

Materials Science Working Group

During the sessions of the Materials Science working group, five projects have been presented and discussed that are suggested to be conducted at the proposed ALFF facility. These can be coarsely divided into i) user projects that utilize the free electron laser to tackle problems centered in other scientific areas and ii) developer projects that focus on the characterization and improvement of the FEL itself. It was found that the work to be performed in both categories is strongly interrelated in the sense that a proper characterization or even control of beam properties like wavelength, intensity and coherence of the emitted radiation is of utmost importance for the success of many suggested user projects. Four of the presentations belonged to the first category, the scientific scope ranging from fundamental studies of particle-solid interaction processes, laser ablation mechanisms, spin dynamics at magnetic nanostructures to more applied fields like the development of mass spectrometric surface characterization methods utilizing desorbed atomic and molecular surface species. One presentation described a project belonging into the second category. In the following, the suggested projects will be briefly summarized.

Fundamental studies of sputtering processes (Bruce King, Andreas Wucher)

Sputtering is the release of particles from a solid surface into the gas phase by ion bombardment. This process is widely employed in thin film deposition and forms the basis for mass spectrometric surface analysis techniques that are used for the chemical characterization of solid surfaces. Although the phenomenon has been extensively investigated for more than three decades, there are still many open questions that need to be understood in order to make efficient use of the process in the applications mentioned above. Since most of the sputtered particles leave the surface in the neutral state, a proper characterization of sputtered fluxes ultimately requires the ionization and mass spectrometric detection of these neutral atomic or molecular species. One ionization mechanism that is particularly suited in this context is photoionization induced by the absorption of one single photon. The great advantage is that in this case the measured photon flux dependence of the ionization efficiency is linear and therefore easy to interpret (see figure 1 for an example), which is very much in contrast to non resonant multiphoton ionization studies conducted on sputtered particles {Kaesdorf, Hartmann, et al. 1992 701 /id}. For the identification of desorbed molecular species, single photon ionization offers the unique possibility to efficiently ionize a molecule without inducing significant photofragmentation. In both cases, it is necessary to irradiate the sputtered particles with photons of an energy greater than the ionization potential. The corresponding wavelengths reside in the UV, VUV and XUV regime that is targeted in the proposed ALFF. In order to obtain quantitative information about photoionization efficiency and, hence, about the desorbed particle fluxes, it is important to have enough photon flux available to drive the photoionization process into saturation (figure 1). In particular for sputtered molecular species, it is moreover extremely important to be able to tune the wavelength of the ionizing laser in the regime around the ionization potential. Due to the high internal excitation of sputtered molecules, it may be advantageous to operate at or even slightly below the IP in order to minimize the excess energy and therefore the photon induced fragmentation. The combination of both requirements is

clearly beyond the capabilities of currently available tabletop VUV laser systems and can only be achieved with the proposed ALFF facility.

As an example of new and surprising results that may still arise from fundamental sputtering experiments, figure 2 shows single photon ionization mass spectra of neutral atoms and molecules sputtered from pure Au and an AuAl₄ alloy surfaces that were taken with the time-of-flight mass spectrometer located at the existing Low Energy Undulator Test Line (LEUTL) at Argonne. Interestingly, the emission of Au atoms, which constitute the major sputtered species for a pure Gold surface, is apparently blocked for the alloy surface. This phenomenon has not been observed before and is currently under further investigation

Surface Analysis by Single Photon Ionization of Atoms and Molecules (Andreas Wucher, Igor Veryovkin)

This presentation reported on the ongoing activities that take place at the existing Low Energy Undulator Test Line (LEUTL) facility at Argonne National Laboratory. Over the last few years, a highly sensitive surface analysis instrument has been designed, built and installed at the LEUTL end station which is based on time-of-flight mass spectrometry of post-ionized sputtered neutral particles. In order to allow the detection of virtually every desorbed atom and molecule released from the surface, the neutral particles are subjected to a non resonant single photon ionization process initiated by either a fixed frequency VUV laser operated at a wavelength of 157 nm or the tunable output of the free electron laser. The instrument has been optimized for utmost detection sensitivity and was recently demonstrated to be able to detect sputtered neutral atoms released from a metal surface with a total efficiency (“useful yield”) of up to 25 % (cf. figure 3). In order to optimize this quantity for different atomic and molecular species, the single photon ionization process must be thoroughly characterized and optimized for each detected particle desorbed from the investigated surface by either ion sputtering or laser ablation techniques. Probably the most important parameter in this context is the single photon ionization cross section and its dependence on the wavelength of the ionizing radiation. As an example, figure 3 shows cross sections of sputtered atoms which have been determined from the saturation behavior of the ionization process at a fixed wavelength of 157 nm. As stated above, tunability of the ionization laser in connection with achievable pulse energy densities of the order of J/cm² constitute an extremely important prerequisite for these experiments which form the basis of many of the proposed applications of the technique in the field of cosmochemistry and biology (cf. the reports of the corresponding working groups).

Materials Processing by Laser Ablation (Libor Juha)

Surface modification of materials by means of laser ablation constitutes an important step in many structuring technologies of modern materials science. The fundamental processes behind the ablation mechanism, however, are still poorly understood. In view of ever decreasing structure dimensions, the VUV wavelength region between 40 nm and 200 nm is particularly interesting for upcoming nano-patterning techniques since it

extends the diffraction limit of conventional UV lasers down into the regime of a few tens of nanometers. Moreover, it is expected that the formation of laser induced periodic surface structures (LIPSS, cf. figure 5) that are commonly observed in laser ablation experiments is strongly wavelength dependent and will therefore be significantly altered in this wavelength range. Unfortunately, next to nothing is known about the fundamental characteristics of the ablation mechanism at VUV wavelengths. From preliminary studies, it was found that the dependence of the ablation rate on the fine structure of the irradiated material may be much less pronounced than in the UV range accessible to conventional excimer lasers (cf. figure 6). Combining VUV wavelength with ultrashort laser pulse duration, it appears feasible to change the ablation characteristics from a mainly thermal process to non-thermal processes. Using the TESLA free electron laser at Hamburg, ablation rates have been measured at a wavelength of 86 nm and a pulse duration of 150 fs and compared to those obtained with nanosecond laser pulses. These studies need to be continued in the future, but the TESLA facility is currently being upgraded and will afterwards not operate in the VUV wavelength range any more. The proposed ALFF will therefore be the only available facility generating tunable, short pulse radiation in the wavelength range needed to pursue these investigations.

Sub-ps Magnetic Domain Imaging (Dave Deavney)

Modern concepts of magnetic storage media and MRAM chips feature nanoscale magnets that are often realized as patterned microarrays of magnetic nanostructures at solid surfaces. In order to optimize the design and performance of such devices, it is necessary to study the magnetization dynamics of magnetic nanostructures with both high lateral and temporal resolution. At present, photoemission electron microscopy (PEEM) in connection with soft X-rays obtained from the APS is used to record images of magnetic microarrays with about 100 nm lateral resolution and good magnetic contrast (cf. figure 6). By means of pump-probe experiments triggered by an electrical pump pulse, the dynamics of magnetic domain modifications can be traced on a 100 ps time scale. In the future, the structure size is expected to significantly drop below the currently achieved lateral resolution, which in turn is mainly determined by chromatic aberrations induced by the width of the kinetic energy distribution of the emitted electrons. It is known that this width may be significantly reduced by changing the irradiating photon energy from the soft X-ray regime (500...3000 eV) to the VUV range (< 10 eV) (cf. figure . Connecting the PEEM to the proposed ALFF facility would therefore greatly enhance the achievable lateral resolution, thus making it possible to image magnetic nanoarrays with structure sizes even below 20 nm. At the same time, the short pulsewidth of the free electron laser opens entirely new possibilities with respect to the time resolution that could be achieved in such experiments. Using pump-probe schemes, it is envisioned to study the correlation of ultrafast spin dynamics occurring on a sub-picosecond time scale within an array of magnetic nanostructures at surfaces.

Spatial Coherence Measurement (David Paterson)

Many of the experiments that are planned to be conducted at ALFF heavily rely on the spatial coherence of the incoming VUV radiation. In order to interpret the results of experiments like LIPSS formation in laser ablation or make experiments like coherent

control of chemical reactions even possible, it is of great importance to exactly know the coherence properties of the free electron laser beam and maybe use this input to drive a feedback mechanism that keeps those properties constant over time. In the proposed project, it is envisioned to measure the spatial coherence by recording a diffraction pattern of a so-called Uniformly Redundant Array (URA, cf. figure 9), i.e., a one or two dimensional array of features that exhibit evenly distributed spacings and, hence, mimics many Young's double slit experiments with different slit spacing. Recording the diffraction pattern of such a device with a VUV sensitive CCD camera allows to determine the complete spatial coherence function on a single shot basis (see figure 10 for an example). This information is extremely valuable for the interpretation of user experiments, since it is expected that the properties of the SASE beam may exhibit significant fluctuations from shot to shot. Moreover, it is conceivable that the information can be inserted into a feedback mechanism stabilizing the beam.

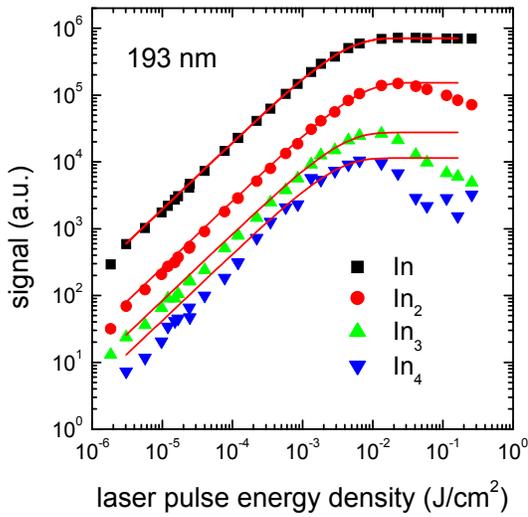


Figure 1: Mass spectrometric signal of neutral atoms and clusters sputtered from an ion bombarded Indium surface and post-ionized using 193 nm UV radiation.

Figure 2: Single Photon Ionization Time-of-flight mass spectrum of neutral atoms and molecules sputtered from a) a pure Au and b) a AuAl₄ alloy surface and post-ionized using $\lambda = 127$ nm radiation from the Argonne free electron laser.

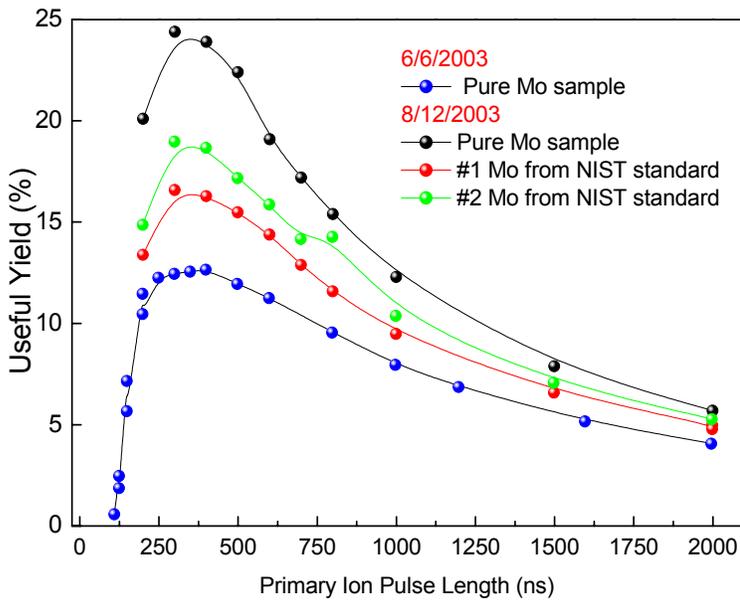


Figure 3: Useful yield for detection of Mo atoms sputtered from a pure Molybdenum surface and post-ionized with $\lambda = 157$ nm radiation vs. pulse duration of the primary ion beam.

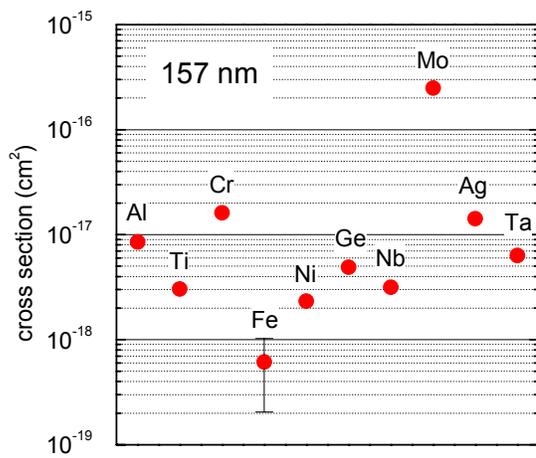


Figure 4: Single photon ionization cross section of various neutral atoms at $\lambda = 157$ nm.



Figure 5: AFM image of ablation crater edge showing the formation of Laser Induced Periodic Surface Structures (LIPSS).

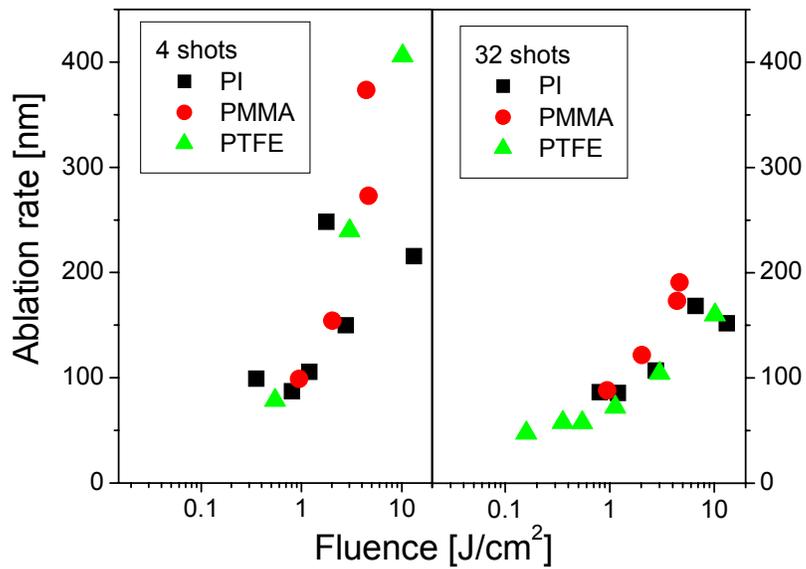


Figure 6: Ablation rate of different polymer surfaces measured using a capillary discharge laser at $\lambda = 46.9$ nm vs. laser power density.

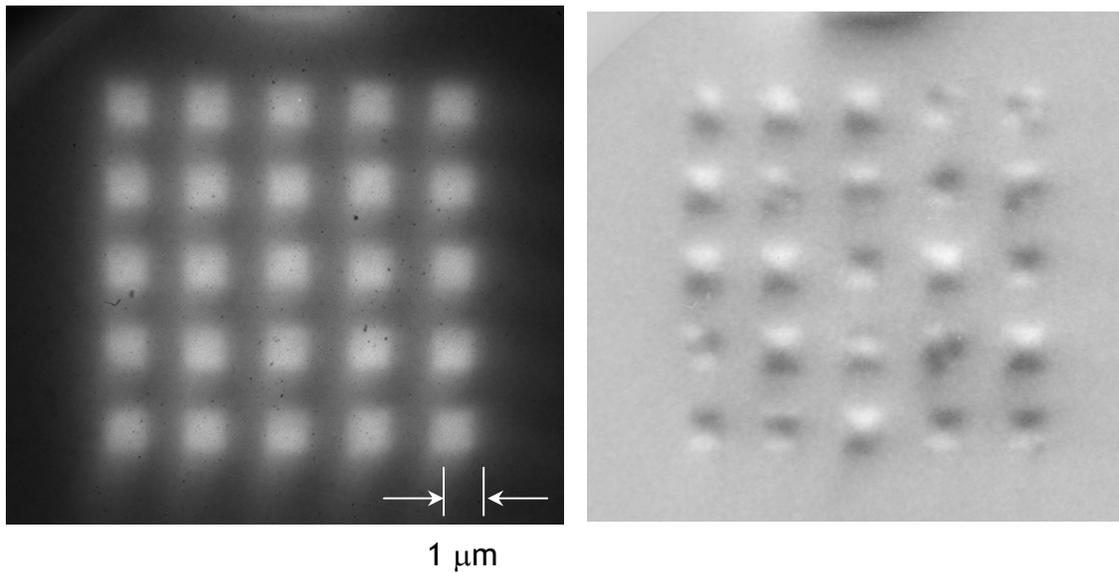


Figure 7: X-ray induced PEEM images of magnetic Co nanostructures on an Al buffered Si surface using a) chemical and b) magnetic contrast at an irradiating photon energy of 778 eV.

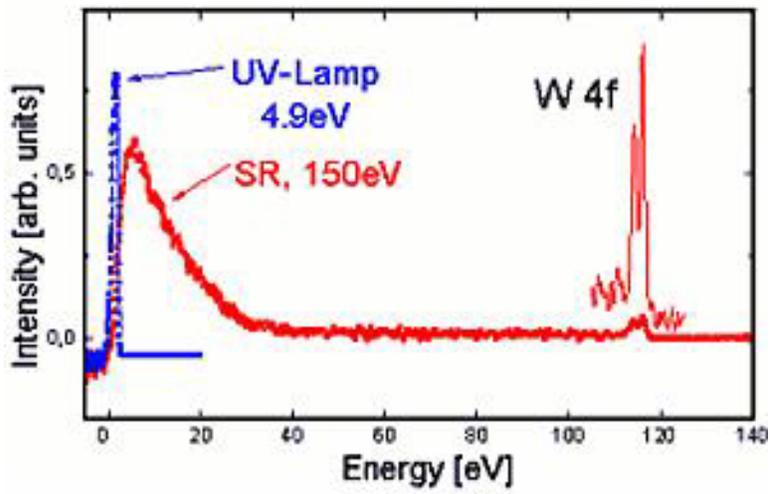


Figure 8: Kinetic energy distribution of electrons emitted under irradiation with soft X-ray and UV photons, respectively.

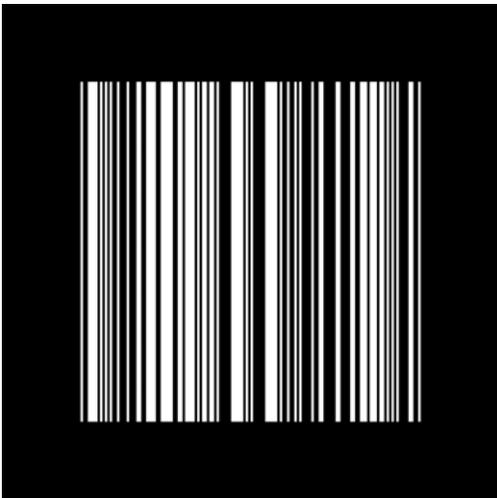


Figure 9: Uniformly Redundant Array used to determine the spatial coherence function

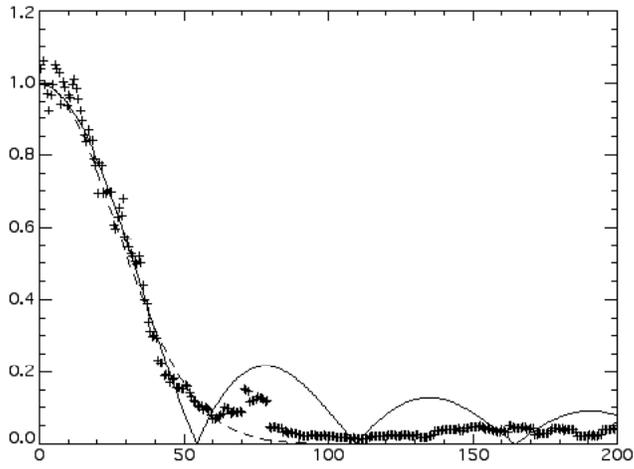


Figure 10: Measured spatial coherence function of soft X-ray radiation ($h\nu = 1500$ eV) emitted from APS Beamline 2-ID-B.