

## SYAO 1: New Avenues in Molecular Alignment and Orientation

Time: Friday 14:30–16:30

Location: HS 1+2

**Invited Talk** SYAO 1.1 Fri 14:30 HS 1+2  
**Ultralong-range Rydberg molecules: Rotational hybridization, control of alignment and orientation, and Rydberg blockade** — ●ROSARIO GONZÁLEZ-FÉREZ — Dpto. Física Atómica, Molecular y Nuclear & Instituto Carlos I de Física Teórica y Computacional, Universidad de Granada, Granada, Spain

Rydberg atoms form exotic ultralong-range molecules when combined with ground-state atoms, ions, or polar molecules, which inherit their exciting properties. In this talk, we will explore the interaction, via anisotropic scattering, of a polar molecule with a Rydberg atom creating a polyatomic Rydberg molecule. The polar molecule is allowed to rotate in the electric fields generated by the Rydberg electron and core. Thus, its rotational structure is significantly affected, and the diatomic molecule becomes oriented and aligned within the Rydberg orbit [1-2]. We also present the first experimental demonstration of the Rydberg blockade due to this charge-dipole interaction between a Rb atom and a RbCs molecule [3]. The atom and molecule are confined in optical tweezers, and for a separation of 310 nm, the charge-dipole interaction provokes the blockade of the transition to the Rydberg state. The observed excitation dynamics are in excellent agreement with the theoretical results obtained using the electronic structure of the Rydberg molecule Rb-RbCs [3]. These Rydberg molecules possess huge electric dipole moments that allow easy manipulation via electric fields.

[1] Gonzalez-Ferez et al, NJP 17, 013021, 2015. [2] Mellado-Alcedo et al, PRA 110, 013314, 2024, [3] Guttridge et al, PRL. 131, 013401, 2023.

**Invited Talk** SYAO 1.2 Fri 15:00 HS 1+2  
**Quantum control of molecular rotation** — ●DOMINIQUE SUGNY — Université de Bourgogne Europe, Dijon, France

We review recent results on the control of molecular rotation for applications in gas-phase alignment and orientation of linear and symmetric top molecules [1]. We show how control techniques can be applied to produce planar alignment and unidirectional rotational motion. The application of optimal control theory in this context will be discussed. Finally, recent extensions of two- and three-dimensional Anderson localization to rotational dynamics will be proposed. Theoretical and experimental results are presented for the different examples.

[1]- Quantum control of molecular rotation C. P. Koch, M. Lemeshko and D. Sugny Rev. Mod. Phys. 91, 035005 (2019)

**Invited Talk** SYAO 1.3 Fri 15:30 HS 1+2  
**Strong-Field Ionization and Electron Rescattering Probabilities in the Molecular Frame** — ●JOCHEN MIKOSCH<sup>1</sup>, MARTIN GARRO<sup>1</sup>, NARAYAN KUNDU<sup>1</sup>, HORST ROTTKE<sup>1</sup>, KILLIAN DICKSON<sup>2</sup>, VARUN MAKHIJA<sup>2</sup>, FEDERICO BRANCHI<sup>3</sup>, FELIX SCHELL<sup>3</sup>, MARK MERO<sup>3</sup>, C P SCHULZ<sup>3</sup>, SERGUEI PATCHKOVSKII<sup>3</sup>, and MARC

VRAKING<sup>3</sup> — <sup>1</sup>Universität Kassel — <sup>2</sup>Univ. of Mary Washington, Fredericksburg, USA — <sup>3</sup>Max-Born-Institut, Berlin

Alignment of molecules can be used to obtain molecular-frame information from a laboratory-frame measurement. We present two different strategies to obtain the strong-field ionization and the elastic electron-recollision probabilities in the frame of an asymmetric top molecule exposed to an infrared light field with linear polarization. Polar angle-resolved, azimuthal angle-averaged information is achieved from a measurement on one-dimensionally laser-aligned molecules. Both polar and azimuthal angle-resolved molecular-frame information is retrieved by analyzing lab-frame coherent rotational wavepacket evolution following a non-adiabatic alignment laser pulse.

By virtue of ion-electron coincidence detection in a reaction microscope, we separate the ground-state ( $D_0$ ) and first excited state ( $D_1$ ) ionization channel. In this way two scattering experiments on the same target are performed simultaneously with two different continuum electron wavepackets. We find that the nodal structure of the ionizing orbitals is more strongly reflected in the electron rescattering rather than the ionization probability. Experimental results are compared with results from a TD-RIS ab-initio simulation.

**Invited Talk** SYAO 1.4 Fri 16:00 HS 1+2  
**Coherent rotational control of gas phase molecular dipoles by concerted Terahertz and Near-IR pulses** — ●SHARLY FLEISCHER — School of Chemistry, Tel Aviv University, Israel — Tel Aviv Center for Light-Matter Interaction, Tel Aviv University, Israel

The orientation and alignment of gas-phase molecular ensembles is a long-pursued goal in physics and chemistry. Short Near-IR (NIR) pulses are commonly used to ALIGN molecules by exerting torque via the anisotropic polarizability tensor of the molecules. Intense single-cycle terahertz fields (THz) ORIENT the molecules via resonant dipole-field interaction. These above-mentioned 'rotational drivers' (NIR and THz) have been vastly utilized selectively, however their concerted application remains very scarce. In this talk I will focus on two uniquely desirable rotational responses provided by judiciously orchestrated rotational excitations by BOTH THz and NIR pulses.

1)Orientation echo spectroscopy: I will present an efficient scheme for orientation echoes where the first pulse (THz) induces rotational dynamics that is rephased by a second NIR pulse. The dynamics of the multi-level rotational system is governed by the interference of multiple transition pathways within the rotational state manifold and will be discussed.

2)Enhanced molecular orientation at ambient temperatures: I will present a practical scheme for enhanced degree of orientation via concerted NIR and THz excitations. Complementary calculations predict far larger enhancements ( $\sim 20$ -fold with respect to that induced by the THz field alone).