphite at 0.6708 nm. Diamond and graphite represent carbon sp⁴ and sp³ bond states respectively, the best known forms of carbon. The delamination arms of the hillocks are approximately aligned with 120° directions and grow away from the buckled sites on the film; the buckles show a roughly 3-fold symmetry. The carbon material made by our deposition is a nano-crystalline film on one level as seen by short range clusters of interatomic planes while on a higher level it is strongly textured towards the growth direction. This allotrope of carbon is non-conductive; it seems to be a polymer-like material with strong anisotropy in the texture-alignment to microstructures especially for the growth directions as determined by x-ray pole figures, high resolution electron microscopy and with optical and laser microscopy.
Laser-induced multi-energy processing (MEP) introduces resonant vibrational excitations of precursor molecules to conventional chemical vapor deposition methods for material synthesis. In this study, efforts were extended to explore the capability of resonant vibrational excitations for promotion of energy efficiency in chemical reactions, for enhancement of diamond deposition, and for control of chemical reactions. The research mainly focused on resonant vibrational excitations of precursor molecules using lasers in combustion flame deposition of diamond, which led to: 1) promotion of chemical reactions; 2) enhancement of diamond growth with higher growth rate and better crystallizations; 3) steering of chemical reactions which lead to preferential growth of \{100\}-oriented diamond films and crystals; and 4) mode-selective excitations of precursor molecules toward bond-selective control of chemical reactions.

Diamond films and crystals were deposited in open air by combustion flame deposition through resonant vibrational excitations of precursor molecules, including ethylene (C\textsubscript{2}H\textsubscript{4}) and propylene (C\textsubscript{3}H\textsubscript{6}). A kilowatt wavelength-tunable CO\textsubscript{2} laser with spectral range from 9.2 to 10.9 µm was tuned to match vibrational modes of the precursor molecules. Resonant vibrational excitations of these molecules were achieved with high energy efficiency as compared with excitations using a common CO\textsubscript{2} laser (fixed wavelength at 10.591 µm). With resonant vibrational excitations, the diamond growth rate was increased; diamond quality was promoted; diamond crystals with lengths up to 5 mm were deposited in open air; preferential growth of \{100\}-oriented diamond films and single crystals was achieved; mode-selective excitations of precursor molecules were investigated toward control of chemical reactions.

Optical emission spectroscopy (OES), mass spectrometry (MS), and molecular dynamic simulations were conducted to obtain an in-depth understanding of the resonant vibrational excitations. Species concentrations in flames without and with laser excitations under different wavelengths were investigated both experimentally and theoretically. Detection of C\textsubscript{2}, CH, and OH radicals, as well as C\textsubscript{x}H\textsubscript{y} species and their oxides (C\textsubscript{x}H\textsubscript{y}O) (x=1, 2; y=0–5) using OES and MS, together with reaction pathway simulations, were used to explain the effect of vibrational excitations of precursor molecules on chemical reactions and on diamond depositions.
Laser-induced diamond graphitization
for high energy nuclear applications

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Synthetic CVD polycrystalline diamond is an attractive material to be used as particle detector in harsh radiation environments, like the experiments in nuclear physics at the next generation of high luminosity colliders. However, the possibility to substitute advanced radiation hard silicon sensors with diamond is currently under study because of the technological problems and the costs involved.

One of the issues to be addressed is the technology for building high quality electrical contacts. Diamond graphitization seems to be an easy and inexpensive solution.

A polycrystalline, thermal grade CVD diamond sample, 10x10x0.2 mm³ was irradiated, on selected rectangular areas, using laser pulses from a KrF (λ = 248 nm) and from an ArF (λ = 193 nm) excimer lasers. Three different types of laser treatments were performed involving (i) a single KrF pulse, (ii) an increasing number of KrF pulses along a strip and (iii) nine ArF pulses. For all the three different areas the laser fluence was set at 5 J/cm². The irradiated areas were analysed by Micro-Raman spectroscopy and scanning electron microscopy observations (SEM). The Raman analysis showed that laser irradiation is effective in inducing structural changes from diamond to disordered graphite and to turbostratic graphite. It has been observed that the features of the structural changes depend heavily on the laser wavelength and on the irradiation method (single or multiple shots). The Micro-Raman findings were confirmed by SEM observations using a field ion microscope (Zeiss Supra-40). A nuclear radiation detector prototype was built by producing graphite electrodes via laser irradiation on a detector grade CVD polycrystalline diamond wafer. The detector was tested with gamma sources and ultra-relativistic ionizing particles before and after intense irradiation (up to about 10¹⁶ particles/cm²) with a 62 MeV proton beam.
Fabrication and characterization of free-standing ultrathin Diamond-like carbon targets for ion-driven fast ignition and laser-driven ion therapy research

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Observation of maximum energy of 185 MeV of carbon ions have been recently reported [1] when free-standing ultrathin Diamond-like Carbon (DLC) targets are being irradiated by high contrast sub-picosecond ultrahigh intensity laser pulses. This result has significant implications for ion-driven fast ignition fusion [2] and laser-driven ion therapy [3]. To enable ion-driven fast ignition fusion and laser-driven ion therapy research, high quality free-standing ultrathin DLC targets are needed.

Here we report the fabrication of DLC thin films using pulsed laser deposition (PLD). PLD is a well-established technique for deposition of high quality DLC thin films. Carbon tape target was ablated using a KrF (248 nm, 25 ns, 20 Hz) excimer laser to deposit DLC films on soap coated substrates. For these depositions laser fluence between 8.5- 14 J/cm² and a target to substrate distance of 10 cm was used. These films were then released from substrates to obtain free standing DLC thin films. Different film thicknesses from 20nm to 200nm thick films were deposited using this technique to obtain free standing targets of up to 1inch square area.

These free standing DLC films were then characterised using different techniques such as AFM, XPS and nanoindentation. AFM was used to obtain the film surface roughness. XPS was utilised to obtain peak ratios of the sp2 and sp3 peaks. Nano-indentation was used to characterise the film strength. Ablation threshold properties of the DLC films were studied using 1064nm ns laser pulses. An ultrahigh intensity sub-picosecond laser pulse typically consists of a lower intensity pre-pulse few ns ahead of the main pulse [4]. This pre-pulse can potentially destroy the DLC target before the arrival of the main pulse. Thus, it is important to characterize the ablation threshold of the DLC targets due to ns pre-pulses. Similar studies were also conducted on the commercially available CVD diamond free standing targets. A comparison of the results from the two types of ultrathin films will be presented.

References
Laser irradiance and CH\textsubscript{4} pressure effect on Carbon plasma dynamics

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Abstract

Carbon plasma plume dynamics study was carried out into methane ambiance in the pressure range of 5x10\textsuperscript{-2} to 5 mbar by ICCD camera fast imaging diagnostic. Carbon plasma was induced by the third harmonic of Nd-Yag laser of 355 nm wavelength and 7 ns pulse duration by ablated graphite target at both laser irradiances 3x10\textsuperscript{9} and 5x10\textsuperscript{9} W/cm\textsuperscript{2}. The expansion of the plasma plume was found to differ much as CH\textsubscript{4} pressure increases. Increase in ambient pressure results in the confinement of the plume and in turn enhance in collisions and fluorescence, slowing down of the expansion plume velocity and a shock front formation. The stratification of the plasma core into two well distinct components at 1 and 5 mbar was also observed for 5x10\textsuperscript{9} W/cm\textsuperscript{2} (see figure 1). Furthermore, at 5 mbar CH\textsubscript{4} pressure, plasma front edge undergone deformation and turbulence which reveals hydrodynamic instabilities formation.

The drag model and the Zel’dovich shock wave model were found to fit one part of the plasma expansion while De Izarra shock wave model interpolate overall plume expansion. From the fit parameters, the mass and the energy of the plasma were estimated and were compared to the ablated mass and laser energy.

Figure 1: Plasma emission at 1 mbar CH\textsubscript{4} pressure for laser irradiances (a) 3x10\textsuperscript{9} W/cm\textsuperscript{2}, (b)5x10\textsuperscript{9} W/cm\textsuperscript{2}
Nd:YAG laser ablation characteristics of thin CIGS solar cell films


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Thin film solar cells based on thin absorber layers such as amorphous silicon (a-Si), CdTe, or CuIn$_{1-x}$Ga$_x$Se$_2$ (CIGS) have several advantages as compared to crystalline silicon (c-Si) solar cells. For example, thin film solar cells can be manufactured at a lower cost than the c-Si counterparts and also, due to manufacturability on flexible substrates, they can be easily integrated into the roof or exterior walls of a building. Among the various types of thin film solar cells, CIGS solar cells offer several attractive properties for practical solar power applications such as high absorption coefficient of the absorber layer reducing the material usage and high photo-conversion efficiency of greater than 20% [1]. Laser processing has critical importance in the development of thin film solar cell manufacturing technologies such as laser scribing and compositional analysis by laser-induced breakdown spectroscopy (LIBS).

In this work, the variations of ablation characteristics of thin CIGS solar cell films with respect to laser parameters are investigated using nanosecond Nd:YAG lasers with different wavelengths. Shadowgraphic images of the ablated CIGS films were taken to examine the in-situ removal phenomena and correlated with the crater morphologies measured with a scanning electron microscope. The more critical laser parameters to accomplish the film ablation conditions required for laser scribing and LIBS are suggested.


Acknowledgment
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Evaporation chemistry of CO₂-laser heated silica

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Evaporation rate data of CO₂-laser heated surfaces of silica is presented, along with a near-equilibrium analysis for heating temperatures ranging from 2500 K - 3000 K. Under reducing gas conditions (hydrogen) rates of evaporation are highest, while oxygen reduces evaporation rates below neutral conditions. The specific influence of these gases on the rates of evaporation is interpreted on the basis of reaction equilibrium at the heated spot, where temperature is measured by infrared imaging. Under “short” laser exposure conditions (≈<0.1 sec), the process of evaporation is purely pyrolytic and alternate solid-gas phase reaction pathways cease to play a role. Measurements under gas flow conditions show that evaporation rates are dependent on forced convection, thus the evaporation rate model presented describes both the transport kinetics via the mass transfer coefficient, and the reactions equilibrium via the temperature-dependent equilibrium constant. Reaction free energies can thus be extracted and compared to published results for validation. In addition, the results of these studies on bulk silica are used to model silica micron-sized particles heating with a CO₂-laser beam to determine the extent of laser-material coupling during particle evaporative shrinking.
Simulations of the melt expulsion in continuous wave laser ablation of aluminum films: Effects of the recoil vapor pressure and external gas flow

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Two-dimensional hydrodynamic simulations of laser ablation of aluminum films by a continuous wave laser are performed. The goal of the computational study is to elucidate the effects of the recoil vapor pressure and external gas flow on the efficiency of material removal from the laser spot. The three-phase modeling of the coupled melt and gas flows and thermal state of the solid material is carried out with an in-house computational code designed for accurate simulations of multiphase flows with complex boundaries. The flows of the melted material and external gas are described by the full incompressible Navier-Stokes equations with the level set method used for tracking the evolving liquid-gas interface. The melting and solidification are described in the framework of the enthalpy formulation. The unsteady Navier-Stokes equations are solved numerically with the artificial compressibility method and high-order upwind difference approximation of convective terms. The simulations are performed in a range of film thicknesses from 0.4 mm to 1 cm, the gas free stream velocities form 0 to 100 m/s, and laser power from the threshold for surface melting up to the value just below the boiling temperature. Transient shapes of the laser crater and rim formation are investigated in the simulations. The time required for melting through the full depth of the film is calculated. The simulation results are compared with predictions obtained with a thermal model of laser ablation based on the one-dimensional heat conduction equation supplemented by a simple model of melt expulsion. Based on the simulation results, boundaries of the regimes where the melt expulsion is dominated by either the recoil effect of the vapor pressure or external gas flow are established in a broad range of irradiation parameters and film thicknesses.

Keywords: Laser melting, laser ablation, continuous wave laser, aluminum film, melt expulsion, shear gas flow, hydrodynamic simulations
Laser-induced microjet injection: wavelength dependence of injection efficiency in the biomedical application

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Keywords: Transdermal drug delivery; Bio-ballistic; Microjet injection; Guinea pig

A desire to eliminate hypodermic needle in transdermal drug delivery may soon be realized. Imaging of the skin after injection of fluorescent probe and biotin via the laser-induced ballistic technique revealed the epidermal and dermal layers which were stained well below 60 µm underneath the abdominal skin of the guinea-pig. An extensive network of cells are shown in the deeper layer of the stained dermis as the distributed fluorescein isothiocyanate (FITC) dose is administered by repeated injection via the laser-based microjet. A significant increase in delivered dose of drugs is reported with multiple pulses of Er:YAG laser beam at lower laser energy than previously obtained by a Nd:YAG system. The new injection scheme at 250 us pulse width generates elongated microjets which effectively penetrates deeper layer of skin at lowered laser energy. A theoretical hydrodynamic analysis shows the upper limit of the jet velocity from the micro injector is ~50 m/s, which is consistent with the measured jet velocity using 1000 mJ/pulse of Er:YAG beam.
Time Resolved Analysis of Femtosecond Laser Impulse Induced in Water and Cell Culture Medium

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When an intense, infrared femtosecond laser is focused inside water or cell culture medium through an objective lens, shockwave and cavitation bubble are generated at the laser focal point by a multiphoton excitation. Because of its effective multiphoton excitation, the threshold of laser pulse energy to induce these generations is lower than that of other pulse lasers. As the result, the stress wave following shockwave and cavitation bubble generations localises at the vicinity of the laser focal point with size of a few tens microns in the condition that the laser pulse energy is tuned to be near the threshold. We are applying the stress wave as an impulsive force to manipulate and to analyze biological cells or tissues [1]. In this application, we have realized to quantify the impulsive force by using atomic force microscopy (AFM). The impulsive force was detected as a bending movement of the AFM cantilever, whose time course was monitored by an oscilloscope directly connected to position sensor (quadrant photo-diode) of the AFM [2].

In this work, we analyzed the time course of the bending movement with assumption that the pushing and retracting forces following cavitation bubble generation and collapse are mainly loaded on the AFM cantilever. In this assumption, the time course of the bending movement was analyzed by an equation:

\[ m\ddot{z} + cz + k\dot{z} = F_1 \cdot \delta(t) - F_2 \cdot \delta(t - \tau), \]

where \( m \) is the equivalent mass of the cantilever system, \( c \) is the linear constant of the viscous damping force of water, and \( k \) is the spring constant of the cantilever. \( F_1 \) and \( F_2 \) mean respectively pushing and retracting forces as a unit of impulse.

The representative result is shown in Fig. 1. Femtosecond laser pulse from regeneratively amplified Ti: sapphire laser (150 fs, 780) was focused through an inverted microscope equipped with 10x objective lens. The distance between laser focal point and the tip of the cantilever was fixed to be 10 µm in XY plane and the bending movement of the cantilever was observed as a function of Z position of the cantilever, i.e. difference of elevation between the laser focal point and the cantilever in optical axis. The pulse energy is 60 nJ/pulse, which is about 1.5 times larger than the threshold energy of the stress wave generation. The transient vibration of the cantilever (Solid lines) was fitted by the equation [1] and reproduced well by the fitting shown as dashed lines of the figure. The time lag (\( \tau \)) between \( F_1 \) and \( F_2 \) was agreement with the time evolution of the cavitation bubble generation and collapse observed by high-speed imaging. The Z position dependence and the laser power dependence of the parameters were investigated in detail and the behaviour of the impulsive force was revealed. On the basis of these results, potential of the femtosecond impulsive force as a tool for biotechnology was discussed.

Ultrafast changes in the lattice symmetry of VO$_2$ probed by coherent phonons

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Measuring the time resolved changes in the optical reflectivity of solids during a photoinduced phase transition has been a simple and powerful tool for studying material dynamics. In this talk, I will discuss how coherent optical phonons, which modulate the reflectivity of a material when excited by a short laser pulse, can be used as an optical probe of structural transitions, provides complimentary information to diffraction based techniques. This is demonstrated on the photoinduced solid-solid phase transition of VO$_2$. Below the phase transition threshold, coherent phonons associated with the low temperature monoclinic phase are observed. On increasing the pump fluence the phonon modes are lost, indicating an ultrafast change in the lattice potential drives the transition to the rutile phase non-thermally.
Time-Resolved Photoluminescence study of ZnO Thin Film Grown by Pulsed Laser Deposition
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Abstract:

Zinc Oxide (ZnO) has attracted tremendous interest due to its direct wide band gap (3.37eV at room temperature) and the large exciton binding energy of 60meV. It has novel optical and electronic properties, such as refractory and chemically stability in air, and UV photoemission at room temperature. These properties make ZnO a promising material for UV lasers, light emitting diodes and fast scintillators.

Pulsed laser deposition (PLD) is an established method to produce complex oxide like ZnO as it allows the control of ambient over a wide range of pressure during the deposition process. ZnO fabricated by this method is naturally n-type due to large concentrations of oxygen vacancies and zinc interstitials, which have low formation energies. By optimizing the growth parameters, the concentrations of these defects can be reduced to fabricate high-quality ZnO thin films. In this study, a KrF (248 nm, 25 ns, 20 Hz) excimer laser is used to ablate a ZnO target (99.9% pure) to deposit thin films on sapphire substrates. The ambient Oxygen pressure during growth is varied from 10-100 mtorr.

The properties of the thin films will be studied using different characterization techniques including XRD, AFM, SEM and photoluminescence (PL). PL studies of the material can give us an insight about its photoemission mechanisms and the various defect levels present in it. The PL of the ZnO thin films will be conducted using different excitation parameters (various wavelengths and the pulse widths). Both CW and pulsed lasers (ns and fs pulses) of different wavelengths will be used to study the material at room temperature. A streak camera (~15 ps resolution) coupling to a grating spectrometer will be used to study the ultrafast PL of the ZnO thin films excited by 266nm 100 fs laser pulses. Results from this study will be presented here.
Generation of THz transients by photo-excited single-crystal GaAs meso-structures

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We report a sub-picosecond photoresponse and THz transient generation of GaAs single-crystal mesoscopic structures excited by femtosecond optical pulses. Our freestanding whiskers and platelets were fabricated by a top-down technique. A high-quality, epitaxial, 500-nm-thick film of semi-insulating GaAs was grown on top of an AlAs sacrificial layer and patterned using optical lithography. Next, the resulting 500-nm-wide and 20-µm-long whiskers and 20×20 µm² platelets were etched away from their native substrate, carefully transferred onto an MgO substrate using a micro-pipette, and incorporated into an Au coplanar strip line. As the high-quality, epitaxially grown crystals, our GaAs photoconductive devices exhibited mobility values as high as ~7300 cm²/V•s, extremely low dark currents, and ~7% detection efficiency. We have carried out a femtosecond, time-resolved electro-optic characterization of our structures and recorded along the transmission line the electrical transients as short as <600 fs, when the whisker was excited by a train of 100-fs-wide, 800-nm-wavelength optical laser pulses. The frequency domain analysis showed that the intrinsic response of our device featured a 320-fs FWHM, corresponding to a 3-dB bandwidth of 1.3 THz. Simultaneously, the platelet structures have been demonstrated to be very efficient generators of free-space propagating THz transients with the bandwidth extending beyond 2 THz. The presented performance of the epitaxial, freestanding GaAs meso-structured photo-devices makes them uniquely suitable for THz-frequency optoelectronic applications, ranging from ultrafast photodetectors to THz-bandwidth optical-to-electrical transducers and photo-mixers.
In this paper we will present results of a study on the time evolution of AC conductivity of non-equilibrium warm dense gold. In the experiment, freestanding 30nm-thick gold foils are excited by 400nm, 45fs (FWHM) laser pulses to energy densities up to ~4MJ/kg. Temporal evolution in AC conductivity of the resulting state is determined from simultaneous measurements of reflection and transmission of a chirped pulse probe at 800nm. This yields two important benchmarks for comparison with theory. The conductivity value at the end of the femtosecond laser pulse is a measure of its dependence on electron temperature as the ions remain cold. Subsequent changes then provide a measure of conductivity as a function of both electron and ion temperatures as thermal equilibration between electron and ion progresses. These data are compared with ab-initio quantum simulations.
Films of organic materials are commonly deposited by laser assisted methods, such as MAPLE (matrix-assisted pulsed laser evaporation), where a few percent of the film material in the target is protected by a light-absorbing volatile matrix. Another possibility is to irradiate the dry organic material directly for film production, as in PLD (pulsed laser deposition), where the film molecules may undergo strong fragmentation. In this presentation we report an alternative surprising mechanism for film deposition of the protein lysozyme in vacuum, when a small amount of residual water drives the ejection and deposition of lysozyme. This can be called an “inverse MAPLE” process, since the ratio of “matrix” to film material in the target is 10:90, which is inverse of the typical MAPLE process where the film material is dissolved in the matrix down to several wt.%.

Lysozyme is a well-known protein which is used in food processing and is also an important constituent of human secretions such as sweat and saliva. It has a well-defined mass (14307 u) and can easily be detected by mass spectrometric methods such as MALDI (Matrix-assisted laser desorption ionization) in contrast to many other organic materials. Also, the thermal properties of lysozyme, including the heat-induced decomposition behavior are comparatively well-known.

The ablation of lysozyme from a dry pressed target in vacuum was measured by weight loss in nanosecond laser ablation at 355 with a fluence of 0.5 to 6 J/cm². Films with a significant number of intact lysozyme molecules have been produced by direct laser irradiation of a pressed target and the number of intact molecules shows a maximum at around 2.5 J/cm². Apparently, there is a certain range of laser fluences when the transfer of intact lysozyme to the film substrate is possible.

The experimental results are explained with the help of molecular-level computer simulations. The simulations show that pure lysozyme cannot ablate without complete fragmentation. However, small pockets of trapped water provide the necessary expansion of the target and the ejection of intact lysozyme molecules above a certain fluence threshold, below which no lysozyme molecules are ejected. For high fluences all molecules are ejected as fragments. For a reasonable concentration of water (10%) the fluence dependence similar to that obtained experimentally is observed in the simulations.

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Matrix-assisted pulsed laser evaporation (MAPLE) offers a gentle mechanism to transfer easy-to-be-decomposed materials from the condensed phase into the vapor phase.

This peculiarity come from the fact that the material of interest is diluted in a volatile, noninteracting (even under laser irradiation) solvent with a typical concentration of a few weight percent and the laser energy is mainly absorbed by the solvent minimizing the laser-solute interaction. Polymers, biological cells, proteins, and even nanoparticles and nanorods were successfully deposited with this technique. Moreover, MAPLE was shown to be a very promising technique for the fabrication of polymeric multilayer device stacks, which are very difficult to realize with the conventional solvent assisted deposition methods. In fact, for example, a poly-(3-hexylthiophene) (P3HT) / [6.6]-phenyl-C61-butyric acid methyl ester (PCBM) bilayer structure was realized by single step matrix-assisted pulsed laser evaporation (ss-MAPLE) technique using the same solvent for both the polymers under vacuum conditions.

In fact, although MAPLE is a solvent-based technique, the idea underlying the ss-MAPLE technique was the fact that a well-optimized, in terms of deposition parameters, MAPLE deposition should in principle let overall solvent vaporization or effectively minimize/eliminate the solvent contamination of the polymeric films. This aspect of the technique is questioned and discussed.
Photoluminescence of Cu:ZnS, Ag:ZnS, and Au:ZnS nanoparticles Applied in Bio-LED

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Abstract
In recent years, the concept of developing green energy and technology is concerned more gradually. Biologic light emission device (Bio-LED) was also developed to replace street lamps to glow our life and trap carbon dioxide at the same time. However, Bio-LED lacks blue light and green light emission. Zinc Sulfide (ZnS) is a material which has a wide band gap characteristics (∼3.66 eV) often use in commercial as a fluorescent powder to emit blue light and also used in the thin film electroluminescence device. When Zinc Sulfide (ZnS) was doped some transition elements, emission spectrum will be shifted, which is a possible aspect to create the green light source.

In this work, transition elements, including Cu²⁺, Ag⁺, and Au³⁺, were used to dope in zinc sulfide by chemical solution synthesis to prepare Cu:ZnS, Ag:ZnS, and Au:ZnS nanoparticles, respectively. Transition elements doping ZnS nanoparticles form the electronic energy level between conduction band and valance band, which will results in the green light emission. Zinc Sulfide (ZnS) emission shift from blue light (∼3.01 eV) to green light (∼2.15 eV). We also found that Au:ZnS nanoparticles will emit green light (∼2.3 eV) and blue light (∼2.92 eV) at the same time because the mechanism of blue light emission was not broken after Au element doped.

Furthermore, we use copper chlorophyll to simulate chlorophyll in Bio-LED. We combined copper chlorophyll with Cu:ZnS, Ag:ZnS, and Au:ZnS nanoparticles by self-assembled method. Then we measure its photoluminescence spectroscopy (PL) and X-ray photoelectron spectroscopy (XPS) to study emission spectrum and bonding mode. We found that Au:ZnS nanoparticles are able to emit green light and blue light to excite the red light emission of copper chlorophyll, which is a potential application of Bio-LED.
The Degradation of a Prototypical Blue Organometallic Phosphor through Photoinduced Disproportionation

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Through the versatility of organic synthesis and device engineering, organic light emitting diodes (OLEDs) have become a principal part of the solid state lighting industry. Phosphorescent OLEDs that emit red¹ and green² light have been fabricated with operational life times that are considered stable for use in applications³ (＞ 10⁴ h). The high energy triplet emitting blue⁴ OLEDs, however, suffer from short lifetimes that have been attributed to the chemical instability of the phosphor. As it has been difficult to characterize the products of chemical reactions that occur in nanoscale thin films, very little is known about the degradation pathways affecting the materials. In this research, we utilize the high sensitivity and soft ionization of laser desorption/ionization mass spectrometry⁵,⁶ (LDI-TOF MS) to investigate photodegradation of the prototypical blue phosphorescent dopant FIrpic⁴ within the high band gap organosilane host UGH3⁴ (100 nm thin film). Irradiation (75 W Xe lamp) of the thin film along the metal-to-ligand charge transfer band of FIrpic under an inert atmosphere (N₂) causes the phosphor to undergo disproportionation. Analysis with LDI-TOF MS identified the photooxidation product of this reaction as [FIrpic – H + O]− (710.1 m/z). The photoinduced disproportionation was completely suppressed when the thin film was irradiated under N₂ + ¹⁸O₂, and the [FIrpic – H + ¹⁸O]− species was generated through a reaction of the phosphor with singlet ¹⁸O₂. A comparison of these two products suggests that photoexcitation is capable of causing the dissociation of FIrpic. One of the products of this decomposition is a reactive oxygen-bearing species capable of oxidizing other compounds in the thin film. Through the use of LDI-TOF MS it can be shown that FIrpic readily undergoes photodecomposition in the solid state a feature that limits its applicability in OLEDs.

References

DESIGNER PULSES FOR OPTIMAL ABLATION: GUIDING HEAT IN ULTRAFAST LASER ABLATION PLASMAS

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The possibility of choosing thermodynamic trajectories for laser excited matter is a determinant factor for controlling a typical laser ablation process, particularly when irradiation on ultrafast timescales is involved. This control specifically concerns the nature and the energetic characteristics of the ablation products subsequent to laser irradiation. In this respect, designing the energy delivery rate using programmable pulse shaping methods in the temporal domain is a powerful way for regulating excitation and thermodynamic relaxation of the material and, equally, its hydrodynamic advance.

We focus here on ultrafast laser irradiation of metals, with the objective of maximizing heat load in the ablation products. Using experimental and theoretical adaptive loops [1,2,3] based on hydrodynamic codes we indicate the temporal shapes of optimal pulses on ultrashort and short timescales required to reach extreme thermodynamic states at limited energy input. These waveforms, particularly impulsive pulses on picoseconds pedestals, affect the excitation level and the energetic content of the ablation products, as well as the balance between thermal and mechanical energy. As the material states rapidly vary from solid to plasma phases, the optimal interaction scenario usually implies light coupling into the incipient material hydrodynamic motion. This triggers transitions to weakly-coupled front plasmas at critical optical density, favoring energy confinement with low mechanical work. Additional collisional heating occurs in denser regions above the critical point, reaching states that spontaneously decompose in excited atomic species.

The consequences are manifold and are particularly visible in the formation of atomic, ionic, and cluster species, their kinetics and spectral emissivities, and in the ejection of nanoscale liquid droplets. A discussion on the nature of these resulting exotic thermodynamic states, mostly implying supercritical paths, will be given. The results are interesting for remote spectroscopy applications, e.g. LIBS and secondary sources, ablation process quality, and for the laser-assisted generation of nanoparticles and PLD.

References:
This paper will review our recent work on the application of pulsed laser processing for novel materials production. Two distinct approaches are reviewed including laser materials modification in controlled gas and liquid media respectively. In particular, it is shown that the artificial surfaces obtained by femtosecond laser texturing of solid surfaces in reactive gas atmosphere exhibit roughness at both micro- and nano-scales that mimics the hierarchical morphology of natural surfaces. Depending on the functional coating deposited on the laser patterned three dimensional structures we can achieve artificial surfaces that are: (a) of extremely low surface energy, thus water repellent and self-cleaned; (b) responsive, i.e., show the ability to change their surface energy in response to different external stimuli such as light, electric field and pH. Moreover, the behaviour of different kinds of cells cultured on laser engineered substrates of various morphologies was investigated. The second part of the paper is focused on the pulsed laser assisted synthesis and functionalization of new types of nanomaterias. Results on the formation of nanoparticles (NP) of Al, Graphene, WS$_2$ and MoS$_2$, using pulsed laser processing in gas and/or liquid media will be presented. Furthermore, a rapid and facile methodology for the photochemical reduction, functionalization and doping of graphene oxide (GO) sheets, based on pulsed UV laser irradiation of GO in liquid or gas media, will be demonstrated. Potential biological and optoelectronic applications of pulsed laser synthesized and modified materials are demonstrated and discussed.
Controlling photocatalystic activities and crystal structures of TiO$_2$ nanoparticles synthesized by reactive pulsed laser ablations

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Nanostructured photocatalysts are attractive because of their chemical activities based on the large specific surface areas. Pulsed laser ablation (PLA) in reactive background gases is a caditate process, where varying simple process parameters, crystal structures and morphologies of deposited oxide nanoparticles can be controlled. Recently, we have demonstrated non-stoichiometric formation of multicomponent oxide (In$_{0.9}$Ni$_{0.1}$TaO$_{4-d}$) nanocrystallites and their visible-light responses$^{1,2}$, and controlling crystal structures (anatase and/or rutile) and morphologies of as-deposited TiO$_2$ nanocrystallites$^3$.

In this study, we introduce rapid thermal processes (RTP), as post annealing to improve the photocatalystic activities as well as crystallinities of the as-deposited TiO$_2$ nanocrystallites.

The third harmonics beam of Nd:YAG laser (wavelength: 355 nm, pulse energy: 10 mJ/pulse) was focused onto a sintered high density TiO$_2$ target, in oxygen back ground gases. Deposition substrates without heating were located at the normal direction to the focusing point. The RTPs (radiation heating) was carried out at temperature range of 400-900°C, for 60 sec., aiming at recovering the surface disordered layers suppressing the sintering diffusions.

We have clarified that 1) Average diameter of the as-deposited primary particles were 5 nm, 2) Crystal structures of the as-deposited species can be controlled to anatase (O$_2$ gas pressure was around 1.0 Torr) or rutile (O$_2$ gas pressure was more than 1.5 Torr), 3) X-ray crystallinities of the TiO$_2$ nanocrystallites were improved, with increasing RTP temperature, 4) In RTP temperatures higher than 700°C, as-deposited anatase structures transformed to rutile structures. Figure 1 shows main diffraction peak intensities of anatase (101) and rutile (110) as a function of the RTP temperature, in the TiO$_2$ nanocrystallites of which as-deposited structure was anatase (O$_2$ gas pressure : 1.0 Torr).

To characterize photocatalystic activities of the TiO$_2$ nanocrystallites, we introduced a methylene blue decomposition method. Fused SiO$_2$ substrates on which the TiO$_2$ nanocrystallites deposited, in methylene blue aqueous solution (0.01 mmol/l) were irradiated by an excitation light (band-pass-filtered Xe lump: 355 nm (FWHM: 10 nm), Irradiation density: 1.0 mW/cm$^2$). These results are shown in Fig. 2. The vertical axis means decrease of main absorption peak (664 nm). It was found that the photocatalytic activities were also increased with increasing RTP temperature. In our TiO$_2$ nanocrystallites, the difference between anatase and rutile was not so obvious, concerning the photocatalytic activities.

Consequently, we have successfully controlled in the TiO$_2$ nanocrystallites not only crystal structures, but also the photocatalystic activities, using a combination of the reactive PLA and RTP processes.

[References]
Comparison of metallic nanoparticle formation processes by ultra-short laser ablation in vacuum, in liquid and by spark discharge in air

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Metallic nanoparticles (NPs) have found numerous applications in medicine, photonics, and other fields due to their unique plasmonic properties. New promising methods of their fabrication include laser ablation and spark discharge. In this study, based on detailed computer simulations, we perfume thermodynamic analysis and elucidate physical mechanisms involved in the formation of metallic NPs.

First, we consider ultra-short laser ablation in vacuum. The developed two-temperature hydrodynamic model is based on a realistic equation of state that includes metastable regions. Far ultra-short laser ablation of metallic targets in vacuum, the majority of NPs are shown to originate from the metastable liquid zone, which is fragmented as a result of void nucleation and rapid material expansion. In the presence of a confining environment, such as air or liquid, the expansion is limited by the ambient pressure. In particular, in the presence of water, fragmentation of the metastable liquid cannot explain nanoparticle formation since the created liquid fragments are pressed back (Fig.1). In this case, for higher laser intensity, the majority of NPs are formed in the unstable liquid-gas region. Here, the size of the created NPs depends on time that material spends in this zone. Furthermore, nucleation of small (10-100nm) liquid particles is also possible in the region of supersaturated vapor. However, the number of these nucleuses cannot be high and spontaneous phase separation taking place in the unstable zone dominates in NPs formation by laser in liquids.

Then, we investigate the processes of NPs formation by spark discharge in air. Here, after efficient photoionization and streamer expansion, the cathode material is sputtered and NPs appear. In multiple discharges, nucleation in supersaturated gas explains the NPs formation. Finally, we compare the efficiency of NPs formation by laser ablation with that by spark discharge in air.

![Figure 1](image)

Phase distributions showing the difference in mechanisms of NPs formation by ultra-short laser ablation of metallic targets in vacuum (a) and in liquid (b).
Femtosecond laser-induced periodic nanostructures

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During the past few years significantly increasing research activities in the field of laser-induced periodic surface structures (LIPSS, ripples) have been reported. This is partly triggered by the availability of commercial fs-laser sources along with the observation that LIPSS with spatial periods ($\Lambda$) significantly smaller than the laser irradiation wavelength ($\lambda$) are solely observed for irradiation of solids by ultrashort laser pulses in the fs-regime. Hence, the generation of LIPSS in a single-step process provides a simple way of surface nanostructuring towards a control of optical, mechanical, or chemical surface properties.

In this contribution we address the current research state in this field. The formation of LIPSS upon irradiation of metals, semiconductors and dielectrics by linearly polarized high-intensity Ti:sapphire fs-laser pulses ($\tau\sim30-150$ fs, $\lambda\sim800$ nm) is studied experimentally and theoretically. The irradiation results obtained in different environments (gas, liquid) are characterized using optical (OM), scanning electron (SEM) and scanning force microscopy (SFM). The SEM micrographs are evaluated by means of one- and two-dimensional Fourier analyses in order to quantify the spatial periods of the structures. Two different types of LIPSS exhibiting very different spatial periods are observed in our experiments, both having a different dependence on the incident laser fluence and pulse number per spot. We demonstrate that for specific conditions, sub-100 nm surface structures can be generated by simple processing in air.

The experimental results are analyzed by means of a theoretical approach, which combines the widely accepted LIPSS theory of J.E. Sipe and co-workers with a Drude model, in order to account for transient changes of the optical properties of the irradiated materials. In semiconductors (silicon), this model is capable of explaining numerous aspects of fs-LIPSS formation, i.e., the orientation of the LIPSS, their fluence dependence as well as their spatial periods and proves the involvement of surface plasmon polaritons.

Double-pulse irradiations are able to reveal those transient excitation stages experimentally. In dielectrics (silica) and semiconductors (silicon), their importance in the LIPSS formation is demonstrated by using (multiple) double-fs-laser-pulse irradiation sequences of variable temporal pulse-to-pulse delays in the fs – ps range. In silica, a characteristic transition of the LIPSS periods from $\Lambda\sim750$ nm$\sim\lambda$ (“metallic” behaviour) to $\Lambda\sim530$ nm$\sim\lambda/n$ (“dielectric” behaviour; $n$: refractive index of silica) is observed for double-pulse delays between one and two ps, indicating the rapid energy relaxation between the two fs-laser pulses and proving the importance of the laser-induced free electron plasma in the conduction band of the solid for the formation of fs-LIPSS.
Femtosecond Laser-driven Waves on Metals

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Abstract: With femtosecond (fs) pulse irradiation, we investigate the structural evolution of a unique type of fs laser-induced periodic surface structure, called nanostructure-covered large scale waves (NC-LSWs), densely covered by iterating stripe patterns of nanostructures and microstructures with a period of tens of microns. We find that the surface morphology of NC-LSWs crucially depends on the fluence of the laser, the number of irradiating pulses, and the incident beam angle. These experimental observations allow us to establish a three-step model for the NC-LSW formation: the formation of laser-induced surface roughness, inhomogeneous energy deposition due to the interference between the refracted light and the scattered light, and nonuniform energy deposition due to shielding by the peaks of LSWs. Moreover, we also reveal that the nonuniform energy deposition induces the selective ablation and subsequent mass transfer on the NC-LSW surface, resulting in the propagation of asymmetric NC-LSWs on metals.
Femtosecond laser pulse train induced periodic surface structures adjustment based on transient localized electron dynamics control

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This study reveals that the periods, areas and orientations of periodic surface structures (ripples) in fused silica can be adjusted by designing femtosecond laser pulse trains to control transient localized electron dynamics and material properties, for which the fundamentals of laser-induced periodic surface structures (LIPSS) formation mechanisms are discussed. It is found that: 1) at the fluences of 1Fth-1.05Fth (where Fth is the single pulse threshold) and 10 bursts, low spatial frequency LIPSS (LSFL) are obtained at the pulse delay of Δt=0fs (as shown in Fig. 1(a)) and split in the peaks at Δt=50fs (as shown in Fig. 1(b)), leading to additional high spatial frequency LIPSS (HSFL) orientated parallel to LSFL and laser polarization, which is due to the second-harmonic generation; 2) the periods of LSFL and those of HSFL are close to the fundamental and second-harmonic wavelengths in the bulk material, respectively, which indicates that the second-harmonic generation is the key in triggering the LIPSS transition at Δt=50fs; 3) at higher fluences of 1.1-1.16Fth and 20 bursts, LSFL, obtained at Δt≤50fs (as shown in Figs. 2(a) and 2(b)), are replaced by orthotropic HSFL at Δt=100-500fs (as shown in Figs. 2(c) and 2(d)), which is attributed to surface plasmons generation and the periodic enhancement of the coupled electric field at the surface. The profiles of the periodic structures in Figs. 2(a) and 2(d) are shown in Figs. 3(a) and 3(b), respectively. Our previous study shows that the subsequent subpulse of the train significantly impacts free electron distributions generated by the previous subpulse(s), which means transient localized electron dynamics can be changed by shaping femtosecond pulse trains. This then adjusts the corresponding mechanisms of photon absorptions and phase changes, which controls the formation mechanisms of ripples and the surface morphology. The dependence of the LIPSS morphology on the pulse delay provides a new method to obtain controllable and smaller nanogratings.

Fig. 1 SEM images of LIPSS morphology evolution on fused silica surface after irradiation with 10 pulse trains (two pulses per train) at a total fluence of 1.05Fth. The polarization direction is indicated by the arrow.

Fig. 2 LSFL and HSFL at a pulse delay of (a) 0fs, (b) 50fs, (c) 100fs and (d) 400fs, after irradiation with 20 pulse trains (two pulses per train) at a total fluence of 1.16Fth.

Fig. 3 (a) AFM profiles of LSFL in Fig. 2(a). (b) AFM profiles of HSFL in Fig. 2(d).
Atomistic modeling and experimental study of single-pulse femtosecond laser ablation, spallation and damage of Ag and Al targets

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The processes responsible for the ablation onset and surface modification in short pulse laser interactions with Ag and Al targets are investigated in large-scale simulations performed with a model combining the molecular dynamics method with a continuum description of laser excitation, electron-phonon equilibration, and electron heat conduction. Although the general mechanisms of photomechanical spallation and ablation of metal targets have been investigated in a number of earlier atomistic simulations and have largely been established, the appearance of recent experimental results on single-pulse laser ablation of Ag and Al provide new opportunities for making a direct link between the computational predictions and experimental observations. The focus of this combined computational-experimental study is two-fold:

(1) to provide a physical explanation of the fluence dependence of the ablation depth in single-pulse laser ablation of Ag and to clarify the controversial issue of the contribution of “ballistic electrons” to the energy redistribution during the time of electron-phonon non-equilibrium in noble metals;

(2) to explain the appearance of a permanent hump (or “swelling”) on the surface of Al and Ag targets irradiated by a 100 fs laser pulse at fluences below the threshold for laser ablation.

Large scale atomistic simulations provide information on the kinetics of melting and resolidification of the surface region, reveal the fluence dependence of the depth of the region affected by the void nucleation and growth in laser spallation, and help to design a predictive analytical model for the description of laser ablation and damage at fluences close to the ablation threshold.

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Self-organized nanoripples on metals surface induced by femtosecond laser

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Abstract

Self-organized nanostructures have been obtained on metal surface during femtosecond laser ablation, the effect of laser fluence, pulse number and laser polarization on nanogratings is investigated in this paper. Under appropriate laser fluence and pulse number, orientation of gratings can be controlled effectively by varying the incident laser polarization. In addition, characterizing approaches including EDAX and XRD are applied to study modified surface compositions. Both detection results indicate that oxygen element in atmosphere is not coupled into the modified surface even the laser irradiation process is performed in ambient air. Our research is expected to offer some valuable clues for manufacturing controllable and high-quality self-organized nanogratings by femtosecond laser.

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Plasmonic structures in close proximity to strongly correlated materials can be used both to control and to probe phase transitions in those materials. In this paper, we demonstrate this concept for nanocomposites comprising a gold nanodisk (ND) array covered by vanadium dioxide (VO$_2$) film. Potential applications for this kind of experiment range from proximal probes of phase-changing materials to enhancing the efficiency of opto-electronic and opto-thermal devices using PCMs.

Arrays of Au nanodisks (NDs) 20 nm thick and 180 nm in diameter, with a pitch of 450 nm, were fabricated on ITO-covered glass substrates by electron-beam lithography. Subsequently, 60 nm of VO$_2$ was deposited on the Au ND array by ablating a vanadium-metal target in an oxygen ambient, yielding amorphous VO$_2$ on the glass substrate. Annealing the composite material at 450˚C for 45 minutes produced a stoichiometric VO$_2$ film covering the Au ND array. The quality of the film on a witness sample was checked by static optical measurements of hysteresis and electron microscopy.

While the IMT was induced by mechanically chopped, low-power laser radiation at 785 nm, the evolution of the IMT as a function of laser power was probed at 1550 nm. At this size, the surface-plasmon resonance of the Au NDs occurs near 785 nm; the NDs can be considered as nanoantennas that scatter the absorbed laser light with increasing efficiency as the IMT progresses. This has the effect of inducing the transition at a substantially faster rate, as illustrated in Figures 1 and 2. The presence of the Au NDs reduced the threshold for the IMT by 37% compared to that of the bare VO$_2$ film produced in the same way, because the plasmonic absorption peak shifts from 1100 nm to 785 nm as the VO$_2$ turns metallic.

The electromagnetic interaction between the Au NDs and the VO$_2$ suggests the possibility of employing the NDs as plasmonic nanoantennas, but in receiving mode, to monitor the electron dynamics in the VO$_2$ film via the plasmon response as the IMT is induced thermally. By measuring the hysteresis during the thermally induced transition and fitting the optical response in the region of strong correlation with a Lorentz-oscillator model, we deduce that the plasmon lineshape is homogeneously broadened, and that the dephasing time $\tau_2$ in the metallic state is reduced by about 30% when the IMT is complete. Possible applications of this nanoantenna effect to the study of chemical-interface damping will be discussed.

Optical Manipulation and Spectroscopy of Macromolecules on Plasmonic Nanostructures

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LSP-based optical trapping is quite intriguing and is currently attracting much attention in nano-photonics and related research fields, since optical trapping based on surface plasmon can potentially overcome several disadvantages of conventional optical trapping technique\textsuperscript{(1)}, (i) the conventional technique requires an intense focused laser light and a complicated optical set up to manipulate a small nanoparticle, and (ii) the spatial resolution in the trapping is, as a matter of course, regulated to be more than several hundreds nanometers by a diffraction limit of an incident light.

In the present study, we have succeeded in optically trap nanoparticles of soft matters such as DNA (l-DNA) and on the basis of resonant excitation of plasmonic nano-antenna. An aspect of such behavior is schematically drawn in Figure 1. Characteristic features involving energetic balance, spectroscopic and dynamic behavior of such plasmon-based optical trapping, i.e., nanoparticle around the metallic nano-gap, will be discussed in detail on the basis of microscopic measurements and theoretical analysis.

\textbf{Figure 1.} Schematic illustration of optical trapping of DNA by surface plasmon.

Theoretical treatment of ultrashort pulse laser processing of transparent materials: What is energetically and mechanically meaningful?

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Ultrashort laser pulses are a powerful tool for modifying the structure and properties of transparent materials. Depending on material properties and irradiation conditions, a wide variety of modifications can be induced such as surface and bulk periodic structures (nanogratings), densification with associated refractive index change, microvoids and void chains, phase transitions, etc. This gives rise to numerous technological applications based on 3D photonic structures in bulk optical materials (waveguides, Bragg gratings, Fresnel zone plates, rewritable optical memories, and others). Among transparent materials, optical glasses are of prime importance for optoelectronics and photonics due to their relatively low cost, processability, and possibility of governing refractive index and inducing optical anisotropy. The physics behind laser-induced glass modifications is extremely rich and involves the multiplicity of the consecutive processes initiated by radiation absorption during the laser pulse and extending to millisecond time scales when the final structure becomes “frozen” in the glass matrix. While tremendous achievements have been made toward laser-writing techniques and assembling integrated optics, the physical mechanisms underlying glass modifications have not been fully understood. The exigency of controllable generation of desired structures requires deeper insight into of the mechanisms and spatiotemporal dynamics of laser-induced glass transformations.

In this report, we will review the physical processes and mechanisms responsible for various forms of glass modification. Different approaches for modeling ultrashort laser pulse propagation in transparent materials will be critically assessed. The dynamics of laser-induced creation of free electron plasma inside bulk glass will be analyzed, depending on the irradiation conditions. A contradictory issue on the free electron density generated in glass materials upon laser irradiation will be addressed with reviewing the existing theoretical results and experimental evaluations based on application of the Drude theory. The results of modeling will be presented obtained on the basis of the Maxwell’s equations supplemented with the equations describing electron plasma generation and the laser-induced electric current. We will demonstrate that the model allows following important features of laser beam propagation in the regimes of tight focusing and dense electron plasma generation when unidirectional approximations such as the non-linear Schrödinger equation do not provide adequate description. Based on this model we have studied spatiotemporal dynamics of laser beam propagation with self-focusing, free electron generation, and plasma-induced defocusing on the example of fused silica glass under particular irradiation regimes employed for laser direct writing. As a result, the geometry of the laser energy absorption zone is determined and the glass temperature is mapped which may be foreseen at the end of electron – glass matrix relaxation. This, in turn, allows estimating the laser-induced stress levels and making conclusions on the routes of glass modification. Finally, based on the performed analysis, we consider the energy balance, matching the free electron energy and temperature with several threshold values (melting, plastic deformation, material failure with void formation, sublimation).

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Ultrafast laser induced index change in glass and new directions in glass photonics


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Localised refractive index changes, varying in size from a few microns to tens of microns, can be induced inside most glass substrates upon exposure to the tightly focussed output of a femtosecond or picosecond laser. The degree with which the index is changed, its sign and the glass chemistry underpinning these changes varies depending on the type of glass irradiated and the irradiation conditions. For example, the index change induced in silica and silicate glasses is typically positive in sign and falls within the range of $10^{-4}$ to $5 \times 10^{-3}$, a range that is similar to that of conventional optical fibre cores. Consequently, ultrafast laser direct writing in silica and silicate glasses can be exploited to produce a diversity of guided wave components such as splitters, Mach Zehnder interferometers, waveguide arrays, vibration sensors, 3D interconnects and integrated waveguide - microfluidic platforms. Phosphate glasses are also compatible with this direct writing technique, however, additional fabrication challenges arise because both negative and positive index changes can be induced [1]. Nevertheless, doped phosphate glasses (and some doped silicate glasses) are an attractive host for directly written active photonic devices [2] because they can accommodate high concentrations of rare earth dopant.

Insights into the nature of the ultrafast laser induced changes to the glass chemistry continue to evolve through techniques such as Raman spectrometry, photo-luminescence measurements, refractive index profilometry, annealing, etching, and optical and electron microscopy [3 -6]. Index change is now known to result from effects such as modification of the fictive temperature, bond breaking and reformation, generation of colour centres, material migration and internal stress. These insights now influence both the engineering of tailored made glass compositions and the laser writing parameters. In this presentation our current understanding of ultrafast laser – glass lattice interactions will be reviewed, I will show how these studies influence new opportunities in 2D and 3D glass photonics.

Optical properties of dielectric materials during femtosecond laser ablation
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A self-consistent model describing the interaction between a dielectric material and ultra-short laser pulses has been developed previously by our group [1]. The model is based on the multiple-rate equations model by B. Rethfeld [2] and has been extended to include propagation of the optical field into the dielectric material, such that it describes the electronic excitation and the light absorption throughout the dielectric material. The optical properties of the dielectric changes during illumination. The complex refractive index determines the specific optical properties and is in the Drude approximation given by

$$\tilde{n} = n + i\kappa = \sqrt{n_0^2 - \frac{\omega_p^2}{\omega_i^2 + i\omega_i \Gamma}},$$

where $n_0$ is the refractive index of the dielectric before excitation, $\omega_p = \sqrt{\frac{e^2 N_e}{m_e \epsilon_0}}$ the plasma frequency, $\omega_i$ the laser frequency, and $\Gamma$ the electron scattering rate. $\epsilon_0$ is the vacuum permittivity, $m_e$ the electron mass, and $e$ the elementary charge. $N_e$ is the free electron density, which increases during the pulse due to multi-photon and tunnel ionization as well as collisional/avalanche ionization. To account for the fact that there is a finite density of electrons in the valance band, the model includes a saturation term, which ensures that $N_e$ does not increase above the initial valence band density. The electron scattering rate, however, also changes during excitation when the electrons are heated via inverse bremsstrahlung. In the model, the scattering rate is assumed to consist of contributions from a (constant) electron-lattice scattering rate and a temperature dependent electron-electron scattering rate (based on a classical gas collision rate). The saturation term and the temperature-dependent scattering rate are omitted in most models, but in the current presentation, it will be shown that these effects must be included in order to reproduce the observed behavior of the optical properties at high laser intensities.

Figure 1 shows the experimentally measured reflectance as a function of fluence together with simulations. The data show that, as expected, the reflectance increases for increasing fluence due to the creation of the free electron plasma. At even higher fluence, the reflectance does, however, decrease again. The reflectance is directly related to the refractive index by $R = \left[1 - \tilde{n}/(1 + \tilde{n})\right]^2$, so this – maybe initially somewhat surprising – behavior can be understood from our model by examining the expression for the refractive index: At high fluences, the majority of the valence electrons are excited to the conduction band during the pulse at which point the plasma frequency saturates. However, the heating of the electrons by plasma absorption continues, which in our description increases the scattering rate and thereby eventually reduces the reflectance. This experimentally observed decrease in reflectance can therefore only be reproduced by the model when including the saturation term and the temperature dependent electron scattering rate.

![Fig. 1. Measured (blue points) and simulated (red line) reflectance of sapphire sample.](image-url)

Ways to assemble nanomaterials from colloidal nanocrystals to obtain novel optical properties are described, along with the use of optical methods to analyze these materials. Specifically, the fabrication of ordered arrays of nanocrystals is probed by using small angle x-ray scattering. Hybrids of nanocrystals and carbon nanotubes are seen to have anomalous photoluminescence Stokes shifts due to hot luminescence caused by FRET. Raman scattering is used to probe the reactivity of ceria-Au nanocrystals.
Scanning Particle Lens Array Laser Micro/Nano Fabrication of User Defined Periodic Patterns

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Abstract

The presentation reports large area surface micro/nano patterning of user defined periodic micro/nano structures on flat and curved surfaces using laser scanning particle lens array system recently developed at the author’s research group. Self-assembled transparent microspheres are used as focusing micro-lenses and an off-axis scanning beam was used to produce micro/nano patterns. To produce patterns on curved or hydrophobic surface, a special technique of particle transfer is used. Sub-surface patterning is also made possible by water immersion.
Colloidal-particle-lens-arrays-assisted
surface nanopatterning by harmonics of femtosecond laser

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The monolayers of colloidal micro-particles can be used for nano-patterning of material surfaces. Here the dielectric nanospheres serve as the near-field lenses when irradiated by the laser pulse [1,2]. Nano-structured surfaces can be employed for numerous applications in photonics and biomedicine. Theoretical considerations and experimental data demonstrate the complex character of laser field distribution caused by the sets of micro-spheres evidencing that the cross-scattering between the spheres is significant [3-5].

In the present communication, we report on formation of periodic pit and/or hillock nanostructures on different substrates when deposited dielectric microparticles arrays are irradiated by the single femtosecond pulses of fundamental frequency (FF), of the second harmonic (SH), and by the bi-chromatic, FF+SH pulses. In our experiments we used titanium sapphire laser system “Spitfire-Pro” in single shot regime. Pulse duration was 50 fs, energy of single pulse was 1.7 mJ, central wavelength was 780 nm, beam diameter was 7 mm. Parabolic lens (focal length was 15 cm) was used for beam focusing. The BBO crystal was used for SH generation with maximal efficiency of 5%. It was put just after the lens to avoid space separation of FF and SH pulses [6]. We demonstrate the accurate 100nm pit structures resulted from irradiation of the close-packed arrays of polystyrene micrometer-sized spheres deposited on polymethylmethacrylate substrates by the SH single pulses. Fluences were changed by moving the sample along the axis of the focused beam. The estimated fluences were in the range of 40-50 mJ/cm² (peak intensity was near 1 TW/cm²).

The similar structures are obtained when irradiating the same spheres-substrate systems by the bi-chromatic pulses with fluence of SH in the range of 20-30 mJ/cm² (peak intensity was 0.5 TW/cm²) and with total fluences of both FF and SH pulses 0.4-0.6 J/cm² (peak intensity was about 10 TW/cm²). In this case, the high-quality hillock arrays on the vitreous-glass substrate were also obtained. The irradiation by only FF beams appeared to be less effective.

We analyzed above experimental results taking into account ionization processes both within the spheres and within the substrates. The corresponding calculations are performed using FDTD (Finite Difference Time Domain) electrodynamic codes and considering bi-chromatic ionization process with the help of approach developed in [7].

Biopatterning through laser induced forward transfer

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The capability to spatially control the deposition of biomolecule containing solutions onto different types of substrates is important for the development of sensors and biosensors, throughput screening apparatuses, biochips, lab-on-a-chip, and tissue engineering. However, the fabrication of microstructures with well-ordered and spatially discrete forms is difficult because of the lack of distinct physical and chemical barriers separating patterns.

In particular, one suitable technique for the deposition of organic, polymeric, and biological materials in liquid phase, with high lateral resolution is laser-induced forward transfer (LIFT). In LIFT, a laser beam is focused through a transparent support plate onto the backside of a metallic thin film coated with the material to be transferred (donor film). Each single laser pulse promotes the transfer of the thin film material onto a receiver substrate that is usually placed parallel and facing the thin film at a short distance.

Up to now, LIFT proved to be well suited for microarray printing, i.e. printing patterns of individual droplets onto flat substrates. However, for the applications presented above it is of paramount importance the production of 2D patterns, i.e. the formation of uniform and stable liquid lines, and printing of such patterns onto different types of substrates.

In this work it is investigated the influence of the main process parameters on the formation of lines of protein solutions though LIFT for the development of odor sensor arrays. 2-port resonator surface acoustic wave (SAW) sensors have been used as receiver substrates, and the recognizing material placed by LIFT as sensing element is bovine odorant protein (BOP). As the functionality and performance of such sensors strongly depends on the uniform coverage of their active surface with the sensing protein, the goal of the study is the determination of the process parameters allowing printing a uniform layer of protein solution which completely covers the active area of the sensors. Once this goal is achieved, the sensor arrays are submitted to functional characterization which proves the feasibility of the technique.
Symmetric damage patterns in glass from highly asymmetric laser beams

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High intensity femtosecond laser pulses can show a spontaneous improvement in spatial mode quality while propagating in air, a process known as “spatial mode cleaning”. It is believed to result from nonlinear processes in the air, such as self-focusing. We have conducted experiments with glass slides placed in the laser focus to “photograph” the spatial mode distribution through the damage patterns produced on the glass. With this technique, we have observed very dramatic mode improvement. A “half round” laser beam is found to still produce nearly perfect circular rings at some points in the focus. Conversely, a laser beam with a vertical or cross-shaped section removed from the center shows very complex dynamics as the slide is translated through the focus. Of note, these results are obtained in a “tight focusing” geometry where self-focusing is expected to play less of a role. These results may have applications for laser-writing on glass, as well as a fundamental tool for studying filament dynamics.
Spatially selective modification of magneto-optical properties and nanostructures in Fe$^{3+}$- and Al-doped transparent glasses by using femtosecond-laser irradiation

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High-intensity ultrashort-pulse laser has attracted much researcher’s attention and widely studied as a powerful tool applicable to control of various physical properties, such as optical, electrical, mechanical and magnetic properties. In particular, space-selective modification of magnetic properties by the direct irradiation with femtosecond (fs) laser is challenging, which provides a unique and expansible advantage of a direct 3-dimensional processing inside transparent materials. In our previous paper, we have demonstrated a spatially selective modification of optical and magnetic properties in Fe- and Al-codoped glasses irradiated with fs-laser. In this paper, we have attempted to investigate the possibility of the magneto-optical changes and to evaluate the nanostructures using transmission electron microscopy (TEM). The glass with the composition of $70\text{SiO}_2\cdot20\text{Na}_2\text{O}\cdot10\text{CaO}$ (mol/%) doped with $0.05\text{Fe}_2\text{O}_3$ and $0.1\text{Al}$ was prepared by melting and quenching method. Near-infrared fs laser ($=775$ nm) was focused 1 mm below the surface of glass samples, resulting in the absorption peaks due to hole-trap centers at around irradiated region. After anneal at 450 °C, the color of the irradiated area inside the glass was changed into faint yellow, which was due to the localized surface plasmon resonance (LSPR) absorption at around 400 nm. In the meanwhile, the magnetization curves at room temperature show that the magnetization was locally enhanced after the irradiation with fs-laser and the subsequent annealing. And then, the magneto-optical Faraday rotation angles were measured as a function of wavelength, showing that the LSPR negative peaks appear at around 400 nm (Fig. 1). This means that a ferrimagnetic Faraday response was enhanced by LSPR due to plasmonic nanoparticles, suggesting a direct coupling between plasmonic and magnetic nanoparticles in a visible region. Figure 2 shows the bright-field TEM image of the glass irradiated and annealed at 550 °C. Two different phases can be observed; one is dark contrasted nanoparticles with the diameter of 2–4 nm. The other one is relatively larger particles with the size of about 5 nm, which shows clear diffraction patterns. It is speculated that the nanocomposites are composed of the Al nanoparticles and ferrimagnetic magnetite nanoparticles. Therefore, the plasmonic enhancement of Faraday response is undoubtedly due to the nanoparticle pair precipitated by irradiation with fs-laser and subsequent annealing.

![Fig. 1](image1.png)  
**Fig. 1** Difference spectra of Faraday effect between the as-prepared glass and the glass irradiated and annealed at 450 °C (a) for 30 min. and (b) for 60 min, and (c) at 550 °C for 30 min.

![Fig. 2](image2.png)  
**Fig. 2** Transmission electron microscopy image of the glass irradiated with fs-laser and annealed at 550 °C.
Polarization dependent femtosecond laser ablation of PMMA

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Abstract: We show that ablation features in PMMA induced by a single femtosecond laser pulse are imposed by light polarization. The ablation craters are elongated along the major axis of the polarization vector and become increasingly prominent as the pulse energy is increased above threshold energy. We demonstrate ~ 40% elongation for linearly and elliptically polarized light in the fluence range of 4 - 20 J/cm² while circularly polarized light produced near circular ablation craters irrespective of pulse energies. We also show that irradiation with multiple pulses erases the polarization dependent elongation of the ablation craters. However, for line ablation the orientation of the electric field vector is imprinted in the form of quasi-periodic structures inside the ablated region. Theoretically, we show that polarization dependence of the ablation features arises due to local field enhancement during light-plasma interaction. Simulations also show that in materials with high nonlinearities such as doped PMMA, in addition to conventional explosive boiling, sub-surface multiple filamentation can also give rise to porosity.
Ultrafast Laser Processing for Fabrication of Microfluidic and Optofluidic Devices

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The rapid development of ultrafast laser is giving rise to advancement in materials processing, and the ultrafast laser processing is becoming a common tool for various applications. One of the most promising and attractive applications is fabrication of microfluidic and optofluidic devices since it can modify the interior of glass in a spatially selective manner due to multiphoton absorption. The laser-irradiated regions inside glass where the chemical property is modified can be selectively etched away by successive wet etching using chemical aqueous solutions such as diluted hydrofluoric (HF) acid, and eventually three-dimensional microfluidics is directly formed inside a glass chip. This two-step process (i.e., ultrafast laser direct writing followed by wet chemical etching) can also be used to integrate free-space optics such as micromirrors and micro-optical lenses inside glass. The ultrafast laser direct writing can also alter the optical properties of the substrate to create a wide range of micro-optical components inside glass materials, including optical waveguides, Mach-zehnder interferometer, optical attenuator, etc. The unique ability of ultrafast laser direct writing to simultaneously alter both the chemical and optical properties permits to integrate functional micro components in a single glass chip without much difficulty in alignment of each component, and thus opens up a new avenue for fabricating a variety of microfluidic and optofluidic devices for chemical and biological analysis. This talk introduces techniques based on ultrafast laser technologies for fabrication of such micro devices. Then, practical demonstrations of the fabricated micro devices such as highly sensitive analysis of the concentrations of liquid samples and determination of the functions of living microorganisms are presented.
Superwicking surfaces produced by direct femtosecond laser ablation

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Using high-intensity femtosecond laser pulses, we create a surface micro-groove patterns that transform regular surfaces of solids to superwicking. Due to the created surface structure, a liquid sprint vertically uphill in a gravity defying way. Our study of the liquid motion shows that the fast self-propelling motion of the liquid is due to a capillary effect from the surface structures we created.
Micro- and nanostructuring of soft organic matter by temporally shaped femtosecond laser pulses

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Processing techniques based on laser irradiation and ablation provide high precision at the micro- and nanoscale and afford accuracy and control to fabricate new smart, functional nanomaterials. In particular, nanofabrication of soft polymeric matter [1,2] is of high interest in technological areas of organic photonics and electronics, biomedicine and bioengineering. The use of ultrashort femtosecond (fs) laser pulses is highly attractive for micro- and nanofabrication because it improves the spatial resolution to enable patterning with nanometer scale precision. Also, optimal energy coupling to the substrate is possible by temporally shape fs pulses to match with the corresponding time scale of processes involved [3]. This offers new avenues for controlling and tailoring the features of the created structures and gives the possibility to guide the material response towards user-designed directions.

In the present study, thin films of the biopolymers gelatine and chitosan, and of the synthetic polymer poly (trimethylene terephthalate) were irradiated using fs pulse shaping techniques combined with a microscope-based setup for material processing. The polymer films were irradiated with laser pulses of 35 fs and a central wavelength of 790 nm provided by an amplified Ti:Sapphire system. The effect of unshaped and temporally shaped pulses, with quadratic and cubic phases, on the micro- and nano-induced morphology was analyzed by characterization of the created surface structures with scanning electron microscopy and atomic force microscopy. We observed different material modification thresholds for temporally asymmetric pulse shapes, and different kinds of induced structures (voids, bubbles, pores and periodic structures or ripples). The results indicate the possibility of control of the generated nanostructures and are discussed in reference to the primary processes induced by ultrafast laser excitation that occur on differentiated fast time scales and involve nonlinear electronic excitation (multiphoton ionization and avalanche ionization), energy transfer to the lattice and phase transitions.

Cold ablation of metals driven by hot electrons

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High-intensity femtosecond laser irradiation of metals heats the electronic subsystem by few electron-volts, while the ionic subsystem remains cold for several picoseconds until electron-ion energy exchange equilibrates both sub-systems. P-V equation of state of such a two-temperature material with $T_e > T_i$ lies higher than the cold pressure curve of metal with $T_e = T_i = 0$. The significant increase of pressure with increase of $T_e$ in a cold lattice can be formally attributed to the growth of “electron” pressure, which is assumed to be zero at $T_e = 0$. The large enough “electron” pressure can result in ejection of cold matter from the irradiated free frontal surface. In order to capture effects of “electron” pressure in molecular dynamics (MD) simulations of femtosecond laser ablation, a novel electron-temperature-dependent EAM potential was developed for Ni. Charge density and embedding energy functions were fitted to electron and ion pressures for different densities and electron temperatures obtained from first-principles density functional theory calculations. The new EAM potential accurately reproduces the mechanical and thermodynamic properties of two-temperature Ni in a wide range of pressures (from -30 GPa to 800 GPa) and electron temperatures (0-5 eV).

Our combined 2T hydrodynamic modeling and MD simulation show that the significant “electron” pressure in a skin layer is produced at early stage of femtosecond laser ablation, thereby creating a strong tensile wave at the film’s frontal surface. If the “electron” pressure effect is great enough, it may cause cold matter to be ejected from the target’s surface on the depth of skin layer. Once electron-ion thermal equilibrium is reached, the conventional thermomechanical mechanism of ablation takes over. It leads to formation of a crater with depth few times larger than the thickness of skin layer.
Thermal response of semiconductor tip during femtosecond laser-assisted field evaporation

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During the laser-assisted field evaporation the atoms are extracted from the surface of the needle-shape specimen (a tip) due to the joint action of the high static electric field and femtosecond laser pulse [1]. At the fixed field, the measured evaporation rate can give information on the time and space temperature distribution of the tip after the interaction with the laser pulse [2].

In this work, we theoretically and experimentally investigate the temperature response of a silicon tip to the femtosecond laser pulse in the presence of high electric field. The developed theoretical model is based on the 2D carrier transport and two-temperature approaches with the 3D laser absorption map, computed by FDTD method.

We show that temporal evolution of the tip temperature depends on the two effects. First, the external static field leads to the hole accumulation and fast high temperature rise after the hole-phonon coupling at the surface of the tip. Second, the bulk carrier distribution and relaxation follows the laser field interference pattern in the tip. This, in turn, leads to the non-homogeneous temperature distribution.

References
Multi-electron dissociative ionization of clusters under ps and fs laser irradiation: the case of alkyl-halide clusters

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The Multi-electron dissociative ionization (MEDI) of alkyl-halide clusters induced by 35 ps (at 266, 532 and 1064 nm) and 20 fs (at 400 and 800 nm) laser pulses is reported. In most cases the MEDI of clusters, is observed at substantially lower laser intensities than those reported for the monomer molecules, while the fragment ions are released with higher kinetic energies Figure 1. For ps laser pulses, single cluster ionization is achieved through multi-photon absorption process. At $\lambda = 266$ nm the dominant dissociation mechanism found to be the AID mechanism, while at 532 and 1064 nm clusters disintegrate due to Coulomb explosion process. In the latter case the combined action of the laser and the internal electric field results to the formation of multiply charged ions at laser intensities far below than those expected from theory and with high kinetic energies. In the fs regime the MEDI of neutral clusters was confirmed for both 400 and 800 nm. As it was expected multi-photon absorption processes found to be favoured for 400 nm while field ionization processes mediated the interaction at 800 nm. For $\lambda=800$ nm field ionization processes resulted in the formation of ions from different fragmentation channels, which also exhibited different angular distributions. Ions with high kinetic energies, originating from the combined action of the laser and the internal electric field exhibit isotropic angular distribution, while those originating from the screening of the internal electric field, due to the collective oscillation of the electron cloud inside the cluster environment, found to possess anisotropic angular distribution perpendicular to the laser polarization vector. On the contrary, ions with low kinetic energies, originating from an electron impact ionization process, are ejected anisotropically, with maximum of their distribution parallel to the laser polarization vector. The comparative study between ps and fs laser pulses revealed that in both cases and for all wavelengths applied, single cluster ionization is achieved through multi-photon absorption process. Furthermore, for fs laser irradiation the cluster geometry remains almost unchanged during the interaction because of the ultra-short duration of the pulse. Due to this, the suppression of the intracluster potential barriers are lower than in the case of ps laser pulses and so the appearance of the same multiply charged fragments at laser intensities two orders of magnitude higher in the case of fs laser pulses is conceivable. Also, the higher charge multiplicities that were recorded for ps laser pulses are interpreted on the same grounds. Finally, it seems that the cluster density is a critical parameter and responsible for the increase of the appearance intensity thresholds as the molecular chain length increases when lighter halogen atoms are participating in the molecular skeleton [1].

Figure 1 Comparative study of the interaction ethyl-iodide and ethyl-chloride clusters in the ps and fs time domain.

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Coherent magnetic vortex motion in optically-formed channels for easy flow in YBa$_2$Cu$_3$O$_{7-x}$ superconducting thin films

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We report our investigation of electric and magnetic properties of partially-oxygen depleted channels for easy vortex motion in the superconducting YBa$_2$Cu$_3$O$_{7-x}$ (YBCO) films. The channels were induced by means of a laser writing technique. Absorption of a focused, low-intensity laser radiation by the YBa$_2$Cu$_3$O$_{7-x}$ material in an inert ambient atmosphere results in the temperature increase in the illuminated area and partial reduction of the oxygen content ($0.5 > x > 0.1$). The latter is called, the laser-writing mode. The laser-written regions exhibit weak superconductivity, i.e., the suppressed superconducting critical temperature $T_C$, critical current density, and the critical first magnetic field, as compared to the laser-untreated areas. However, if the laser power is above a certain threshold, the same processing process leads to the irreversible orthorhombic-to-tetragonal phase transition and the YBCO becomes non-superconducting (laser-patterning mode). Using the above laser processing method, we have patterned 50-µm-wide and 100-µm-long YBCO microbridges crossed with either one or two, 5-µm-wide laser-written channels. In the case when the microbridge was either current biased or exposed to the external magnetic field, these weak-superconductivity channels were penetrated by the magnetic field in a form of Abrikosov magnetic vortices. Increasing the bias current, the self-produced magnetic field created the Lorentz force that exceeded the pinning force and the vortices could move coherently along the laser-written channels. A coherent motion of vortices was confirmed by a direct observation of the quasi-Josephson-like current steps on the bridge’s current-voltage characteristics in a narrow temperature range from 0.95 $T_C$ to 0.97 $T_C$. Our results demonstrate that the laser-writing technology can be very successfully used for forming artificial channels of easy vortex penetration in high-temperature superconductors, allowing for a precise control of nucleation and motion of vortices in pre-assigned places of the superconducting YBCO film.
Formation of brookite-type TiO\textsubscript{2} layer by irradiating laser pulses onto a titanium plate in high-temperature, high-pressure water

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We have found that a TiO\textsubscript{2} layer with a brookite-type crystal structure is formed when a titanium plate is irradiated by YAG laser pulses in high-temperature, high-pressure water.

We used a special chamber which was resistant to a high pressure up to 35 MPa and a high temperature up to 500 °C. We installed a titanium plate in the chamber which was filled with distilled water. The chamber had five windows. We injected YAG laser pulses at a wavelength of 1060 nm via one of the windows, so that the titanium plate became irradiated by the YAG laser pulses from the normal direction. We analyzed the surface of the laser-irradiated titanium plate by Raman scattering, X-ray diffraction, and scanning electron microscopy.

It was observed that the surface of the titanium plate was oxidized significantly when it was placed in high-temperature, high-pressure water. The oxidized layer had a rutile-type crystal structure in the case of supercritical water at a pressure of 30 MPa and a temperature of 430 °C, while in the case of subcritical water at the same pressure, the crystal structure of the oxidized layer was anatase. No significantly-oxidized layers were formed when a titanium plate was placed in the water at temperatures lower than 300 °C.

When a titanium plate was irradiated by YAG laser pulses in the water at a pressure of 30 MPa and a temperature of 230 °C, we observed the formation of a brookite-type TiO\textsubscript{2} layer. It was observed in the inside and just outside (<0.5 mm) of the laser-irradiated area. The outside region of the laser-irradiated area had an anatase-type crystal structure when the water temperature was higher than 300 °C. On the other hand, we observed a rutile-type TiO\textsubscript{2} layer in the laser-irradiated area when a titanium plate was irradiated by YAG laser pulses in supercritical water. It should be mentioned that no significantly-oxidized layers were observed even in the laser-irradiated area, when a titanium target was irradiated by YAG laser pulses in water at 0.1 MPa and at room temperature.

In summary, we have found a laser-aided method for synthesizing brookite-type TiO\textsubscript{2} layers on titanium plates. This is a remarkable synthesis method since it is a “one-step” process which needs no chemicals.

\textbf{Figure 1} Raman spectrum of a titanium plate irradiated by YAG laser pulses in subcritical water at a pressure of 30 MPa.
Laser-Based Guided Acoustic Waves Propagating at Surfaces (2D) and Edges (1D)

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The interest in ultrasonic guided waves covers a wide field, from nondestructive evaluation (NDE) and structural health monitoring (SHM) of industrial systems to basic research on high-quality low-dimensional waveguides. In conventional applications of ultrasonics to macroscopic objects such as pipes and rails there is a tendency to replace bulk waves by guided waves, which is connected with reduced cost, less inspection time, and greater coverage. These ultrasonic waves are constrained by the boundaries of the complete structure and consist, for example, of torsional wave modes in the case of a tube. Permanent sensor installation for guided long distance diagnostics in rails, is a new approach in SHM. On the other hand, also sophisticated small waveguide systems are of increasing interest, where the surface (2D) or the edge (1D) of the struture and not the whole structure guides the non-dispersive ultrasonic wave.

In recent years photoacoustics opened the door to new applications of 2D linear surface acoustic waves (SAWs), e.g., nondestructive evaluation (NDE) of surface-breaking cracks. Real partially closed cracks of micrometer size have been analyzed. Pulsed laser excitation of solitary elastic surface pulses and their detection with a continuouswave probe laser has been achieved, by generating dispersion with a thin film coating that introduces a length scale. In addition, such laser-based pump-probe experiments allow the excitation of short nonlinear SAW pulses developing steep shock fronts that fracture brittle materials such as silica or silicon. It is possible to measure the fracture strength of materials and compare the critical failure stress with ab initio calculations of the ideal strength of perfect single crystals. While linear SAWs have found widespread use in NDE, there are currently no industrial applications of laser-based nonlinear SAWs.

The excitation and detection of 1D edge or wedge waves propagating along a wedge formed by two planar surfaces that meet at the apex of the wedge or wedge tip has been performed by laser irradiation. The characteristic features of the non-dispersive linear wedge waves such as their small phase velocity below the Rayleigh velocity, the very high degree of localization of the displacement field and elastic energy at the wedge tip, and their existence for certain geometries in anisotropic media such as silicon could be verified by laser-based experiments. Despite the expected strong nonlinearity of certain edge-localized modes, 1D solitary waves and strongly nonlinear wedge waves could not be detected up to now. Future applications include monitoring of defects of cutting tools or turbine blades, sensor applications measuring the velocity change upon surface modifications, ultrasonic motors, stirring and acoustic streaming in fluidics, and aquatic propulsion.
Enhancement of laser-induced ultrasound (LIUS) signal using reduced graphene oxide coating

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Nanosecond pulse laser generates acoustic wave on the water-material interface. Absorbed beam energy heats the material and results in thermoelastic expansion. The thermal stress of a material is dependent on the absorbance and expansion coefficient of a material. In this work, we coated reduced graphene oxide to help absorption of beamed energy on a thin aluminum film of high thermal expansion coefficient. A laser shadowgraph shows enhanced acoustic wave propagation at ~1500 m/s under water. The measured pressure of laser-induced ultrasound is enhanced by a factor of 80 compared to the measurement with an aluminum film alone. The effect of reduced graphene oxide coating is examined with different thicknesses and wavelengths. The frequency of laser-induced ultrasound is determined by thermoelastic response. Strong intensity and broad bandwidth of laser-induced acoustic wave suggest enhancement in repetition time and high resolution for biomedical imaging.

Fig. 1 The pressure of a laser-induced acoustic wave is increased with the laser energy and the thickness of reduced graphene oxide coating.
Designs of photonic crystal nanocavities for stimulated Raman
scattering in diamond

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Abstract: Diamond is a good candidate for producing Raman laser due to its high first order Raman gain coefficient. Since its Raman shift (~1332.5cm⁻¹) is large compared to other solid-state materials, it is possible to produce a Raman frequency converter using diamond crystals. Photonic crystals can be employed for confining photons within periodic structures, the scale of which is on the order of the incident wavelength, making it convenient for integrating all-optical circuits. Combining the merits of both diamond and photonic crystals, we present two designs of photonic crystal nanocavities (in hexagonal and square lattice patterns) which can produce stimulated Raman lasing with low threshold power. After optimizing the photonic bandgaps, triple resonant modes with high $Q$ and small modal volume are realized in each design by tuning the radii of dot defects in nanocavities. As an example Figure 1 below shows the hexagonal photonic crystal and its Raman spectrum when pumped by a 532nm source. Numerical simulations show that for such a design, the threshold power of the pump ranges from tens to hundreds of nano-Watt for generating Raman lasers. Similar results are obtained for photonic crystals of square lattice. The flexibility of the designs together with the biocompatibility of diamond may make our designs applicable in bio-sensing systems.

Figure 1. (Left) A hexagonal photonic crystal lattice of diamond containing triple resonant modes with high $Q$ and small modal volume; (Right) Stimulated Raman spectrum giving Stokes and anti-Stokes lines when pumped by a 532nm laser source.
Light-Matter Interactions: Applications in Photonics and Biophotonics

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Focussed ultrashort laser pulses can modify the local refractive index of certain materials, significant research has been expended into using ultrafast lasers to fabricate integrated optical devices. Integrated optical waveguides – the optical analogue of wires – can be simply fabricated by translating the sample in the path of such short optical pulse trains, which effectively amounts to writing the desired optical circuit in a controlled way in that sample. This direct-write approach offers several key benefits over conventional fabrication techniques. It neither requires use of expensive clean room facilities, nor involves complex film deposition and subsequent etching processes. This technology can also yield 3D structures, unachievable through conventional techniques.

In my talk I will present how the ultrafast laser inscription technology can be used to develop components like switches, splitters/combiners, amplifiers, etc. I will also describe how to exploit new materials (laser and highly nonlinear materials) such that several all-optical devices could be monolithically integrated on the same substrate in the form of optical integrated circuits for biophotonic applications.
An Experimental and Simulation Study on Femtosecond Laser Tuning of Silicon Microring Resonators

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We have recently demonstrated femtosecond (fs) laser pulses can be used as a tool for post fabrication tuning of silicon on insulator (SOI) microring resonators [1]. This technique is able to tune the operational wavelength of the SOI resonators bi-directionally to a desired wavelength by varying the laser fluences to induce amorphization or ablation in the silicon waveguide. In this paper, we will present detailed studies on this technique.

In order to obtain a desired resonant operational wavelength, step by step multiple shot tuning of a resonator is required. The growth of amorphous silicon (a-Si) under multiple shot laser excitation is studied by measurement of optical properties and Raman spectroscopy, and the ablation threshold on a-Si will be compared with that of crystalline silicon (c-Si). In our previous study the fs tuning did not introduce significant change in the quality (Q) factor of the SOI resonator [1]. However the SOI microring used in the previous study had only a moderate Q factor. In this work SOI microrings with higher Q factors are studied to help us understand the effect of ablation and/or amorphization on resonator loss.

The experimental results on the evolution of amorphization and ablation threshold of silicon will also be compared with numerical simulations [2] for a better understanding of the processes involved.

References


High temporal contrast femtosecond petawatt Ti:sapphire laser facility and its applications

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Abstract:

The recent technical progress on the femtosecond petawatt Ti:sapphire laser are presented, which include the peak power and temporal contrast enhancement. With an upgraded pump energy of 110J in the final Ti:sapphire amplifier, the output energy of 50.8J was achieved, corresponding to the conversion efficiency of 46.1%. The peak power of 1.26PW could be obtained with the pulse duration of 29fs and compression efficiency 72%. Meanwhile, pulse clean technique combining optical-parametric amplification (OPA) and second-harmonic generation (SHG) was applied to improve the temporal contrast of the PW laser facility. The ASE contrast was promoted from original $\sim 10^8$ to $\sim 10^{11}$ (measurement-limited).

Besides the laser technologies developments, the PW laser facility, which was built in 2007, has been applied widely in many research fields, such as ultra-high-power femtosecond laser propagation and filamentation in air, table-top fusion and brilliant neutron source generation, laser driven cascaded wakefield acceleration of electron beams and laser wakefield acceleration of electron beams with capillary discharge waveguide. The significant applications achieved from the petawatt laser facility are summarized.

At present, based on the high contrast femtosecond petawatt Ti:sapphire laser facility, proton generation experiments with foil targets are being carried out. The efficient protons generation driven by this laser can be expected in the near future.
Writing gradient index lenses in ophthalmic polymers using near-infrared femtosecond laser pulses

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Abstract: Near-infrared femtosecond laser pulses were used for micromachining in ophthalmic polymers. Refractive index changes up to 0.05 were obtained, and lateral gradient index lenses were written inside flat polymers, inducing astigmatism up to 0.8 diopters.

OCIS codes: (140.3390) Laser materials processing; (160.5470) Polymers; (320.7110) Ultrafast nonlinear optics.

Femtosecond laser micromachining is unique in the capability to write three-dimensional microstructures inside bulk materials, such as glass or polymers. When femtosecond laser pulses are tightly focused inside transparent material, the laser intensity inside the focal volume is so high that it can cause nonlinear effects and modify the material locally without causing collateral damage. Refractive index changes in glass have been induced for the application of writing three-dimensional photonic devices. The refractive index changes obtained previously were in the range of $1 \times 10^{-4}$ to $1 \times 10^{-2}$ [1-2]. In 2006, it was found that large refractive index changes up to +0.06 could be induced in some hydrogel polymers for ophthalmic applications, by using a high-repetition-rate low-pulse-energy femtosecond laser oscillator [3].

Here we report our recent studies on femtosecond laser micromachining in one of the ophthalmic polymers that is FDA-approved for vision applications. We obtain large refractive index changes at high scanning speeds in this polymer, and demonstrate writing of lateral gradient index lenses inside flat polymer samples.

![Fig. 1. Experimental setup for femtosecond laser micromachining](image1.jpg)

A mode-locked Ti:Sapphire laser (Mai Tai HP, Spectra Physics) is used in the present experiment setup. The laser produces femtosecond laser pulses with 100 fs pulse width at 80 MHz repetition rate tunable around 800 nm wavelength. The average power of the laser output is up to 2.5 W, and is attenuated by a high-energy zero-order attenuator. With some proper beam shaping, the near-infrared laser pulses at 800 nm are further attenuated by a metallic variable attenuator, and finally focused through a high NA microscope objective into the sample (Fig. 1). The objective is set to compensate spherical aberration and create a nearly diffraction-limited focal spot at various depths into the sample. The average laser power at the objective focus is typically reduced to below 400 mW. The sample is a type of ophthalmic hydrogel polymer (Akreos®, Bausch & Lomb), one of the materials for intraocular lenses (IOL) currently on the market. The polymer sample was immersed in Balanced Saline Solution (BSS, Bausch & Lomb) overnight before any experimental procedure is performed. During the femtosecond laser micromachining process, the sample is sandwiched between a glass slide and a coverslip, and maintained in BSS solution (Fig. 1). Then the sandwich structure is mounted horizontally on a three-dimensional scanning stage platform. The laser pulses are focused at about 150 µm depth, and we write three-dimensional structures inside the polymer sample, based on calibrated refractive index change profile as a function of scanning speed (Fig. 2). By optical modeling and fabricating a gradient index lens, we have achieved cylindrical power up to 0.8 diopters inside flat polymer samples.

![Fig. 2. Plot of refractive index change induced by femtosecond laser micromachining as a function of scanning speed.](image2.jpg)

A Twyman Green interferometer and Shack-Hartmann wavefront sensor was used to measure the cylindrical power of the gradient index lens fabricated by femtosecond laser micromachining.

References
Laser synthesis of QDs from precursors inside electro-conductive polymeric films

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The inorganic/organic composites with quantum dots (QDs) and organic functional molecules are promising materials for their applications in light emitting diodes (LEDs) due to their size-tunable optoelectronic properties [1]. Recently, a novel method of quantum dots (QDs) formation directly inside a polymer matrix using a laser source has been demonstrated [2]. This method is particularly attractive to obtain the nanocomposite material within selected regions of a polymer required for the device design.

The realization of functional devices needs to overcome several problems facing the preparation of the polymer/precursor film and the correct laser processing parameters to initiate the QDs formation. To verify the formation of the QDs inside the polymeric matrix several combinations of polymer/precursors films were treated with a picosecond laser at wavelength of 266 nm. Precursors for CdS and CdSe QDs were used in experiments.

The structural studies of laser-irradiated samples carried out by means of transmission electron microscopy (TEM) showed the QD formation. The QDs were well distributed in the film. However, they were collected to clusters including 10-60 QDs of different size. The mean size of QDs was less than 10nm. The size of QDs and the clusters depended on the laser irradiation dose transferred to the film.

The optical analysis carried out by means of UV-VIS and optical microscopy confirmed the formation of the QDs after laser processing. The time-resolved photoluminescence revealed the energy transfer from the organic host to QDs. However, the charge separation was present due to a certain energy level alignment. Modification of the polymer/precursor blends is still required to prevent imbalance of carrier injection to QDs. Photo-luminescent spectroscopy and fluorescence microscopy have revealed that even if the QDs are not emissive, in certain polymer/QDs combinations the PL emission of the polymer is restored after laser treatment.

Surface nano-structuring produced by ultrashort infrared and X-ray laser pulses

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Short laser pulse with wavelength from infrared to X-ray produces pressure in a thin surface layer. This pressurized layer decays acoustically into vacuum from the frontal boundary of a target and into the bulk side of a target. The decay and corresponding expansion of matter of the pressurized layer produce tensile stress $\tau$ which stretches matter of this layer during acoustic decay time $t_s = d_T / c_s$, where $d_T$ is thickness of the pressurized layer. Stress $\tau$ increases when absorbed energy $F_{abs}$ increases. Spallation layer separates from a target if stress $\tau$ overcomes material strength $\tau_{str}$. Simulations show that there is a molten layer with thickness $\sim d_T$, when an illumination is powerful enough to achieve the threshold condition $\tau = \tau_{str}$. Therefore, nucleation of voids takes place in liquid. A spallation layer is located between a nucleation zone and vacuum boundary. This layer does not separate shortly after nucleation. In our case the thickness $d_T$ of the spallation layer and its inertia (proportional to mass per unit of surface) are so small that after nucleation the deceleration of a spallation layer continues. This means that the cavitation bubbles continue to exist and therefore their walls continue to resist to stretching through surface tension. Near threshold $\tau = \tau_{str}$ the resisting stage is so long in time that cooling due to heat conduction into cold bulk freezes the bubbles. The undersurface bubbles, frozen jets with drops, and remnants of walls of broken bubbles form random nanorelief at a target vacuum boundary. This conclusion follows from our recent experimental results and numerical simulations.
Ultrafast diagnostics of photo-excited processes in solid using femtosecond laser-based soft x-ray pulse sources


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A variety of applications of the ultrashort pulse laser-solid interaction to the laser-processing technology has stimulated great interest in a comprehensive understanding of the initial stage of the photo-excited dynamics induced by a high-intensity ultrashort laser pulse [1]. Recent progress in high-brightness ultrashort pulse sources in the wavelength region from the extreme ultraviolet (EUV) to the soft x-ray based on high-intensity femtosecond laser has opened the door to new ultrafast diagnostic techniques for exploring such photo-excited processes. Both short wavelength and ultrashort pulse duration of such sources are expected to provide complementary information in the photo-excited processes to conventional time-resolved ultrafast laser spectroscopy techniques, while experimental realization of such techniques is still challenging.

Here, we present a series of dynamical studies for the photo-excited processes in semiconductors and metals by using various time-resolved soft x-ray spectroscopy techniques. We used two kinds of the laser-based ultrashort soft x-ray sources generating a region of wavelength approximately from 10 to 20 nm: femtosecond-laser-produced plasma and high-order harmonic sources. Typically, the former source shows a quasi-continuous emission spectrum with a picosecond pulse duration, while that of the latter one shows a sharp spectrum with a femtosecond duration. Therefore, we developed various time-resolved soft x-ray spectroscopy techniques by utilizing the above characteristics of each source appropriately.

For the dynamics of laser melting process, which typically proceeds on picosecond time scale, we developed the time-resolved x-ray absorption fine structure (TR-XAFS) spectroscopy based on the laser-produced plasma soft x-ray source [2]. The quasi-continuous spectrum is suitable for the absorption spectroscopy. Since XAFS provides information on the local electronic state, which is sensitive to a chemical bonding or a local atomic structure, TR-XAFS has the potential to become a powerful tool for investigating both laser melting and laser ablation processes where phase transformation, bond breaking, and particle ejection occur instantaneously. For the laser ablation process, in particular, the process of the ejection of ablated particles, we developed the TR-XAFS imaging technique by combining the TR-XAFS spectroscopy technique and x-ray microscope [3]. These techniques enabled us to measure a bond length expansion during the melting process, and clearly identify each ablation particle in the ablation plume and generation of nanoparticles [4].

In addition, we successfully measured a photo-excited electron and hole relaxation dynamics by developing a femtosecond time-resolved photoelectron spectroscopy system based on the 59th harmonic pulse (13.5 nm) source [5]. This technique provided us a transient change of the surface potential due to the spatial separation of the electron-hole pair generated by a laser excitation, and clarified dynamical properties of photo-excited electron and hole transport and their recombination on surface. Thus, ultrafast diagnostic techniques based on ultrashort EUV and soft x-ray pulse sources are promising tools for investigating dynamical behavior of the photo-excited processes in solids.

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References
Time- and angle-resolved photoemission spectroscopy using a femtosecond high-harmonic light-source

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Rapid progress in ultrafast X-ray science worldwide, both in high-harmonic and X-ray free electron laser sources, has paved the way for a completely new generation of real time experiments investigating ultrafast processes in all areas of science. Femtosecond and attosecond pulses are now available spanning the extreme ultraviolet and soft X-ray regions of the spectrum that are perfectly synchronized to a pump laser pulse.

After an introduction to high-harmonic generation (HHG) and survey, the impact of using these table-top light-sources for the study of ultrafast material science will be shown. For instance, the laser assisted photoelectric effect (LAPE) could be demonstrated for IR-XUV excitation of a Pt(111) surface [1] as well as laser assisted Auger decay (LAAD) [2]. Recent scientific breakthroughs employing X-ray pulses from HHG in the areas of surface dynamics [3], correlated-electron materials [4], and heterogeneous magnetic materials [5,6] will be discussed. The review concludes with a summary and an outlook to the feasibility of real time studies of photo-induced phase transitions in a broad class of advanced correlated materials.

High harmonic generation spectroscopy of molecules in excited states

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High Harmonic Generation (HHG) for molecules presents a rich set of new physical phenomena, as both the ionization and electron recombination steps of HHG are dependent on the particular symmetry of the active orbital and its orientation with respect to the laser field. Because of this orientation dependence, HHG in molecules provides a unique time resolved ‘probe’ of the electronic orbital structure. Recent work has shown that the HHG spectrum from aligned molecular samples can be used to determine the molecular structure for small molecules and can be used for imaging of the electron rearrangement in the dissociating bromine molecules.

This contribution is devoted to detailed studies of HHG process in molecules in excited states, beyond ‘single active electron’ approximation. We study the influence of multielectron effects on HHG spectra and phases of harmonics. We analyze in detail the ellipticity of harmonics, compare the results with the ground state results and present the influence of different orientation of the molecule with respect to the polarization of the laser electric field. Finally we discuss the influence of the laser pulse length, wavelength and intensity on the process of HHG in molecules. We gratefully acknowledge funding from National Science Foundation (award PHY-1068706).
Spatial resolution in multiphoton laser polymerization

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Multiphoton polymerization is a powerful tool for 3D nano-structure fabrication by means of direct fs laser writing. Here, the sharp, threshold-like response of a material to laser exposure provides opportunity to create nanofeatures with sizes that are smaller than the diffraction limit. The percolation-like transition is a possible physical mechanism that allows such a nonlinear spatial confinement in the case of laser polymerization.

Monte-Carlo 3D modeling of percolation with the spherically symmetric occupation probability distribution corresponding to the laser beam [1] shows the dramatic increase in the fluctuations of the size and position of the largest connected cluster (voxel) when attempting to decrease its size below the critical scale. For laser polymerization, this provides the natural fluctuation-managed limitation of the minimal size of a nanofeature. We present the analytical estimation of the critical size of the largest cluster, which fits well with the data obtained from the numerical experiments.

We investigate the role of diffusion of mobile particles participating in the laser polymerization process.

We theoretically analyze the limitations imposed by diffusion of small active radicals created due to initiation process on the formation of two separate nanofeatures such as voxels, rods and plates. We develop a theoretical approach by consecutively taking into account diffusion of radicals of growing length [2]. This approach allows us to estimate a characteristic spatial scale which determines the spatial resolution for the particular polymerizing system.

In the present communication, we show that contrary to diffusion of active radicals, the diffusion of quencher molecules can be used for significant improvement of spatial resolution of laser polymerization technique. We present a new single-beam method for increasing the resolution of direct fs laser writing by multiphoton polymerization, based on quencher diffusion. This method relies on the combination of a mobile quenching molecule with a slow laser scanning speed, allowing the diffusion of the quencher in the scanned area and the depletion of the multi-photon generated radicals [3]. We show that this method provides opportunity to minimize both the size of the elementary feature and the distance between the features. The material we use is an organic-inorganic hybrid, while the quencher is a photopolymerizable amine-based monomer and part of the polymer backbone upon fabrication of the structures. We use this method to fabricate woodpile structures with a 400 nm intralayer period. This is comparable to the results produced by two-beam direct laser writing based on stimulated-emission-depletion (STED) microscopy, the method considered today as state-of-the-art in 3D structure fabrication [4]. We optically characterize these woodpiles to show that they exhibit well-ordered diffraction patterns and stopgaps down to near-infrared wavelengths.

Diffuse reflectance spectroscopy from photon-excited doped anodized aluminium

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Optical properties of anodized aluminium alloys were determined by optical diffuse reflectance spectroscopy of such films. Samples with different concentrations of dopants were excited with a white-light source combined with an integrating sphere for fast determination of diffuse reflectance. This diffuse UV-VIS reflectance of Ti-doped and Zr-doped anodized aluminium films was measured over the wavelength range of 200 nm to 900 nm. Titanium doped-anodized aluminium films with 5-15 wt% Ti, and zirconium-doped anodized aluminium with ~10-15 wt% Zr were characterized. Changes in the diffuse light scattering of doped anodized aluminium films, and thus optical appearance, with doping are discussed. Both titanium and zirconium doping decreases the reflection intensity and the color of the titanium doped films become more bluish with doping. Using the Kubelka-Munk model [1] on the diffuse reflectance spectra of such films, the bandgap $E_g$ of the oxide alloys can be determined.

Pulsed Laser annealing of nanoparticles for thin film solar cells: 
Multiphysics simulation

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Abstract

Nanoscale size dependence of laser nanoparticles interactions is first discussed in this chapter. Nanoparticles (NPs), due to their extremely small physical appearance, are quite different from their bulk counterparts in physical properties, including melting point, heat capacity, thermal and electrical conductivities which have been discussed in this chapter. Finite element analysis (FEA) multiphysics model coupling electromagnetic (EM) sub-module and thermal heat transfer (HT) sub-module is built to reveal the mechanism of laser-NPs interactions. EM module represents laser-NPs interactions and HT module simulates the heat conduction among NPs. Accordingly experiments are carried out to verify the validity of EM-HT models. Both simulation and experimental results showed that smaller NPs have stronger interactions with incident laser beams higher Plasmon resonance; extremely small NPs (<50 nm) usually have significant lower melting points compared with their bigger counterparts. Direct pulsed laser crystallization (DPLC) of photoactive materials, such as copper indium diselenide (CIS) and cadmium telluride (CdTe) and direct pulsed laser recrystallization (DPLR) of transparent conductive oxide (TCO) materials, such as alumina-doped zinc oxide (AZO) are examined from the theoretical aspect. Both DPLC and DPLR could be regarded as interactions between laser and photoactive materials, laser and transparent conductive oxide materials are examined in this chapter. It is found that due to size effects, (1) smaller nanoparticles have lower melting point than their bulk counterparts thus lower processing temperature is needed to complete DPLC and DPLR processing when smaller nanoparticles; (2) Smaller nanoparticles have stronger interactions with incident laser beams which ends up with higher resulting temperature field. This indicates that smaller nanoparticles require smaller laser fluence in DPLC and DPLR processes; (3) Selectivity of DPLC/DPLR processing is demonstrated by EM-HT simulation. When target materials experience high temperature, other underlying materials (such as Molybdenum, supporting substrates) only have low temperature up to 500K in most cases studied. This indicates that DPLC and DPLR are promising in processing polymer and/or paper based flexible solar cells or other electronic devices. Without the loss of generalness, the nanoscale size effects on melting points, thermal and electrical conductivities, specific heat capacity found in this article could be extended to many other materials including metals, semiconductors, and polymers.
High-sensitivity electro-optic polymer probing system using photo-isomerization and Fabry-Perot effects

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The external electro-optic (EO) probe technique has been a very useful tool for the characterization of high-speed devices and circuits. For most external EO probing techniques, inorganic EO crystal materials, GaAs, LiTaO₃, and BSO, are used. These materials have many disadvantages such as large dielectric constants, low EO coefficients, high cost, and are difficult to machine. As EO polymer material offers a solution to these problems, they have become much more attractive and have been utilized in EO probing applications [1]. However, the EO effect of the polymer material may decay and hence sensitivity of the polymer probe may degrade due to molecular dipole orientation relaxation. We have experimentally demonstrated that using a pumping laser to induce the photo-isomerization effect in a pre-poled EO polymer can enhance and maintain the noncentrosymmetric molecular orientation [2]. In this paper, this effect is further combined with the Fabry Perot effect by using a tunable laser. Experimental results of an EO sensor made of DR1/PMMA show that this combination effect can greatly improve the sensitivity. The experimental set-up is as shown in Fig. 1. The EO polymer prober is consisted of a DR1/PMMA film on a high reflection mirror for the probing tunable laser. The pumping laser is incident on the DR1/PMMA film from the backside of the prober to induce the photo-isomerization effect. The electric-field was applied to the EO polymer by a pair of ITO electrode. Experimental result is as shown in Fig. 2. Wavelength of the tunable laser was varied from 682 nm to 683 nm. It can be seen that the Fabry-Perot effect produce the peak EO signal at the resonance wavelength and the pumping laser can greatly further improve the signal magnitude. Therefore, sensitivity of this EO polymer prober system is enhanced.

Reference

Growth and characterizations of nanostructured tungsten oxides

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Tungsten oxide (WO₃) nanostructures as nanostructured thin films and clusters-assembled are attractive for sensor applications. We report on WO₃ nanostructures deposited by radio-frequency (RF) assisted laser ablation technique. A tungsten oxide ceramic target was irradiated at different wavelengths; the depositions have been carried out in reactive atmosphere (oxygen gas or a gas mixture of oxygen and argon) on heated alumina substrates up to 600°C. The gas pressure varied between 0.01 mbar and 7 mbar. The influence of laser wavelength, substrate’s temperature, gas pressure, gas composition and RF power on properties of obtained nanostructures was investigated by Atomic Force Microscopy, X-ray Diffraction, Secondary Ion Mass Spectroscopy and spectroellipsometry.
Reduction and cohesion of thick fog by infrared light resonance with H$_2$O molecular vibration using excited carbon materials.

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In this work, we report on the results of thick fog dispersion using high thermal conductivity carbons (HTCCs) (Rahima, TEIJIN) by infrared light irradiation. The thick fog affects some negative things such as traffic accidents increase and damage to farm goods. It is important to discover the technique of thick fog dispersion, however, efficient processes are unreported. In order to disperse thick fog efficiently, we focused on two types of infrared light processes. One is the cohesion and adhesion effect by resonance process. The infrared light is irradiated to the HTCCs. It is noteworthy that HTCCs are excited by infrared light irradiation, and excited HTCCs are emitted characteristic wavelength as a resonator source with H$_2$O molecular vibration. Excited HTCCs can expect to trap H$_2$O molecules in the thick fog and disperse them high efficiency. The other is reduction effect by thermally process. The infrared light is irradiated to the thick fog for dispersion by thermally influence. In this experiment, we prepared the HTCC nets and set up in a chamber. The thick fog (particle size: 12 μm, flow rate: 0.83 ml/sec) was generated by pressured water, and flowed into the chamber. An infrared light source (wavelength: 2.5 μm, power: 0.16 mW/cm$^2$) set in front of the issue of flowing fog at the chamber. The infrared light was irradiated to the flowing fog and HTCC nets as dispersion and excitation sources. Laser diodes (wavelength: 650 nm) were set up in the fog flowed chamber for transmittances measurement. Visibilities in the fog flowed chamber were calculated from transmittances amounts. As results, transmittances and visibilities were improved from 6.64 to 27.80 %, and 8.21 to 19.46 m by infrared light irradiation, respectively.
Abstract for ICPEPA 8

Optical properties of one-dimensional metallo-dielectric photonic crystal (1D MDPC) in ultraviolet region

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Abstract

In this paper, we investigate the transmission properties of a one-dimensional metallo-dielectric photonic crystal (1D MDPC) operating near the plasma frequency of the metallic layer using transfer matrix method. With the permittivity of the metallic layers approaching zero in ultraviolet frequencies, extra-ordinary optical properties were observed. A very narrow passband with a split at the plasma frequency is observed only for the case of TM excitation. The dependence of the transmission spectra on layer thickness of the MDPC and the incident angle is discussed in detail.

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Abstract for ICPEPA 8

Experimental measurement in transmission characteristics of a waveguide based on low frequency spoof surface plasmon polaritons

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Abstract

In this paper, a kind of plasmonic waveguide, which is made of a periodic subwavelength square array of metallic pillars, is investigated both theoretically and experimentally. Based on the guiding mechanism of spoof surface plasmon polaritons (spoof SPPs), the transmission characteristics of this waveguide are controllable by altering the geometric parameters of the periodic structure. Microwave experimental measurement in the transmission coefficient verifies the high efficiency of wave guiding in such open waveguide. Numerical simulations are made to show the consistence with measured results.
Multilaminate and chilarity control of carbon nano-materials by photo excitation light assisted thermal decomposition.

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Carbon nano materials had good electrical properties and attract acceptable attentions for future nano-electrical device use. We had discovered the carbon nanotube (CNT) and carbon nanofiber (CNF) photo assisted growth method in our previous examination. Previous results showed photo assisted CNF samples were linear and thicker than non assisted samples. This indicated photo assisted techniques for carbon nano material production had some impacts to control multilaminate and chilarity of CNTs. In order to develop photo assisted processes more, we use the optical parametric oscillator (OPO) laser as a photo excitation assisted source for controlling multilaminate and chilarity of CNTs.

Main CNTs and CNFs growth method was thermal decomposition. First, Fe particles were deposited on Si substrate by pulsed laser deposition as a metal catalyst for CNTs growth. Second, Si substrate set up into a vacuumed chamber, and passed an electrical direct current (~ 10 A) as a thermal generation source. Third, ethanol gas was flowed into the chamber and decomposed on heated Si substrate. Finally, CNTs and CNFs were grown on the Si substrate by ethanol thermal decomposition. The photo assisted processes were OPO laser irradiation. OPO laser which wavelengths were dependent on chilarity absorptions of CNT were irradiated to the heated Si substrate. In this study, we report on the relationships between generation forms of carbon materials and photo assisted wavelength, and controllability of multilaminate and chilarity of CNTs by photo assisted light irradiation.
Mg-Al based hydrotalcite-like materials and their derived mixed oxides deposited by pulsed laser deposition

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Abstract

Layered double hydroxides (LDHs) also known as hydrotalcite-like (HT-like) materials have been studied extensively in the last decades especially for the perspective of being used as anion exchangers. As thin films they can also be used for applications as catalytic supports, chemical sensors, corrosion-resistant coatings, optoelectronics etc.

We report on the deposition of thin films of Mg-Al layered double hydroxide (LDH) and their mixed oxides by pulsed laser deposition technique (PLD). Dry-pressed pellets of Mg-Al LDH powder (with Mg/Al ratio of 2 or 3), prepared by co-precipitation, were used as targets for PLD experiments.

The formation of a LDH thin film depends strongly on the laser wavelength due to the different optical absorption properties. The surface investigation revealed films with high roughness, therefore the films are attractive in applications where a high surface area is a demand.

The structural and morphological properties of the deposited LDH films were examined by X-Ray Diffraction, Atomic Force Microscopy, Scanning Electron Microscopy combined with energy dispersive X-ray analysis and Secondary Ions Mass Spectrometry. Catalytic measurements were performed for the reaction of cyanoethylation of ethanol with acrylonitrile.
OPTIMIZATION OF SURFACE CHEMICAL MICROPATTERNs FOR SELECTIVE CELL ADHESION EXPERIMENTS USING LASER IRRADIATION

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Naturally occurring chemical gradients are important for cell growth, adhesion and other biological processes. To study them and separate the effects of different molecules, one needs to create controlled chemical gradients [1]. There are a lot of different methods which allow fabrication of surfaces with controlled chemical composition, but most of them have disadvantages preventing efficient structure manufacturing. The aim of this work was to fabricate the gradient self-assembled monolayer (SAM) structures on gold surfaces using the method of submerged laser ablation [2]. The localized laser heating releases molecules from a substrate and molecules from the solution can be adhered. Therefore, by irradiating SAM on a gold surface submerged in a thiol solution we form monolayer structures from different molecules. The gradient structures are formed using solutions with different molar ratios of two different thiols. Using a nanosecond pulsed laser as an irradiation source, gradients of biotin-thiol (Fig. 1) and thiols with carboxylic functional groups were formed in the ethylene-glycol-thiol SAM background layer. The latter gradients were further modified to form fibronectin gradients. Using varying laser parameters, such as the pulse energy and the beam diameter, the SAM structures of desired dimensions presenting a gradient of functional groups can be manufactured.

References:


Deposition of La doped SrTiO$_3$ Films for Water Splitting Photocatalytic Reaction on Flexible Substrates by Laser Induced Forward Transfer

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A photocatalytic reaction is one of the photo excitation processes. In several photocatalytic reactions, a water-splitting reaction is expected to discover for fuel cells and other devices. The Honda-Fujishima effect is a famous as a water-splitting reaction which can generate hydrogen and oxygen gasses from H$_2$O by ultraviolet light irradiation. This reaction is used TiO$_2$ (Eg: 3.2 eV, anatase) powder as a water-splitting source and Pt electrode as a promoter metal. In several water-splitting materials, we had discovered La doped SrTiO$_3$ (Eg: 3.2 eV, SrTiO$_3$) (LSTO) thin films. LSTO thin films can generate hydrogen gases by UV light irradiation. Amount of hydrogen gasses which generated from LSTO films is higher than generated from TiO$_2$ films. In addition, it is important to note that absorption wavelength of LSTO thin films are shifted from UV light area (350 nm) to visible light area (430 nm). LSTO can excite by visible light and is expected to visible light water-splitting. In order to apply this material, we focus on the laser induced forward transfer (LIFT). LSTO films were deposited on base plate by pulsed laser deposition. Nd: YAG laser (wavelength: 532 nm, energy: 0.5 ~ 20 μJ) was irradiated to the back of base plate which deposited LSTO films, and LSTO films were discharged. Discharged materials were transferred onto flexible receiver substrates. Features of LIFT that this method is able to transfer under the atmospheric pressure, and deposited substrates use various. In this report, we discuss about results of LSTO films condition on flexible substrates by LIFT and relationships between hydrogen gasses generation rate and films conditions.
Time-resolved carrier lifetime measurements in thin-film Si-on-glass photovoltaic absorbers

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We have developed a novel, time-resolved spectroscopy system for characterization of amorphous and microcrystalline silicon (a-Si and mc-Si) thin films grown on glass substrates for applications as absorbers for solar cells. Efficiencies of a-Si–mc-Si tandem cells have been reported to be as high as 11.9%, due in part to optimizations to the cell design and the use of light management techniques throughout the device stack. Further improvements, however, require a better control of the growth process of the a-Si and mc-Si films, in order to enhance of their electron-hole recombination time, via reduced defect formation, improved grain-boundary passivation, and reduced unwanted contaminants. We have implemented an all-optical, femtosecond pump-probe spectroscopy method and collected a large family of normalized reflectivity change (ΔT/T) waveforms of various a-Si and mc-Si films, deposited under different conditions, to determine the optimum deposition parameter space. The studied Si absorbers exhibited carrier lifetimes on the order of 10s of picoseconds to nanoseconds; thus, any traditional technique would not be capable of accurately resolving their carrier lifetime dynamics. In order to understand the physical mechanisms behind the observed phenomena, we have adopted a theoretical model based on three, coupled rate equations, describing time relaxation of photo-excited carriers and the efficacy of the traps. The main two channels of relaxation were identified as the carrier trapping and the Shockley-Read-Hall recombination and were implemented in the model. The model fitted the measured ΔT/T transients extremely well, demonstrating a correlation between the growth-induced hydrogen content in the film and the trap concentration, as in a-Si, hydrogen can be present as both Si-H and Si-H₂ species.
Pulsed Laser Deposition of Epitaxial Fe\textsubscript{38.5}Pd\textsubscript{61.5} Ordered Films

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Thin films of 3d-4d/5d metallic alloys such as Fe-Pt, Co-Pt, and Fe-Pd are of technological interest due to their ordered L1\textsubscript{0} tetragonal phase which exhibits high magnetocrystalline anisotropy comparable to that of 3d-4f rare earth magnets. A combination of hard magnetic properties with ductility and corrosion resistance makes this family of alloys ideal for applications including micro-electro-mechanical systems and ultra-high-density magnetic storage. These alloys are known to develop unique microstructures, including a novel strain-induced chessboard eutectoid microstructure featuring exchange coupling effects that has been found between the hard L1\textsubscript{0} and soft L1\textsubscript{2} magnetic phases of the Co-Pt system. Within this class of materials, Fe-Pd alloys possess a somewhat lower magnetocrystalline anisotropy compared to Co-Pt and Fe-Pt, but the Fe-Pd phase diagram showing considerably lower order-disorder transition temperatures renders them well-suited for nanostructured magnetic applications and study.

Epitaxial films of Fe\textsubscript{38.5}Pd\textsubscript{61.5} at the L1\textsubscript{2}-L1\textsubscript{0} eutectoid composition have been grown on MgO 001 oriented substrates by pulsed laser deposition. These films exhibit atomic ordering with increasing temperature, transitioning from the disordered A1 (FCC) phase to the ordered L1\textsubscript{2} phase. Fe\textsubscript{38.5}Pd\textsubscript{61.5} films grown at 550°C have been found to possess a two-phase microstructure of prismatic 50-100 nm disordered A1 secondary phases with 110 oriented facets embedded within an ordered L1\textsubscript{2} matrix. These secondary phases exhibit single domain magnetic axis rotation, while the easy magnetic axis of the ordered L1\textsubscript{2} matrix lies in plane due to strain induced by epitaxy. The growth these two-phase films has been studied as a function of deposition time. The films grown in this study were characterized by x-ray diffraction, vibrating sample magnetometry, atomic and magnetic force microscopy, and high resolution scanning electron microscopy.
Dry patterning of fluorine-doped tin oxide thin films by using square top-hat UV laser beams

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The fluorine with tin oxide (FTO) dopant thin films has been attracted much attention to meet the requirements of high transparency and low sheet resistance. Because the FTO thin films contain no expensive indium (In) element, the cost of this transparent conductive material is in general less than other transparent conductive oxides (TCOs) containing In. The FTO thin films, due to their strong price competition, are accepted to apply as the conductive material for the flat panel displays, touch panels, flexible electronics, dye-sensitized solar cells (DSSC), light-emitting diodes (LEDs), and other optoelectronics products. Furthermore, the traditional electrode patterning of thin films involves photolithography processes. The photolithography equipment is expensive, and requires environment harmful chemicals and complicated steps in the fabrication process.

This study aims to establish a square top-hat beam shaper in an ultraviolet (UV) laser system for the electrode patterning and to investigate the interaction between square laser beams and FTO thin films deposited on glass substrates. The processing parameters including laser fluences, pulse repetition frequencies, and feeding rates of a motorized platform are used to ablate out the FTO films. The laser pulse repetition frequency and the feeding rate of the motorized platform are applied to calculate the overlapping rate of laser spot and to discuss the patterning quality. The surface morphology, edge quality, and three-dimensional (3D) topography for the patterned FTO films are measured and analyzed by a confocal laser scanning microscope. This novel process can reduce the fabrication steps and the quantity of chemical solutions and can improve the removal efficiency of FTO films.
Epitaxial growth and electrical properties of Sb-doped SnO₂ thin film grown by ArF excimer laser assisted metal organic deposition

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Tin oxide is a promising material for next generation electronic devices because it offers good properties, such as high conductivity, transparency, and chemical stability. It is also an abundantly available natural resource. Epitaxial growth of the oxide material is of considerable practical concern because its electrical and optical properties strongly depend on the orientation of the thin film. Therefore, controlling the orientation of the electrode thin film for functional oxide materials is very important. However, in most cases, the processes for oxide epitaxial growth require both vacuum and high temperature, making device production expensive. To decrease the processing temperature, we developed an excimer laser-assisted metal organic deposition (ELAMOD) method. By using ELAMOD, we prepared the epitaxial Sb-doped SnO₂ film on TiO₂ substrate by using the KrF and XeCl laser [1, 2, 3]. In this paper, we tried to prepare on the Sb-doped SnO₂ film by using ArF laser. Also, the effects of the crystal orientation and Sb concentration on the crystal growth and electrical properties were investigated.

Fig. 1 shows XRD patterns of the Sb-doped SnO₂ film prepared by ELAMOD using ArF laser. As can be seen from figure 1, an epitaxial Sb-doped SnO₂ film was obtained by laser irradiation at a fluence more than 100mJ/cm². In addition, all the films on the (110), (001) and (100) TiO₂ substrates was found to be epitaxially grown whereas the lattice mismatching between film and different crystal orientation of the TiO₂ substrates.

The electrical resistivity and carrier concentration and mobility of the film were measured. The resistivity of the (001) Sb-doped SnO₂ film was found to be lower than that of the (001) and (110) oriented Sb-doped SnO₂ films on TiO₂ substrate as show in Fig. 2. In order to control the resistivity, the effect of the concentration of the Sb-doping on the electrical properties was investigated. When the 2% Sb was doped to SnO₂, the lowest resistivity of the (001) Sb-doped SnO₂ films on (001) TiO₂ substrate is 2.80×10⁻³Ωcm. Detail electrical properties such as mobility and carrier concentration will be presented.

Fig. 1 XRD patterns of the Sb-doped SnO₂ film prepared by ELAMOD.

Fig. 2 Resistivity of the (001), (110) and (100) Sb-doped SnO₂ film by ELAMOD.

Pinning of fullerene LUMO edge at the interface with standing up Copper Phthaloacyanine

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Abstract

The electronic structure evolution of interfaces of fullerene (C₆₀) with copper phthalocyanine (CuPc) on highly oriented pyrolitic graphite (HOPG) and on native silicon oxide have been investigated with ultra-violet photoemission spectroscopy (UPS) and inverse photoemission spectroscopy (IPES). The LUMO edge of C₆₀ was found to be pinned at the interface with CuPc on SiO₂. A substantial difference in the electron affinity of CuPc on the two substrates was observed as the orientation of CuPc is lying flat on HOPG and standing up on SiO₂. The ionization potential and electron affinity of C₆₀ were not affected by the orientation of CuPc due to the spherical symmetry of C₆₀ molecules. We observed band bending in C₆₀ on the standing-up orientation of CuPc molecules, while the energy levels of C₆₀ on the flat lying orientation of CuPc molecules were observed to be flat. The observation points to a dependence of photoexcited charge transfer on the relative molecular
orientation at the interface.
Pulsed laser deposition and annealing of superhard tetrahedral amorphous carbon, cubic boron nitride and nanocrystalline diamond films

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Tetrahedral amorphous carbon (ta-C) cubic boron nitride (c-BN) and nano-crystalline diamond (n-D) films were prepared by pulsed laser deposition using a KrF excimer laser for the ablation of the film forming species. The conditions under which the various films form will be presented. The growth rates, microstructure and mechanical properties of those films will be discussed and compared to other deposition methods and the special advantages of the pulsed laser deposition method will be emphasized.

It will be shown that ta-C, c-BN and n-D films with high hardness in the range of 45 – 65 GPa can be deposited at high growth rates. As especially ta-C and c-BN films show high internal stresses due to the high energy of the film forming particles, which results in poor adherence, the preparation of micrometer thick films of those materials requires a suitable method of stress reduction during the deposition process. We developed a special pulsed laser annealing technique, which is applied alternating to the deposition process of thin sub-layers. It will be shown that stress-free ta-C films with 80 to 85 % sp³ bonds, 60 – 65 GPa hardness and thicknesses in the µm-range can be prepared by using a KrF-excimer laser of 248 nm wavelength for annealing. In the case of c-BN films, a F₂-laser of 157 nm wavelength has to be used for annealing, where the stresses could be reduced by some 50 %, so far.

The great advantage of the method in comparison to conventional thermal annealing is the short time it requires to completely remove stress (the process itself requires only a few µs) and that it is possible to go over directly from deposition to annealing and vice versa. The latter is of particular significance for the deposition of ta-C films as the substrate temperature during actual film growth must not exceed 90 °C.

Some examples of coated components and tools will be shown and the industrial potentialities of the method will be discussed taking into account the commercially available lasers as well as the costs.
The Effect of Electrolytic Oxyhydrogen Gas Addition on LPG Combustion:

a Laser Induced Breakdown Spectroscopy Study

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Keywords: Laser induced breakdown spectroscopy (LIBS); Electrolytic oxyhydrogen (EOH); Exhaust reduction; Combustion

The electrolytic oxyhydrogen (EOH) gas derived from water by electrolysis is added to the liquefied petroleum gas (LPG) flame. Figure 1 shows the flame images for increasing EOH flow rates at a fixed LPG concentration. The flame structure is changed and the light scattering from the exhaust decreases with increasing EOH flow rate. We conducted the emission spectroscopy for analyzing the flame and flame exhaust. Several key properties of a combined LPG and EOH flame, such as combustion species, flame exhaust gases, and flame temperature were measured using the LIBS procedure, a conventional gas analyzer, and a thermocouple. Here we confirm significant reduction of exhaust emissions when the EOH is added to a LPG flame from the LIBS analysis.

Fig. 1 Flame images for varying EOH gas flow rates at a fixed LPG concentration (10 L/min). (a) 1 L/min (b) 2 L/min (c) 3 L/min (d) 4 L/min (e) 5 L/min (f) 6 L/min (g) 7 L/min.
Dynamic study of charge asymmetric dissociation in strong laser fields

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In this study of molecular excitation, ionization and dissociation by strong laser irradiation, we compare the dynamics of double-ionization induced charge asymmetric dissociation (CAD) in two isoelectronic diatomic molecules, CO and N\textsubscript{2}. With ultrahigh temporal resolution time-of-flight measurements, a strong intensity dependent kinetic energy release (KER) in the charge-neutral CAD channel C\textsuperscript{2+}+O is observed. This is interesting because one of the two fragments is a neutral atom. In comparison, its counterpart channel N\textsuperscript{2+}+N has nearly constant KER. Further analysis shows that the C\textsuperscript{2+}+O channel is predominantly produced through a sequential process whereas the N\textsuperscript{2+}+N channel involves a nonsequential transition, and their different dynamics mainly originate from their different detailed electronic structures.
Applications of Transparent Conducting Oxides in Plasmonic/Optical Modulators

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Due to the poor electro-optic (EO) properties of conventional materials, ultracompact EO modulators have become one of the critical technical bottlenecks that impeding the wide applications of on-chip optical interconnects. These conventional modulators usually have dimensions on the order of tens of micrometers or even millimeters. However, on-chip optical interconnects require EO modulators at the nanoscale. Recently we found that light absorption can be greatly enhanced in epsilon-near-zero-slot structure even when the slot width is less than 1nm [1], in which graphene works as tunable epsilon-near-zero (ENZ) material. ENZ materials are promising EO materials as they have advantages: (1) sharply enhanced absorption can be achieved in an ultrathin slot; (2) the ultrathin slot does not introduce a large insertion loss; (3) the ENZ material often has tunable optical properties because a small change in carrier density may result in a significant change in dielectric constant.

Based on the experimental results of Ref. [2], we demonstrate that ultracompact plasmonic and optical modulators can be achieved in a slot waveguide structure by employing indium tin oxide (ITO) as the ENZ materials filled in the 10nm-thick slot region. Without external voltage, the carrier concentration of the ITO is \( N_1=1.0 \times 10^{21}/\text{cm}^3 \), and the dielectric constant \( \varepsilon_1 \), which is far-from-zero at a specific wavelength. With a suitable voltage, we assume that the ITO is divided into two 5nm-thick layers, the carrier concentration in the 5nm layer closer to the insulator increases to \( N_2 \), and the other layer keeps the carrier concentration as \( N_1 \). The accumulated carriers result a close-to-zero dielectric constant \( \varepsilon_2 \) at the same wavelength. Note that the magnitude of the dielectric constant has changed \( \varepsilon_2/\varepsilon_1 \approx 23.4 \) times by only 1.0V voltage. From the results of three-dimensional finite difference time domain (FDTD) simulations, a 3dB modulation depth can be achieved in a 150nm-long plasmonic waveguide and a 200nm-long dielectric waveguide, as shown in Fig 1, respectively. The tunable ENZ-slot waveguides may enable EO modulation at nanoscale and an optical modulator can be made at the scale of a transistor.

![Figure 1](image-url)

Figure 1: (a) The illustration of an EO modulator embedded in a plasmonic rib waveguide. (b, c) The 3D simulation of light propagation between a plasmonic rib waveguide and the EO modulator at \( N_1=1.0 \times 10^{21}/\text{cm}^3 \) and \( N_2=1.65 \times 10^{22}/\text{cm}^3 \), respectively. (d) The illustration of an EO modulator embedded in a dielectric rib waveguide. (e, f) The 3D simulation of light propagation between a dielectric rib waveguide and the EO modulator at \( N_1 \) and \( N_2 \), respectively.

References
Femtosecond laser-induced porosity in PMMA

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Femtosecond laser ablation of polymers is a promising technique for technological and biomedical applications such as surface patterning to enhance implant union and cell adhesion. In this study, femtosecond laser, with wavelength of 800nm, pulse duration of 40 fs, and with fluence near the ablation threshold, has been utilized to change the surface properties of Poly(methyl methacrylate) (PMMA) for the purpose of cell biology. The surface of PMMA has been ablated to make periodic micro-holes, as well as to induce nanopores by drawing lines on the surface of PMMA. In this study, influence laser parameters such as pulse energy, number of pulses, and laser polarization on the shape, depth of pores, and their distribution is studied.
Optimization of direct femtosecond laser written holograms

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The feasibility of writing computer-generated holograms directly on surfaces by ultrashort laser pulses has been demonstrated previously by e.g. our group [1]. The ultrashort laser pulse duration provides a high writing resolution due to minimized heat-propagation effects [2]. In addition, the technique is highly versatile, since the method can be applied on all flat, reflecting materials. The current presentation describes a systematic investigation of changes in the hologram properties when structural properties of the laser-written areas are controlled.

Holograms are written on polished copper surfaces using 800 nm, ~120 fs laser pulses from a 1-kHz-repetition-rate laser system. The sample motion is done by computer-controlled stages. The writing speed has increased significantly compared to Ref. 1 by applying in one direction a fast, precise, magnetically-driven air-bearing stage. Using the position-sensitive output from the stage controller, the individual pixels of the computer-generated hologram can be written by a single laser pulse during the stage motion. The positional accuracy is better than one micrometer.

An example of an image reconstructed from a laser written hologram is shown in Fig. 1(a). If the hologram efficiency is characterized by the intensity of the holographic reconstruction [i.e. the intensity of the pattern in Fig. 1(a)], this efficiency can be measured as a function of the hologram parameters. For instance, Fig. 1(b) shows the intensity versus the diameter of individual laser holes for a fixed pitch of 6 μm. The red line represents a fit to a model, which assumes that the efficiency is dependent on the relative area covered by the laser-written pixels. The maximum intensity in Fig 1(b) corresponds to more than 20% of the intensity in the central (0th order) peak being transferred into one image.

In the presentation, we will also show the dependence on the hologram pitch as it is varied from 2 to 8 μm, while the effect of applying phase randomization will be discussed qualitatively. Finally, we will discuss the potential of the method for marking and anti-counterfeiting applications.

Fig. 1. (a) An example of a reconstructed image used to characterize the efficiency. (b) Measured intensity of the holographic reconstruction as a function of the hole size.

A two-stage physics-based model for plasma produced by intense nanosecond laser ablation in vacuum under external magnetic field

Authors: Sha Tao, Benxin Wu, Yun Zhou, and Gary J. Cheng

Abstract: In this paper a two-stage physics-based model has been developed to study the evolution of plasma produced by high-intensity nanosecond laser ablation in vacuum under external magnetic field (where the laser beam is sufficiently intense to drive the target surface above the thermodynamic critical temperature). This kind of model has been rarely reported in literatures to the authors’ best knowledge. In the early stage, the laser-target interaction, and the induced plasma generation and its short-term evolution are simulated by solving one-dimensional (1D) hydrodynamic equations. An equation of state (EOS) that can cover the density and temperature range in the whole physical domain has been applied to supplement the hydrodynamic equations. In the later stage, the plasma long-term evolution is simulated by solving 2D gas dynamic equations. The two-stage model can predict the spatial distributions and temporal evolutions of plasma temperature, density, velocity and other parameters. The model is used to study and discuss the effects of strong external magnetic fields on the plasma evolution. It provides an useful guiding tool for related fundamental studies and practical applications.
Magnetic Field-Assisted Laser Drilling

Authors: Chane Ye, Sha Tao, Benxin Wu, Gary J. Cheng

A magnetic field-assisted laser drilling process has been studied, where nanosecond laser ablation is performed under an external magnetic field. This kind of process has been rarely studied before in literatures. The study shows that the magnetic field-assisted laser drilling process produces deeper drilling depth and more confined drilling debris, as compared to laser drilling without magnetic field. The difference is analyzed and a hypothesized explanation has been proposed based on the effect of the magnetic field on the plasma produced during laser ablation.
Femtosecond laser-induced surfaces structures of silicon: thermal and optical study of formation mechanisms

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Abstract

Femtosecond laser induced surface modification allows to functionalize surfaces at a micro and nanometer scale using a clean and dry process [1, 2]. The infrared laser interaction with Si target using fluences near ablation threshold lead to the formation of three different types of structures depending on the number of applied pulses:

1. After one or several pulses, resonant, quasi-parallel and periodic structures appear. They are spaced by a distance near laser wavelength, and result from laser excitation of surface plasmon polaritons if laser fluence is high enough.

2. After more laser irradiations, non-resonant “beads” structures (or “grooves”) are formed as a result of melting and thermo-capillary wave excitation before their resolidification.

3. After a large number of laser irradiations, cones are formed as a result of an amplification of the bead structures.

The proposed mechanisms are demonstrated by a comparison of experiments with numerical simulations.

References


Studies of Ultrafast Laser Interaction with SiO₂-Si Samples

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We present investigations of ultrafast laser interaction with samples of silicon dioxide on silicon, and compare the outcomes with analogous experiments on pristine silicon.

SiO₂-Si samples with variable oxide thickness are irradiated with 800-nm-wavelength ultrashort light pulses under stationary and translational irradiation conditions. Single shot irradiation led to qualitatively different results for the oxide samples compared with irradiation of pristine silicon surfaces, with a propensity for radial cracks in the underlying silicon for the SiO₂-Si samples. Translational irradiation of the oxide samples led to a wide variety of target modifications, often revealing periodic structures. Ripple formation in SiO₂-Si targets has been investigated using a variety of techniques, including high-resolution optical microscopy, scanning electron microscopy, atomic force microscopy (AFM), and transmission electron microscopy (TEM) utilizing focused ion beam (FIB) sample preparation techniques. Etching of the SiO₂ layer using HF facilitates AFM analysis of the buried silicon surface. The AFM measurements combined with TEM analysis of FIB-prepared specimens has provided detailed information on the buried periodic structures formed in the silicon substrate.

Future work will involve the investigation of a wider range of parameters including the influence of the laser wavelength, the effects of pulse length, the role of pulse repetition rate and sample scan speed, and expanding the values of oxide layer thickness.

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Abstract: Through femtosecond (fs) laser pulse irradiation, we study two dimensional quasi-periodic arrays of nanostructure-covered conical microstructures (NC-CMs) on Ni, and find that a significant amount of nickel oxide covers NC-CMs by the interaction of fs laser pulses with Ni in ambient air. Moreover, we create various shapes of NC-CMs by controlling the polarization and the incident angle of the laser beam, and discover that the size of nanostructures is distributed asymmetrically on the CMs only at off normal incidence. We suggest that nonuniform energy deposition induced both by the polarization and the incident angle of laser beam plays an important role in the asymmetric nanostructure distribution and shape of CMs on Ni.
Evaluation of controllability of femtosecond laser-induced impulse in water utilizing atomic force microscopy

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When a near-infrared femtosecond laser is focused in water through an objective lens, shockwave and cavitation bubble are generated at the laser focal point resulting from efficient multiphoton absorption. Generation of the shockwave and cavitation bubble induces a transient stress wave, which localizes in a micron-sized area around the laser focal point. Since the stress wave practically acts as an impulsive force on a micro object, we have lately demonstrated that the impulsive force is widely available as an external force for revealing and manipulating cell function in single cellular level e.g. estimation of adhesion force of the animal cell[1], injection of the biomolecules into the animal cell[2]. However, characteristics of the impulsive force are not well understood since it was quite difficult to quantify such a localized impulsive force. In this work, the controllability of the impulsive force was evaluated as a function of the laser pulse energy with the local force measurement system utilizing atomic force microscopy which we previously developed[3].

A single shot femtosecond laser pulse (800nm, 150fs, 60-150 nJ/pulse) was focused in water through 20x objective lens (N.A=0.40). The laser focal point was adjusted at image plane of the objective lens and the laser focal position in optical axis (Z position) was mechanically adjusted near an AFM cantilever, which was set in water (Fig. 1). Immediately after the laser irradiation, the AFM cantilever was oscillated upon the impulsive force propagating from the laser focal point. The oscillation was detected as differences of voltage between top and bottom photodiodes of a quadrant photodiodes (Fig. 2). By analyzing magnitude of the oscillation, the impulsive force loaded on the cantilever (F_{AFM}) was quantified. F_{AFM} dynamically varied depending on Z position of the laser focal point. To fit the Z position dependence, we estimate the total impulsive force generated at the laser focal point (F_0) in an assumption that the impulsive force spherically propagates as a short wave packet. The results are shown in Fig. 3 as black diamonds. The total impulsive force F_0 almost linearly increased with the pulse energy in a region between 60-110 nJ/pulse, and then reached the saturation at the pulse energy of 150 nJ/pulse. In the linear region, experimental error of the impulsive force was estimated to be less than $10 \times 10^{-12}$ [Ns] because the pulse energy can be tuned by a 5 nJ/pulse. This precision would be comparable or smaller than uncertainty of the biological samples. Furthermore, behaviour of the impulsive force was investigated to observe an appearance after the laser irradiation using a high-speed imaging. As a result, we found that behavior of the impulsive force was cooperated with that of the cavitation bubble size (Fig. 3). This suggests that a stress wave due to expansion and collapse of the cavitation bubble would mainly contribute to induction of the impulsive force.

Freezing of nanocavities beneath the surface of metal film irradiated by femtosecond laser pulse

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Ultrafast energy deposition of femtosecond laser pulse in a surface layer of metal results in a sequence of phenomena including supersonic melting of heated layer and formation of compression and rarefaction waves. The rarefaction/stretching wave propagates into the bulk of target and can produce the large enough stretching of the melt to cause the bubble nucleation and cavitation. It has been shown by atomistic simulation that due to the fast cooling of the surface layer by electron thermal conduction, a layer of supercooled liquid is formed at later times and resolidification front starts to propagate toward the surface. The bubbles in Al melt may have not enough time to fully collapse before they will be frozen in the form of disk-shaped nanocavities by the resolidification front.

It has been confirmed experimentally that nanocavities have been formed inside a resolidified surface layer of Al after irradiation by a femtosecond laser pulse. The shapes of nanocavities are agreed well with those obtained in simulations, which indicates that nanocavities were indeed produced by freezing of the collapsing bubbles in experiment.
Progressive formation of fine and coarse ripples on SiC surface by repeated irradiation of femtosecond laser pulses

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The formation of ripples on solid surfaces during femtosecond laser ablation has been an issue among researchers due to the uncertainty of the physical origin of those ripples and also to the unique optical properties of rippled structures or the potential applicability of those ripples for nanostructuring. The size of ripples produced by femtosecond laser ablation is roughly classified into two groups, namely, the fine ripples for which the spacing between adjacent ripples is below half of the laser wavelength and the coarse ripples for which the spacing is close to the laser wavelength. The interference model has been rather well accepted to explain the coarse ripples. However, the mechanisms responsible for the formation of fine ripples still remain elusive and a number of theories such as second harmonic generation [1], surface plasmon [2], nanoplasma [3] have been recently proposed to explain the formation of fine ripples. Furthermore, there have been many studies that reported simultaneous observation of fine and coarse ripples [4].

In this work, we report the progressive formation of first nanoparticles, next fine ripples, and eventually coarse ripples during irradiation of SiC surfaces with increasing number of femtosecond laser pulses (λ=515 nm, τ=250 fs). At laser fluence greater than the single pulse ablation threshold, nanoparticles were produced on the surface by the first few pulses over which fine ripple patterns overlapped for increased pulses. As the pulse number was further increased, the surface was gradually transformed into a coarse ripple covered one. At laser fluence below the threshold, however, only fine ripples were formed nonuniformly. These results confirmed that laser pulse energy and pulse number are critical factors determining the ripple types. Also, the measured reflectance of the SiC surface covered with fine and coarse ripples is presented in comparison with that of the original surface.


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Laser-induced breakdown spectroscopy (LIBS) for detection of low concentration standard reference materials under low pressure conditions

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Keywords: LIBS, low pressure, plasma, low concentration

We carried out quantitative analysis of LIBS signals of Standard Reference Materials (SRM) at low pressure conditions for precise determination of minor elements. A Q-switched Nd:YAG laser (1064 nm with a pulse duration of 5-7 ns at 70 mJ) is focused onto the surface of a target placed inside of a vacuum chamber. The gate delay is varied from 0.3 to 1 µs, and the gate width is set to 1.05 ms. The targets were mounted on a XYZ stage inside the chamber of 760 to 10⁻³ torr to utilize the pressure dependence of the plasma. In order to minimize error due to a matrix effect, we used 21 SRMs that belong to different categories of food, clay, sludge, steelmaking alloy, geochemical and agricultural materials. Further to discriminate and identify the unknown materials, principal component analysis (PCA) was used. We establish a benchmark procedure for detection of the element at low concentration via the low pressure LIBS approach.
Water-assisted cutting of glass with picosecond lasers

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Most of thin-film devices,- solar panels, displays, smart phones,- are using glass as a support material. The main obstacles in applying of laser technology for the glass cutting are the debris and micro-crack formation [1]. Cracking and thermally-induced effects could lead to degradation of the mechanical and bending strength of the glass. Some studies have shown increased ablation quality and efficiency in underwater or water-assisted laser machining [2, 3]. The effect of water in the laser processing is mostly manifested by partial conversion of the light energy to a mechanical pulse which transports debris away from the working area and cooling down the workpiece more effectively than gases.

Laser ablation experiments were carried out with a diode-pumped picosecond laser Atlantic (Ekspla) generating pulses of 10 ps duration with a pulse energy ranging from 10 to 60 µJ. In the experiments, laser radiation with 532 and 1064 nm wavelengths was used. Water mist from the airbrush spray formed a thin water film on the sample surface. Soda-lime-silica glass plates with thicknesses of 1.3 and 0.5 mm were used as the samples.

During the experiments, the nozzle distance to the sample surface and the inclination angle of the airbrush were optimized. Width and depth of trenches ablated by a single scan with the laser beam depended in a non-trivial way on the laser pulse energy and the scanning speed. This indicates importance of the water film parameters to the processing outcome.

The better scribe quality was obtained with the water-assisted processing. The crack size and melted areas near the edges of the scribe were significantly reduced as well as the glass surface was debris-free compared to the trenches processed in the ambient air. Bottom of the trenches was flat. At low scanning speeds (up to 32 mm/s), the water-assisted laser ablation improved the edge sharpness, straightness of the trench walls and ablation rate. Whereas, the processing in ambient air generated cracks parallel to the scribe center when the scanning speed was up to 64 mm/s.

Water assistance in glass cutting applications is a perspective technology, capable of increasing the ablation quality and also optimize the cutting process.

Surface delamination of polyimide using 355nm nano second pulse laser

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ABSTRACT
In this paper we have investigated the phenomena of surface delamination of polyimide (PI) using photochemical effects between 355nm nano second laser and PI. Under threshold fluence of photochemical ablation, that is, the earliest step of ablation, the surface is starting to be delaminated layer by layer with its nano scale thickness. As the number of laser pulses with a low fluence is increasing, the surface delamination is happening much more. These phenomena result from the laser-induced pressure wave generated by instant and transient temperature change of PI surface with respect to the absorption depth of laser wavelength that is related with a thickness of delaminated layers. And in chemical characteristics from XPS and XRD measurements, the O/C and N/C atomic ratios increase and crystallinity decreases despite of amorphous structure of PI.

Fig. 1 PI surface delaminated by UV laser

Fig. 2 XRD patterns of PI, (a) before surface delaminated and (b) after surface delaminate by UV laser
A NEW METHOD OF SELECTIVE MOLECULAR DISSOCIATION THROUGH RESONANT, ONE-COLOR, SUCCESSIVE VIBRONIC TRANSITIONS

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Abstract

This paper presents the theoretical derivation of a new method of selective molecular excitation through interaction with a LASER field. The new mechanism proposed for this interaction consists of successive vibronic excitations promoted by multiphoton, one-color LASER interactions. The excitations are resonant under specific conditions calculated here, a particular window of physical parameters and quantum numbers, characteristic of the system studied. A new dissociation method is proposed, based on this mechanism, which could open a new selective and efficient way of action upon molecules. The probability of dissociation as a result of successive absorption of energy in the multiphoton laser field is derived. The yield and selectivity of this method are superior to those of other classical methods of dissociation through bombardment with particles having energies close to the dissociation energy of the molecule to be dissociated.
Computational investigation of the formation of metallic nanoparticles by laser ablation of metalorganic precursor solutions

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A computational investigation of the fundamental mechanisms of the formation of metal nanoparticle in a technique based on laser ablation of an aqueous solution of metalorganic precursors is performed. This technique has a promise of providing an efficient method for dry synthesis and deposition of clean nanoparticles with composition tailored to the requirements of applications and narrow size distributions. A coarse-grained computational model for molecular dynamics simulations is developed to describe the processes leading to the ablation of a frozen solution and nanoparticle formation. The internal degrees of freedom of the molecules that are missing in the coarse-grained model are accounted for through the “internal heat bath” approach that associates an internal energy variable with each dynamic particle in the system and allows for the energy exchange between the internal (implicit) and dynamic (explicit) degrees of freedom in the model. As a result, the model reproduces the experimental heat capacities of solvent and solute molecules and allows for a more straightforward comparison to experimental data. The model is parameterized for an aqueous solution of palladium acetate and simulations are performed for several laser fluences close to the ablation threshold.

The results of the simulations suggest that the metal atoms released as a result of photodissociation of palladium acetate diffuse in the transiently heated matrix and form clusters. The absorption of laser light by the precursor molecules and the products of photodissociation heats up the surrounding water, leading to the explosive boiling and ablation of the surface region of the target. The metal clusters and atoms are entrained in the expanding ablation plume and are ejected from the target. The evolution of the composition of the target is considered in the simulations.
Kinetic simulations of burning of Al vapor in high-power CW laser ablation of an aluminum target in a shear air flow

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The presence of an oxidizing gas flow can substantially modify the characteristics of laser ablation of metal targets and may affect both the efficiency and dominant mechanisms of material removal. In the present work, the effect of aluminum vapor burning in a shear air flow on high-power continuous wave laser ablation of aluminum targets is investigated. Simulations are performed with a model that incorporates a kinetic model of aluminum oxidation in air into the direct simulation Monte Carlo method capable of a realistic representation of multi-component non-equilibrium gas dynamics in a wide range of pressure. The simulations reveal that the effect of the aluminum vapor burning on the flow structure and formation of the alumina film in the vicinity of the laser pot essentially depends on pressure in the external air flow.

At small pressure in the external flow, the molecular diffusion is responsible for transport of alumina and oxygen to the spot through the stream of evaporating material. In this regime, the growth of the oxide film on the surface of the laser spot is dominated by heterogeneous oxidation of the melted Al and direct deposition of alumina formed through gas-phase reactions.

At large pressure in the external flow, the molecular diffusion is found to be unable to deliver either oxygen nor alumina directly to the laser spot. In this regime, the steady-state evaporation from the target surface into the shear gas flow induces a complex unsteady flow pattern that includes multiple shock waves and vortex regions. The flow structure oscillates due to instability of the vortex region upstream from the laser spot. Contrary to an intuitive expectation, it is found that the aluminum vapor can efficiently propagate by diffusion-convection in the direction upstream the external gas flow. In this regime, the simulations suggest a fairly complicated scenario of the alumina film formation. It includes deposition of alumina trapped in the circulation zones upstream the evaporation spot, effective delivery of oxygen to the upstream edge of the evaporation spot, and possible growth alumina film from the upstream edge of the spot in the downstream direction.

**Keywords:** Laser ablation, evaporation, continuous wave laser, aluminum, oxidation, redeposition, external shear air flow, direct simulation Monte Carlo
Superwetting human enamel and dentin surfaces produced by femtosecond laser pulses

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Abstract

Good wettability of enamel and dentin surfaces is an important factor in enhancing adhesion of restorative materials in dentistry. In this study, we developed a femtosecond laser surface texturing approach that makes both the enamel and dentine surfaces superwetting. In contrast to the traditional chemical etching that yields random surface structures, this approach produces engineered surface structures. The surface structure engineered and tested here is an array of parallel microgrooves that generates a strong capillary force. Due to the powerful capillary action, water is rapidly sucked into this engineered surface structure and spreads even on a vertical surface.
On ripple formation in various metals and super-hard tetrahedral amorphous carbon films in consequence of femtosecond laser irradiation

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Ripple formation in consequence of ultrashort laser pulse irradiation of materials is a well-known phenomenon. We, too, have investigated the formation of ripples in various metals, i.e. steel, tungsten carbide hard metal, copper, brass and gold, as well as in superhard ta-C films, where we used femtosecond laser pulses of 775 nm, 388 nm and 258 nm mean wavelength and 130 fs pulse duration. The aim was to investigate how the ripple parameters depend on irradiation parameters, and if such ripples have a potentiality for applications.

In the paper, we will show that on smooth surfaces the ripple orientation is perpendicular to the electric field vector of the linearly polarized laser beam, as is well-known, and that in the presence of grooves a certain orientation along these grooves takes place. Moreover, it will be shown that the ripple period decreases with decreasing laser wavelength and/or increasing angle of incidence of the laser beam on the substrate, that ripples form already after only one laser pulse and that their depth and shape vary with the number of pulses per area.

By using optimum parameters large areas of the materials and films can be rippled swiftly, which would be important for applications. For instance, we investigated their use as diffraction gratings for the generation of optical effects, a few examples of which will be presented. The improvement of frictional and wear behavior of tribologically stressed surfaces by ripples was investigated on ta-C coated steel and WC hard metal surfaces.