Promovierendensymposium (SYPS)
Velocity Map Imaging: Focusing on intra- and interatomic dynamics

jointly organized by
the Working Group 'Young DPG' (AGjDPG) and
all divisions of the section AMOP

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The introduction of Velocity Map Imaging (VMI) by Eppink and Parker in 1997 has had a great impact in a variety of fields in atomic- and molecular physics. It has become a powerful tool for studying photoelectron spectroscopy, molecules in strong laser fields, femto- and attosecond pump-probe spectroscopy as well as photodissociation and chemical reaction dynamics. Still, imaging techniques are continuously being further developed and refined as illustrated by new applications of VMI spectrometers, ranging from surface scattering experiments to spatial mapping of biological tissues.

The symposium shall present the VMI technique and demonstrate the diversity of dynamic processes that can be studied as well as the depth of insight that can be gained. The joint technique connects the research presented and, in this way, facilitates understanding of what drives a central topic in AMOP physics: the determination of intra- and interatomic dynamics.

Overview of Invited Talks and Sessions

(Lecture room: Audimax)

Invited Talks

SYPS 1.1	Thu	14:10–14:40	Audimax	**Oxygen and imaging, a perfect match** — •David Parker
SYPS 1.2	Thu	14:40–15:10	Audimax	**Attosecond imaging** — •Marc Vrakking
SYPS 1.4	Thu	15:25–15:55	Audimax	**Applications of the fast imaging Pixel Imaging Mass Spectrometry camera** — •Mark Brouard
SYPS 2.1	Thu	16:30–17:00	Audimax	**Unraveling the dynamics of state- and conformer selected molecules fixed in space with the VMI** — •Jochen Küpper
SYPS 2.3	Thu	17:15–17:45	Audimax	**Velocity map imaging: from molecules to clusters, nanoparticles and aerosols** — •Michal Farnik, Viktoriya Poterya, Jozef Lengyel, Andriy Pysanenko, Pavla Svrckova, Jaroslav Kocisek
SYPS 2.5	Thu	18:00–18:30	Audimax	**Velocity map imaging studies of quantum state resolved scattering at gas-solid and gas-SAMs surfaces** — •David J. Nesbitt, Monika Gruetter, J. Robert Roscioli, Carl Hoffman, Daniel J. Nelson

Sessions

SYPS 1.1–1.4	Thu	14:00–15:55	Audimax	**Velocity map imaging - focusing on intra- and interatomic dynamics 1**
SYPS 2.1–2.5	Thu	16:30–18:30	Audimax	**Velocity map imaging - focusing on intra- and interatomic dynamics 2**

SYPS 1: Velocity map imaging - focusing on intra- and interatomic dynamics 1

Time: Thursday 14:00–15:55 Location: Audimax

Welcome and introduction

Invited Talk SYPS 1.1 Thu 14:10 Audimax
Oxygen and imaging, a perfect match — •David Parker — Radboud University, Department of Physics, FNWI-IMM-MLF, NIjmegen, The Netherlands

Molecular oxygen, O_2, is fascinating molecule. Despite this, a full understanding of the photodynamics of molecular oxygen is lacking due to the complex electronic structure and the forbidden nature of almost all optical transitions of O_2. Over the past decade our group has been able to reveal many new aspects of O_2 photodynamics due in part to the development and application of the velocity map imaging technique. In this talk I will highlight our past work on the photodissociation of: O_2 super-excited states , the Herzberg continuum6 of O_2, singlet oxygen b-state, the Schumann-Runge continuum, and O_2-isoprene clusters. I will mainly describe new work on the photodissociation of the singlet oxygen a $^1\Delta_g$ -state. Velocity map imaging will be shown to be particularly well-matched to the study of O_2 photodynamics.

Invited Talk SYPS 1.2 Thu 14:40 Audimax
Attosecond imaging — •Marc Vrakking — Max Born Institute (MBI), Berlin

The natural timescale for electron dynamics reaches down to the attosecond domain. Following the discovery of attosecond laser pulses, about a decade ago, attosecond science has developed into a vibrant, new research field, where the motion of single or multiple electrons and, in molecules, the coupling of electronic and nuclear motion, can be investigated, on attosecond to few-femtosecond timescales. Attosecond experiments require suitable observables. In my talk I will describe how "attosecond imaging", basing itself on kinetic energy and angle-resolved detection of photoelectrons and fragment ions using a velocity map imaging (VMI) spectrometer, has been exploited in a number of pump-probe experiments. The use of a VMI spectrometer in attosecond experiments has allowed the characterization of attosecond pulse trains and isolated attosecond pulses, the elucidation of continuum electron dynamics and wave packet interferometry in atomic photoionization and the observation of electron localization in dissociative molecular photoionization.

SYPS 1.3 Thu 15:10 Audimax
Insight in chemical dynamics by three-dimensional momentum imaging — •Robert Siemering[1], Eric Wells[2], Itzik Ben-Itzhak[3], Matthias Kling[4], and Regina de Vivie-Riedle[1] — [1]Ludwig-Maximilians-Universität, München, Germany — [2]Augustana College, Sioux Falls, USA — [3]Kansas State University, Manhattan, USA — [4]Max Planck Institute of Quantum Optics, Garching, Germany

The description of laser pulse interaction with molecules have different constrains for theory and experiment. While adaptive feedback in experiments can manipulate dynamics in molecular systems, the finding of the underlying mechanism or the forming of an appropriate theoretical model is difficult. This is especially the case when the feedback is limited to a single observable. By rapidly inverting velocity map images of ions to recover the three-dimensional photofragment momentum distribution and incorporating that feedback into the control loop, the specificity of the control objective is dramatically increased. As an example in the isomerization of acetylene ($C_2H_2^{2+} \rightarrow CH_2^+ + C^+$) the angle-resolved ratio of CH^+/CH_2^+ was controlled in the experiment[1]. With the experimentally obtained pulses on-the-fly-trajectories were performed to unveils the underlying barrier suppression mechanism. The theoretical model can explain the observed shift to lower kinetic energy release values for the CH_2^+ fragments.

[1] Wells, E., *et al.*, Adaptive Strong-field Control of Chemical Dynamics Guided by Three-dimensional Momentum Imaging, *Nature Communications*, DOI:10.1038/ncomms3895

Invited Talk SYPS 1.4 Thu 15:25 Audimax
Applications of the fast imaging Pixel Imaging Mass Spectrometry camera — •Mark Brouard — Department of Chemistry, University of Oxford

Recent work on the development of a fast imaging camera, known as the Pixel Imaging Mass Spectrometer (or PImMS) camera will be described [1,2]. The talk will focus on example applications of the PImMS camera. These range from studies of molecular photofragmentation [3] using correlation imaging techniques, progress towards three-dimensional velocity map ion imaging, through to imaging mass spectrometry of surfaces [4]. Future potential developments and applications will also be discussed.

[1] A. Nomerotski, M. Brouard, E. Campbell, A. Clark, J. Crooks, J. Fopma, J.J. John, A.J. Johnsen, C.S. Slater, R. Turchetta, C. Vallance, E. Wilman and W.H. Yuen, JINST 5, C07007, (2010). [2] J J John, M Brouard, A Clark, J Crooks, E Halford, L Hill, J W L Lee, A Nomerotski, R Pisarczyk, I Sedgwick, C S Slater, R Turchetta, C Vallance, E Wilman, B Winter and W H Yuen, JINST 7, C08001, (2012). [3] A. T. Clark, J. P. Crooks, I. Sedgwick, R. Turchetta, J. W. L. Lee, J. J. John, E. S. Wilman, L. Hill, E. Halford, C. S. Slater, B. Winter, W.H. Yuen, S. H. Gardiner, M. L. Lipciuc, M. Brouard, A. Nomerotski, and C. Vallance, J. Phys. Chem. A 116, 10897, (2012). [4] M. Brouard, E. Halford, A. Lauer, C. S. Slater, B. Winter, W. H. Yuen, J. J. John, L. Hill, A. Nomerotski, A. Clark, J. Crooks, I. Sedgwick, R. Turchetta, J. W. L. Lee, C. Vallance, and E. Wilman, Rev. Sci. Instrum. 83, 114101, (2012).

SYPS 2: Velocity map imaging - focusing on intra- and interatomic dynamics 2

Time: Thursday 16:30–18:30 Location: Audimax

Invited Talk SYPS 2.1 Thu 16:30 Audimax
Unraveling the dynamics of state- and conformer selected molecules fixed in space with the VMI — •Jochen Küpper — Center for Free-Electron Laser Science, DESY, Hamburg — Department of Physics, University of Hamburg — The Hamburg Center for Ultrafast Imaging, Hamburg

Velocity-map imaging (VMI) provides a powerful detection scheme for the dynamics of complex molecules. In this tutorial, I will introduce the methods to spatially separate different species present in molecular beams, to fix these molecules in space, and to investigate their structures and dynamics using VMI.

Inhomogeneous electric fields enable the spatial separation of conformers (structural isomers), nuclear spin isomers, and individual quantum states. These experiments exploite the neutral-molecule analogues of the electric bender, the ion guide, and the linear accelerator. The created state-selected molecular samples provide unprecedented options to fix molecules in space. The VMI spectrometer allows to unravel the resulting rotational dynamics of molecules in the applied electric and laser fields of vastly different strength and duration. The detailed analysis of these rotational dynamics and the prepared pendular states is an instructive example of quantum control.

VMI is also used to image photoelectron angular distributions (PAD). Utilizing the described controlled samples one can observe molecular frame (MF) PADs that provide direct images of electronic and geometric structures of molecules, potentially with femtosecond time resolution.

SYPS 2.2 Thu 17:00 Audimax
Quantification of the Photoelectron Circular Dichroism from Multiphoton Ionization with Femtosecond Laser Pulses — •Christian Lux[1], Stefanie Züllighoven[1], Cristian Sarpe[1], Matthias Wollenhaupt[2], and Thomas Baumert[1] — [1]Universität Kassel, Institut für Physik und CINSaT, D–34132 Kassel, Germany — [2]Carl von Ossietzky Universität Oldenburg, Institut für Physik, D–26129 Oldenburg, Germany

The asymmetry of photoelectron angular distributions from randomly oriented enantiomers of chiral molecules in the ionization with circularly polarized light arises in forward/backward direction with respect

to the light propagation. This effect was termed Photoelectron Circular Dichroism (PECD) and so far investigated using synchrotron radiation [1]. In our recent publication [2] we have demonstrated that PECD is accessible via a Resonance Enhanced Multi-Photon Ionization (REMPI) using femtosecond laser pulses. We observed highly structured asymmetries in the range of ± 10%. Attributed to the MPI high order odd Legendre polynomials appear in the measured PECD. In this talk we show our recent findings on the bicyclic Ketones Camphor, Fenchone and Norcamphor. In order to quantify this data we want to introduce quantitative measures. These measures will be used to distinguish the three bicyclic Ketones and to quantify ellipticity dependences on Camphor as well as the enantiomeric excess in mixtures of R- and S-Fenchone.

[1] I. Powis in S. A. Rice (Ed.): Adv. Chem. Phys. **138**, 267-329 (2008)

[2] C. Lux et al., Angew. Chem. Int. Ed. **51**, 5001-5005 (2012)

Invited Talk SYPS 2.3 Thu 17:15 Audimax

Velocity map imaging: from molecules to clusters, nanoparticles and aerosols — •Michal Farnik, Viktoriya Poterya, Jozef Lengyel, Andriy Pysanenko, Pavla Svrckova, and Jaroslav Kocisek — J. Heyrovsky Institute of Physical Chemistry, ASCR, Dolejskova 3, 18223 Prague 8

We will present several experiments with large clusters and nanoparticles performed with our cluster beam (CLUB) apparatus which has recently been upgraded with velocity map imaging (VMI) system. The unique and versatile apparatus allows for various experiments besides VMI including, e.g., high resolution mass spectrometry, particle cross section measurements etc. Combination of these experiments can lead to unprecedented detailed information about the dynamics of photochemistry and photophysics even in large clusters. This will be illustrated for systems of atmospheric relevance such as hydrogen halides and freons on/in ice nanoparticles.

SYPS 2.4 Thu 17:45 Audimax

Imaging Cold Molecules on a Chip — •Silvio Marx[1], David Adu Smith[1], Mark Abel[1], Thomas Zehentbauer[1], Gerard Meijer[1,2], and Gabriele Santambrogio[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Radboud University of Nijmegen, Nijmegen, The Netherlands

We recently reported the manipulation of the external and internal degrees of freedom of cold molecules using a chip-based Stark decelerator. This comprised the trapping, guiding and deceleration of packets of polar molecules as well as the excitation of the molecules' rotational and vibrational degrees of freedom while they were on the chip. Now we present the final crucial component for a fully integrated molecule chip: on-chip detection.[1] By means of resonance-enhanced multiphoton ionization (REMPI) and ion optics we image the molecules in the microtraps of our chip and use this resolution to analyze the phase space distribution of the molecules. This is done by taking time-resolved snapshots of the molecules' ballistic expansion after release from their traps, in a similar fashion as for the time-of-flight imaging of atomic ensembles on atom chips. Moreover, with this detection method we investigate the effect of a phase-space manipulation sequence applied to the trapped molecules.

[1] S. Marx et al., Phys. Rev. Lett. 111, 243007 (2013)

Invited Talk SYPS 2.5 Thu 18:00 Audimax

Velocity map imaging studies of quantum state resolved scattering at gas-solid and gas-SAMs surfaces — •David J. Nesbitt[1], Monika Gruetter[3], J. Robert Roscioli[2], Carl Hoffman[1], and Daniel J. Nelson[1] — [1]JILA/University of Colorado, Boulder, CO, USA — [2]Aerodyne Research Inc., 45 Manning Road, Billerica, MA, USA — [3]University of Goettingen, Goettingen, Germany

This talk describes results from a novel surface-scattering technique which combines resonance enhanced multiphoton ionization (REMPI) with velocity-map imaging (VMI) to yield quantum-state and 2D velocity component resolved distributions in the scattered molecular flux. We will discuss work in hyperthermal scattering (Einc = 21(5) kcal/mol) of jet cooled HCl from i) Au(111) on flat mica surfaces and ii) -CH3 terminated self-assembled monolayers (SAM). These first data establish an exciting new class of experimental tools for exploring energy transfer and reactive scattering events on SAMs, liquid, and metal interfaces with quantum state resolved information on correlated internal and translational distributions.