High-power laser science at the Advanced Laser Light Source

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The Advanced Laser Light Source (ALLS) is a unique user facility located at INRS-EMT (Varennes, Canada) counting on 40M CDN$ of investment since 2002. Since 2019, this facility has joined the LaserNetUS network and is now funded as a national research infrastructure by the Canada Foundation for Innovation – Major Science Initiatives. These fundings ease access to the facility for academic and government users. In the first part of my talk, I will give an overview of the facility's capabilities including the most powerful laser in Canada with 750 TW. In the second part, I will discuss recent results on electron acceleration under tight focusing [1] and for high dose-rate of MeV electron beam from ambient air [2], as well as a high flux neutron beamline based on laser wakefield acceleration of electrons [3].

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Exploring molecular dynamics using molecular dynamics at ALLS

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We will provide an overview of the recent developments at the Advanced Laser Light Source (ALLS) to image molecular dynamics and to make use of molecular dynamics for the development of novel femtosecond light sources based on molecular gases.

Keywords - ultrafast laser sources, reaction microscope, COLTRIMS, molecular dynamics, molecular movies, femtosecond laser, hollow core fiber

Imaging molecular dynamics

Coulomb explosion imaging (CEI) is a powerful tool to measure changes of geometrical structure and to link it to electronic rearrangements and to track a broad variety of molecular dynamics; even if they occur in a non-concerted manner and require single-molecule detection sensitivity.

Example cases from the small hydrogen molecule H2 [1] up to the heterocyclic 12-atomic molecule 2-thiouracil [2] are being discussed. Upon photoexcitation of a molecule, it will break apart. We can see fragments following direct, conventional dissociation paths, as well as fragments deviating from this minimum energy path. The latter are called roaming fragments and explore the potential energy landscape in a statistical manner. This roaming phenomenon observed in formaldehyde [3]. This hard-to-grasp yet almost universal statistically occurring hindered dissociation process can be observed using a CEI thanks to its single molecule sensitivity.

The presentation will start with an overview on recent developments of the COLTRIMS/reaction microscope beamline at the Advanced Laser Light Source (ALLS) user facility obtained in collaboration with the University of Waterloo and the National Research Council Canada. We use CEI in combination with high repetition rate Ytterbium laser systems.

Generating new light sources using molecular dynamics

Ytterbium (Yb) laser systems are leading the third generation of high-energy femtosecond laser technology, providing high average powers at pulse durations ranging from 0.1 to 1 ps, which are typically longer than those of Ti:Sapphire laser systems used in the past decades. Obtaining the ultrashort pulses necessary to investigate molecular dynamics from such sources requires implementing post-compression techniques, for example, via nonlinear spectral broadening in gas-filled waveguides, so-called hollow core fibers (HCF). While noble gases are commonly used as nonlinear media in such waveguides, recent developments have shown that molecular gas-filled HCFs provide efficient post-compression from sub-ps to ps pulses (see e.g. [4]) via self-phase modulation (SPM) in the molecular gas methane. We will discuss the strength of methane (CH4) as a nonlinear medium for HCF-based pulse compressors to achieve high average power few-cycle pulses of sub-16 fs duration with >70% transmission and up to 250 kHz repetition rate in a compact setup and present it as a cost-effective alternative to highly expensive and scarcely available noble gases such as xenon, krypton or neon.

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The Hyper Spectral Ultrafast Source - HYPUS

Bruno E. Schmidt
few-cycle Inc.

We present an Yb laser based ultrafast spectroscopy platform offering multiple, simultaneous output beams between 200nm to 12µm wavelength. Our "OPA-free" approach supports ~1fs timing jitter between the UV and IR channels

Probing femtosecond-to-attosecond dynamics in matter with high-harmonic generation and free-electron laser sources

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High-harmonic generation (HHG) and free-electron lasers (FELs) provide complementary radiation sources for investigating ultrafast dynamics in atoms, molecules, and solids across a broad range of energies. Both enable femtosecond-resolution studies of electronic and structural relaxation, while also offering routes to attosecond sensitivity through interferometric and polarimetric observables that do not require isolated attosecond pulses.

FEL-based experiments enable element- and site-specific access to ultrafast molecular dynamics with sensitivity to core-level processes. I will present time-resolved pump–probe photoelectron spectroscopy studies of femtosecond electronic relaxation in molecules using site-selective detection. Beyond this regime, interferometric photoelectron spectroscopy provides sub-cycle sensitivity through phase-resolved measurements. I will discuss an interferometric FEL experiment on chiral molecules where coherent control of the photoelectron signal reveals attosecond chiral dynamics.

In parallel, HHG provides a coherent broadband source of extreme-ultraviolet and soft X-ray radiation for transient absorption spectroscopy. In our laboratory, we routinely generate harmonics up to ~220 eV, enabling table-top access to the EUV/soft X-ray regime for core-level studies. Beyond its role as a source, HHG can also be used as a spectroscopic tool to study nonlinear electron dynamics driven by the laser field during the generation process. I will present an experiment where polarization-resolved HHG spectroscopy reveals interference between electronic pathways in the emission process, providing sensitivity to attosecond electron dynamics in solids.

Spin-orbital excitations encoding the magnetic phase transition in the van der Waals antiferromagnet FePS3

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Magnetic van der Waals (vdW) materials offer new avenues for exploring exotic magnetic phases and collective behavior. [1] Recent research using optical spectroscopy reveals the high sensitivity of acoustic phonons to the magnetic states even in few-layer structures. [2] FePS3, whose interlayer exchange interaction is very small, is an S = 2 zig-zag quasi-two-dimensional antiferromagnetic insulator with a honeycomb lattice. [3] Here we performed resonant inelastic X-ray scattering (RIXS) at Fe L3-edge as a function of temperature across the magnetic transition to elaborate the spin-orbital excitations of FePS3 and their relation to magnetism. We identify that the multiplet response of Fe L3 RIXS has a large sensitivity to the magnetic state. By comparing with ligand-field-theory calculations, we identify the essential role of the trigonal lattice distortion and negative metal-ligand charge transfer to account for the low-energy spin-orbital excitations. We reveal the robustness of these elementary excitations down to the few atomic layer limit following the persistent magneto-optical excitation and phonon modes in monolayers measured by Raman scattering. [4] This provides crucial insight into how the spontaneous magnetic symmetry-breaking stabilizes in the quasi-two-dimensional limit of the vdW magnet FePS3. Our study highlights the crucial role of lattice and orbital anisotropy for stabilizing the quasi-two-dimensional magnetism in magnetic van der Waals materials. [5]

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Liquids from the Perspective of Photoelectrons: Scattering and Interfacial Properties

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Liquid-jet photoelectron spectroscopy (LJ-PES) has become a standard tool for studying the electronic structure and dynamics of chemically, biologically, and atmospherically relevant solutes in their natural aqueous environment. LJ-PES provides direct access to both the electronic structure and interfacial characteristics of aqueous solutions. The kinetic energies of both primary and secondary electrons can be accurately determined, enabling a detailed characterization of explicit solution surface properties, including molecular / chemical structure and charge distributions. In addition, non-local relaxation processes and electron delocalization upon core-level excitation provide access to rich bulk-solution dynamical information. This includes proton, electron, and charge transfer, and enables explicit insight into first-solvation shell energetics and composition.
In this presentation, I will discuss the capability of LJ-PES to resolve solute distributions at the solution-vacuum interface, with emphasis on the absolute probing depth into solution. This is an ongoing challenge due to the insufficiently understood electron scattering lengths in liquids, particularly at low energies. For the first time, we combined LJ-PES with He-scattering experiments to quantify interfacial compositions and associated electron scattering lengths over a wide range of electron energies, fully based on experiment. Furthermore, I will address near-ionization-threshold processes, in particular the Coulomb-interaction of the direct photoelectron and the Auger electron in the vicinity of the ionized species, i.e., post collision interaction (PCI). PCI can be exploited to directly probe core-hole lifetimes and screening of the local solvation environment and thus may provide a sensitive tool for studying electron stabilization and delocalization within the first few solvation shells.

In-situ scanning XMCD imaging of hysteretic domain evolution in piezo-actuated perpendicularly magnetized Co/Ni multilayers

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Strain-mediated electric-field control of magnetism promises energy-efficient alternatives to current-driven switching, yet direct nanoscale observation of how strain reshapes magnetization reversal remains scarce. We imaged magnetization reversal in Ta/Pt/[Co(0.3 nm)/Ni(0.7 nm)]×12/Pt multilayers with perpendicular magnetic anisotropy by scanning x-ray magnetic circular dichroism (XMCD) microscopy, while a piezoelectric stage applied nominal in-plane tensile strain in situ. Helicity-paired images with 50 nm resolution revealed a domain-wall network only within a limited field interval of the descending out-of-plane sweep; near zero field the domain pattern faded into the background although magnetometry indicated incomplete bulk reversal. Piezo actuation shifted the field at which the domain pattern was most pronounced and modified its morphology, suggesting magnetoelastic modulation of effective perpendicular anisotropy. A phenomenological model with spatially distributed irreversible switching thresholds qualitatively reproduced this transient domain-visibility window, pointing to history-dependent switching across a piezo-modified reversal landscape. These results establish piezo-actuated scanning XMCD microscopy as a direct probe of electrically driven magnetization reversal and highlight the interplay between strain and magnetic history in low-power magnetic devices.

Probing Ultrafast Electron Motion with Attosecond XFELs

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The ultrafast motion of electrons is a frontier problem for photochemical processes. X-ray Free Electron Laser (XFEL) facilities, such as the Linac Coherent Light Source (LCLS), produce high-brightness, ultrashort pulses, with wavelength continuously tunable across the x-ray regime, and have recently been demonstrated to produce pulses with attosecond duration. I will present an overview of the first experiments performed with the high average power FEL driven by the new super-conducting accelerator at SLAC leveraging attosecond pulses to probe ultrafast electron motion.

Imaging Ultrafast Electronic and Nuclear Dynamics in Gas-Phase Molecules

Daniel Rolles
Department of Physics, Kansas State University

Structure-sensitive methods based on femtosecond light or electron pulses are now making it possible to measure how molecular structures change during light-induced processes on a femtosecond time scale. I will present the results of a combined experimental and theoretical study of an ultrafast UV-induced ring-opening reaction to discuss the strength and weaknesses of different experimental methods for studying ultrafast processes in the gas phase and highlight the benefits of multimodal studies for obtaining a complete picture of the reaction dynamics and underlying reaction mechanisms. I will also present recent progress on applying machine learning techniques and neural networks to aid with the analysis and interpretation of multidimensional experimental data.

Uptake and Release of Trace Gases at Liquid-Vapor Interfaces Investigated by X-ray Photoelectron Spectroscopy

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The interfaces of aqueous solutions with air drive many important processes in the atmosphere and the environment, such as the uptake and release of trace gases by aerosols and the sequestration of carbon dioxide by the oceans. Laboratory studies have provided a wealth of information on the rates and product distribution of heterogeneous reactions at solution-vapor interfaces, but direct information on the physicochemical properties of these interfaces during the reaction are still scarce.
X-ray photoelectron spectroscopy (XPS) is an excellent method for the investigation of liquid-vapor interfaces due to its inherent chemical and interface sensitivity.[1] In this talk we present the fundamental challenges and experimental approaches for the measurement of liquid-vapor interfaces under moderate vacuum conditions using liquid jets, as well as under equilibrium and reaction conditions using static liquids in combination with ambient pressure XPS (APXPS).[2] We will also discuss case studies of the application of XPS to the investigation of the cooperative interaction of surfactants and ions at the interface [3]; apparent shifts in the tautomer ratios [4] and acid-base equilibria at the interface compared to the bulk of the solution [5]; the precise determination of the difference in the depth of solvation of protonated and deprotonated carboxylic acid [6]; as well as the direct observation of the uptake and reaction of trace gases at liquid-vapor interfaces. The presentation concludes with an outlook on future directions, including multimodal studies of liquid interfaces by simultaneous X-ray and IR-based spectroscopies. [7]

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Tracking ultrafast electron dynamics in individual nano-samples with Coherent Diffractive Imaging

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Can a dielectric nanostructure be switched from transparent to absorbing and back to transparent within a single optical cycle? Can we track and control charge transport within a nanoparticle in a light field at the time resolution frontier? These fundamental questions with far-reaching consequences can now be addressed for the first time thanks to the rapid development of X-ray free-electron lasers (FELs) and high harmonic generation (HHG) sources, which deliver well-controlled, intense short-wavelength pulses of few femtoseconds and even attoseconds duration. They open up a way to visualize and track ultrafast electron dynamics in nanoscale matter on their relevant inherent time scale with single-pulse single-particle coherent diffractive imaging (CDI).

In its basic version, CDI allows to retrieve the structural properties of a nanoparticle from the amplitude of the scattered light field [1]. But in fact, CDI is also sensitive to changes in the particle's electronic structure. At near-resonant photon energies, ionization and plasma formation in an otherwise homogeneous nanoparticle can be visualized [2]. Using well-controlled, extremely intense HHG pulses we could resolve a reproducible switching of the scattering response in dielectric nanoparticles on a femtosecond time scale [3]. The upcoming two-color capabilities of XFELs, combined with recent algorithmic development [4] will enable us to turn the inherent sensitivity of CDI to the target's electronic structure into a near-background-free, spatio-temporally resolved probe for electron dynamics in nanoscale matter with potential impact in many fields from nanoplasmonics and nonlinear X-ray optics to Petahertz optoelectronics.

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Revealing two-dimensional electronic states in an ultrawide-bandgap semiconductor by microfocused synchrotron ARPES

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Ultrawide-bandgap semiconductors are promising platforms for next-generation power electronics, yet their surface and interfacial electronic structures remain poorly understood because direct momentum-resolved measurements are experimentally challenging. Here, we use microfocused angle-resolved photoemission spectroscopy (micro-ARPES) at the beamline BL06U of NanoTerasu to investigate electron-doped β-Ga2O3, a representative ultrawide-bandgap oxide semiconductor. The high photon flux and micron-scale beam size enable us to selectively probe high-quality cleaved regions and directly visualize the two-dimensional electron system formed at the surface. The observed Fermi surface is nearly circular in the surface plane, revealing an almost isotropic in-plane electronic structure despite the low-symmetry monoclinic crystal structure. By tuning the sheet carrier density, we further resolve a systematic evolution from broad spectral weight with a high-binding-energy tail at low density to a well-defined parabolic quasiparticle band at higher density. This behavior is consistent with a crossover from a polaronic regime toward a Fermi-liquid-like electronic state. Notably, the quasiparticle effective mass increases with carrier density, indicating an unconventional many-body renormalization beyond a simple polaron-to-Fermi-liquid crossover. These results demonstrate how advanced microfocused synchrotron photoemission can reveal emergent electronic states in future ultrawide-bandgap materials and establish NanoTerasu as a powerful platform for nanoscale electronic-structure studies.

Continuous flow hydrothermal synthesis of ultrasmall metal oxide particles

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Continuous flow hydrothermal synthesis is a powerful methodology for fabricating metal oxide nanoparticles. In high temperature compressed water, the solubility of metal ions decreases significantly, leading to a high degree of supersaturation and an accelerated nucleation rate. Furthermore, in-situ surface organic modification can be achieved immediately following particle formation because organic molecules become miscible in high-temperature compressed water.

Recently, precision synthesis has enabled the fabrication of metal oxide nanoparticles within the 1–10 nm range via the continuous flow hydrothermal route [1]. These ultrasmall metal oxides exhibit extraordinary structural distortion, which triggers the emergence of unique properties, most notably valence fluctuation. Theoretical studies have further corroborated these findings, revealing unusual electronic states in such nanoparticles [2].

Another compelling feature of organically modified metal oxides is facet control. The modification allows for the exposure of thermodynamically unstable surfaces, thereby enhancing the reactivity of the metal oxides. This facet control has yielded high oxygen storage capacity, making these materials promising oxygen carriers for low-temperature applications.

In addition, these organically modified nanoparticles can be formulated into highly concentrated, stable nanofluids. This high dispersibility is crucial for advanced heat transfer applications. In this presentation, the structural and electronic characteristics of these organically modified metal oxides based on synchrotron measurement data will be discussed.