Parity Violation in Molecules

850. WE-Heraeus-Seminar

29 March - 01 April 2026

at the Physikzentrum Bad Honnef, Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 850. WE-Heraeus-Seminar:

Fundamental symmetry breaking is important to understanding the formation of our universe including baryogenesis and the homochirality of living organisms. It plays an important role in finding a more complete theory than the Standard Model of particle physics. In recent years, molecules have become a leading platform to search for such symmetry breakings through precision metrology as they exhibit enhanced sensitivity and in particular, their many degrees of freedom can be leveraged to isolate violations of fundamental symmetries from the background. However, in recent years, the molecular precision spectroscopy community has become focused on simultaneous parity and time reversal symmetry violation, overlooking the equal importance of time-reversal symmetry even, parity violating effects. For example, despite its prediction in the 1960s, the parity violating energy difference between mirror-images of chiral molecules is yet to be observed, which is crucial for our understanding of molecular chirality. Only recently several individual works rediscovered the unique opportunities of molecular parity violation, indicating even its undiscovered potential in revealing new physics. Moreover, recent advancements in molecular quantum control and molecular theory may bring the detection of molecular parity violation now in reach for the first time.

This workshop will use the momentum of these recent advances to refocus the attention on molecular parity violation experiments and foster collaboration in the field.

Introduction

Scientific Organizers:

Prof. Dr. Dmitry Budker	Helmholtz Institut Mainz, Germany
Dr. Konstantin Gaul	Helmholtz Institut Mainz, Germany
Ass. Prof. Dr. Yuval Shagam	Technion, Haifia, Israel

Administrative Organization:

Dr. Stefan Jorda
Marion Reisinger

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Introduction

Venue:

Physikzentrum
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Registration:

Marion Reisinger (WE Heraeus Foundation)
at the Physikzentrum, reception office
Sunday (17:00 h – 21:00 h) and Monday morning

Program

Program

Sunday, 29 March 2026

17:00 - 21:00	Registration	
18:00 - 20:00	*BUFFET SUPPER and informal get-together*	
20:00 – 20:30	Dmitry Budker Konstantin Gaul Yuval Shagam	**Welcoming Note & Video Presentation: "About the Wilhelm and Else Heraeus Foundation"**
20:30 - 21:00	Peter Schwerdtfeger	**Towards the First Measurement of Parity Violation in Chiral Molecules – New Attempts and Future Prospectives**
From 21:00	***Musical jam session*** *(feel free to bring instruments)*	

Program

Monday, 30 March 2026

07:30 - 09:00	*BREAKFAST*	
Chiral Molecules I		
09:00 – 09:30	L.D. Barron (to be presented by J. Crassous)	**Cosmic Chirality both True (Parity Violation) and False (CP Violation)**
09:30 - 10:00	Benoît Darquié	**Toward a low-energy test of the parity symmetry via precise mid-IR spectroscopy of cold chiral neutral molecules**
10:00 - 10:30	*COFFEE BREAK*	
10:30 - 11:00	Anastasia Borschevksy	**Molecules as probes of the Standard Model: theoretical perspective**
11:00 - 11:15	Wenhao Sun	**Exploring Molecular Chirality Using Microwave Spectroscopy**
11:15 - 11:45	Jeanne Crassous	**Synthesis of chiral molecules for fundamental spectroscopic measurements**
11:45 – 12:00	Lukas Pasteka	**Selected molecular candidates for parity violation measurements investigated by electronic structure calculations**
12:00 – 12:25	DISCUSSION TIME	
12:25 – 12:30	CONFERENCE PHOTO (in front of the main entrance)	
12:30 – 14:00	*LUNCH*	

Program

Monday, 30 March 2026

Ions & Traps

14:00 - 14:30	David Leibrandt	**Towards precision quantum-logic spectroscopy of chiral molecular ions**
14:30 - 15:00	Jonas Stricker	**Production of multiply charged molecules as versatile probes of fundamental physics**
15:00 – 15:15	Yair Rajmiel	**Generation of cold $CHDBrI^+$ by VUV ionization for precision measurement of PV in molecules**
15:15 – 16:00	*COFFEE BREAK*	
16:00 – 16:30	Michael Drewsen	**Prospects of photon recoil spectroscopy of complex molecular ions**
16:30 – 16:45	Monika Leibscher	**Sympathetic rotational cooling of trapped molecular ions**
16:45 – 18:00	POSTER FLASH	
18:00 – 18:30	DISCUSSION TIME	
18:30 – 20:00	*DINNER*	
20:00 - 22:00	POSTER SESSION	
From 22:00	***Musical jam session*** *(feel free to bring instruments)*	

Program

Tuesday, 31 March 2026

07:30 - 08:45	*BREAKFAST*	
BSM Physics I		
08:45 - 09:15	Victor Flambaum (online)	**Parity Violation and New Physics**
09:15 – 09:45	Yotam Soreq	**P and CP Violation In Molecules as a Probe Of New Range Hadronic Forces**
09:45 - 10:15	*COFFEE BREAK*	
Small Molecules & Atoms I		
10:15 – 10:45	Tim Langen	**Laser Cooling of Molecules for Precision Tests of Fundamental Symmetries**
10:45 – 11:00	Luke Caldwell	**Towards YbCaF molecules for probing nuclear CP violation**
11:00 – 11:30	DISCUSSION TIME	
Chiral Interactions and Chiral Control		
11:30 – 12:00	Yossi Paltiel	**Dynamic Breaking of Mirror Symmetry in Spin-Dependent Electron Transport through Chiral Media**
12:00 – 12:30	Sandra Eibenberger Arias	**Coherent Control of Chiral Molecules**
12:30 – 14:00	*LUNCH*	

Program

Tuesday, 31 March 2026

Small Molecules & Atoms II

14:00 – 14:30	Mikhail Kozlov	**Parity-Nonconserving Spin-Spin Vector Coupling In Diatomic Molecules**
14:30 – 15:00	Ronald Fernando Garcia Ruiz	**Towards Parity Violation Measurements of Single Molecular Ions in a Penning Trap**
15:00 – 15:30	Svetlana Kotochigova	**Relativistic study of rare-earth-containing molecules in search for parity-violating effects**
15:30 – 16:00	*COFFEE BREAK*	

In Solution & NMR

16:00 - 16:30	Malcolm Levitt	**Prospects for NMR detection of parity violation in molecules**
16:30 – 17:00	Giulia Lavarda	**From Molecular Design to Light-Driven Function in Chiral Supramolecular Polymers**
17:00 – 17:15	Erik Van Dyke	**Detecting molecular parity violation with nuclear magnetic resonance**
17:15 – 17:30	Imre Tóth	**High resolution thallium-205 NMR in coordination chemistry**
17:30 – 18:00	DISCUSSION TIME	
18:30	*HERAEUS DINNER*	
	(social event with cold & warm buffet with complimentary drinks)	

Program

Wednesday, 01 April 2026

07:30 - 08:45	*BREAKFAST*	
Small Molecules & Atoms III		
08:45 - 09:15	Debayan Mitra	**Polyatomic Molecular Approach to Parity Violation**
09:15 - 09:45	Emine Altuntas	**ZOMBIES: Towards a measurement of the parity-violating nuclear anapole moment of ^{137}Ba in BaF**
09:45 – 10:15	*COFFEE BREAK*	
BSM physics II		
10:15 – 10:45	Pedro Bargueno (online)	**A simple model for long range parity violation between chiral molecules**
10:45 – 11:00	Cedric Quint	**Stringent Constraints on New Pseudoscalar & Vector Bosons from Precision Hyperfine Splitting Measurements**
11:00 – 11:30	DISCUSSION TIME	
Chiral molecules II		
11:30 – 12:00	Robert Berger	**Parity violating effects in NMR and rotational spectroscopy of chiral molecules**
12:00 – 12:30	Martin Quack	**Fundamental Symmetries and Symmetry Violations in Chiral Molecules: the Quantum Dynamics of Parity Violation and Tunneling from High Resolution Spectroscopy**
12:30 – 12:35	Scientific Organizers	**Closing Remarks**
12:35 - 14:00	*LUNCH*	

End of the seminar and departure

For participants leaving on Thursday a self-service breakfast will be provided on Thursday morning

Posters

Poster Session Monday 30 March 2026

Lennard Marek Arndt	**Tailored targets for precision spectroscopy with multiply charged actinide molecules.**
Roi Ben Zion	**Investigation of Chiral-Induced Spin Selectivity (CISS) in Rubidium-Chiral Molecules interfaces**
Luke Caldwell	**Towards YbCaF molecules for probing nuclear CP violation**
Rohan Chakravarthy	**Effect of a Large Nuclear Moment in ^{173Y}b**
Lei Cong	**Spin-dependent exotic interactions**
Cathy Darling	**Towards the association of triatomic YbCaF**
Simon Diewald	**Production of molecular actinide ions for precision spectroscopy**
Eduardus Eduardus	**Enhanced Vibrational Parity Violation Effects in Chiral Carbon Tetrahedral Cations**
Tatsam Garg	**Precision metrology with molecules in an optical lattice**
Namrata Gohain	**Sum rules in chiral systems**
August Gude	**An entanglement protocol to measure atomic parity violation at sub 0.1% precision**
Raphael Hahn	**Experimental prospects for the spectroscopic measurement of parity violation in chiral molecules**
Timur Isaev	**Prospects of laser-coolable polyatomic molecular ions**
Karol Koziol	**Estimating the QED corrections to energy levels and NMR shielding constant of atoms and diatomic molecules**
Monika Leibscher	**Sympathetic rotational cooling of trapped molecular ions**

Poster Session Monday 30 March 2026

Zheng Li	**Detection of the Parity Violation Energy Difference in Molecules Based on Alternating-Pulse Quantum Sensing**
Mathieu Manceau	**High-resolution mid-infrared molecular spectroscopy for PV measurements**
Marjan Mirahmadi	**Locally Chiral Molecular Rotations Induced by Electronic Transitions**
Klaus Muller Dethlefs	**A Periodic Many-body Quantum Matter Produced in an Ultra-cold Quantum Degenerate Plasma: Direct Observation of Fractional Elementary Charges**
Lukas Pasteka	**Selected molecular candidates for parity violation measurements investigated by electronic structure calculations**
Eifion Prinsen	**Ab Initio Calculations of the Polarizability of BaOH: Towards a Competitive eEDM Measurement**
Cedric Quint	**Stringent Constraints on New Pseudoscalar & Vector Bosons from Precision Hyperfine Splitting Measurements**
Yair Rajmiel	**Generation of cold $CHDBrI^+$ by VUV ionization for precision measurement of PV in molecules**
Mohamed Sabba	**The Veritable $Rh(acac)^3$ and its Chiral Shifts: Rhodium-103 NMR as a Probe of Molecular Parity Violation?**
Yuval Shagam	**An ion trap with velocity map imaging for PV search in chiral molecular ions**
Chiara Schleif	**Investigating small molecular ions using high resolution action spectroscopy methods**
Wenhao Sun	**Exploring Molecular Chirality Using Microwave Spectroscopy**
Ankush Thakur	**P and T-odd interaction constants of YbX (X : Cu, Ag and Au) molecules using KRCI method**

Poster Session Monday 30 March 2026

Imre Toth	**High resolution thallium-205 NMR in coordination chemistry**
Andrzej Tyc	**Ultracold high-spin Σ-state polar molecules for new physics searches – YbCr vs RaCr**
Sophia Vadachkoria	**Electronic Spectroscopy of $YbNH_2$**
Wietze van Boven	**Fock-Space Coupled cluster on parity-violating frequency difference of WHNClF+ vibrations**
Erik Van Dyke	**Detecting molecular parity violation with nuclear magnetic resonance**
Rishi Varan	**Photoelectron Velocity Map Imaging Spectroscopy of MF^- (M = Ba, Yb)**
Nick Vogeley	**A Cold Beam Source for Precision Experiments with Chiral Molecules**
Nevo Werner Reiss	**Novel ESST methods for PV measurements in Chiral Molecules**
Andrey Yachmenev	**Magnetic effects and enantiomer-selectivity in isotopically chiral molecules**
Shilpa Yadav	**Towards full controllability of enantiomer-specific state transfer despite orientational degeneracy**
Chen Zhang	**A theoretical feasibility study on amplifying molecular parity-violation signals using coherent pulse sequences**
Nick Zobel	**Production of Radioactive Molecular Ions via Electron Impact Ionization**
Clara Zyskind	**Testing Fundamental Physics with Trapped Ions**

Abstracts of Talks

(in alphabetical order)

ZOMBIES: Towards a measurement of the parity-violating nuclear anapole moment of ^{137}Ba in BaF

M. Bhattarai[1], V. Chen[1], R. Xi[1], E. Altuntas[2] and D. DeMille[3,4]

[1] University of Chicago, *Chicago, USA*
[2] University of Oklahoma, *Norman, USA*
[3] Johns Hopkins University, Baltimore, *USA*
[4] *Argonne National Laboratory, Lemont, USA*

Parity, one of the fundamental symmetries of nature, is violated by the weak interactions. In atoms and molecules this manifests as nuclear spin independent and nuclear spin dependent parity violation (NSD-PV). While the nuclear spin independent effects have been measured with high precision, NSD-PV effects remain poorly characterized. To date, the only nonzero atomic measurement was performed in Cs, with large uncertainty and disagreement with other data.

We study NSD-PV effects using diatomic molecules, which provide closely spaced rotational/hyperfine levels that enhance parity-violating signals. By bringing opposite-parity rotational levels near degeneracy with a strong magnetic field, we amplify the NSD-PV signal by several orders of magnitude. The NSD-PV interaction matrix element is measured using a Stark-interference technique.

We report progress from the ZOMBIES experiment using BaF molecules, building on a proof-of-principle measurement in ^{138}BaF that demonstrated sensitivity sufficient to measure the predicted NSD-PV effect in ^{137}BaF at the 10% level [1, 2]. Recent upgrades include a cryogenic buffer gas beam source and improved laser frequency stabilization, leading to increased molecular flux and experimental stability. We also discuss ongoing efforts to enhance the low abundance ^{137}BaF signal using magnetic focusing and rotational and hyperfine cooling, as well as recent studies of the hyperfine and Zeeman structure relevant for state preparation and detection [3].

These developments place molecular BaF experiments at the forefront of precision measurements of NSD-PV and open a path toward a direct measurement of the nuclear anapole moment in ^{137}Ba.

References

[1] E. Altuntas, J. Ammon, S. B. Cahn, and D. DeMille, *Phys. Rev. Lett.* 120, 142501 (2018)
[2] E. Altuntas, J. Ammon, S. B. Cahn, and D. DeMille, Phys. Rev. A., 97, 042101 (2018)
[3] F. Kogel, Y. Chamorro, M. Bhattarai, M. Rockenhäuser, T. Garg, D. DeMille, A. Borschevsky, T. Langen, Phys. Rev. A 112, 042807 (2025)

A simple model for long-range parity violation between chiral molecules

D. Martínez-Gil[1], S. Miret-Artés[2] and P. Bargueño[1]

[1]Departamento de Física, Universidad de Alicante, Campus de San Vicente del Raspeig, E-03690 Alicante, Spain

[2] Instituto de Física Fundamental, Consejo Superior de Investigaciones Científicas, Serrano 123, E-28006, Madrid, Spain

In this talk, we develop a system plus environment approach for interacting chiral molecules under a quantum-classical description of the spin-spin model using a Caldeira-Leggett-like coupling. After presenting the interacting model, we show that a long-ranged parity-nonconserving interaction, encoded within a nonlinear Schrödinger equation, produces an energy difference between the two enantiomers of the central chiral molecule when it interacts with a chiral environment. Three examples of such interactions are considered, with particular focus on Z^0-photon vacuum polarization. Finally, we reveal a chirality transmission effect phenomenon, where the time-averaged population difference of the central molecule is amplified when the chiral asymmetry of the environment is considered.

References

[1] D. Martínez-Gil, P.Bargueño and S. Miret-Artés, Phys. Rev. A **112**, 022810 (2025)

Cosmic Chirality both True (Parity Violation) and False (*CP* Violation)

L.D. Barron (to be presented by J. Crassous)

University of Glasgow, Glasgow G12 8QQ, UK

Chirality is a central and unifying theme in modern science. It links the properties of the universe and its constituent elementary particles, through organic stereochemistry, to the structure and behaviour of the molecules of life (1,2). The discrete symmetries of parity *P*, time reversal *T* and charge conjugation *C* may be used to characterize the properties of chiral systems. The concepts of 'true chirality' (time-invariant enantiomorphism) and 'false chirality' (time-noninvariant enantiomorphism) that emerge extend Lord Kelvin's definition to situations where motion is an essential ingredient thereby clarifying, *inter alia*, the nature of physical influences able to induce absolute enantioselection. A truly chiral influence may induce absolute enantioselection in all circumstances; whereas a falsely chiral influence may only induce absolute enantioselection in a process far-from-equilibrium via a 'ratchet effect' associated with a new principle of enantiomeric microscopic reversibility (2). Truly chiral systems support time-even pseudoscalar properties like optical rotation, circular dichroism and Raman optical activity (1,2); falsely chiral systems support time-odd pseudoscalar properties like topological insulators and the elusive magnetic monopole. Parity violation and *CP* violation are manifestations of true and false cosmic chirality, respectively, and may have roles in abiotic enantioselection. Parity violation lifts the degeneracy of (truly) chiral enantiomers, the strict enantiomer (exactly degenerate) now being the mirror-image composed of antiparticles, with strict enantiomers interconverted by the *CP* operation. It is speculated that infiltration of *CP* violation into ordinary matter via an axion-mediated interaction (3), for example, may induce, via the ratchet effect associated with enantiomeric microscopic reversibility analogous to that induced by *CP* violation in elementary particle processes, a tiny yet ubiquitous 'enantioselective push' in all chiral molecular processes far from equilibrium.

References

[1] L.D. Barron, Molecular Light Scattering and Optical Activity, 2nd edn, CUP (2009).

[2] L.D. Barron, Isr. J. Chem. **61**, 517 (2021).

[3] J.E. Moody, F. Wilczek, Phys. Rev. D **30,** 130 (1984).

Parity violating effects in NMR and rotational spectroscopy of chiral molecules

R. Berger[1]

[1]Philipps-Universität Marburg, Fachbereich Chemie,
Hans-Meerwein-Str. 4, 35032 Marburg, Germany

Electroweak interactions and parity-violating interactions with cosmic fields can induce splittings in resonance frequencies of enantiomers. On the low-frequency end, these lead to line-splittings in NMR and rotational spectra. In this presentation, I intend to present some recent activities related to model developments as well as theoretical predictions of parity-odd NMR frequency shifts in heavy-elemental chiral molecules. Additionally, I plan to discuss numerical studies on parity-violating effects in chiral systems amenable to microwave spectroscopy.

Molecules as probes of the Standard Model: theoretical perspective

A. Borschevsky

The Van Swinderen Institute for Particle Physics and Gravity, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

Search for violation of fundamental symmetries provides a unique opportunity for testing the Standard Model. Atomic and molecular experiments offer a low energy and comparatively inexpensive alternative to high energy accelerator research in this field. As the observable effects (such as parity violation, PV) are expected to be very small, highly sensitive systems and extremely precise measurements are required for the success of such experiments. Atomic and molecular theory can provide crucial support for these experiments.

An important task of theoretical research is to identify optimal molecular and atomic systems for measurements and to understand the mechanisms behind the enhanced sensitivity, which is strongly dependent on the electronic structure. Thus, accurate computational methods are needed in order to provide reliable predictions rather than estimates, and to obtain the various parameters that are required for the interpretation of the experiments.

I will present the results of our recent investigations of molecules in the context of search for parity violating effects. An overview of the theoretical methods will be provided, including the recently developed scheme for assigning error bars on theoretical predictions. Then, I will focus on showcasing the different types of systems (diatomic, triatomic, and chiral molecules) that are promising candidates for experiments that aim to test the Standard Model and perhaps detect new physical phenomena [1-3].

[1] Y. Hao, P. Navratil, E. B. Norrgard, M. Ilias, E. Eliav, R.G.E. Timmermans, V. V. Flambaum, and A.Borschevsky
Nuclear spin-dependent parity-violating effects in light polyatomic molecules
Phys. Rev. A **102**, 052828 (2020)

[2] Eduardus, Y. Shagam, A. Landau, S. Faraji, P. Schwerdtfeger, A. Borschevsky and L. F. Pasteka
Large vibrationally induced parity violation effects in CHDBrI
Chem. Commun., **59**, 14579 (2023)

[3] M. R. Fiechter, P. A. B. Haase, N. Saleh, P. Soulard, B. Tremblay, R. W. A. Havenith, R. G. E. Timmermans, P. Schwerdtfeger, J. Crassous, B. Darquié, L. F. Pašteka, and A. Borschevsky
Toward Detection of the Molecular Parity Violation in Chiral Ru(acac)3 and Os(acac)3
J. Phys. Chem. Lett. **13**, 10011 (2022)

Towards YbCaF molecules for probing nuclear CP violation

L. Caldwell[1], B. Fox[1], S Haswell[1], C. Darling[1], H. Sewell[1]

[1]Department of Physics & Astronomy, University College London, London, WC1E 6BT UK

Searches for symmetry-violating nuclear moments provide a powerful probe of physics beyond the Standard Model. The Yb-173 nucleus has a quadrupole-deformed shape, which strongly enhances the size of its magnetic quadrupole moment and makes it an attractive candidate for such studies. We are developing a new experiment that combines laser-cooled Yb atoms with laser-cooled CaF molecules in optical tweezers to assemble triatomic YbCaF molecules. These molecules are predicted to be deeply bound, with a strongly polarised electronic structure around the Yb nucleus, leading to enhanced sensitivity to nuclear symmetry-violating effects. In addition, YbCaF is expected to have a non-linear geometry that gives rise to parity doublets in the electronic and vibrational ground states. These closely spaced opposite-parity states enable efficient lab-frame molecular alignment using modest electric fields and provide powerful tools for suppressing dominant classes of systematic errors. We report on our current experimental progress toward assembling ultracold YbCaF molecules and discuss the key challenges associated with producing and controlling ground-state triatomic molecules for precision measurements.

Synthesis of chiral molecules for fundamental spectroscopic measurements

J. Crassous

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Chirality is encountered at all scales, in both inert and living systems. From a chemical perspective, it represents an everyday concern for industry, notably in pharmaceuticals, agribusiness, and cosmetics. As a result, chirality encourages chemists to develop efficient processes for the preparation of new chiral molecules. In recent years, growing interest has also emerged in materials science, where chiral systems are involved in specific interactions with light or with electron spins, opening the way to novel optoelectronic and spintronic devices.

In fundamental research, parity violation (PV) in chiral molecules has attracted the attention of physicists and chemists for several decades. Measuring the extremely small energy difference (ΔE_{PV}) between the two enantiomers of a chiral molecule remains a major experimental challenge and would constitute a significant breakthrough in our understanding of the underlying physics.

In this lecture, I will describe our efforts to prepare suitable molecules for the prospective experimental observation of parity violation using ultra-high-resolution infrared spectroscopy, in collaboration with physicists and theoreticians [1-5].

References

[1] B. Darquié, C. Stoeffler, A. Shelkovnikov, C. Daussy, A. Amy-Klein, C. Chardonnet, S. Zrig, L. Guy, J. Crassous, P. Soulard, P. Asselin, T. R. Huet, P. Schwerdtfeger, R. Bast, T. Saue, *Chirality* **22***,* 870 (2010).

[2] N. Saleh, R. Bast, N. Vanthuyne, C. Roussel, T. Saue, B. Darquié, J. Crassous, *Chirality* **30**, 147 (2018).

[3] B. Darquié, N. Saleh, S. K. Tokunaga, M. Srebro-Hooper, A. Ponzi, J. Autschbach, P. Decleva, G. A. Garcia, J. Crassous, L. Nahon, *Phys. Chem. Chem. Phys.* **23**, 24140 (2021).

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[5] Q. Sallembien, L. Bouteiller, J. Crassous, M. Raynal *Chem. Soc. Rev.* **51**, 3436 (2022).

Toward a low-energy test of the parity symmetry *via* precise mid-IR spectroscopy of cold chiral neutral molecules

A. Bonifacio[1], S. Viel[1], R. Hahn[1], M. N. Ngo[1], M. Saffre[1], Y. Liu[1], W. Dong[1], E. Cantin[1], O. Lopez[1], A. Amy-Klein[1], M. Manceau[1], B. Darquié[1]

[1]Laboratoire de Physique des Lasers, Université Sorbonne Paris Nord, CNRS, Villetaneuse, France

There is an increasing demand for precise molecular spectroscopy, in particular in the mid-infrared fingerprint window, whether it be for modelling our atmosphere, interpreting astrophysical spectra or testing fundamental physics.

I will present our efforts towards building new-generation mid-infrared spectrometers specifically designed for precision vibrational spectroscopy of complex polyatomic molecules in the gas phase. This includes amongst other things producing gases of polyatomic species cooled to a few kelvins in cryogenic buffer-gas cells [1], developing frequency stabilised mid-IR lasers calibrated to some of the world's best frequency standards [2] and explore the opportunities offered by cutting-edge mid-IR photonics technologies [3]. The proposed technologies are at the forefront of cold molecule research and frequency metrology and have allowed us to measure absolute frequencies of a variety of species with record up to 12-digit accuracies [2].

This opens possibilities for using polyatomic molecules to improve tests of fundamental physics and precision measurements in general. I will focus on our ongoing effort towards measuring the tiny energy difference between chiral enantiomers [4] expected to result from electroweak interactions [5] and to be a sensitive probe of dark matter and Standard Model extensions [6].

References

[1] S. K. Tokunaga *et al*, New J. Phys. **19**, 053006 (2017)

[2] B. Argence *et al*, Nature Photon. **9**, 456 (2015); R. Santagata *et al*, Optica **6**, 411 (2019); D. B. A. Tran *et al*, APL Photonics **9**, 030801 (2024); D. B. A. Tran *et al*, arXiv:2502.08201 (2025)

[3] B. Chomet *et al*, Optica **11**, 1220 (2024); Y. Wang *et al*, New J. Phys. **27**, 023038 (2025); M. Manceau *et al*, Laser Photonics Rev., e00879 (2025)

[4] N. Saleh *et al*, Chirality **30**, 147 (2018); A. Cournol *et al*, Quantum Electron. **49**, 288 (2019); M. Fiechter *et al*, J. Phys. Chem. Lett. **13**, 10011 (2022); Eduardus *et al*, arxiv:2509.26407 (2025)

[5] M. Quack, Angew. Chem. Int. Ed. **41**, 4618 (2002); J. Crassous, C. Chardonnet, T. Saue and P. Schwerdtfeger, Org. Biomol. Chem. **3**, 2218 (2005).

[6] K. Gaul, M. Kozlov, T. Isaev and R. Berger, Phys. Rev. Lett. **125**, 123004 (2020)

Title

Prospects of photon recoil spectroscopy of complex molecular ions

Speaker

Michael Drewsen
Department of Physics and Astronomy
Aarhus University

Abstract

Trapped, single complex molecular ions have in the past been demonstrated to be sympathetically cooled translationally to near the Doppler laser cooling limit through the Coulomb interaction with laser cooled and co-trapped single atomic ions [1]. Furthermore, simpler molecular ions have been sympathetically cooled of to near their motional ground state through resolved sideband cooling of the atomic ions [2,3], and their internal state have been prepared either by buffer gas cooling [4] or probabilistic state detection [5-7]. In the talk, I will discuss, our efforts to extend ground state cooling of complex molecular ions with photon recoil spectroscopy in both the sideband unresolved [8] and resolved regime [9] with the aim - among others - of measuring the chirality of single molecules through a photon recoil spectroscopy version of the schemes presented in in Ref. [10]. Finally, to illustrate the very diverse potential of recoil spectroscopy, an example of how to test for potential extensions to quantum mechanics with charged macromolecules [11] will briefly be discussed.

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[2] G. Poulsen, PhD thesis, Aarhus University, 2011.
[3] R. Rugango *et al.,* New J. Phys. **17,** 035009 (2015).
[4] K. Hansen *et al*., Nature **508**, 76 (2014).
[5] Wolf, F. et al., Nature **530**, 457 (2016).
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[7] M. Sinhal *et al*., Science **367**, 1213 (2020).
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Coherent control of chiral molecules

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Chiral molecules are ubiquitous in nature and they are of great importance in many biological and chemical processes. Over the last two decades there have been important developments in fundamental chiral research and new experimental methods have emerged.

In my presentation, I will introduce our recent developments in the experimental quantum state control of chiral molecules using Enantiomer-Specific State Transfer (ESST) [1,2]. Beyond chiral analysis, ESST enables the control and manipulation of chiral molecules at the quantum level. Using tailored microwave fields, a chosen rotational state can be enriched for a selected enantiomer. This holds great promise for a number of fundamental applications, including measuring parity violation in chiral molecules. Our group has developed a new experimental approach utilizing both ultraviolet and microwave radiation and spectroscopy, overcoming previous limitations in transfer efficiency due to thermal state population and orientational degeneracy [3-5]. I will present details of our recent work together with discussions of the dependence of ESST on various experimental parameters.

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Parity violation and new physics

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We consider effects of parity violation produced by extra Z' boson, axion, long-range parity violating interaction mediated by two light fermions, dark matter and dark energy fields.

Towards Parity Violation Measurements of Single Molecular Ions in a Penning Trap

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This talk will present the developments of an experimental approach based on single molecular ions confined in a cryogenic Penning trap. The strong magnetic field of the trap allows molecular states of opposite parity to be Zeeman shifted into near degeneracy, increasing sensitivity to parity-violating nuclear effects by more than eleven orders of magnitude [1,2]. I will describe the current status of the apparatus, and progress towards a proof-of-principle using SiO^+ ions. The plans for extending this method to different isotopes and complementary molecular systems will be discussed.

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Relativistic study of rare-earth-containing molecules in search for parity-violating effects

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We present a theoretical study of the low lying adiabatic relativistic electronic states of lanthanide monohydroxide (Ln-OH) molecules near their linear equilibrium geometries. In particular, we focus on heavy, magnetic DyOH and ErOH relevant to fundamental symmetry tests. We use a restricted-active-space self-consistent field method combined with spin-orbit coupling as well as a relativistic coupled-cluster method to determine ground and excited electronic states.

Analysis of the results from both methods shows that the dominant molecular configuration of the ground state is one where an electron from the partially filled and submerged 4f orbital of the lanthanide atom moves to the hydroxyl group, leaving the closed outer-most 6s2 lone electron pair of the lanthanide atom intact. The linear DyOH and ErOH molecules have multiple states with the non-zero projections of the total electron angular momentum along the internuclear axis. In absence of external magnetic and electric fields, we are looking for Ω-doubling in rotational states of molecules. These opposite parity states can be used to observe (nuclear) spin-dependent parity violations.

Parity-nonconserving spin-spin vector coupling in diatomic molecules

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For the diatomic molecules the nuclear spin-dependent parity-nonconserving (PNC) electron-nuclear interaction induces PNC vector coupling between two nuclear spins $\boldsymbol{I}$ and $\boldsymbol{S}$:

$$H^{\mathrm{PNC}} = 2\pi\, \boldsymbol{J}^{\mathrm{PNC}} \cdot (\boldsymbol{I} \times \boldsymbol{S})\,,$$

where vector $\boldsymbol{J}^{\mathrm{PNC}}$ is directed along the molecular axis. For heavy nuclei this effect is dominated by the PNC nuclear anapole moment, which was observed only once with 14% uncertainty [1]. For heavy nucleus with charge Z the coupling $\boldsymbol{J}^{\mathrm{PNC}}$ scales faster than Z^3 and for $Z{\sim}80$ can be in the mHz range [2]. In particular, a comprehensive relativistic coupled-cluster calculation for the TlF molecule gave [3]:

$$J^{\mathrm{PNC}}(\mathrm{TlF}) = -3.03\; g_{\mathrm{Tl}}\;\mathrm{mHz}\,,$$

where g_{Tl} is the anapole moment constant for Tl nucleus. Experimental observation of this effect can be made within the framework, used to search for hadronic interactions that violate both time-reversal (T) and parity (P) invariance with the dc electric and magnetic fields replaced by rf fields resonant with a nuclear spin-flip transition. Using the parameters of the proposed CeNTREX experiment on the TlF molecule [4] we estimate that the measurement of the anapole moment constant g_{Tl} with S/N=100 requires accumulation time about 4 hours [3].

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Laser Cooling of Molecules for Precision Tests of Fundamental Symmetries

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Molecules are emerging as powerful platforms for precision measurement science, offering unique sensitivity for high-resolution spectroscopy and tests of fundamental symmetries. These experiments can probe energy scales and interactions that complement searches at high-energy collider facilities, opening new avenues for discovering physics beyond the Standard Model in previously unexplored regimes.

Achieving the required level of control over molecules demands precise manipulation of both internal quantum states and external motion, as well as long coherence times. Laser cooling provides a versatile route to meet these requirements [1].

While well-established techniques enable control over molecular vibrational and rotational degrees of freedom, extending laser cooling to heavy species remains challenging. In particular, complex hyperfine structures arising from multiple nuclear spins significantly complicate the realization of efficient optical cycling. Yet it is precisely these heavy, complex molecules that are especially promising candidates for searches for nuclear parity and time-reversal violation.

In this talk, I will introduce novel laser-cooling strategies designed to address these challenges [2,3]. I will present experimental implementations of these techniques and their application to several isotopologues of barium monofluoride (BaF) [4], and discuss their scalability and prospects for cooling other molecular species - including radioactive ones - relevant to precision tests of fundamental symmetries [5,6]. Finally, I will outline a new experiment based on laser-cooled BaF molecules aimed at probing nuclear-spin-dependent parity violation and the weak interaction.

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From Molecular Design to Light-Driven Function in Chiral Supramolecular Polymers

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Organic materials offer a powerful platform to address challenges in sustainable energy conversion, where function arises from the interplay between molecular structure and supramolecular organization. In this context, chirality is emerging as a fascinating yet still underexplored design principle, capable of influencing molecular packing as well as excited-state and spin dynamics.

Our research explores how chirality encoded at the molecular level transfers across length scales to shape the structure and properties of self-assembled materials. We employ supramolecular polymers, formed through the non-covalent assembly of photoactive building blocks, as model systems to study energy-conversion processes. Through tailored molecular design, we introduce chiral motifs that direct assembly into targeted supramolecular architectures, where molecular arrangement and dynamics govern light-induced processes. Within these supramolecular frameworks, we investigate phenomena highly relevant to organic optoelectronics, including singlet fission, aggregation-induced emission, and exciton transport. These examples illustrate how chiral supramolecular order can be harnessed to control energy, charge, and spin dynamics, providing design rules for next-generation photoactive soft materials.

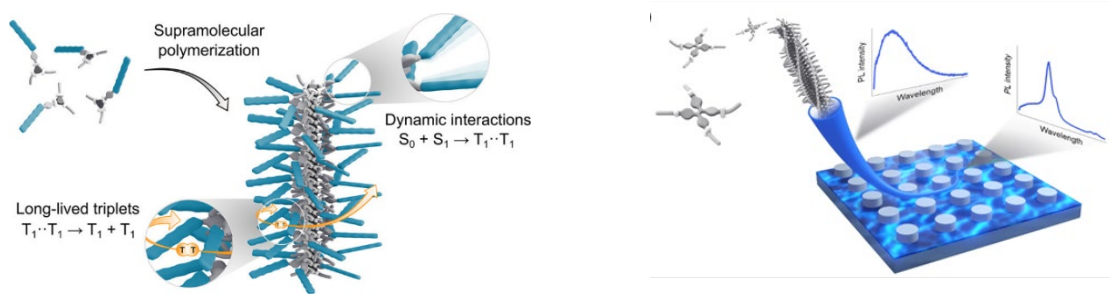

Figure: Long-lived triplets from singlet fission in supramolecular polymers (left) and tunable emission from supramolecular polymers in optical nanocavities (right).

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Towards precision quantum-logic spectroscopy of chiral molecular ions

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A revolution is underway in quantum control of molecular ions based on quantum-logic spectroscopy (QLS). In these experiments, a single molecular ion of interest is co-trapped together with a single atomic "logic" ion, which is used for sympathetic laser cooling of translational motion and quantum-logic readout of internal molecular states. Because this readout technique is non-destructive, it can be used to prepare pure vibrational, rotational, and hyperfine states of the molecule in a probabilistic but heralded fashion [1]. A wide spectrum of coherent molecular transitions can be driven as two-photon stimulated-Raman-transitions using a single CW laser or femtosecond laser optical frequency comb [2,3]. These sources can be hundreds of terahertz away from resonance with any molecular transitions, and no resonant lasers are required for the molecule. Thus, a broad class of molecular ion species can be studied in a single experimental apparatus using only a few lasers, unlocking new opportunities in the search for parity violation and physics beyond the Standard Model. Thus far, however, these techniques have only been demonstrated on diatomic molecules.

This talk will present a new experiment being constructed at UCLA for QLS and precision measurements of polyatomic molecules. The primary obstacle to QLS of polyatomics is that many more states are populated by thermal radiation. We have designed a cryogenic vacuum chamber optimized to suppress the power spectral density of thermal radiation near molecular transition frequencies, and designed and simulated new machine-learning based QLS algorithms that reduce the number of measurements necessary for preparation of pure quantum states [4]. The current status of the apparatus as well as plans for parity violation searches with chiral polyatomic molecules will be discussed.

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Sympathetic rotational cooling of trapped molecular ions

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Molecules, with their rich internal structure, are emerging as a powerful platform for fundamental physics, including precision measurements of parity violation. In particular, their rotational degrees of freedom provide access to a large, well-isolated Hilbert space with strong anharmonicities, enabling a high level of quantum control. A prerequisite for realizing this level of control is the ability to trap and cool the molecules.

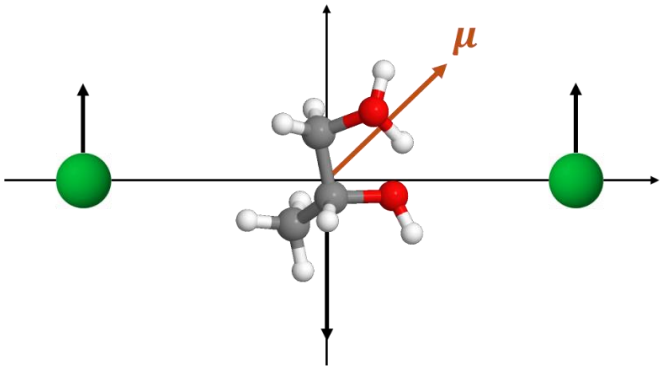

Figure: Protonated propanediol co-trapped with two Yb+ ions in a linear Paul trap. The black arrows indicate a motional mode of the particles in the trap. The molecular dipole μ couples the rotational degrees of freedom of the molecule to the normal modes of the trap, making the rotational subspace amenable to sympathetic sideband cooling.

We propose a protocol for the sympathetic cooling of a molecular asymmetric-top rotor co-trapped with laser-cooled atomic ions, based on resonant coupling between the molecule's electric dipole moment and a shared motional normal mode of the ion crystal [1]. We demonstrate the feasibility of resonant dipole–phonon coupling across a broad class of molecular ions and evaluate it for heavy diatomic species, namely ThF^+, and polyatomic asymmetric-top molecules. Moreover, by combining rotational sideband cooling with coherent microwave control, we show that the rotational state distribution can be engineered, either by depleting arbitrary rotational subspaces or by cooling the full manifold into a single rotational state. This enables the exploitation of the rotational Hilbert space of molecules for applications in quantum information processing and high-precision spectroscopy.

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Prospects for NMR detection of parity violation in molecules

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Nuclear Magnetic Resonance (NMR) spectroscopy is a very high-resolution form of spectroscopy which is applicable to substances in any physical state (gas, liquid, solid) and which is capable of detecting minute perturbations of the electronic environment of atomic nuclei. In favourable cases, NMR can detect frequency shifts and spin-spin couplings in the range of mHz or less[1]. The observation of long-lived nuclear spin states with decay times exceeding 1 hour in room-temperature solution[2] suggest the possibility of extending the frequency resolution even further. Furthermore, active electronic feedback has been claimed to improve the frequency discrimination of NMR signals by at least one order of magnitude[3].

Parity violation has been predicted to induce chemical shift differences of the order of μHz between the enantiomers of some chiral molecules[4,5]. Detection of such tiny differences in NMR interactions poses formidable challenges. I will attempt to assess the feasibility of detecting minute parity violation effects in NMR, including the use of bespoke chemical synthesis and advanced NMR methodology, such as the exploitation of geometric quantum phases[6].

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Polyatomic Molecular Approach to Parity Violation

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Parity violation beyond the Standard Model is necessary to explain the absence of antimatter in the universe. Several tests of parity violation have been performed over the decades using both table-top experiments and particle accelerators [1]. However, we are yet to measure significant deviation from the Standard Model. Most notable is the lack of low energy measurements, a sector where table-top experiments outperform accelerators. In this talk, I will describe how mercury-based polyatomic molecules offer new handles for symmetry tests. Nuclear-spin-dependent parity violation (NSD-PV) in molecules is enhanced in open-shell, high Z species with closely spaced levels of opposite-parity, where electric-field tuning enables Stark-interference detection [2]. Furthermore, the existence of unique rotational modes and vibrational structure can be exploited to reduce systematics while maintaining large internal electric fields and strong sensitivity to NSD-PV. I will describe our plan to spectroscopically study mercury-based molecules of increasing complexity, from HgF to $HgNH_2$ to $HgOCH_3$. We will investigate the feasibility of laser cooling to produce slower beams and possible 3D traps. For complex polyatomic species, some non-laser-cooling based techniques must be developed to overcome the challenges posed by molecular structure. I will describe our roadmap for the measurement of NSD-PV using the novel platform of polyatomic molecules.

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Dynamic Breaking of Mirror Symmetry in Spin-Dependent Electron Transport through Chiral Media

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Two fundamental questions have puzzled scientists for more than 150 years. The first is, "how did life become homochiral?" The second, related question is, "why was this specific handedness selected?" By homochiral, we mean that all living organisms contain chiral molecules, i.e., molecules that exist in two forms (enantiomers), like left and right hands. However, in life on Earth, we predominantly find only one form: the L-form for amino acids, and the D-form for sugars and nucleic acids such as RNA and DNA.

Some models have been proposed to explain the development of homochirality during evolution. Recently, it has been shown that homochirality could have emerged through the enantioselective interactions of molecules with magnetic substrates due to the asymmetric crystallization of an RNA precursor on a magnetite substrate, abundant on early Earth. This phenomenon is based on the chirality-induced spin selectivity (CISS) effect [1,2]. Despite its robustness, this model, like some other previously proposed ones, could not provide an answer to the second question: why one specific handedness (D for RNA) was selected. This difficulty stemmed from the assumption that two mirror-imaged forms of a chiral molecule are perfectly identical.

Here we demonstrate that spin-involving processes can have different outcomes in the two enantiomers of chiral molecules.

We show by direct measurements, theory, and *ab initio* calculations that dynamic spin processes in chiral molecules could result in different efficiencies of spin-related phenomena, including the interaction of chiral molecules with magnetic surfaces. The findings may lead to new venue for control enantiospecific interactions, applying electrons' spin polarization, and may provide an explanation for the specific homochirality in nature.

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Fundamental Symmetries and Symmetry Violations in Chiral Molecules:

the Quantum Dynamics of Parity Violation and Tunneling from High Resolution Spectroscopy

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The quantum dynamics of chiral molecules were central to the discovery of quantum tunneling shortly after the development of quantum mechanics in the form of the Heisenberg equations of motion and the Schrödinger equation (1-3). Later in the 20th century they were again crucial in understanding the fundamental role of parity violation for stereochemistry and in introducing electroweak quantum chemistry (4-6). In the 21st century chiral molecules may become the key to the discovery of extraterrestrial life by using small chiral molecules as tracers for homochirality in astrophysical spectroscopy. In the lecture we shall start with a short historical and conceptual introduction and then report about recent work of our group in Zurich. After an effort of 40 years we are close to finally measuring the parity violating energy difference D_{pv} between the enantiomers of chiral molecules, which has been well studied by theory but so far not yet observed in any experiment(4-6). Our current experimental setup has an estimated detection limit for D_{pv} of about 100 aeV (7). Major current efforts concern the understanding of the complex high resolution spectroscopy and quantum dynamics of chiral molecules and isotopic chirality as governed by the interplay of rotation, vibration, tunneling, and parity violation which is important both for the design of the experiment for detecting molecular parity violation and also for astrophysical spectroscopy (See(8), www.ir.ETHz.CH and references therein)

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Stringent Constraints on New Pseudoscalar & Vector Bosons from Precision Hyperfine Splitting Measurements — •Cedric Quint[1], Fabian Heisse[1], Joerg Jaeckel[2], Lutz Leimenstoll[1], Christoph H. Keitel[1]Zoltán Harman[1] — [1]Max Planck Institute for Nuclear Physics — [2]Institute for Theoretical Physics, Heidelberg University

Axion-like particles and similar new pseudoscalar as well as vector bosons coupled to nucleons and electrons are predicted to lead to spin-dependent forces in atoms and ions. We argue that hyperfine structure measurements in hydrogen- and lithium-like charge states are a sensitive probe to this effect. Employing specific differences of these splittings reduces uncertainties due to nuclear effects in hyperfine structure calculations and measurements. Using this, we show that existing measurements on Be provide competitive limits in the region $m_\phi \gtrsim 100\,\mathrm{keV}$, improving upon existing constraints by up to a factor of 2 for pseudoscalar couplings. We also find that future measurements on Cs have a further factor of $2-2.5$ improved discovery potential for pseudoscalars and an order of magnitude for new vector bosons when compared with the corresponding current constraints.

Reference: arXiv:2506.03274

Generation of cold $CHDBrI^+$ by VUV ionization for precision measurement of PV in molecules

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Measurement of Parity Violation (PV) in trapped chiral molecular ions requires the molecules to be internally cold. Our main candidate molecule $CHDBrI^+$ is promising due to its predicted PV shift of 1.8 Hz in the C-H bend mode, while maintaining a high dissociation threshold of 1.29 eV with an 8 second radiative lifetime [1]. Although it is common to prepare cold molecular ions in ion traps through UV REMPI, the UV radiation leads to fragmentation of halomethane molecules. We will present how ionization using VUV light avoids these dissociative processes. Additionally, we will demonstrate that utilizing autoionizing molecular Rydberg states in the ionization process enables the selection of the final vibrational state. However, this method does not provide rotational state selectivity. We plan to incorporate an IR rovibrational resonant transition in the first step of the ionization to filter specific rotational quantum states in this 1+1' scheme. The second VUV photon will be tuned to a specific vibrational end state to achieve rotational and vibrational selectivity in the resulting molecular ions. To this end, we have performed rovibrational IR spectroscopy of the neutral precursor candidate CHDBrI, along the C-H stretch mode.

Our method allows the preparation of various excited long-lived vibrational states in the molecular ion. This will allow probing of hot band transitions which may show enhanced sensitivity to PV. This has been predicted to be the case in combination modes in other halomethane molecules [2].

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Towards the First Measurement of Parity Violation in Chiral Molecules – New Attempts and Future Prospectives

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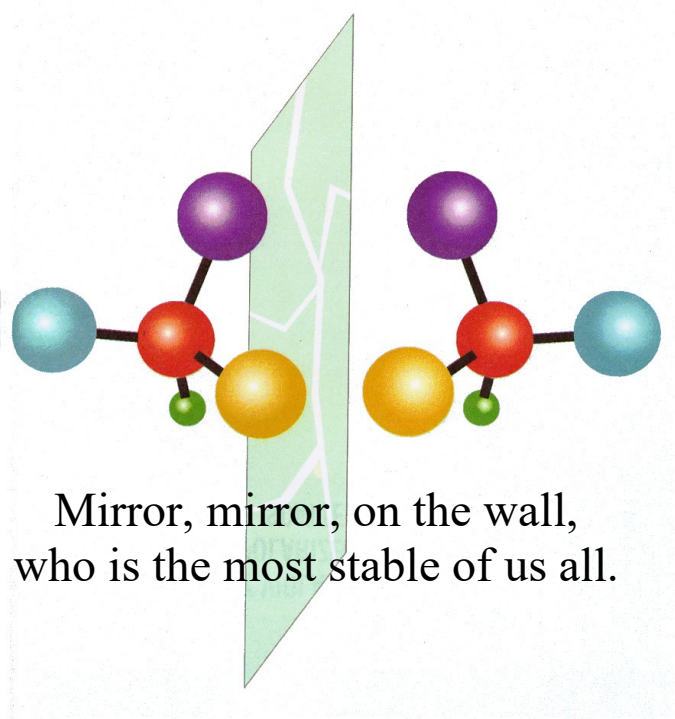
A fundamental discovery of this century is that our Universe is "left" handed (Weinberg-Salam-Glashow theory), the electro-weak interaction (parity-odd) gives rise to primarily left-spinning electrons during beta decay. In 1957 Lee and Yang discovered parity violation (PV) in the K^+ decay, which was confirmed shortly after by Wu et al. for the b-decay. In the last decade PV effects in atomic transitions have been measured and calculated to high accuracy confirming the so-called standard model in particle physics. PV through Z-boson exchange between electrons and nucleons, leads to a small energy difference between enantiomers of chiral molecules although there is no experimental verification yet of this distinct symmetry breaking effect despite many attempts. Current high resolution optical spectroscopy carried out in the CO_2 laser frequency range (878-1108 cm^{-1}) at LPL in Paris achieves resolutions of about 1 Hz. Recent calculations in our group applying the standard model show that PV effects in vibrational transitions of chiral methane derivatives CFXYZ (X,Y,Z= H, Cl, Br, I) are in the mHz range and below the detection limit. Our research group is therefore searching for molecules including heavy elements (because of the PV Z^5-scaling) to achieve enhanced PV effects in the Hz range. New promising candidates are presented in collaboration with the French PV initiative, which aims at a 100 mHz resolution in Ramsay-Fringes experiments using quantum cascade lasers. Another future alternative is single-molecule spectroscopy in traps at ultra-cold temperatures.



P and CP violation in molecules as a probe of new range hadronic forces

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The searches for P and CP violating effects in molecules are a power full probe of physics beyond the standard model. In this talk we focus new long range interactions between nucleons which break partiy or CP. We demonstrate the sensetivity of di-atomic and chiral molecules to these new interaction. In paricualr, we show that electron EDM measurements in di-atomic molecules provide the most sensetive terrestical bound on new CPV spin-0 long range interaction. Future parity violaton of chiral molecules, direct comparision sbetween the left and right handed moleculs are promising probe of new PV forces at molecular scale. The talk is based on Refs. [1] and [2].

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Production of multiply charged molecules as versatile probes of fundamental physics

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We present two efficient and versatile approaches to produce multiply charged lanthanide- and actinide-containing molecules based on high fluence laser ablation and electron-beam ion trapping.

High fluence laser ablation of chemically tailored targets such as ThF_4 and metallic uranium produces a broad range of molecular species, including ThF_x^{n+} ($x = 0 – 3$, $n \leq 3$) and UO^{n+} up to $n = 4$. Molecules of particular relevance for searches for nuclear Schiff moments, such as ThF^{2+} and UO^{3+}, are readily accessible, while UO^{4+} approaches the limit of chemical stability [1, 2]. High fluence laser ablation of well-prepared targets provides a reliable and reproducible method to produce multiply charged molecules, and enables access to species such as PaF^{3+}.

In addition to high fluence laser ablation, we discuss the production of multiply charged molecular ions through electron impact ionization. For controlled ionization and charge state selection of these fragile species, neutral molecules from laser ablation are injected into a Heidelberg compact electron beam ion trap (HC-EBIT) [3]. The tunability of the EBIT enables non-destructive ionization via electron impact and selective preparation of desired charge states, establishing the system as a high-efficiency source for precision spectroscopy. This production and ionization scheme constitutes a key element of the CHArged Radioactive Molecule Spectroscopy (CHARMS) project, which aims to probe fundamental symmetries and physics beyond the Standard Model.

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Exploring Molecular Chirality Using Microwave Spectroscopy

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High-resolution microwave spectroscopy has emerged as a powerful and versatile tool for the investigation of chiral molecules in the gas phase. By exploring precise rotational fingerprints, this technique enables unambiguous spectral characterization and structural determination, even for complex and heavy-atom–containing species. When combined with phase-sensitive methods such as microwave three-wave mixing, microwave spectroscopy allows for sensitive chiral recognition and enantiomer-specific measurements. Beyond characterization, tailored microwave fields can be used to manipulate molecular populations and coherences in selected rotational states, opening pathways toward the quantum control of chiral molecules and the exploration of fundamental chiral dynamics.

In this contribution, I will present a series of studies conducted by our group over the past few years, including the characterization of chiral molecules, particularly heavy-atom–containing species, chiral recognition, quantization, and population control using microwave three-wave mixing spectroscopy. I will also discuss our most recent progress on the quantum control of tunneling motions that interchange mirror-image structures.

High resolution thallium-205 NMR in coordination chemistry

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Thallium is known for the layman from its (well-overrated) poisonous behavior. Tl has two natural stable isotopes, both ^{203}Tl (29.5 %) and ^{205}Tl (70.5 %) are half spin nucleus, I = ½, with outstanding sensitivities. ^{205}Tl is more sensitive, receptivity relative to ^{1}H NMR is 0.142. The resonance frequency (57.68 MHz at 2.35 T) is outside the range of most NMR probes, only very laboratories can record $^{203/205}$Tl NMR spectra using selective Tl-probe.

The classic book by A. G. Lee is a gold mine of the rich Tl-chemistry [1], our short summary of Tl-inorganic chemistry [2] might help someone getting an overview.

Tl has +1 or +3 oxidation state, Tl^{+} resembles to K^{+} (and Ag^{+}), while $Tl(H_2O)_6^{3+}$ is a strong oxidant and extremely hydrolysable in aqueous solution. In contrary to the lighter elements of Group 13 (Al^{3+}, Ga^{3+} and In^{3+}) Tl^{3+} is a typical “soft” ion. The coordination number and geometry of Tl(III)-complexes are different depending on the structure of multidentate organic ligands and/or size of the monodentate ones. The presentation deals with selected examples of Tl(III)-complexes studied in our laboratories by ^{205}Tl NMR: e.g. Tl(III)-cyanides [3], $[Tl_2Na_2(H_2O)_2\ (P_2W_{15}O_{56})_2]^{16-}$[4], and $[(CN)_5Pt\text{-}Tl(CN)_n]$ (n= 0,1,2,3) [5] focusing on equilibrium, ligand exchange kinetics and structure.

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Detecting molecular parity violation with nuclear magnetic resonance

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Abstract

The weak interaction has been shown experimentally to exhibit parity nonconservation (PNC) in both nuclear and atomic systems, yet no experiments have been able to observe PNC in a molecular system. This work seeks to build on previously demonstrated use of co-sensing and diastereomeric splitting in high-field NMR spectroscopy to resolve energetic differences between chiral molecules from nuclear-spin-dependent contributions of the weak force to the chemical shift tensor [1,2]. Residual chemical shift differences scale with Z^n $(2 < n < 5)$ and are expected to be on the order of mHz between enantiomers containing high Z, spin ½ nuclei such as ^{195}Pt, ^{205}Tl/^{203}Tl, and ^{207}Pb [2,3,4].

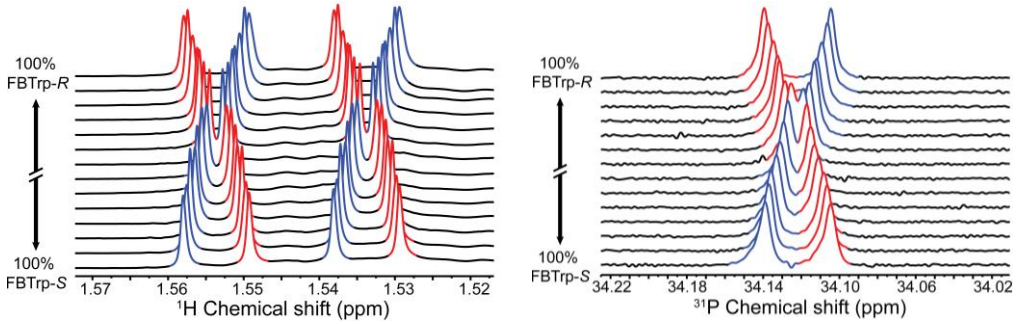

Figure 1. Titration of a chiral solvating agent undergoing rapid, reversible binding (k_{ex} >> R_2) with a sensor molecule generates a variable diastereomeric splitting in NMR spectra. Co-sensing is employed by comparing diastereomeric splitting in a light nucleus (^{1}H, ^{13}C, ^{15}N, …), which is insensitive to PNC, and a heavy nucleus (^{195}Pt, ^{205}Tl, ^{207}Pb) which is PNC sensitive. Combining these measurements mitigates systematic error to resolve PNC-induced splitting in the NMR spectrum of the heavy nucleus, which is orders of magnitude smaller than the linewidth.

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Abstracts of Posters

(in alphabetical order)

Tailored targets for precision spectroscopy with multiply charged actinide molecules.

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Multiply charged molecular actinide ions are attractive candidates for precision spectroscopic tests of fundamental symmetries. Such molecules can be produced via laser ablation of metallic foils (in case of Th and U) or chemically tailored targets containing the required elemental constituents. [1] While the ablation of metal foils is comparably simple and well reproducible, the elements serving as ligands of the desired molecular ions required for spectroscopy (e.g., fluorine or oxygen) are usually only present in trace amounts. Therefore, we demonstrate the production of suitable ablation targets using I) chemical treatment of metal foils, such as oxidation via H_2O_2, II) direct synthesis of salt-based targets and III) electrochemical deposition on conductive substrates. [2,3] Chemical treatment of metal foils can enhance the amount of the desired ligand elements but is limited to abundant isotopes which can be cast into metal foils. On the other hand, tailored salt-based targets of rare isotopes can be produced by direct synthesis and subsequent electroplating. Depending on the application, the targets can be chemically designed to already contain all elements required for the efficient formation of the desired molecular species or to preferentially provide atomic ions for in-flight reactions. Comparative studies of a variety of chemically prepared ablation targets can thus form the basis for physics and spectroscopy experiments to explore the limits of the Standard Model. Examples of produced laser ablated ions from tailored targets include ThF^{2+}, PaF^{3+}, UF^{4+}, ThO^{+}, and UO^{3+} which are all considered as promising probes in the hunt of new physics. [1-4]

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Investigation of Chiral-Induced Spin Selectivity (CISS) in Rubidium-Chiral Molecules interfaces

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The Chiral-Induced Spin Selectivity (CISS) effect describes how electron transport through chiral molecules becomes spin-dependent, favouring a particular spin orientation. While CISS has been extensively explored in solid-state platforms, the broader impact of chiral molecular interfaces on atomic spin systems remains largely unexplored[1].

In this work, we investigate the interaction between atomic vapors and chiral molecules by fabricating vapor cells with a chiral coated inner surface, and comparing the nonlinear magneto-optical rotation (NMOR) spectroscopic response of chiral-coated (for two enantiomers), and non-coated rubidium vapor cells. Measurements are performed in a magnetically shielded environment, under well-defined magnetic fields applied using coils in Helmholtz configuration. By analysing polarization-rotation signals from coated and uncoated cells under comparable experimental conditions, we explore whether chiral coating induces a surface and enantiomer-dependent modifications to the vapor.

Our measurements indicate chirality-dependent differences in the NMOR response between coated and uncoated cells,including a narrowing of the lineshapes in chiral-coated vapor cells, as well as intriguing power dependent modifications of the NMOR response. In addition, we observe indications that the response to applied transverse magnetic field differs between opposite molecular handness in chiral-coated vapor cells. This approach establishes a controlled methodology for exploring chirality-induced spin selectivity in atomic systems and provides a pathway toward integrating chiral molecular effects into vapor-cell-based precision measurements.

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Effects of a large nuclear moment of ^{173}Yb

January 7, 2026

We report a measurement of the hyperfine state and nuclear spin dependent quenching of the lifetime of the excited $4f^{13}6s^2 : F_{7/2}$ state in $^{173}\mathrm{Yb}^+$ measured by coherently exciting the highly forbidden $4f^{14}6s : S_{1/2} \rightarrow 4f^{13}6s^2 : F_{7/2}$ transition [1]. We also report a measurement of the hyperfine structure of the $4f^{14}5d : {}^2D_{5/2}$ state in $^{173}\mathrm{Yb}^+$ using the $4f^{14}6s : {}^2S_{1/2} \rightarrow 4f^{14}5d : {}^2D_{5/2}$ electric quadrupole (E2) transition at 411 nm in trapped $^{173}\mathrm{Yb}^+$ ions and preliminary results on the resolution of the higher order hyperfine structure C coefficient. The measurement involves coherent excitation of the atom with an ultrastable laser to the excited hyperfine states and the measurement of the absolute frequency of the transitions with a frequency comb referenced to an ultrastable silicon cavity and a hydrogen maser. This measurement, along with the planned measurement of the hyperfine structure of the $4f^{13}6s^2 : {}^2F_{7/2}$ state will lead to the resolution of the higher order nuclear moments predicted in Yb.

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Title: Spin-dependent Exotic Interactions

Authors: Lei Cong[+,]*, Wei Ji[#,]*, Pavel Fadeev, Filip Ficek, Min Jiang, Victor V. Flambaum, Haosen Guan, Derek F. Jackson Kimball, Mikhail G. Kozlov, Yevgeny V. Stadnik, Dmitry Budker

Institute: Helmholtz-institute Mainz and so on

The fifth force may arise due to “new physics” beyond the Standard Model. We focus on the spin-dependent fifth forces [1] that are mediated by new particles, such as spin-0 particles (axion and axion-like particles) and spin-1 particles (e.g., light Z’ particle or massless paraphoton). These new ultralight particles are also candidates for dark matter and dark energy, and may also break fundamental symmetries. Spin-dependent interactions between fermions have been extensively searched for in experiments, employing methods such as comagnetometers, nitrogen-vacancy spin sensors, and precision measurements of atomic and molecular spectra [2, 3, 4]. Our research involves a theoretical reassessment of exotic spin-dependent forces [5]. It produces a systematic and complete set of interaction potentials expressed in terms of reduced coupling constants. We conduct an extensive analysis of the existing body of experimental literature on spin-dependent fifth forces, which produces systematic exclusion plots. This leads to a comprehensive understanding of the current research landscape and provide insights for further research.

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Towards the association of triatomic YbCaF

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Our experiment aims to search for CP symmetry-violating nuclear moments by embedding the quadrupole-deformed Yb-173 nucleus within triatomic YbCaF. YbCaF is predicted to be bent in its ground state, which may lead to closely spaced parity doublets, favourable for use in precision measurement experiments. Work is currently being undertaken towards the cooling and trapping of both the Yb-173 atoms, as well as CaF molecules. A beam of Yb is Zeeman-slowed and captured in a 2D MOT, before being transferred to a 3D MOT. CaF molecules will be produced by laser ablation of calcium in a cryogenic helium buffer-gas cell, forming a cold molecular beam that is slowed using frequency-chirped light before capture in a molecular MOT. Single Yb atoms and CaF molecules will then be loaded into separate optical tweezers, which will be merged. We will then investigate atom-molecule collisions, with the aim of identifying a Feshbach resonance which can be used to associate YbCaF.

Production of molecular actinide ions for precision spectroscopy

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Actinide molecular ions are of direct relevance for high-precision spectroscopy in the search for effects of physics beyond the Standard Model [1]. The production of molecular actinide ions is an experimental challenge and requires a starting material that contains all relevant elements as well as a suitable method for extracting the molecular ions from the source. Established approaches include the ISOL technique and hot cavity ion sources combined with resonant ionization (RILIS) for the production of atomic ions, followed by in-flight chemical reactions in a buffer gas to form molecular species [2]. Laser ablation from tailored ablation targets has proven to be a viable and often underestimated alternative for samples and isotopes available in microgram to sub-microgram quantities. Especially in cases where molecular ions in charge states above 1+ are required, laser ablation has demonstrated its versatility for both the generation and investigation of such species.

In this contribution, we present the production of multiply charged actinide fluoride molecular ions using high-fluence laser ablation of salt-based targets of UF_4, NpF_5, and PaF_x [3]. Following the successful production of multiply charged ThF_x^{n+} (with x = 0-3, n =1-3) including ThF^{2+} molecular ions [4], UF^{4+} molecular ions were produced from UF_4 targets, and PaF^{3+} was observed as a prominent molecular species from PaF_x targets. In addition, a possible indication for the production of NpF^{5+} is discussed [3]. The molecular ions ThF^{2+}, PaF^{3+}, UF^{4+}, and NpF^{5+} are all isoelectronic to the well-known RaF molecule [5] and hence of interest in the search for physics beyond the Standard Model. Thus, high-fluence laser ablation of actinide fluoride salts provides a viable pathway toward generating such RaF-like molecular ions and establishes an experimental foundation for future high-precision spectroscopic studies.

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Enhanced Vibrational Parity Violation Effects in Chiral Carbon Tetrahedral Cations

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Recently, the utilization of molecular ions has led to a new approach for measuring parity violation (PV) frequency differences in chiral molecules[1]. In addition to several experimental advantages, molecular ions are predicted to exhibit amplified PV frequency differences compared to their neutral counterparts. Notably, the simple carbon-centered tetrahedral cation $CHDBrI^+$ is predicted to exhibit a PV frequency difference comparable in magnitude[2] to that predicted for large period-6 metal complexes[3,4]. Further investigations of related halomethane tetrahedral cations demonstrate similar enhancement, as shown in Figure 1. These findings provide greater experimental flexibility in selecting suitable molecular ion candidates for future PV measurements.

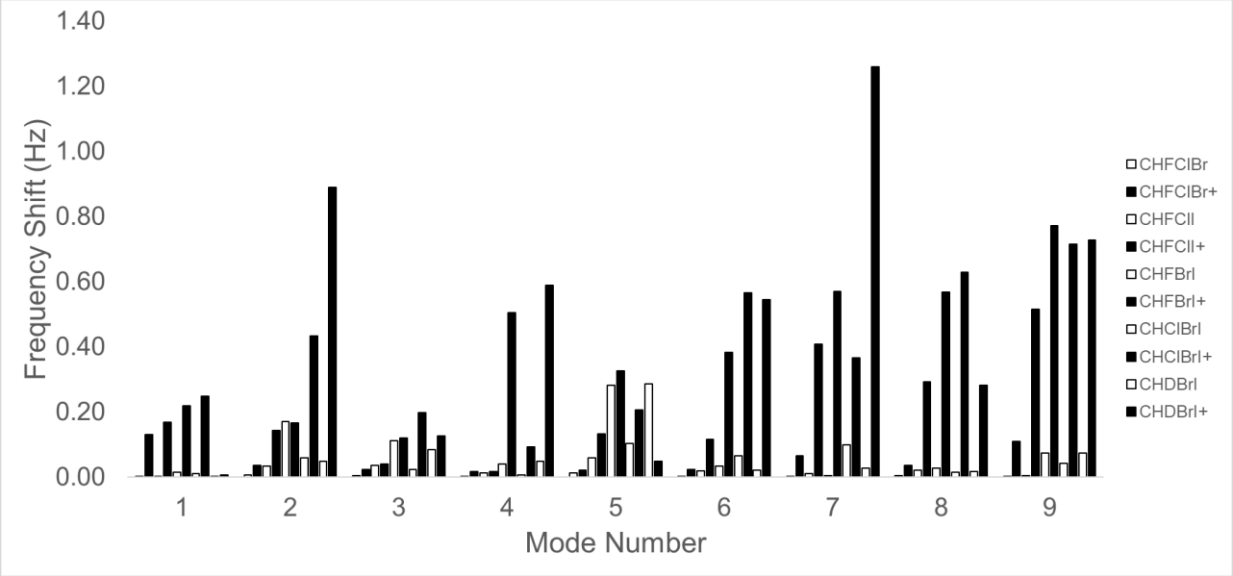

Figure 1. PV frequency differences in several halomethane tetrahedral molecules in the neutral (white) and cation (black) states.

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Precision Metrology with Molecules in an Optical Lattice

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Ultracold molecules have recently drawn widespread interest as exquisite platforms for precision measurements to probe fundamental physics. In particular, the internal structure of molecules with a heavy nucleus, large electric dipole moment, and several bosonic and fermionic isotopologues leads to vastly improved sensitivities to symmetry-violating physics. The current generation of precision measurement experiments is using fast cryogenic molecular beams, and thus, is significantly limited in the achievable duration of coherent measurement and degree of quantum state control. The natural next step is to borrow the techniques of atomic laser cooling and trapping, and extend them to molecules. While the rich internal structure complicates this realization in heavy dipolar molecules, particularly in fermionic species with complex hyperfine structure, there has been significant progress towards this goal over the last two decades. Building upon these advances, I will describe our progress towards preparing a precision metrology platform by laser cooling and trapping different isotopologues of barium monofluoride (BaF) molecules in an optical lattice to study symmetry-violating physics. While trapping and ultracold temperatures will allow for record-long coherence times and better control over the internal quantum states, the ability to switch between different bosonic and fermionic isotopologues will enable access to varying scenarios of symmetry violations. The techniques developed along the way are advancing us in the direction of achieving denser ensembles of ultracold heavy molecules where interactions begin to play a crucial role, and thus, facilitate sensing schemes that use entanglement to surpass the standard quantum limit, paving the way to true quantum metrology with molecules.

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Sum rules in chiral systems

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Sum rules offer an analytical tool for analysing physical properties of atoms and molecules. The Reiche-Thomas-Kuhn sum rule for oscillator strengths, for instance, provides a check for canonical commutation relation between position and momentum [1-3]. Related rules have also been investigated for optical activity in chiral molecules [4]. In this contribution, we explore the use of sum rules for other operators probing chirality, e.g., $\hat{H}_{PV}$ and γ_5, to arrive at simplified interpretations of the above chirality descriptors.

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An entanglement protocol to measure atomic parity violation at sub 0.1% precision

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Measurements of atomic parity violation (APV) provide low-energy tests of the electroweak electron–hadron interaction and enable precise determinations of nuclear weak charges. Despite their importance for the physics of fundamental symmetry breaking, APV measurements have stagnated at the 0.35% level due to challenging systematic effects [1]. We are developing a new APV experiment targeting sub-0.1% precision using two entangled barium ions prepared in a decoherence-free subspace [2]. The entangled ions serve as a differential quantum sensor insensitive to common-mode measurement noise, thereby suppressing systematic error sources. We will measure vector light shifts that carry a parity-violating signal by confining ions in a surface Paul trap and driving optical transitions between atomic states of opposite parity. Beyond improving constraints on the electroweak interaction and several beyond-Standard-Model scenarios, our experiment introduces novel control and noise-rejection strategies that are broadly applicable to other parity-violation measurements in quantum systems.

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Experimental prospects for the spectroscopic measurement of parity violation in chiral molecules

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We present our current plans to measure parity violation (PV) in neutral chiral molecules by comparing vibrational frequencies of left and right enantiomers, following Lethokov [1]. The quest for the ideal target molecule to successfully probe this effect is still ongoing. The experiment performed in our group on CHFClBr in 1997-2000 [2,3], to this date the closest to a successful measurement of PV, is used as a starting point. From there we explore two main routes for the choice of the molecule of interest: either light and simple (< 6 atoms); or complex (> 10 atoms) and centred around one heavy element. The former kind of molecules typically exhibit lower PV shifts, but their higher vapour pressure and simpler spectrum allows for easier experiments, and thus better statistics. The complex molecules can exhibit rather large PV effects (> 1 Hz) [5,8], at the cost of spectral and instrumental complexity.

In this work we summarize some of the criteria that the ideal molecule should fulfil (amplitude of PV, accessible transitions, stability, …), recall the most promising species from the literature [4-8], and lay out possible paths to a successful measurement of PV in chiral molecules. These paths require close collaborations between and with theoretical and synthetic chemists, extension of frequency metrology to the whole of the mid-infrared range (6-20 µm), the development of dedicated cold molecules sources and of new, sensitive, detection methods. A critical assessment of potential systematic effects is also necessary and detailed in this work.

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Prospects of laser-coolable polyatomic molecular ions

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Among molecular systems amenable for direct cooling with lasers, laser-coolable polyatomic molecular cations present particular interest. Even the design of such species is challenging. In small molecules (up to six atoms) just increasing the charge of optical circulation center (OCC) carrying atom, whether in the core of ligand, normally leads to compounds with forbidden optical transitions between ground and first excited electronic states [1-3]. In Ref. [4] we proposed using OCC-functionalized zwitterions as laser-coolable molecular cations with allowed working optical cooling transitions, providing examples of such compounds. We are discussing properties of these compounds and their prospects for use in searches for effects of space parity- and time-reversal symmetry violations.

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Estimating the QED corrections to energy levels and NMR shielding constant of atoms and diatomic molecules

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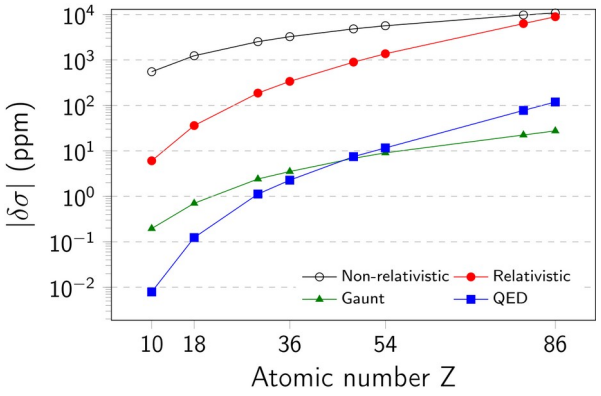
Estimating the quantum electrodynamics (QED) corrections, such as vacuum polarization and self-energy, is important in highly accurate calculations of parity violation in molecules [1]. Last years, the effective model to estimate the QED effect on NMR shielding constant for multi-electron atoms and diatomic molecules has been developed [2-4]. In this contribution we present our recent results in this field. The re-parametrization of Flambaum–Ginges radiative potential for self-energy correction to energy levels is also presented [5].



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Detection of the Parity Violation Energy Difference in Molecules Based on Alternating-Pulse Quantum Sensing

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Our research proposes a novel quantum control-based method to detect the minuscule parity violation (PV) energy difference between molecular enantiomers. The PV effect, originating from the weak interaction via the exchange of Z^0 bosons, induces an energy difference ($E_{PV} \approx -E'_{PV}$) between left- and right-handed chiral molecules, albeit of an extremely small magnitude (~10^{-20} eV). The core of the proposed method is the application of a sequence of laser pulses with alternating phases (e.g., π, –π, π, –π, ...) to a Λ-type three-level system. This technique is highly sensitive to static detuning deviations (δ), which correspond to the PV energy splitting, while being robust against errors in the Rabi frequency. Crucially, the scheme is designed for use with a naturally mixed chiral sample (1:1 ratio of enantiomers), thereby eliminating the need for prior chiral separation. After the pulse sequence, the PV-induced population difference between enantiomers in the ground state is amplified. This difference can then be measured using highly sensitive techniques to determine the enantiomeric excess. While acknowledging significant challenges—in particular, that the projected detection limit (~0.1 Hz with ion traps) remains far above the expected PV difference (~10^{-15} Hz)—this note provides a qualitative theoretical framework for a potential direct detection pathway for PV effects in mixed chiral samples, which would represent a significant advancement for testing fundamental symmetry violations.

High-resolution mid-infrared molecular spectroscopy for PV measurements

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Quantum cascade lasers stabilized on optical frequency combs (QCLs/OFC) with traceability to primary frequency standards represent a major advance for high-precision mid-infrared spectroscopy. This capability has recently been demonstrated at the Laboratoire de Physique des Lasers **[1]**. QCLs/OFC provide an unprecedented level of frequency accuracy and spectral resolution in the mid-infrared molecular fingerprint region, enabling ultra-precise measurements of molecular transitions. Such ultimate frequency control is particularly crucial for tests of molecular parity violation in chiral species. These studies require extremely accurate and stable laboratory reference frequencies, as the expected parity-violating effects are exceedingly small. Current experimental sensitivity is largely limited by the availability of suitable spectroscopic tools combining high resolution, wide tunability, and absolute frequency traceability, as well as by the difficulty of producing dense and cold samples of complex molecules of interest for PV measurements.

We present our latest experimental efforts to overcome these limitations by extending the spectral coverage and tunability of QCL/OFC systems, while simultaneously developing approaches to prepare cold, dense molecular samples using buffer-gas cooling **[2]**. This combination aims to enable frequency-metrology-based spectroscopy of a broader range of complex chiral, molecules **[3]**. Ultimately, this work seeks to provide broadband, sensitive, and ultra-precise mid-infrared spectroscopy tailored to molecular parity violation measurements.

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Locally Chiral Molecular Rotations Induced by Electronic Transitions

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Although chirality is typically considered a spatial property of an object and depends on its geometrical symmetries such as point inversion and reflection, local chirality arises from a vector tracing a chiral trajectory in time. For instance, locally chiral Lissajous figures of light are known to generate strong enantiosensitive signals in HHG [1,2]. Inspired by this concept, we introduce locally chiral molecular rotations via rotational wavepackets constructed of few lower rotational states, in contrast to previously demonstrated rotational chirality by using an optical centrifuge. In this picture, the molecular rotational dynamics can be visualized as the molecular axis tracing a chiral trajectory, or more precisely, a 3D Lissajous curve in space, drawing an analogy to synthetic chiral light over one full rotation period.
Probing the prepared molecular state with an ultrafast pulse, we explore the interplay of two locally chiral structures: the induced polarization vector of chiral electron currents [3] and the rotational trajectory of a molecule. We formally investigate how controlled molecular rotation affects the electronic excitations in different ultrafast electronic process. Moreover, including the electron-rotational couplings based on Hund's cases (a) and (b), we provide deeper insight into how electronic currents effect this newly introduced form of rotational chirality.

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A Periodic Many-body Quantum Matter Produced in an Ultra-cold Quantum Degenerate Plasma: Direct Observation of Fractional Elementary Charges

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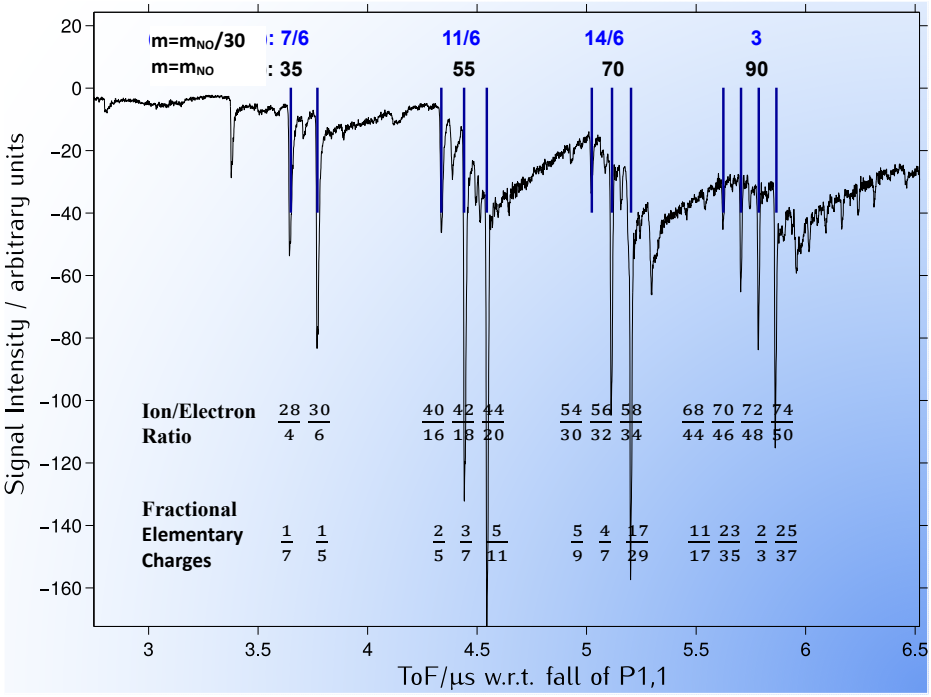
We report the experimental observation of a very striking periodicity in a many-body system: an ultra-cold plasma. A long life-time (>0.3 ms) ***quantum degenerate*** molecular Rydberg plasma is generated in the high-density region of a pulsed supersonic jet expansion by two-colour resonant excitation of nitric oxide (10%) in neon (5bar) into the high-*n* Rydberg threshold region close to the ionization limit. Experimentally, two synchronous UV laser pulses produce the plasma a few mm away from the jet nozzle. For plasma densities of > 10^{16} cm^{-3} reached in our experiments the electrons should become quantum degenerate, *i.e.* the **electron *de Broglie wavelength*** becomes larger than the Wigner-Seitz radius ***a*** relevant to describe the mean distance between the particles.

A time-of-flight (ToF) mass spectrometer collinearly with the jet expansion is used to analyse the plasma by applying two successive high-voltage pulses of 3.6 kV with a 0.2μs gap. According to the well-known law, the ToF's of the observed positively charged objects are proportional to the square roots of the ***mass to charge ratios m/z***, resulting in a strictly reproducible progression of (m/z) from 35 to 92.5 (black) or *w.r.t.* $m(NO^+)$ = 30u from 7/6 to 3 (blue)). From the m/z ratio numbers one obtains the corresponding ***ion to electron ratios*** of the 12 observed peaks, from 7/1 to 37/25 (ratios are given in even ion/electron numbers!).



In conclusion, we observe a many-body quantum matter consisting of a series of topologic bodies that contain magic numbers of ions and electrons for which the ion/electron ratio follows a periodicity. These objects are manipulated by fields in the ToF spectrometer without being destroyed, which shows that they behave as objects with a centre of mass. The 12 many-body states of ion/electron ratios of **28/4, 30/6; 40/16, 42/18, 44/20; 54/30, 56/32, 58/34; 68/44, 70/46, 72/48; 74/50**
are equivalent to the directly observed ***Fractional Elementary Charges*, *FEC*'s** (*i.e.* electron/ion ratios) of **1/7, 1/5; 2/5, 3/7, 5/11; 5/9, 4/7, 17/29; 11/17, 23/35, 2/3; 25/37**.

This observed progression follows fractions derived from the so-called ***Jain Series*** describing the filling factors for the ***Fractional Quantum Hall Effect*** observed in planar solid-state semiconductors close to 0K and in a high magnetic field, typically 10T. An interpretation for the observations presented here is the initial formation of a ***Wigner-Abrikosov Ion Crystal*** in the quantum degenerate plasma, i.e. a ***Solid State*!** The application of the very fast (10ns) rising and falling high electric field pulses then produces the ***planar topological bodies*** which are showing very surprising, but clearly evidenced features of the Jain Series of the Fractional Quantum Hall Effect that one would not necessarily expect to observe in a "***gas-phase supersonic jet experiment***" at twelve orders of magnitude lower density than solid state.

Selected molecular candidates for parity violation measurements investigated by electronic structure calculations

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The Z-boson mediated parity violation (PV) interactions between the electrons and the nucleons are predicted to cause a small difference between the energy levels of left- and right-handed enantiomers of chiral molecules [1]. Despite the firm theoretical foundation, large potential significance, and considerable efforts, this has not yet been experimentally observed [2,3]. We present several recent advances in identifying and predicting properties of promising molecular candidates in direct collaboration with experimental groups building the next generation experiments for detection of molecular PV in vibrational spectra [4–6]. Selected systems include axially/helically chiral organometallic compounds (e.g. Fig. 1) and isotopically chiral methane derivatives. We used relativistic density functional theory and coupled cluster calculations to identify optimal vibrational modes with expected PV effects exceeding by up to 3 orders of magnitude the projected instrumental sensitivity of the experiments under construction. Behavior of the effect was analyzed in terms of Z-scaling, electronic structure, chiral density and vibrational motion

Figure 1: Enantiomers of $Os(acac)_3$ breaking the mirror symmetry.

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Ab Initio Calculations of the Polarizability of BaOH: Towards a Competitive eEDM Measurement

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In recent years, di- and triatomic molecules have become increasingly popular for high-precision experiments testing the validity of the Standard Model, as molecular electronic structure may provide strong enhancement of charge (C), parity (P), or time-reversal (T) symmetry-violating effects [1]. One of such CP-odd effects arises from the interaction between the electron electric dipole moment (eEDM) of the unpaired electron and the internal electromagnetic fields in a molecule. The eEDM is being measured with rapidly increasing precision, with the current upper bound at $\mathcal{O}(10^{-29})$ e cm [2]. The NL-eEDM collaboration is currently designing an experiment to lower this limit by making use of barium monohydroxide molecules (BaOH) [3]. BaOH is laser-coolable and MOT-trappable and allows the suppression of certain systematic effects. For optimal laser-cooling and trapping, the electronic structure and properties of BaOH must be known to high precision. We present high precision *ab initio* calculations of the static and dynamic polarizabilities of BaOH, employing the state-of-the-art coupled cluster computational method (CCSD(T)). Uncertainties on the calculated values are estimated by investigating dependence on basis set, complete basis set limit, CCSD(T) active space, and treatment of relativistic and QED effects [4].

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The Veritable $Rh(acac)^3$ and its Chiral Shifts: Rhodium-103 NMR as a Probe of Molecular Parity Violation?

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Rhodium-103 carries the distinction of being one of only 4 diamagnetic spin-1/2 nuclei with a natural abundance of 100% (alongside ^{19}F, ^{31}P, and ^{89}Y). Despite this attractive quality, the extremely low gyromagnetic ratio of ^{103}Rh, and the ensuing experimental challenges, have historically placed ^{103}Rh within the list of Brian Mann's "Cinderella nuclei" - obscure and neglected spin-1/2s that are scarcely measured.

This bleak picture has changed. Today, rhodium NMR is easily measurable on commercial probes without special modifications. The supreme importance of rhodium in the field of catalysis, and the extraordinary responsiveness of ^{103}Rh nuclear spin parameters to the subtlest changes in chemical environment, has led to an explosion of ^{103}Rh NMR studies in the last 5 years alone. But beyond this exciting chemistry, rhodium-103 has a hidden propensity to support exotic spin physics.

We provide an overview of our group's adventures with rhodium NMR, which include (i) accessing $^{103}Rh_2$ nuclear singlet states in rhodium paddlewheel complexes, enabled by isotope-induced symmetry breaking via selective ^{18}O labelling, (ii) using broken symmetry to observe a striking solvent-dependence of homonuclear ^{103}Rh-^{103}Rh J-couplings in homoleptic complexes (iii) measuring hitherto undescribed chirality-induced shifts of the helically chiral $Rh(acac)_3$ complex, motivated by a search for the elusive phenomenon of molecular parity violation.

Investigating small molecular ions using high resolution action spectroscopy methods

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High resolution spectroscopy is an important tool to reveal and understand intramolecular energetic structures and dynamics.
Using a 22-pole cryogenic ion trap, rotationally resolved vibrational and electronic spectra of small molecular ions are measured. As the low concentration of trapped ions does not enable direct spectroscopy, in-house customised/developed action spectroscopy methods are applied, predominantly: Zero-background “kick-out” Laser Induced Reaction (LIR) [1] and Leak-Out Spectroscopy (LOS) [2]. While LOS is a very universal tool, where parameters like temperature and collision partner can be adjusted in a way the method is theoretically applicable to any molecule, a cleverly chosen LIR scheme enables almost unprecedented sensitivity.
The most recent projects dealt with the symmetric top molecule H_3O^+ [3] and the linear isomers HCN^+/HNC^+ [4], all of them known to play fundamental roles in chemical processes in the interstellar medium (ISM). Rotationally resolved mid-infrared spectra of all three molecules were measured, unveiling overtone bands for H_3O^+ and HNC^+, while for HCN^+ the resulting spectrum contained both a combination band and a rovibronic transition to the low-lying first electronically excited state $A^2\Sigma^+$**.**
Furthermore, the first electronically excited state $A^2\Pi$ of the isomer HNC^+ was investigated using an optical dye laser, rotationally resolving several quanta of the CN stretching mode and additional combination bands in the visible photon range.
In parallel to this work, both isomers have recently been investigated also in further spectral ranges, characterising the rotational spectrum [5] and the first electronic transition of HCN^+ [6] and several rovibrational bands of both isomers [7].
This contribution will give an overview of the experimental setup, insight into the applied action spectroscopy methods, and the analysis of the obtained spectra.

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An ion trap with velocity map imaging for PV search in chiral molecular ions

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Experimentally comparing left- and right-handed enantiomers is a powerful way to isolate parity-violating (PV) forces due to the unique asymmetry of chiral molecules (Fig. 1). We are building trapped ion version of the search focusing on CHDBrI+ [1,2] to detect PV arising from the nuclear weak force for the first time. Precision spectroscopy of such complex polyatomic molecular ions requires both cold generation and state-selective detection. With respect to the former, we aim to prepare cold CHDBrI+ through state-selective ionization in the VUV [1].

Our approach to probe the PV signature uses a mixed handedness ensemble of trapped chiral molecular ions to suppress technical noise in the measurement. To this end, we have proposed a variant of vibrational Ramsey spectroscopy which is combined with enantiomer separation through three-wave mixing [3].

These experiments require state-selective detection techniques to measure the internal state populations of chiral molecular ions. Our plan is to use direct velocity map imaging (VMI) of the photofragments in single photon dissociation [1,3] or a state-selective variant. We will present our implementation of an RF ion trap that is integrated with a VMI detector and present preliminary results showing a resolution below 10 m/s. This should be sufficient to resolve individual rotational states in the molecule. We also present new results involving the heating dynamics of the Yb+ ions where the heating rate for each axis can be measured independently.

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P and T-odd interaction constants of YbX (X : Cu, Ag and Au) molecules using KRCI method

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In pursuit of physics beyond the Standard Model of elementary particles, extensive efforts have focused on observing violations of parity (P) and time-reversal (T) symmetries [1]. An electron electric dipole moment (eEDM) is a direct consequence of such symmetry violating effects and is therefore of considerable interest for constraining new theories proposed to explain the baryon asymmetry in the Universe. An additional contribution to P and T-violating effects arises from the scalar-pseudoscalar (S-PS) nucleon-electron neutral current interactions. There are currently several ongoing experiments in the search for the eEDM using heavy diatomic polar molecules. Accurate determination of P and T-odd interaction constants, such as W_d, which is related to the eEDM, and W_s, which characterizes the nucleon-electron S-PS interaction, is required to interpret experimental results. These constants can only be obtained through calculations employing relativistic many-body theories.

Among the plethora of proposed molecules, YbX (where X = Cu, Ag and Au) molecules have recently been investigated for eEDM searches [2, 3]. In this work, we have performed relativistic calculations of P and T-odd interaction constants for the aforementioned molecules. These computations have been carried out using the Kramers-restricted configuration interaction method limited to single and double excitations. To this end, the generalized active space (GAS) technique has been employed. Within the GAS model, the active orbital space is divided into three subspaces: paired, unpaired, and virtual orbitals. The uncontracted Dyall core-valence basis sets of double-, triple-, and quadruple-zeta quality, in conjunction with Gaussian charge distribution for the nuclei, have been utilized. In addition, we have computed the parallel and perpendicular components of the magnetic dipole hyperfine structure constants for different isotopes of atoms constituting the molecules considered in this work.

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Ultracold high-spin Σ-state polar molecules for new physics searches – YbCr vs RaCr

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In our previous work [1], we demonstrated that high-spin Σ-state polar molecules assembled from ultracold atoms constitute a promising platform for probing charge-parity (CP) violating physics beyond the Standard Model. In particular, we identified YbCr as a prime experimental candidate and predicted a statistical sensitivity to the electron electric dipole moment (eEDM) at the level of $\left(6 \times 10^{-31}/\sqrt{n_{day}}\right) e \cdot cm$, placing it among the most sensitive eEDM measurement schemes to date.

In the present study, we extend this framework to the heavier molecule RaCr, which also belongs to the class of high-spin Σ-state polar molecules. The presence of the octupole nuclear deformation in Ra leads to an increased sensitivity of RaCr to nuclear CP-violating effects. This enhancement is particularly relevant for hadronic CP-violating observables, while retaining strong sensitivity to leptonic sources of CP violation. Our results indicate that RaCr represents an exceptionally promising candidate for future searches for new physics using ultracold molecules.

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Electronic Spectroscopy of $YbNH_2$

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$YbNH_2$, as a heavy metal-containing polyatomic molecule, is of interest for precision tests of fundamental symmetry violations by virtue of possessing closely spaced states of opposite parity in the |K| = 1 manifold of states [1]. In addition, an unpaired valence electron in ground state, and naturally occurring isotopes of ytterbium with nonzero nuclear spin render the molecule relevant to nuclear-spin dependent parity-violation (NSD-PV) studies [2]. We report on initial gas-phase production methods and the spectroscopic characterization of $YbNH_2$, combining medium resolution laser-induced fluorescence (LIF) measurements with electronic structure calculations. LIF spectroscopy of the Ã–X̃ and B̃–X̃ band systems reveals highly diagonal vibrational branching, critical for laser-cooling efforts and high-precision measurements [3]. Ongoing results from high-resolution LIF spectroscopic studies will be presented - the foci of which will be the characterization of closely spaced parity doublets, hyperfine structure, and applied electric field studies of $YbNH_2$ - to assess its suitability for experiments probing parity-violating effects.

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Fock-Space Coupled cluster on parity-violating frequency difference of $WHNClF^+$ vibrations

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Parity-violating (PV) effects were first measured in nuclei, and later in atoms. Parity-violating effects are also expected to be present in larger systems, such as molecules, but have not yet been measured. This has made molecular systems an excellent candidate to study the weak force at larger scales.

The goal of the presented research was to apply Fock-Space Couple Cluster with different numbers of orbitals included in the auxiliary space on $WHNClF^+$, which has been shown to have a large PV frequency shift by Figgen et al. [1]. This could determine whether multireference effects need to be taken into account during calculations on this system.

The parity-violating frequency shift showed a strong dependence on the number of orbitals included in the auxiliary space, as the PV frequency shift could vary by an order of magnitude.

Therefore, multireference effects cannot be neglected in this system to obtain accurate results, and applying FSCC to other open shell systems may hence lead to more accurate results as well.

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Photoelectron Velocity Map Imaging Spectroscopy of MF^- (M = Ba, Yb) (sample)

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The $^2\Sigma_{1/2}$ and $^2\Pi_{1/2}$ states present in certain diatomic molecules have been suggested as having characteristics useful for investigations of nuclear spin dependent parity violation (NSDPV) quantities. By Slow Electron Velocity-map Imaging (SEVI), we have measured the electron affinities (EAs) of BaF (6787 ± 36 cm^{-1}) and YbF (9088 ± 5 cm^{-1}). By thermodynamic considerations, we have been able to place lower bounds on the bond dissociation energies (BDEs) of these species (D_0^- (BaF^-) > 28117 cm^{-1}; D_0^- (YbF^-) > 23796 cm^{-1}). As BDE > EA for these species, these data function as experimental verification of the viability of the experimental scheme devised by Gaul et. al to use RaF^- to detect NSDPV effects[1], which proposes that these anions - easy to manipulate by the application of electric fields - may be used as precursors to cold molecular beams of neutral MF to facilitate laser cooling efforts and high-precision measurements. BaF^- and YbF^- are significantly safer to handle than radioactive RaF^- but still offer significant enhancement of parity violation effects by virtue of being diatomic radicals with heavy nuclei. Additionally, we will make comparisons with data obtained for the lighter AeF^- (Ae = Mg, Ca, Sr) and comment on methods of gas-phase production of these anions, for experimental considerations. Electronic structure calculations on YbF^-/YbF, which compare and contrast the use of relativistic Hamiltonians versus relativistic pseudopotentials in describing states of multireference character, will also be presented.

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A Cold Beam Source for Precision Experiments with Chiral Molecules

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Performing precision measurements on chiral molecules requires a cold and slow beam source to minimize Doppler broadening and keep the occupation of rotational and vibrational states within reasonable levels [1][2][3][4]. We report on the simulation and construction of a cryogenic buffer gas source which can provide beams of chiral molecules at T<3K with ~100 m/s forward velocity. We have investigated and characterized the source's performance with Yb atoms and recently verified predications from published numerical simulations on the diffusion behaviour through the inert buffer gas [5]. In the next step, we plan to interface the cold beam to photoelectron circular dichroism (PECD) measurement for enantio-sensitive characterization of the beam [6]. We believe that this source will enable experiments of chiral effects on the single molecular level and is applicable to various chiral species.

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Novel ESST methods for PV measurements in Chiral Molecules

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Chiral molecules have two mirror image configurations for their atoms known as enantiomers. Many naturally occurring chiral molecules are found to be in a single handedness, and their enantiomers exhibit different smells, flavors and biological functions. However, the root cause for the specific handedness found in nature remains a mystery, but it is hypothesized that the asymmetry is caused by weak force Parity Violation (PV) on a molecular scale. Due to the miniscule magnitude of the expected effect, new experimental techniques must be developed for the search. Enantiomer-Specific State Transfer (ESST) Three-Wave Mixing schemes use the anti symmetry of electric dipole triple product of chiral molecules to control state populations in each enantiomer. This allows the signal from each enantiomer to be detected separately in a mixed sample. These ESST schemes were proposed to measure the PV effect in Chiral molecules by extending them from the MW regime to the mid-IR regime. We explore applications of frequency combs for expanding chiral molecule ESST into the near-IR, as well as new schemes for PV measurements in chiral molecular ions and searches for new physics beyond the Standard Model.

Magnetic effects and enantiomer-selectivity in isotopically chiral molecules

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We investigate the role of nuclear-spin hyperfine interactions in the rotational dynamics of isotopically chiral molecules, using $CHF^{35}Cl^{37}Cl$ as an example. In particular, we study how isotope-induced chirality manifests in the interaction with microwave radiation and a magnetic field in a three-wave mixing scheme. We show that symmetry breaking arising from the different masses and nuclear quadrupole moments of ^{35}Cl and ^{37}Cl induces a weak electric-dipole transition between hyperfine-split rotational states. Together with two stronger allowed electric-dipole transitions, this transition closes a three-level loop. Our simulations demonstrate enantiomer-selective excitation in the presence of an additional magnetic field. When the magnetic field is collinear with the microwave electric field, a falsely chiral field configuration is formed, enabling absolute enantiomer selectivity in rotational excitations.

Towards full controllability of enantiomer-specific state transfer despite orientational degeneracy

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We present recent experimental advances towards full enantiomer-specific control of quantum states of chiral molecules using enantiomer-specific state transfer (ESST). Early ESST experiments achieved only a few percent of state-specific enantiomeric enrichment, primarily limited by thermal population and spatial (M-state) degeneracy [1,2]. Our group has overcome the thermal population limitation and demonstrated near-complete control of the enantiomers in a triad of rotational states including the absolute ground state with J = 0 [3].

We further address the limitation imposed by spatial degeneracy in rotational state triads not including the absolute ground state, where M-state dependent Rabi frequencies limit efficient population transfer using conventional pulse schemes. We incorporate theoretically tailored microwave pulse sequences [4] to demonstrate enhanced enantiomer-specific control despite degeneracy. Our work provides experimental capabilities relevant for future precision studies of parity violation in chiral molecules [5].

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A theoretical feasibility study on amplifying molecular parity-violation signals using coherent pulse sequences

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There are assumptions stating that the weak force can induce a flavour in handedness via asymmetrical β decay [1]. This effect is in induces a small energy difference, in the order of 10^{-16}, between the different enantiomer states [2]. The extremely small magnitude of this shift makes direct spectroscopic detection challenging, motivating the exploration of amplification schemes based on coherent dynamics.

In this preliminary theoretical project, we investigate whether pulse sequences developed for quantum sensing, particularly alternating-phase π-pulse trains in Λ-type systems, can enhance the detectability of small detuning differences arising from parity-violating energy splitting. Using the analytical propagator structure derived from Cayley-Klein parameters, we compare population evolution in three-level models under static detuning deviations.

These schemes have been shown to accumulate small phase errors nonlinearly with the number of pulses, suggesting a possible route to amplifying PV-induced asymmetries in mixed chiral samples.

We outline a qualitative strategy in which a racemic molecular ensemble is driven by alternating-phase pulses, resulting in slightly different excitation probabilities for the two enantiomers due to ΔE_{PV}. We further discuss possible detection methods, such as isolating ground-state populations or employing nonlinear optical probes, such as high-harmonic generation.

This study aims to clarify whether coherent-control techniques could serve as a complementary pathway toward future PV measurements in molecular systems.

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Production of Radioactive Molecular Ions via Electron Impact Ionization

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The Charged Radioactive Molecule Spectroscopy (CHARMS) project aims to probe fundamental symmetries and search for new physics, particularly CP violation, by measuring nuclear Schiff moments in heavy nuclei. High-efficiency production and ionization of specific molecular ions are essential to achieve these measurements. Building upon recent successes in generating actinide molecules via laser ablation[1], we present our investigations on an injection and ionization scheme utilizing a Heidelberg Compact Electron Beam Ion Trap (HC-EBIT) at the Max-Planck-Institut für Kernphysik.

In this configuration, a laser ablation target can be positioned close to the electron beam to enable the immediate capture and ionization of molecules released from the ablation process. The system can be used with precious, chemically tailored micro-targets of down to 10^{12} atoms[2]. A distinct advantage of the HC-EBIT configuration is its tunability, which allows for the production of selective charge states. For the production of heavy molecular ions, the EBIT needs to be operated at energies far below common operating conditions, which introduces specific constraints. Finally, we discuss design considerations for a optimized source that will utilize both laser ablation and electron impact ionization to deliver tailored radioactive molecules in selected charge states, serving as a high-efficiency source for a precision spectroscopy trap.

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Testing Fundamental Physics with Trapped Ions

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We report a measurement of the isotope shifts in a chain of Yb^+ isotopes with a precision of 4×10^{-9}. This measurement, along with a precise determination of the mass ratio using highly charged Yb^{42+} ions, allows us to extract higher-order changes in the nuclear charge distribution along the Yb isotope chain and to set bounds on a hypothetical fifth force [1].

We also report the most stringent bounds to date on possible violations of local Lorentz invariance by constraining the symmetry-breaking coefficients of the Standard Model Extension Lagrangian to the 10^{-21} level. This was achieved by performing composite pulse Ramsey spectroscopy in the $F_{7/2}$ manifold of a single trapped Yb^+ ion [2].

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