

# Coherent Optical Metrology Beyond Electric-Dipole-Allowed Transitions

THIS SESSION HAS BEEN ORGANISED IN COLLABORATION WITH THE  
SFB COMB.AT

Friday, 22.08.2025, Room Erika Weinzierl Saal

Time	ID	COHERENT OPTICAL METROLOGY BEYOND ELECTRIC-DIPOLE-ALLOWED TRANSITIONS (COMB.AT) <i>Chair: Mikhail Lemeshko, Institute of Science and Technology Austria</i>
11:00	671	<p><b>Theory of angular momentum transfer from light to molecules</b></p> <p><i>Mikhail Maslov<sup>1</sup>, Georgios M. Koutentakis<sup>1</sup>, Mateja Hrast<sup>1</sup>, Oliver H. Heckl<sup>2</sup>, Mikhail Lemeshko<sup>1</sup></i>  <sup>1</sup> Institute of Science and Technology Austria (ISTA), Klosterneuburg, Austria,  <sup>2</sup> Optical Metrology Group, Faculty of Physics, University of Vienna, Austria</p> <p>Structured light carrying orbital angular momentum is instrumental to quantum control, spectroscopy, and sensing, yet a general theoretical framework for its interaction with complex particles has been missing. We present a symmetry-based Hamiltonian—applicable to arbitrary beam profiles and point-group symmetries—that couples spherical gradients of the light field to a particle's multipole moments. In ro-vibrational spectroscopy, our model quantifies how spin and orbital angular momenta of light are transferred to the molecular rotation and its center-of-mass motion, and reveals that transitions forbidden for plane waves can be strongly enhanced. Our analytical approach offers practical guidance for beam shaping and advances the use of light's angular momentum in precision spectroscopy, quantum technologies, and chiral sensing.</p>
11:15	672	<p><b>Optical Vortex–Induced Orbital Angular Momentum Transfer in Ro-Vibrational Spectroscopy</b></p> <p><i>Georgios Koutentakis<sup>1</sup>, Mikhail Maslov<sup>1</sup>, Monika Bahl<sup>2</sup>, Mateja Hrast<sup>1</sup>, Tom Jungnickel<sup>2</sup>, Timo Gaßen<sup>2</sup>, Oliver H. Heckl<sup>2</sup>, Mikhail Lemeshko<sup>1</sup></i>  <sup>1</sup> Institute of Science and Technology Austria (ISTA), Klosterneuburg, Austria,  <sup>2</sup> Optical Metrology group, Faculty of Physics, University of Vienna, Austria</p> <p>Optical vortex beams carry both orbital (OAM) and spin angular momentum. We demonstrate that their helical phase excites ro-vibrational transitions that are forbidden with conventional light. In particular, the gradients of the structured electric field couple to molecular multipoles, permitting quadrupole and higher-order OAM exchange. Analytical and numerical studies reveal that, beyond modal purity quantified by the R-index, OAM transfer relies on the interplay of intrinsic and extrinsic orbital angular momentum. This allows us to classify all paraxial beam profiles in terms of a single figure of merit and identifies checkerboard modes as optimal for light-molecule OAM interaction. Such structured beams enable routes to manipulate molecular dynamics and herald future OAM-enabled spectroscopy for chiral molecules, biomolecules, and complex nanoparticles.</p>
11:30	673	<p><b>The R-Index metric for evaluating OAM Content and mode purity in optical fields</b></p> <p><i>Monika Bahl<sup>1</sup>, Georgios M. Koutentakis<sup>2</sup>, Mikhail Maslov<sup>2</sup>, Tom Jungnickel<sup>1</sup>, Timo Gaßen<sup>1</sup>, Oliver H. Heckl<sup>1</sup></i>  <sup>1</sup> University of Vienna, Austria,  <sup>2</sup> Institute of Science and Technology Austria (ISTA), Klosterneuburg, Austria</p> <p>Light fields can carry orbital angular momentum (OAM), enabling rotational motion through light-matter interactions. Manipulating this momentum has driven major technological advances. Recent work by Maslov et al. on OAM interactions with molecules has renewed interest in using spectroscopy to observe otherwise forbidden transitions. We aim to identify optical fields with optimal OAM that could induce quadrupole transitions in cold molecules. This work introduces the R-index, a novel metric that quantifies the intrinsic canonical momentum or the OAM content in optical fields. The index is also used to assess the purity of a field, offering insight into the fidelity and robustness of OAM generation.</p>

11:45	674	<p><b>Technical Developments of Multi-Pulse CPA for Nonlinear Spectroscopy</b></p> <p><i>Vinzenz Stummer<sup>1</sup>, Edgar Kaksis<sup>1</sup>, Matthias Schneller<sup>1</sup>, Aref Imani<sup>1</sup>, Paolo Carpeggiani<sup>1</sup>, Hongtao Hu<sup>1</sup>, Audrius Pugžlys<sup>1,2</sup>, Andrius Baltuška<sup>1,2</sup></i></p> <p><sup>1</sup> TU Wien, Institut für Photonik, Vienna, Austria, <sup>2</sup> Center for Physical Sciences &amp; Technology, Vilnius, Lithuania</p> <p>Millijoule-energy, picosecond-spaced ultrashort pulses are advancing nonlinear spectroscopy, including <sup>229</sup>Th optical nuclear spectroscopy. Recent burst-mode amplification progress to boost spectral brightness for applications is reported.</p>
12:00		<b>END</b>
12:30		<b>Poster Awards and Closing Ceremony</b>

ID	COMB.AT POSTER
681	<p><b>Shaped light in spectroscopy: how using light carrying OAM can enhance molecular spectroscopy</b></p> <p><i>Timo Gaßen<sup>1</sup>, Mikhail Maslov<sup>2</sup>, Georgios M. Koutentakis<sup>2</sup>, Mirela Encheva<sup>1</sup>, Tom Jungnickel<sup>1</sup>, Monika Bahl<sup>1</sup>, Oliver H. Heckl<sup>1</sup></i></p> <p><sup>1</sup> University of Vienna, Austria, <sup>2</sup> Institute of Science and Technology Austria, Klosterneuburg, Austria</p> <p>Tailoring light to a spectroscopic application is common practice in high performance molecular spectroscopy this includes changing the properties of light by altering its frequency profile or pulse duration. However, changing the spatial structure of light is a practice underutilized in modern spectroscopy. Spatially shaped light could therefore open a new avenue in spectroscopy, studying fundamental light-molecule interactions. One such interaction was proposed in a publication of Maslov et al. where light carrying orbital angular momentum (OAM) modifies the underlying selection rules of the observed transitions. An experiment studying the influence OAM can have on the selection rules was brought forth, with the potential to not only learn about the molecule but also the nature of light carrying OAM.</p>
682	<p><b>Towards the measurement of orbital angular momentum-enabled transitions in molecules</b></p> <p><i>Tom Jungnickel<sup>1</sup>, Monika Bahl<sup>1</sup>, Timo Gaßen<sup>1</sup>, Georgios M. Koutentakis<sup>2</sup>, Mikhail Maslov<sup>2</sup>, Oliver H. Heckl<sup>1</sup></i></p> <p><sup>1</sup> University of Vienna, Austria, <sup>2</sup> Institute of Science and Technology Austria, Klosterneuburg, Austria</p> <p>To measure orbital angular momentum enabled-transitions in the ro-vibrational spectrum of molecules sets unique requirements for the preparation of these molecules. OAM-enabled transitions only affect the magnetic quantum number and to split these degenerate transitions, we use the Stark effect. Furthermore, we utilise buffer gas cooling to limit the Doppler broadening of these transitions. The molecules will then interact with a mid-IR optical frequency comb inside of an enhancement cavity which will determine the transition positions using a Fourier-transform spectrometer. The symmetry of the OAM-carrying beams (e.g. Laguerre Gaussian beams) requires the electric field in the cell to be parallel to the beam. This work combines different simulations to design an optimal experimental setup for this complex experiment.</p>
683	<p><b>Bottom-up Analysis of Ro-Vibrational Helical Dichroism</b></p> <p><i>Mateja Hrast, Georgios M. Koutentakis, Mikhail Maslov, Mikhail Lemeshko</i> <i>Institute of Science and Technology Austria, Klosterneuburg, Austria</i></p> <p>Helical dichroism (HD) is a proposed method for the resolution of molecular chirality, employing the orbital angular momentum (OAM) of light and hypothesized to arise from electric-dipole–electric-quadrupole interactions. Going beyond the conventional assumptions, we propose a rigid theoretical framework for the analysis of HD, based on molecular symmetries and rotational eigenstates. Using our recently developed model of molecule-light interaction Hamiltonian we derive the rotational selection rules, which clearly establish that HD only emerges from spin-orbit coupled light, even for beams without far-field OAM or spin. Our findings refine the conditions for observing HD, shedding light on the outcome of prior experiments and guiding future designs for chiral sensing using structured light.</p>

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Coherent Optical Metrology Beyond Electric-Dipole-Allowed Transitions (COMB.AT) is a Special Research Program targeting disruptive advances in precision spectroscopy. We combine optical frequency combs carrying orbital angular momentum (OAM) with mid-IR and VUV spectroscopy to access dipole-forbidden transitions. The University of Vienna group (Heckl) pioneers OAM-enabled molecular spectroscopy; ISTA (Lemeshko) develops the theory of OAM–molecule interactions; TU Wien (Baltuška) provides ultrafast tailored light sources; TU Wien (Schumm) leads laser-driven nuclear spectroscopy with <sup>229</sup>Th; and Uni Würzburg (Palffy-Buß) advances nuclear excitation theory. In the long term, COMB.AT aims to test variations of fundamental constants—such as the fine-structure constant and proton-to-electron mass ratio—and to lay the groundwork for future molecular and nuclear clocks as precision tools beyond the Standard Model.