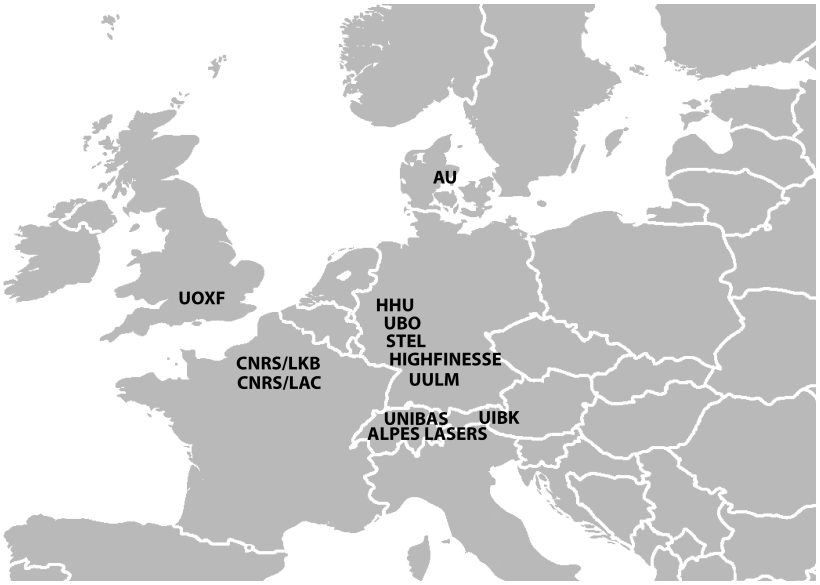
Cold Molecular Ions at the Quantum Limit

A Marie Curie Initial Training Network



Introduction to the network

COMIQ (COld Molecular Ions at the Quantum limit) will investigate how cooling, trapping, and control techniques applied to molecular ions can expand the realm of quantum technology, enhance precision meaurements on molecular systems and lead to chemistry at the ultracold quantum limit.

The network is supported by the European Commission (FP7) under the sub-programme PEOPLE (Marie Curie Actions). 13 early stage researchers and one experienced researcher will obtain training in all relevant topics of the field; molecular physics, quantum optics, ion trapping and cooling, chemical reactions, spectroscopic techniques, theoretical and computational chemistry, and physical chemistry. The training will be provided by 9 universities/laboratories and 3 private companies from 6 different countries in Europe.

The aim of COMIQ is to investigate and control a variety of molecular ion processes at the very quantum limit. COMIQ will work on establishing cold molecular ions as new quantum objects for applications in quantum technology, precision measurements, and controlled chemistry. The network is highly interdisciplinary, combining quantum optics, quantum information sciences, molecular physics, and chemical physics in a novel and original fashion.

The network consists of ten host institutions, both academic and industrial, and all early stage researchers will be seconded at least once during the fellowship at another partner site. The 13 research projects which are in short described in this publication with indication of planned place of secondment are available from November 1st, 2013.

This material is also available at out website: www.itn-comiq.eu

ESR1: State-prepared cold molecular ions: Lifetime measurements and reaction studies

Supervisor: Michael Drewsen
Host Institution: Aarhus University
Duration: 36 months
Planned secondment: University of Oxford

Description:
The focus of this project is to investigate and advance various techniques for state-preparation of molecular ions. In a room temperature trap, laser induced rotational cooling of sympathetically cooled molecular ions [Nature Phys. **6**, 271 (2010)] will in conjunction with novel probabilistic state preparation techniques be applied for rovibrational ground state preparation of polar molecular ions such as MgH^+ and CaH^+. Based on this initial state preparation specific excited rotational states will be prepared trough Raman transitions induced by a fs laser [Phys. Rev. Lett. **104**, 140501 (2010)]. Furthermore, by applying a cryogenically cooled trap, the rovibrational degrees of freedom of translationally sympathetic cooled molecular ions will be pursued through helium buffer gas cooling at temperatures of 5-10 K. By separating the mechanism for translational cooling (sympathetic cooling) from the rovibrational cooling scheme, rovibrational buffer gas cooling at helium pressures several orders of magnitude lower than usually applied should be feasible, and enable subsequent manipulation of the rovibrational degrees of freedom by light fields. For instance, via rotational and vibrational excitations, lifetimes of specific rovibrational states will be measured. Finally, the state prepared molecular ions will be applied in state specific reaction experiments with cold state selected reaction partners.

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ESR2: Quantum technology and precision spectroscopy with state-selected cold molecular ions

Supervisor: Stefan Willitsch
Host Institution: University of Basel
Duration: 36 months
Planned secondment: Aarhus University

Description:
The aim of the project is to develop a toolbox for the coherent manipulation and precision spectroscopy of the rovibrational states of apolar sympathetically cooled molecular ions such as N_2^+ and O_2^+. Building on the Basel group's recently developed methods for the quantum-state initialization of cold molecular ions [Phys. Rev. Lett. **105** (2010), 143001], apolar molecular ions will be prepared in well-defined ro-vibrational states. These molecular states exhibit extremely long radiative lifetimes and promise the implementation of magnetically insensitive qubits, thus forming an ideal basis for applications in quantum information science and molecular precision measurements. Rovibrational molecular ion qubits and the technology for their coherent manipulation will be implemented, benefitting from the development of the next-generation quantum-cascade-laser technology within COMIQ. New protocols for the readout of molecular quantum states will be developed and the properties of the qubits will be characterized. As a first application, precision spectroscopy of dipole-forbidden, extremely narrow rovibrational transitions and and highly accurate measurements of state lifetimes will be performed. This project is performed in collaboration with the University of Aarhus to which a 6-months secondment is planned.

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ESR3: Quantum logic spectroscopy with a state selected simple molecular ion

Supervisor: Laurent Hilico
Host Institution: CNRS Laboratoire Kastler Brossel
Duration: 36 months
Planned secondment: Heinrich Heine University of Düsseldorf

Description:
The hydrogen molecular ion H_2^+ is a quasi ideal quantum system allowing for direct optical determination of an important fundamental constant: the proton to electron mass ratio m_p/m_e. Indeed, recent theoretical progress in relativistic, QED and hyperfine corrections for the three body Coulomb problem show that two-photon vibrational spectroscopy at the 6×10^{-11} relative accuracy level is possible [1], and will lead to an significant improvement with respect to the 2012-CODATA recommended value. The group is setting-up an experiment aiming at observing the first Doppler free two-photon vibrational transition at 9.17 µm in state selected H_2^+ ions by REMPD and at measuring its frequency to determine m_p/m_e.

The tasks will consist in
- implementing a state selected H_2^+ REMPI ion source
- cooling the H_2^+ ions by sympathetic cooling with a laser cooled Be^+ ion crystal in a linear trap
- performing H_2^+ two-photon spectroscopy using our frequency controlled 9.17 µm laser source [2]

to prepare quantum logic spectroscopy by
- designing and implementing a "precision trap" for H_2^+/Be^+ ion pair sympathetic side band cooling
- implementing femto-second comb stabilization of the 9.17 µm laser source to reach unprecedented accuracies

The team comprises 3 permanent researchers, PhD and Master's degree students and closely interact with other Kastler Brossel laboratory teams. It is supported by electronics, mechanics and optics workshops.

References:
[1] V.I. Korobov, L. Hilico, J.-Ph., calculation of the relativistic Bethe logarithm in the two-center problem, Phys. Rev. A **87**, 062506 (2013).
[2] F. Bielsa, A. Douillet, T. Valenzuela, J.-Ph. Karr and L. Hilico, Narrow-line phase-locked quantum cascade laser in the 9.2 micron range, Opt. Lett. **32**, 1641 (2007).

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Laboratoire Kastler Brossel
Physique quantique et applications

ESR4: Reactive collisions between internally cold neutrals and laser cooled ions

Supervisor: Tim Softley
Host Institution: University of Oxford
Duration: 36 months
Planned secondment: University of Innsbruck, 6 months, Modelling reactive collisions between internally cold molecules and laser cooled ions. University of Basel, 3 months, Molecular ion state selection and characterization.

Description:
Building on previous experiments [Willitsch2008a, Gingell2010, Bell2009], the aim of this project is to work with two sources of cold neutral molecules in combination with laser-cooled or sympathetically cooled ions to study reactive collisions down to milliKelvin temperatures. The first such source is the Stark decelerator which generates a velocity tuneable, internally state-selected pulsed beam of polar molecules such as ND_3 or OH. Target systems to study include reactions of ND_3 with N_2^+, $C_2H_2^+$ and DCl^+. The second source is an electrostatic quadrupole guide combined with a cryo-cooled He (or Ne) buffer gas source at 6K or 17K. The continuous source works with a wider range of cold polar molecules than the Stark decelerator and the molecules have a thermal rotational distribution at 6K or 17K. Target reactions using this source include collisions of methyl halides with Ca^+; and reactions of HDO or isotopologues of ammonia (e.g., NH_2D or ND_2H) with sympathetically cooled ions such as NH_3^+, NH_4^+, CH_3^+. The Fellow will collaborate with Basel in the use of molecular ion state selection by photoionization/REMPI/pulsed field ionization and with Aarhus in the use of optical cooling of reagents to study reactions of state-selected or rotationally cold ions with state-selected neutrals. The project also will involve learning and applying quantum scattering approaches to ion molecule collisions in collaboration with University of Rome.

References:
[Willitsch2008a] S. Willitsch, M. T. Bell, A. D. Gingell, S. R. Procter, and T. P. Softley, Phys. Rev. Let. **100**, 043203 (2008)
[Gingell2010] A. D. Gingell, M. T. Bell, J. M. Oldham, T. P. Softley, and J. N. Harvey J. Chem. Phys. **133**, 194302 (2010)
[Bell2009] M. T. Bell, A. D. Gingell, J. M. Oldham, T. P. Softley and S. Willitsch, Faraday Discuss. **142**, 73 (2009)

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ESR5: High-efficiency preparation of a single quantum state of a molecular ion and a high-precision spectroscopic determination of a fundamental mass ratio

Supervisor: Stephan Schiller
Host Institution: Heinrich Heine University of Düsseldorf
Duration: 36 months
Planned secondment: Aarhus University

Description:
In our group, we are focusing on a particular molecular ion, HD^+. This is a useful model system for testing techniques that can be of more general utility in the field of heteronuclear molecular ions, but it is of significant interest also in its own right, beign a fundamental quantum system. HD^+ is a three-body bound quantum system that can be accurately described ab initio by Quantum Electrodynamics, using as input certain fundamental constants, in particular the Rydberg energy and the two mass ratios of the three constituent particles. A comparison between experimental HD^+ transition frequencies and the ab initio results therefore provides a test of the validity of theoretical treatments, and/or a determination of these fundamental constants. At present, the experimental inaccuracies of the transition frequency measurements is still higher than the theoretical or fundamental constants inaccuracies, resulting in an on-going experimental challenge.

We have performed many seminal experiments on cold molecular ion spectroscopy over the last decade, including laser-based rotational cooling, resonance-enhanced multi-photon dissociation, pure rotational excitation, fundamental vibrational spectroscopy. We also demonstrated addressing of individual hyperfine states of ro-vibrational levels by excitation of individual hyperfine transitions, and controlled transfer of population into a selected hyperfine state. On the theory side, we have worked extensively on the systematic frequency shifts of HD^+, a very important topic if precision measurements are pursued. In the open position, developments will be pursued that are aiming at increasing significantly the efficiency and accuracy of precision spectroscopy of HD^+.
In detail, the task will be:

- Develop an ion trap for storing a single atomic and a single molecular ion, sympathetically cooled.
- Integrate a laser-based scheme for population effectively a single hyperfine state
- Integrate a scheme for reading out efficiently the population of a single hyperfine state
- Apply preparation and read-out techniques to various forms of high-resolution spectroscopy (radio-frequency, pure rotational, and ro-vibrational spectroscopy)
- Compare the measured transition frequencies with the ab-initio theory values in order to validate the ab-initio theory calculations, and combine the results of the various spectroscopic measurements to determine an improved value of the mass ratio electron mass - reduced nuclear mass.

The work will be done in a team comprising several PhD and Master's students, a senior researcher and supported by a team of electronics engineers.

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ESR6: Quantum dynamics of ion-molecule reactive and inelastic processes from μK to K

Supervisor: Franco Gianturco
Host Institution: University of Innsbruck
Duration: 36 months
Planned secondment: CNRS Laboratoire Aimé-Cotton, Orsay

Description:
The work for the suitable candidate will focus on the developments of both numerical methods for the analysis of experimental data and also on the quantum study (with existing computational codes in the receiving research group and with the aim of partly developing new quantum models that start from such codes) of a variety of dynamical / reactive processes involving molecular ions (anions and cations) of current intense scrutiny in astrophysical groups due to the developments of new observational sources in large radiotelescopes and satellite missions.

In particular, the accepted candidate will work in close contact with experimental groups that are carrying out laboratory experiments on such astrophysical ions in order to familiarize himself/herself on the actual interfaces that are required to analyse and interpret the collected data. Furthermore ,this candidate will also interact very closely with theoreticians in the same group and will become proficient with the use and understanding of the relevant quantum codes that will endeavor to understand the nanoscopic mechanisms acting during the experimental observations, linking them with the astrophysical observations.

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ESR7: Formation and reactions of cold $(BaRb)^+$ and Rb_2^+ molecules

Supervisor: Johannes Hecker Denschlag
Host Institution: Ulm University
Duration: 36 months
Planned secondment: University of Bonn, Aarhus University, University of Basel

Description:
In our BaRbIe experiment we immerse a single cold trapped Barium (Ba) [or Rubidium (Rb)] ion in an ultracold gas of neutral Rb atoms. In this very unique and novel set-up we can study for the first time interactions of ions and atoms at ultralow temperatures. Compared to the short ranged van der Waals interactions between neutral particles, atom ion interactions are long range due to the $1/r^4$ polarization potential. We investigate how ions and atoms collide and react in the cold regime.

An important goal is to transfer quantum control techniques that have in recent years been developed for ultracold neutral quantum gases to atoms and ions. For example, we want to demonstrate Feshbach resonances in collisions between ions and atoms, which can be used to control the interactions of the particles and to form cold ionic molecules. We then want to study the properties of these molecules and measure how quickly they relax to more deeply bound states when they collide with cold atoms. The applicant will join these efforts to move forward the frontier of ultracold chemistry between atoms and ions.

Literature:

- Population distribution of product states following three-body recombination in an ultracold atomic gas. A. Härter, A. Krükow, M. Deiß, B. Drews, E. Tiemann, and J. Hecker Denschlag, Nature Physics **9**, 512 (2013)
- A single ion as a three-body reaction center in an ultracold atomic gas , A. Härter, A. Krükow, A. Brunner, W. Schnitzler, S. Schmid, and J. Hecker Denschlag, Phys. Rev. Lett. **109**, 123201 (2012)
- Dynamics of a cold trapped ion in a Bose-Einstein condensate, Stefan Schmid, Arne Härter, Johannes Hecker Denschlag, Phys. Rev. Lett. **105**, 133202 (2010)

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ESR8: Coherence and decoherence of a single-ion qubit immersed into an environment

Supervisor: Michael Köhl
Host Institution: University of Bonn
Duration: 36 months
Planned secondment: University of Basel

Description:
In quantum information processing it is important to manipulate a qubit in a coherently controlled way. Trapped atomic ions have been successfully employed in this field, since they are very well isolated from the environment and therefore offer long coherence times. However, when an isolated ion is coupled to an environment, it will become gradually entangled with many degrees of freedom and therefore eventually decoheres [1,2,3]. Therefore the question remains of how quantum computing could work under realistic conditions (i.e. when there is a finite coupling between qubit and the environment), and how it could potentially be optimised.

In this project, we plan to investigate decoherence mechanisms of a single or a few trapped ions controllably coupled to an environment. Particular focus will be on the question whether the environment could also be used as a resource to generate or protect entanglement between ions. We plan to investigate this process by carefully tailoring the properties of the environment, for example its dimensionality or the coupling strength between ion and environment. To this end, the project is concerned with the design, buildup and operation of the next generation of hybrid ion traps, where the tools for fast preparation and detection of the ion's spin coherence are already integrated into the trap. Specifically, we plan to employ recently developed optical fiber cavities [4] as an integral part of a new setup.

References:
[1] L. Ratschbacher et al., Phys. Rev. Lett. **110**, 160402 (2013).
[2] L. Ratschbacher et al., Nature Physics **8**, 649 (2012).
[3] C. Zipkes et al., Nature **464**, 338 (2010).
[4] M. Steiner et al., Phys. Rev. Lett. **110**, 043003 (2013).

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ESR9: Development of ultra high precision wavelength meters in the MID-IR range

Supervisor: Thomas Fischer
Host Institution: HighFinesse
Duration: 36 months
Planned secondment: Aarhus University

Description:
The PhD project is devoted to the development of precise wavelength meters in the mid-infrared range. The PhD candidate will develop interferometric wavelength measurement techniques and he will characterize optical components and detectors in the mid-IR range. The goal is the integration of these system components into a single device. The performance of the mid-IR wavelength meter will be characterized using narrow molecular ion transition lines in the experiments of COMIQ partners.

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HighFinesse
Laser and Electronic Systems

ESR10: Development of quantum cascade lasers and THz sources for state manipulation and spectroscopy of molecular ions

Supervisor: Richard Maulini
Host Institution: Alpes Lasers
Duration: 36 months
Planned secondment: University of Basel

Description:
Quantum cascade lasers and THz sources for state manipulation and spectroscopy of molecular ions.
The development of the next-generation quantum-cascade-laser technology within COMIQ will contribute to the implementation of the necessary technology for coherent manipulation of rovibrational molecular ion qubits. The design of QCLs with the integration of targeted multiple functions require the development of complex predictive models using smart optimization and analysis algorithms such genetic algorithm applied to simulation models.

However, the use of such new development and design tools require an increasing amount computational power not directly accessible with traditional computing hardware. The availability of cloud computing as a service is a new and incentive way to implement new modeling architectures at an affordable cost. Cloud computing does not only allow to compute simulations faster but it also enables taking advantage of the possibility offered by genetic algorithms to explore new research perspectives like the design of dedicated optical cavities and gain medium that were unaccessible before.

Within the framework of this development, the PhD student will initially continue to deploy a cloud architecture that enables the execution of sophisticated simulation workflows by redesigning existing models for execution on a computer cloud. Concurrently she/he will develop a variety of genetic and other bio-mimetic algorithms to exploit the cloud architecture and apply the newly developed tools to state-of-the-art research problems. The identified theoretical solutions will then be fabricated and experimentally assessed by the measurement of fundamental properties of the laser structures such as the optical gain.

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ESR11: Structure and dynamics of cold molecular ions: formation and destruction processes

Supervisor: Oliver Dulieu
Host Institution: CNRS Laboratoire Aimé-Cotton, Orsay
Duration: 36 months
Planned secondment: University of Ulm

Description:
Several experimental teams of the COMIQ consortium recently demonstrated the cooling of atomic ions (Yb^+, Ba^+) immersed in an ultracold Rb buffer gas [1, 2], and the formation of molecular ions in a merged trap of cold atoms (Rb) and cold ions (Ca^+, Ba^+). Such experiments open the way to a novel ultracold chemistry dominated by pure quantum effects (quantum tunneling, barrier reflection) which could be controlled at the single quantum state level.

Reactive dynamics with electronically excited atomic ions is a key element for the formation of molecular ions sympathetically cooled by trapped atomic ions. In this theoretical project, elastic and inelastic collisions as well as elementary chemical reactions will be modeled in the ultracold regime (T < 1mK), and in the cold regime (1mK < T < 1K), with both ground state and excited species. Resonances in the cross sections are very likely because the energy of rotational transitions of collision complexes is comparable with translational energies.

The work will include the determination of potential energy surfaces (PES) for ground and electronically excited reagents, and the modeling of the associated dynamics using various methods (time-independent close-coupling, Discrete variable representation…). Results will be constrained by state-of-the-art measurements in particular through a secondment at UULM where the $BaRb^+$ and Rb_2^+ ions are considered. Such calculations will make clear the structural and dynamical features in order to choose appropriate molecular systems and conditions. The insights gained from $RbBa^+$ and will help the theoretical analysis of other systems of interest, such as e.g. $RbCa^+$/Rb and MgH^+/Rb.

References:
[1] C. Zipkes, S. Palzer, C. Sias, and M. Köhl, Nature **464**, 388 (2010); C. Zipkes, S. Palzer, L. Ratschbacher, C. Sias, and M. Köhl, Phys. Rev. Lett. **105**, 133201 (2010).
[2] S. Schmid, A. Härter, J. Hecker Denschlag Phys. Rev. Lett. **105**, 133202 (2010)
[3] F.H.J. Hall, M., N. Bouloufa-Maafa, O, Dulieu, and S. Willitsch, Phys. Rev. Lett. **107**, 243202 (2011); F.H.J. Hall, M. Aymar, M. Raoult, O. Dulieu, and S. Willitsch, Mol. Phys. **111**, 1683 (2013); F. H.J. Hall, P. Eberle, G. Hegi, M. Raoult, M. Aymar, O. Dulieu, S. Willitsch Mol. Phys. DOI: 10.1080/ 00268976.2013.780107 (2013)

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ESR12: Cold collisions between ultracold atoms and state-selected cold molecular ions

Supervisor: Stefan Willitsch
Host Institution: University of Basel
Duration: 36 months
Planned secondment: CNRS Laboratoire Aimé-Cotton, Orsay

Description:
The recent progress in the cooling of ions, atoms and molecules has paved the way to study collisional and chemical processes in a new physical regime at extremely low energies. Such experiments pave the way to elucidate the quantum character of collisions, to accurately characterize molecular interaction potentials and to study the quantum-mechanical details of chemical reaction mechanisms with an unprecedented precision [Int. Rev. Phys. Chem. **31** (2012), 1]. The aim of the project is to explore quantum effects in cold collisions of ultracold atoms and molecules with quantum-state-selected, cold molecular ions in an ion-neutral hybrid trap [Phys. Rev. Lett. **107** (2011), 243202]. Building on recent pioneering experiments on cold collisions of molecular ions performed by the Basel group [Phys. Rev. Lett. **109** (2012), 233202], reactive collisions between ultracold atoms and state-selected molecular ions will be studied to characterize the effect of the internal quantum state in cold ion-atom collisions. Subsequently, these experiments will be extended to cold neutral molecules, enabling for the first time the study of collisions and chemical reactions between neutral molecules and molecular ions at millikelvin temperatures. This project will benefit from an intense theory collaboration with Laboratoire Aimé-Cotton du CNRS in Orsay near Paris to which a 6-months secondment is planned.

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ESR13: Development of fs-laser (frequency comb) techniques for ultrahigh resolution spectroscopy of molecular ions

Supervisor: Michael Drewsen
Host Institution: Aarhus University
Duration: 36 months
Planned secondment: Heinrich Heine University of Düsseldorf

Description:
The aim of the project is mainly to develop fs laser techniques for ultra-precise measurements of the rotational splitting in molecular ions. For instance, using a previously developed fs frequency comb technique for atomic state manipulations [Phys. Rev. Lett. **104**, 140501 (2010)], transitions between the low lying rotational states of the electronic and vibrational ground state of the MgH^+ ion will be investigated. The attainable resolution is even expected to allow for observation of the hyperfine splitting of these rotational levels. Through coherently driven Raman transitions it is furthermore the aim to demonstrate Rabi oscillations between two distinct rotational states, and evaluate the prospects of using such states as alternative qubit states for an ion trap based quantum computer.

The ultimate spectral resolution will be achieved using Quantum Logic Spectroscopy (QLS) [Science **309**, 749 (2005)] on a single molecular ion prepared in the quantum mechanical ground state with respect to their external motional modes. Sympathetic sideband cooling will be an important ingredient for achieving this. Through the additional application of adiabatic cooling [G. Poulsen and M. Drewsen, arXiv:1210.4309], it is furthermore the intension to extend QLS to direct vibrational spectroscopy of MgH^+ molecules.

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