

MANIPULATION OF TRANSLATIONAL MOTION OF POLAR MOLECULES BY STRONG RESONANT MICROWAVE FIELDSWei Zhong, Manish Vashishta, Pavle Djuricanin, and Takamasa Momose

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The ultimate resolution of spectroscopy is limited by the duration of the interaction time between molecules/atoms and radiation fields, also known as transit-time broadening. In order to achieve sub kHz resolution or better, it is indispensable to make translationally slow molecular ensembles. Spectroscopy of such cold and ultracold molecules are expected to offer completely new possibilities beyond those achieved by cold atoms in the study of fundamental and practical problems. These include the measurement of the electric dipole moment of the electron [1,2] and the measurement of the energy difference between right-handed and left-handed forms of chiral molecules [3].

We have been developing various techniques to make cold molecular ensembles below 1 K. One promising method for controlling the translational motion of polar molecules is to use the AC Stark force induced by microwave electric fields nearly resonant to a pure rotational molecular transition [3,4]. By periodically switching the standing waves of microwave fields in a well-designed microwave cavity, it is possible to decelerate molecular beams down to a few mK [6] and trap them. With this technique, we have successfully manipulated the translational motion of the $J = 0$ rotational ground state of acetonitrile and acetone by the microwave fields nearly resonant to the $J = 1 \leftarrow 0$ pure rotational transition. Our latest progress on microwave deceleration will be discussed.

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A VERSATILE METHOD FOR PRODUCTION OF COLD POLYATOMIC POLAR MOLECULES

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Dense ensembles of translationally slow and internally cold polar molecules offer a powerful tool for exploration of fundamental physics, e.g., measurement of the electric dipole moment of the electron, and open new avenues for research in diverse fields, from quantum many-body physics through quantum information science to controlled chemistry. Producing such samples, however, still remains a challenge, in particular for polyatomic molecules, since it requires simple and reliable but yet versatile techniques for deceleration, trapping, and cooling of molecules.

Towards this end, we have demonstrated the production and electrostatic guiding [1] of cold molecular beams from a cryogenic buffer-gas cell [2] and the deceleration of fast polyatomic molecules by the centrifugal force in a rotating frame [3]. RF depletion spectroscopy in the electrostatic guide is employed to probe the internal states of the molecules from the cryogenic buffer-gas cell. The combination of the cryogenic buffer-gas source with the centrifuge decelerator is expected to deliver intense beams of both internally cold and translationally slow molecules. Trapping and accumulation can take place in an electrostatic trap [4], and subsequent further cooling in the trap can be realized via a recently demonstrated opto-electrical Sisyphus effect, involving infrared, microwave, and RF transitions between internal states of the molecules [5]. The combination of the above individual techniques holds promise to yield dense samples of slow molecules with high internal-state purity.

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**TOWARDS MEASURING PARITY VIOLATION IN CHIRAL MOLECULES
USING VIBRATIONAL SPECTROSCOPY**

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Parity violation (PV) effects have so far never been observed in chiral molecules. Caused by the weak nuclear force, PV should lead to frequency differences in the rovibrational spectra of the two enantiomers of a chiral molecule. However the effect is small, making its observation a very difficult experimental challenge. The measurement of PV in molecules is interesting for a range of subjects across the board. It is a probe of the weak interaction, serving as a test of the standard model of physics. It has also been suggested to be related to biomolecular homochirality, a strong quantity imbalance observed on earth between left- and right-handed biomolecules. We have been working towards measuring this difference using Ramsey interferometry in the mid-infrared (at around 10 μm) using ultra-narrow line width CO₂ lasers referenced to atomic clocks in Paris via an optical link. We expect to reach a fractional sensitivity at around 10⁻¹⁵ (~ 10 mHz) on the frequency difference between enantiomers [1].

We present the results of preliminary investigations conducted on methyltrioxorhenium (MTO), an achiral test molecule whose chiral derivatives, which have recently been synthesized, are expected to show a ~ 10⁻¹⁴ level PV effect [2]. We report on the high-resolution spectroscopy of MTO [3, 4], both in cells and in molecular beams. This work has enabled us to identify several key elements of the current experiment needing improvement prior to making a PV measurement. The first is the lack of tunability of our CO₂ lasers. We present our on-going work towards their replacement with quantum cascade lasers (QCLs) [5] the very latest mid-infrared laser technology which offers broad and continuous tuning. Secondly, the current molecular beam source only yields a modest flux for species such as MTO which are solid at room temperature. We plan to overcome this by developing a buffer-gas-beam source and report on our latest efforts to implement buffer-gas cooling on polyatomic species such as MTO.

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**QUANTUM INTERFERENCE ASSISTED MEASUREMENT OF ABSOLUTE
ABSORPTION CROSS SECTIONS**

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Quantum interference experiments with molecules open the path to study the quantum superposition principle with complex and massive particles.

We discuss the current state of the art of successful matter-wave interference experiments [1,2].

The phase and visibility of the molecular interference fringes is highly sensitive to external perturbations. In our experiments we exploit this for gaining information on various internal molecular properties, such as electric susceptibilities and dipole moments [3] and conformational dynamics.

We demonstrate in particular the measurement of absolute optical absorption cross sections using Kapitza-Dirac-Talbot-Lau interferometry [4,5].

The absorption of single photons from a probe laser imparts a momentum recoil onto the molecules inside the interferometer. This leads to an effective reduction of the quantum fringe visibility from which we deduce an absolute absorption cross section.

Our method is independent on the knowledge of the molecular vapour pressure and it is applicable to dilute molecular beams. It is minimally invasive and can therefore be applied to a large class of neutral molecules, clusters and nanoparticles.

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HIGH-RESOLUTION ROTATIONAL SPECTROSCOPY IN A CRYOGENIC 22-POLE ION TRAP

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We present a novel action-spectroscopic scheme for high-resolution rotational spectroscopy of mass-selected, cold molecular ions. First introduced for performing electronic spectroscopy [1], and termed LIICG (Light Induced Inhibition of Complex Growth), we have recently demonstrated the versatility of the new technique by recording ro-vibrational spectra of CH_5^+ [2] and purely rotational spectra of C_3H^+ [3]. LIICG is based on the method of light induced reactions (LIR) [4], but instead of promoting a bimolecular reaction process by resonant excitation it utilizes a change of the rate of termolecular He attachment to molecular ions depending on their internal excitation, making it a very general method seemingly applicable to a large class of cations. Rotational spectra are recorded by storing only a few thousand mass-selected ions in the presence of a high number density of He (around 10^{14} cm^{-3}) in a cryogenic (4 K) 22-pole ion trap, and counting the formed ion-He complexes as a function of excitation frequency after a fixed storage time.

Here we will give details on the rotational LIICG scheme and our understanding of its underlying kinetics based on measurements of well known rotational transitions of CO^+ , HCO^+ , and CD^+ . We will also present first rotational data on the astronomical relevant ions CH_2D^+ , CD_2H^+ , and NH_3D^+ .

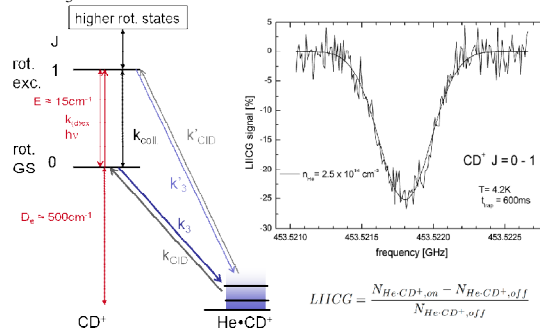


Fig. 1: **Left:** LIICG excitation and reaction scheme for CD^+ in the presence of a high number density of He. **Right:** The rotational ground state line of CD^+ recorded with LIICG by storing about 4000 CD^+ in a cold and dense helium bath for 500 ms and continuous irradiation with tunable narrow-band mm-wavelength radiation (VDI multiplier chain, 100 μW output power). Upon resonant excitation the number of formed CD^+ -He complex ions decreases, so that the rotational spectrum of the bare ion can be recorded by counting the complex ions as a function of excitation frequency.

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**USING MOLECULAR ALIGNMENT TO TRACK THE COLLISIONAL
RELAXATION OF A GAS SAMPLE**G. Karras¹, F. Billard¹, J. Houzet¹, E. Hertz¹, B. Lavorel¹, J.M. Hartmann², O. Faucher¹¹ Laboratoire Interdisciplinaire Carnot de Bourgogne (ICB), UMR 6303 CNRS-Université de Bourgogne, 9 Avenue Alain Savary, BP 47 870, 21078 Dijon Cedex, France² Laboratoire Interuniversitaire des Systèmes Atmosphériques (LISA), UMR 7583 CNRS-Université Paris Est Créteil, Université Paris Diderot, Institut Pierre-Simon Laplace, 94010 Créteil Cedex, France

One of the most effective ways used to induce post-pulse alignment in a molecular ensemble is by irradiating it with a short laser pulse. In the simplest scenario, a linearly polarized laser pulse with moderate intensity, far from any resonance with the molecular energy levels, is capable of aligning a molecule along its polarization axis. Since for most linear molecules their electric polarizability is anisotropic with respect to the plane of the molecular axis, its coupling with the electric field of the laser leads to their alignment through two-photon Raman-like transitions. The present work takes advantage of the polarization technique and demonstrates the use of molecular alignment as a tool for probing dissipative environments. The theoretical framework was originally proposed by Ramakrishna and Seideman [2] and developed further by Hartmann and Boulet [3]. Experimentally it has been already shown that under ambient and high pressure conditions [4], collisions between the molecules affect differently the permanent and the transient components of the alignment. Here, a preheat pulse is used to increase the rotational temperature of the molecules, which subsequently dissipate this energy to their translational degrees of freedom through elastic and inelastic collisions. As it is shown, due to rotational-translational R-T transfers, the averaged rotational energy is progressively reduced whilst the average translational is increased. The dissipation dynamics are tracked in a time interval of 500 ps, enough for the system to reach full Boltzmann equilibrium, by recording the induced molecular alignment.

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**TWO PHOTON ROTATIONAL ACTION SPECTROSCOPY
OF COLD OH⁻ IN A 22-POLE TRAP**

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Action spectroscopy of cations in 22-pole traps has seen many improvements since the introduction of laser induced reaction (LIR) [1]. Recently the technique of He attachment to cold (~ 4 K) species was introduced (LIICG – laser induced inhibition of cluster growth [2]), removing the need of a suitable endothermic reaction. Moreover, this method is also applicable to pure rotational spectroscopy [3], which was feasible only for few species with LIR. We present an alternative two photon scheme for direct rotational spectroscopy, applicable to anions and cations. In this case an IR LIR process (endothermic anion molecule-reaction, see Fig. 1. Eq. 3.) is used to probe a rovibrational excitation of OH⁻ by counting the reaction product H⁻ and a THz excitation is used to alter the population of OH⁻ rotational states at the same time. Although the rovibrational and rotational transitions of OH⁻ were measured before [4,5], this technique allows to obtain the rotational transition with unprecedented 1 ppb accuracy.

Two photon sequence:

1. OH⁻ ($\nu = 0, J = 0$) + $h\nu_1 \rightarrow$
OH⁻ ($\nu = 0, J = 1$)
2. OH⁻ ($\nu = 0, J = 1$) + $h\nu_2 \rightarrow$
OH⁻ ($\nu = 1, J = 1$)
3. OH⁻ ($\nu = 1, J = 1$) + H₂ \rightarrow
H⁻ + H₂O

$h\nu_1$ – THz photon $h\nu_2$ – NIR photon

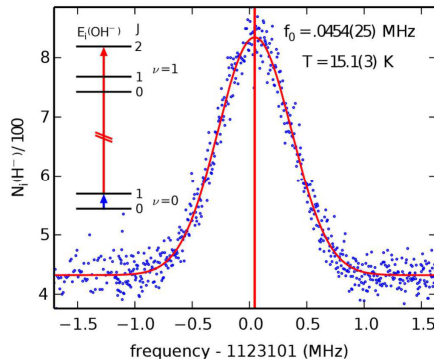


Figure 1. Two photon LIR excitation consisting of the rotational and rovibrational transitions. The rovibrational transition is kept at resonance. The nominal temperature of 10 K leads to moderate OH⁻ ($J=1$) population and to stable background. As soon as the rotational transition is scanned into resonance, the population of $J=1$ state increases and additional product counts can be detected.

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