

A new experiment to test parity symmetry in cold chiral molecules using vibrational spectroscopy

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Abstract. We present a brief review of our progress towards measuring parity violation in heavy-metal chiral complexes using mid-IR Ramsey interferometry. We discuss our progress addressing the main challenges, including the development of buffer-gas sources of slow, cold polyatomic molecules, and the frequency-stabilisation of quantum cascade lasers calibrated using primary frequency standards. We report investigations on achiral test species of which promising chiral derivatives have been synthesised.

Keywords: parity violation, mid-IR high-precision laser spectroscopy, frequency metrology, Ramsey interferometry, molecular beams, buffer-gas cooling, chiral molecules, quantum cascade laser.

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1. Introduction

Many molecules are chiral, coming in left- and right-handed versions known as enantiomers. Enantiomers are mirror images of one another. The potential energy surface of a chiral molecule shows two minima separated by an interconversion barrier, which one may associate with the left- and right-handed enantiomers. Strikingly, enantiomers, or chiral states, are not the eigenstates of the symmetric molecular electromagnetic Hamiltonian and nutation from one to the other can happen by quantum tunnelling through the potential barrier. However, interaction with the environment, or decoherence, tends to hold the molecule in the left- or right-handed enantiomer. When the interconversion barrier between enantiomers is very high, which is the case for the molecules considered here, the left- and right-handed states can then be considered to a good approximation as energy eigenstates.

In addition to the electromagnetic interaction, the weak interaction, one of the three other fundamental forces, is also at play in molecular systems, notably in the interaction between electrons and nuclei. According to the Standard Model, the energy levels of enantiomers should be slightly different because of the parity-violation (PV, the left-right symmetry breaking) inherent in the weak force [1]. We aim to make the first ever measurement of this symmetry-breaking energy difference. This measurement will serve as a sensitive probe of the weak force and the limits of the Standard Model [2], and might shed light on the mystery of biological homochirality [3].

Atomic physics experiments carried out in Novosibirsk in 1979 found that neutral weak currents violate parity [4]. Since then, scientists have dreamt of measuring the PV energy difference between chiral enantiomers, but no experiment has ever reached the required sensitivity. A number of techniques have been proposed for the observation of PV in chiral molecules, including rotational, rovibrational, electronic, Mössbauer and NMR spectroscopy, as well as crystallisation and solubility experiments, and optical activity measurements (see [2, 5–8] and references therein). However, to our knowledge very few other groups currently pursue an experimental realisation. Dmitry Budker's group has recently proposed a measurement using NMR spectroscopy [9], and Martin Quack's group is currently pursuing a measurement based on the time evolution of parity in chiral species after a parity selection step [10]. The latter is an optical technique, but one quite different from our approach. It has distinct experimental challenges, and requires working with substantially different chiral species. This group has published a proof of principle using an achiral molecule [11] and has also proposed promising candidate species for this experiment [12].

The parity-violating energy difference in turn leads to frequency differences between the rovibrational spectra of left- and right-handed enantiomers, which we aim to measure for the first time using high-precision mid-IR spectroscopy [13]. The most precise experiment, carried out around 2000 at the Laboratoire de Physique des Lasers by one of the co-authors, measured the 30 THz vibrational frequency of the C–F stretch mode of CHFClBr molecules using saturated absorption spectroscopy [14, 15]. This reached a precision of 8 Hz, a great achievement but still a factor of 3000 larger than the ~ 2 mHz predicted frequency difference between the left- and right-handed enantiomers of that molecule [16]. To improve on this, we are building a more sensitive instrument and are learning to work with molecules where the energy difference is expected to be far larger.

2. Molecules under study

The PV energy difference scales strongly with nuclear charge [17], so we consider molecules with heavy atoms near the chiral centre. We have already worked with theoretical and experimental chemists to find the best chiral species for measuring the PV energy difference. This led to the successful synthesis of solid oxorhenium organo-metallic compounds that have vibrational transitions around 30 THz and PV frequency shifts as large as ~ 1 Hz (see [5, 18–21] and references therein), up to 1000 times larger than in CHFClBr. Several other tracks are currently being followed and other molecules are being prepared. Our collaborators have recently found a uranium compound ($N \equiv \text{UHFI}$) with a record ~ 20 Hz predicted PV frequency shift [22]. While synthesising and isolating such compounds has not been demonstrated so far, this could be possible in the future.

To gain insight in the apparatus and know-how required for performing precise spectroscopic measurements on such complex species, we have conducted high-resolution mid-IR spectroscopy of the achiral precursor methyltrioxorhenium (MTO, CH_3ReO_3) in both room temperature cells and cold supersonic beams [5, 18, 20, 23, 24]. Based on these first results, we are designing and constructing a state-of-the-art instrument for high-resolution vibrational spectroscopy of chiral molecules.

3. Experimental setup

Figure 1 illustrates the setup of this new experiment. A slow, cold molecular beam of the rhenium species of interest is made by producing the molecules in the gas phase inside a buffer-gas cell cooled to a few kelvins, and then extracting the molecules into a beam. For molecules which are solid at room temperature, such buffer-gas beams formed in a cryogenic cell, one of the latest molecular beam source technologies, exhibit both low velocity and some of the highest beam fluxes to date [25], making them very attractive for precise spectroscopic measurements. The molecules then pass through a Ramsey interferometer sensitive enough to measure tiny changes in the vibrational frequency of the molecules [26]. To reach the required frequency, which is in the mid-IR region of the spectrum, we use quantum cascade lasers (QCLs). To achieve the required frequency stability, we stabilise the laser to a frequency comb, which is itself locked to an ultra-stable $1.5 \mu\text{m}$ laser signal, ultimately referenced to primary frequency standards, potentially a Cs fountain clock which realises the International System of Units standard of time

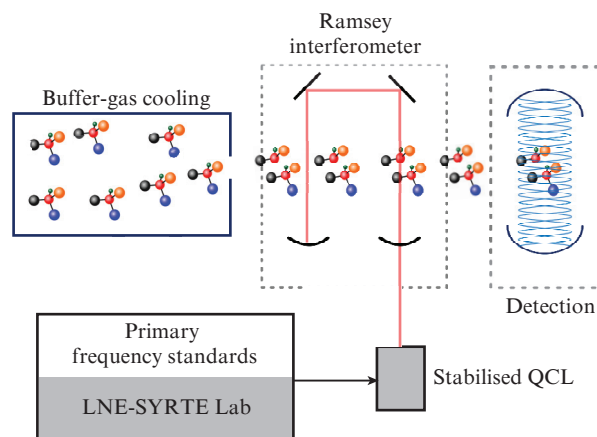


Figure 1. Schematic of the proposed experiment to measure parity-violating rovibrational frequency differences between enantiomers of cold chiral molecules using a stabilised quantum cascade laser (QCL).

[27, 28]. QCLs offer broad and continuous tuning, and available wavelengths cover the entire mid-IR region, allowing the study of a considerable number of candidate species and thus providing invaluable flexibility.

Molecules must then be detected with high sensitivity. We are currently investigating detectors based on cavity-enhanced schemes recording mid-IR absorption or the microwave field molecules emit when they rotate. To measure the PV vibrational frequency shift, the vibrational frequency of each enantiomer must be measured in this new apparatus. Because this is a differential measurement between the two enantiomers, most systematic frequency shifts will cancel out. We project a measurement precision below 0.1 Hz [5], an improvement of at least two orders of magnitude compared to the CHFClBr experiment.

4. Buffer gas cooling

We have already made much progress in the development of methods to cool the complex and heavy rhenium species of interest for PV measurements to a few kelvins. This is achieved through collisions with a buffer gas of 4 K helium inside a copper cell mounted on the cold stage of a cryo-cooler. As illustrated in Fig. 2, we have demonstrated cryogenic buffer-gas cooling of the first organo-metallic species, MTO [24, 29]. The molecules are produced with a rotational temperature of approximately 6 K by laser ablation of a MTO pellet inside the buffer-gas cell. This development extends the technique of buffer-gas cooling to a new class of molecules.

We have also learned how to bring such species into the gas-phase by laser ablation, a method previously used only for diatomic molecules. Many polyatomic molecules have little vapour pressure at easily accessible temperatures, so the ability to introduce them by laser ablation is important. Our work shows that rhenium species of interest for PV measurements survive the ablation process and that the translational and rotational degrees of freedom cool efficiently through collisions with helium, which is very promising for the production of buffer-gas-cooled molecular beams. We have also demonstrated the first precise spectroscopic measurements of buffer-gas-cooled molecules in the mid-IR molecular fingerprint region around $10 \mu\text{m}$, obtaining rotational and hyperfine-resolved absorption spectra (see Fig. 2), and allowing the determination of the nuclear quadrupole coupling of the

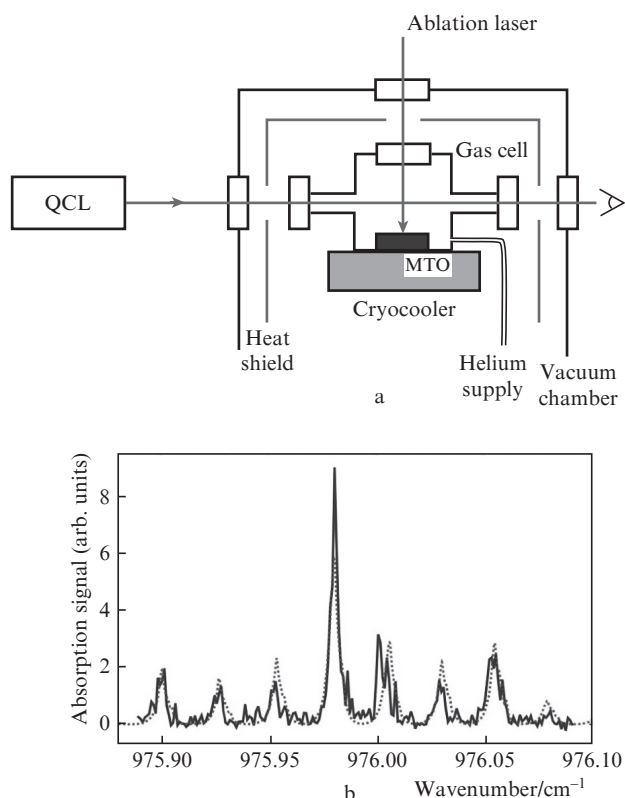


Figure 2. (a) Experimental setup for cryogenic buffer-gas cooling of methyltrioxorhenium (MTO): a copper cell is filled with $\sim 10^{-2}$ mbar of helium and is cooled to 6 K using a cryocooler; the cell is surrounded by aluminium radiation shields at a temperature of about 40 K, and is housed in a vacuum chamber; a target containing MTO is ablated with pulses from a Nd:YAG laser at 1064 nm; light from a free-running quantum cascade laser (QCL) emitting at ~ 976 cm^{-1} is sent through the cell to record absorption spectra. (b) Spectrum of buffer-gas-cooled molecules showing the rotational contour of the Q branch of the antisymmetric Re = O stretching mode of the ^{187}Re isotopologue; the dashed curve is a fit to the data from which a rotational temperature of 6 ± 3 K is inferred [29].

excited vibrational state, which is unprecedented for such a complex molecule.

We have also demonstrated cryogenic buffer-gas cooling of another polyatomic molecule, 1,3,5-trioxane [(CH₂O)₃, a cyclic trimer of formaldehyde]. Although it is a solid at room temperature, it has enough volatility to be injected into the buffer-gas cell through a room temperature tube. We have recorded saturated absorption spectra of trioxane, which constitute the first sub-Doppler spectroscopic measurements of buffer-gas-cooled molecules in the fingerprint region and the first sub-Doppler spectra of polyatomic molecules inside a buffer gas cell.

Challenges remaining in the source development are the construction of a versatile buffer-gas source for beam production. Our work has focussed on developing methods of laser ablation and capillary loading. All the measurements were performed inside the cell. The next step will be to make a molecular beam, and optimise the cell configuration to produce a slow, high-flux beam of cold complex molecules. Given that we do not yet know which chiral molecule we will use for our measurement, it is beneficial to design a versatile cell that can be used with a range of loading mechanisms. Capillary loading is well-suited to molecules with a high volatility, whereas low volatility species are better introduced via an

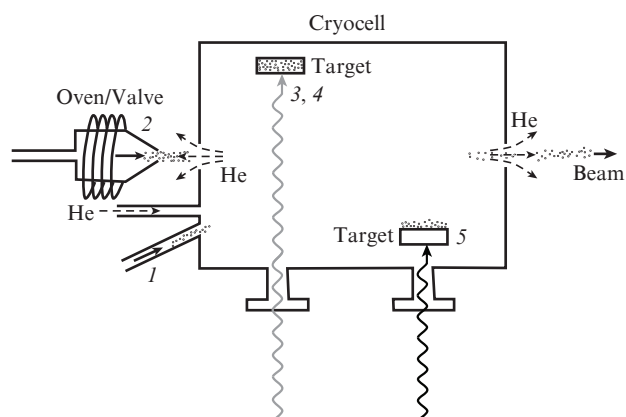


Figure 3. Schematic of a buffer-gas cell, showing different molecule-loading mechanisms: (1) capillary loading; (2) loading from a valve or oven; (3) laser ablation; (4) matrix-assisted desorption; (5) laser-induced acoustic desorption. Small points show molecules of interest; dashed arrows represent helium flow; the grey wavy line represents a laser beam for ablation or desorption of a solid target containing the molecules of interest; the black wavy line represents laser radiation for laser-induced acoustic desorption of a solid target containing the molecules of interest.

oven or through laser ablation. Figure 3 shows different loading mechanisms that we aim to investigate.

The development of the molecular beam will require sensitive detection. We will build an optical cavity around the beam-line and perform cavity-enhanced absorption spectroscopy in the mid-IR range. Combining this with wavelength modulation spectroscopy will allow weak absorption signals to be recorded with a large signal-to-noise ratio, ensuring great sensitivity for the optimisation of the beam flux.

5. Laser stabilisation for high-precision vibrational spectroscopy

Regarding the development of laser techniques for our new instrument, we have been able to phase-lock a 10 μm QCL to the secondary frequency standard of this spectral region, a CO₂ laser stabilised on a saturated absorption line of OsO₄ [23, 30]. The excellent spectral features of the ultra-stable CO₂ laser were transferred to the QCL, resulting in a linewidth on the order of 10 Hz, about two orders of magnitude narrower than the previously published narrowest QCL linewidth. More recently, we have developed a widely tunable frequency-stabilised QCL with direct traceability to primary frequency standards. Our method allows us to lock any mid-IR laser to a frequency comb stabilised to a near-IR reference, given by the optical phase of an ultra-stable 1.55 μm laser located at LNE-SYRTE (the French national metrology institute). This signal is monitored against atomic frequency standards and transferred via a 43-km long optical fibre cable with correction of the propagation-induced phase noise. The stability of the reference is transferred to the mid-IR source, which therefore exhibits a relative frequency stability lower than 2×10^{-15} between 1 and 100 s. Moreover, thanks to the traceability to the primary standards of LNE-SYRTE, its absolute frequency is known with an uncertainty below 10^{-14} (potentially as low as the accuracy of the Cs fountain clock, 3×10^{-16}). This stabilisation method was demonstrated with a CO₂ laser [31, 32] and a QCL [27, 28].

Stabilising the laser this way frees us from having to lock the QCL to any particular reference (either another mid-IR

laser or a molecular transition), which would constrain the laser's operating frequency range. It also results in a ~ 0.1 -Hz linewidth. To our knowledge, this is by far the narrowest and most accurate QCL reported. The setup allows the QCL to be scanned over a few hundred megahertz while maintaining the highest stabilities and accuracies by scanning the near-IR reference using an electro-optic modulator [33].

To demonstrate the QCL's potential for state-of-the-art high-precision spectroscopic measurements, saturated absorption spectra of OsO_4 as narrow as 25 kHz were recorded using a Fabry–Perot cavity, allowing centre frequencies to be determined with an uncertainty of a few tens of hertz [27, 28]. As illustrated in Fig. 4, we have also conducted saturated absorption spectroscopy of several methanol lines in a multipass cell, and have determined central frequencies with an uncertainty of a few kilohertz, an improvement of 3 to 4 orders of magnitude over previous measurements.

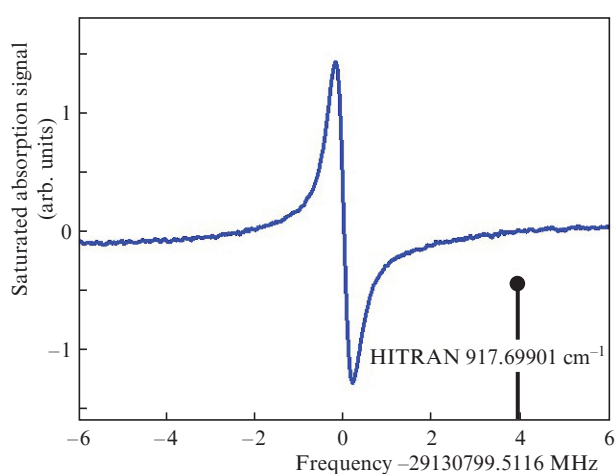


Figure 4. Saturated absorption spectrum of the P(A,co,0,0⁺,33) rovibrational line of methanol, belonging to the P branch of the ν_8 C–O stretch vibrational mode, recorded in an astigmatic Herriott multipass cell (Aerodyne Research, AMAC-36 model, 20 cm base path, 182 paths) using frequency modulation and first-harmonic detection. Light from a stabilised QCL is split into a pump beam and a probe beam of power incident to the cell of around 1.5 mW and 0.8 mW, respectively. Experimental conditions are as follows: pressure, 1 Pa; modulation frequency, 20 kHz; frequency modulation excursion, 50 kHz; frequency step, ~ 15 kHz; averaging over five pairs of scans of the opposite frequency sweep direction; and total integration time per point, 1 s. The P(A,co,0,0⁺,33) line frequency position reported in the HITRAN database [34] is also shown.

We have so far demonstrated ultra-stable QCLs in a limited spectral window around 10 μm , but the resonances of many promising molecules are not at 10 μm . Challenges in the laser system development are the construction of fully operational spectrometers based on QCLs stabilised to the comb anywhere in the mid-IR range while taking full advantage of the wide tunability offered by QCLs. To provide a high level of flexibility, the spectrometer should be designed in such a way that QCLs can be easily interchanged, to allow us to stabilise QCLs at any frequency.

6. Conclusions

We have described the progress towards our new experiment to measure signatures of parity violation in chiral molecules.

We have discussed recent developments made in the production of cryogenically cooled complex molecules, and in laser stabilisation of quantum cascade lasers. Together, the advances reported here demonstrate some of the methods needed for a measurement of parity violation in chiral molecules. Beyond PV, they also open up new possibilities for measuring and controlling complex molecules and for using them in a variety of precise measurements.

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