華東師範大學
EAST CHINA NORMAL UNIVERSITY

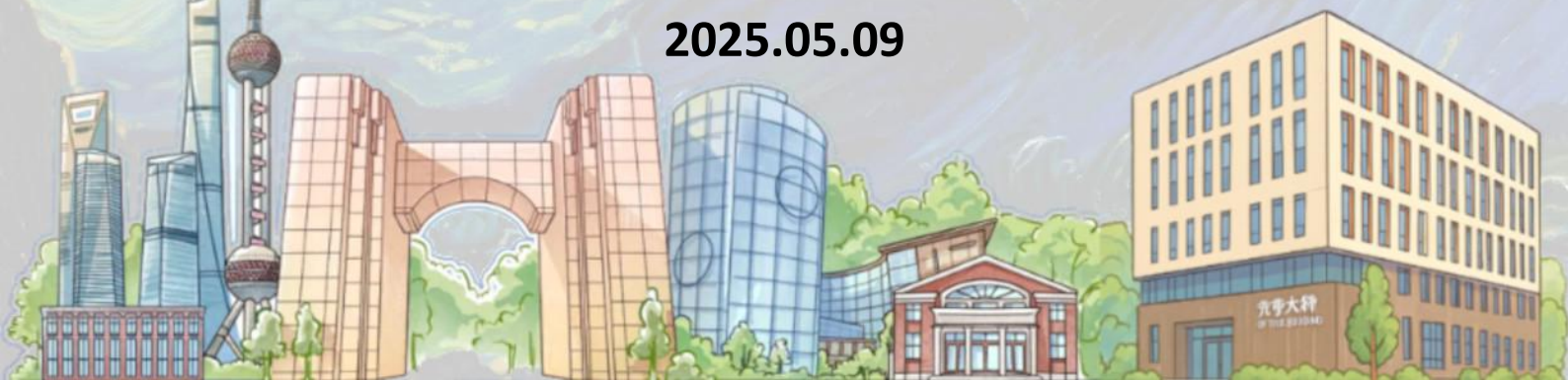
# Joint Seminar on Ultrafast Science

## PROGRAM

**State Key Laboratory of Precision Spectroscopy**

**East China Normal University**

**2025.05.09**



# General Information

## Time and Location

**Time**: 2025.05.09, 08:30-20:00 CST
**Location**: Room B325, Optics Building, East China Normal University

## Pick-up Schedule (Destination: ECNU)

| Guest | Pick-up Time | Pick-up Location | Contact Person |
|---|---|---|---|
| Prof. Stapelfeldt | 08:00 | Marriot ECNU | Wenbin Zhang |
| Prof. Rohringer | 08:50 | Aloft ShanghaiTech | Hui Jiang |
| Prof. Ivanov | 10:00 | SHA Airport T2 | Hongcheng Ni |
| Dr. Yi | 10:00 | PVG Airport T2 | Hongcheng Ni |

## Send-off Schedule (Destination: ShanghaiTech)

| Guest | Depature Time | Depature Location | Contact Person |
|---|---|---|---|
| Prof. Rohringer | 13:00 | Xiayu Hall | Shengzhe Pan |
| Prof. Stapelfeldt | 20:00 | Keyuan Chujian | Hongcheng Ni |
| Prof. Ivanov | 20:00 | Keyuan Chujian | Hongcheng Ni |
| Dr. Yi | 20:00 | Keyuan Chujian | Hongcheng Ni |

## Program Committee

Chair: Prof. Jian Wu (East China Normal University)
Co-Chair: Prof. Kiyoshi Ueda (Tohoku University)

## Contacts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|---|---|---|
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| Dr. Hui Jiang | 13861201577 | 1286338181@qq.com |

# Program Overview

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|---|---|
| Session 1 | Chair: Jian Wu, East China Normal University |
| 08:30 — 08:35 | Opening Address<br>Jian Wu, East China Normal University |
| 08:35 — 09:55 | The Primary Steps of Ion Solvation in a Liquid Helium Nanodroplet<br>Henrik Stapelfeldt, Aarhus University |
| 09:55 — 10:00 | Coffee Break |
| Session 2 | Chair: Hongcheng Ni, East China Normal University |
| 10:00 — 11:20 | X-ray Superfluorerscence<br>Nina Rohringer, University of Hamburg & DESY |
| 11:20 — 11:50 | Ultrafast Dynamics of Small Molecules in Helium Nanodroplets<br>Wenbin Zhang, East China Normal University |
| 11:50 — 12:00 | Group Photo |
| 12:00 — 14:00 | Lunch |
| Session 3 | Chair: Shengzhe Pan, East China Normal University |
| 14:00 — 15:00 | Lab Tour |
| 15:00 — 16:20 | Quantum Optics of High Harmonic Generation: From Atoms to Solids and Waveguides<br>Misha Ivanov, Max Born Insititute |
| 16:20 — 16:30 | Coffee Break |
| 16:30 — 17:00 | Photoelectron Spin Texture in Tunneling Ionization Induced by a Linearly Polarized Laser Pulse<br>Pei-Lun He, Shanghai Jiao Tong University |
| 17:00 — 18:00 | Free discussions |
| 18:00 — 20:00 | Dinner |
| 20:00 | Departure for WUFXS2025 @ ShanghaiTech |

# The Primary Steps of Ion Solvation in a Liquid Helium Nanodroplet

Henrik Stapelfeldt
Aarhus University

Solvation is an omnipresent process both in our daily life and in the natural sciences. At the atomic level, the primary steps of solvation are the attraction and binding of ions or molecules of a solute to molecules or atoms of a solvent. Until recently, these steps had not been observed in real time.

I will present experimental results that have enabled us to observe the solvation dynamics of a single alkali cation in liquid helium with atomic resolution and on the natural femtosecond time scale [1,2]. A single $Li^+$, $Na^+$, or $K^+$ ion is created instantly at the surface of a nanometer-sized droplet of liquid helium and we measured in real time the gradual attachment of individual He atoms to the ion. In the case of $Na^+$, we found that the binding of the first 5 He atoms occurs at a rate of 1.8 atoms/ps, which is consistent with time-dependent density-functional-theory simulations.

In addition, we determined the time-dependence of the energy dissipated from the local region around the alkali ion during its solvation. We found that it follows Newton's law of cooling for the first 5 ps [2].

The experimental methodology applied opens opportunities for exploring in real time the noncovalent bonding of cations to molecules.

[1] S. H. Albrechtsen, C. A. Schouder, A. Viñas Muñoz, J. K. Christensen, C. Engelbrecht Petersen, M. Pi, M. Barranco, and H. Stapelfeldt, Nature 623, 319 (2023).

[2] S. H. Albrechtsen, J. K. Christensen, C. Engelbrecht Petersen, C. A. Schouder, P. J. Carchi-Villalta, I. Sanchez-Perez, M. Bartolomei, T. Gonzalez-Lezana, F. Pirani, and H. Stapelfeldt, submitted (2025), https://arxiv.org/abs/2502.11783

# X-ray Superfluorerscence

Nina Rohringer
University of Hamburg & DESY

X-ray free-electron lasers (XFELs) open new avenues towards studying collective x-ray emission and nonlinear x-ray matter interaction. In this talk I will give a retrospective on the phenomenon of collective spontaneous x-ray emission following ultrafast inner-shell photoinization (x-ray superfluorescence). Theory as well as experimental achievements will be discussed. X-ray superfluorescence has been demonstrated in atomic gases in the soft x-ray range [1], in rare-gases [2] and clusters [3] in the XUV, and in solids and liquids in the hard x-ray range [4,5]. As opposed to the XFEL pulses that are based on the process of self-amplified spontaneous emission (SASE) and have limited temporal coherence, x-ray superfluorescence produces phase-stable, ultrabright x-ray pulses of fs and sub-fs duration [6,7]. Using this principle of inner-shell x-ray lasing, I discuss two new proposals for further tailoring x-ray pulses: By combining collective x-ray emission and Bragg scattering in a crystal --- an x-ray analogue of a distributed feedback laser --- it is possible to create phase-stable pairs of x-ray pulses in the hard x-ray spectral region. Furthermore, we propose a set-up for an x-ray laser oscillator that combines a Bragg cavity, serving as monochromator, and a multi-pass superfluorescent emission [8], thereby offering the opportunity to create x-ray pulses of unprecedented brightness and narrow bandwidth.

[1] N. Rohringer et al., Nature 481, 488 (2012).
[2] L. Mercadier et al., Physical Review Letters 123, 023201 (2019).
[3] A. Benediktovitch et al., Physical Review A 101, 063412 (2020).
[4] T. Kroll et al., Physical Review Letters 120, 133203 (2018).
[5] T. Kroll et al., Physical Review Letters 125, 037404 (2020).
[6] M. D. Doyle et al., Optica 10, 1602 (2023).
[7] C. Weninger and N. Rohringer, Physical Review A 90, 063828 (2014).
[8] A. Halavanau et at., Proceedings of the National Academy of Sciences 117, 15511 (2020).

# Ultrafast Dynamics of Small Molecules in Helium Nanodroplets

Wenbin Zhang
East China Normal University

Superfluid helium nanodroplets, cooling embedded molecules to 0.37 K, serve as exceptional nanoreactors for studying light-induced interactions between cold molecules and their helium environment. To explore the ultrafast dynamics of molecules within these droplets triggered by femtosecond laser pulses, we developed helium-nanodroplet target recoil ion momentum spectroscopy (HeNTRIMS). This advanced reaction microscopy technique measures electrons and ions ejected from a molecule inside the droplet in coincidence, providing detailed insights into molecular behavior.

Using HeNTRIMS in femtosecond pump-probe experiments, we observed laser-induced, field-free molecular alignment dynamics of $D_2$ within helium nanodroplets [1]. Our findings reveal that $D_2$ molecules rotate as if isolated in the gas phase for at least 100 ps, equivalent to more than 500 rotational periods, exhibiting behavior distinct from larger molecules in droplets. Furthermore, we captured the real-time, collision-induced ultrafast dissipation of vibrational nuclear wave packet dynamics in $D_2^+$ ions embedded in helium nanodroplets [2]. Unlike neutral molecules, charged ions strongly interact with the helium solvent through ion-He collisions, resulting in rapid vibrational decoherence within approximately 140 fs. We also investigated how these strong ion-He interactions influence light-induced molecular bond breaking. Through analysis of above-threshold multiphoton ionization of $H_2$ in superfluid helium nanodroplets [3], we found that low-vibrationally excited $H_2^+$ ions in droplets are more susceptible to dissociation compared to the gas phase, where only highly vibrationally excited $H_2^+$ dissociates. These discoveries enhance our understanding of light-induced molecular dynamics in helium nanodroplets and offer pathways for precise control.

Additionally, by modeling the molecule as a de Broglie wave interacting with the droplet's confining potential, we studied the spatial extent of a molecule's matter wave relative to the nanodroplet [4]. Analysis of angular nodal structures in photoelectron momentum distributions (PMDs) revealed preserved nodal patterns, enabling us to differentiate the delocalization of lightweight $H_2$ molecules from the localization of heavier $D_2$ and $O_2$ molecules within the droplet.

[1] J. Qiang et al., Femtosecond Rotational Dynamics of D2 Molecules in Superfluid Helium Nanodroplets, Physical Review Letters 128, 243201 (2022).

[2] J. Qiang et al., Femtosecond Collisional Dissipation of Vibrating $D_2^+$ in Helium Nanodroplets, Physical Review Letters 132, 103201 (2024).

[3] L. Zhou et al., Enhancing Strong-Field Dissociation of $H_2^+$ in Helium Nanodroplets, Physical Review Letters 130, 033201 (2023).

[4] Z. Ye et al., Localization and Delocalization of a Single Molecule in a Helium Nanodroplet, Physical Review Letters (under review).

# Quantum Optics of High Harmonic Generation:

# From Atoms to Solids and Waveguides

Misha Ivanov
Max Born Institute

For more than three decades, high harmonic generation has been the corner stone of attosecond technology. The quantum nature of the material response to intense driving light has never been in doubt, in spite of the many nearly classical aspects of this response. However, during these past three decades, the nature of the generated light was universally assumed to be classical. Recently, this view has started to change. The first reason for the change is the availability of bright quantum sources of incident light, which are already intense enough to enable harmonic generation on their own. These sources can also be mixed with intense classical light, providing well-controlled "hybrid" light at the input that can generate quantum light at the output. The second reason is the possibility of correlated measurements of light, where multiple generated light frequencies are measured in coincidence with the measurement of the transmitted incident light, possibly inducing correlations in the measured photons.

But what about the quantum dynamics of the material system? Can the quantum properties of these material dynamics, triggered and controlled by the classical incident light, be mapped onto the quantum properties of the generated harmonic light? It appears that the answer is "yes", and that achieving this goal might be simpler than one could have expected.

I will present our latest results on controlled generation of quantum light in resonant atomic gases and in quantum systems coupled to structured photonic continua, such as a two-level system inside the waveguide. Our results suggest that the dream of generating multiple harmonics of the incident laser light entangled across multiple octaves, from the infrared (IR) to the extreme ultraviolet (XUV) range, is completely realistic.

# Photoelectron Spin Texture in Tunneling Ionization Induced by a Linearly Polarized Laser Pulse

Pei-Lun He
Shanghai Jiao Tong University

The spin polarization of photoelectrons in tunneling ionization is investigated using numerical solutions of the time-dependent Schrödinger equation in companion with our analytic treatment via the spin-resolved strong-field approximation and classical trajectory Monte Carlo simulations. We demonstrate a nontrivial spin texture of photoelectrons in momentum space, exhibiting a vortex structure relative to the laser polarization axis. The momentum-resolved polarization stems from the emergence of spin-correlated quantum orbits in the continuum. For direct electrons in few-cycle pulses, the nonvanishing initial transverse velocity of the electron is responsible for the polarization, while in long pulses, the spin texture is essentially shaped by recollisions. Furthermore, the interference between direct and rescattering ionization leads to spin-polarized electron holography, offering an alternative method to extract atomic fine structural information.

[1] P.-L. He et al. Photoelectron Spin Texture in Tunneling Ionization Induced by a Linearly Polarized Laser Pulse, Physical Review Letters 134, 163201 (2025).