Exploring new Fundamental Physics by Precision Spectroscopy of Cold Molecules

«WHAT NEXT» MEETING

Pasquale Maddaloni



Firenze - May 4, 2015

Starting from *Puzzles*

Electron's electric dipole moment

Why does the Universe have so little anti-matter?



Why are life macromolecules homochiral?



Variation of fundamental constants

Can Gravity be unified with other interactions?

$$G = 6.673 \times 10^{11} \text{ Nm}^2 \text{ kg}^2$$

$$c = 3 \times 10^8 \text{ ms}^{-1}$$

$$h = 6.626 \times 10^{-34} \text{ Js}.$$

Axion dark matter

What is dark matter?



Electron's EDM measurement



ThO \implies $|d_e| < 8.7 \times 10^{-29} e \text{ cm} (90\% \text{ confidence})$

Extensions to the Standard Model, such as weak-scale supersymmetry, posit the existence of new particles and interactions that are asymmetric under time reversal (T) and nearly always predict a small yet potentially measurable electron electric dipole moment in the range of 10⁻²⁷ to 10⁻³⁰ e·cm. The size of the EDM is also intimately related to the question of why the Universe has so little antimatter. If the reason is that some undiscovered particle interaction breaks the symmetry between matter and antimatter, this should result in a measurable EDM







L. Santamaria et al., *in preparation*

Parity violation in chiral molecules







CHFClBr @ 9.3 μ m \Rightarrow 5·10⁻¹⁴ B. Darquie et al., Chirality 22, 870 (2010)

The potential energy surface of a chiral molecule shows two minima (as a function of the inversion coordinate q), that are the left "L" and right "R" enantiomers. In the presence of a weak force between nuclei and electrons, a small parity violation energy difference is expected

between the ground states (as well as excited states) of the enantiomers: R- and L-handed molecules cease to be exact mirror images of each other. For CHFClBr, $\Delta v_{PV} = v_L - v_R = (\Delta E^*_{PV} - \Delta E_{PV})/h$ is such that $\Delta v_{PV}/\nu \sim 10^{-16}$.

ALTERNATIVE APPROACHES

Study time-dependent oscillations in the optical activity of chiral molecules: the presence weak interactions would be revealed by oscillation of the optical activity around a non-zero value III M. Harris & Stodolsky, Phys. Lett. **78B**, 313 (1978)

Look for violation of the selection rules in the time evolution of states of well-defined initial parity: 1) preparation of molecular states of well-defined parity; 2) free evolution of the isolated molecule with initial well-defined parity; 3) observation of the population states of the forbidden parity (created by the parity violating interactions) as a function of time \square M. Quack, Chem. Phys. Lett. **132**, 147 (1986)

Constancy of constants

- Seneralized Kaluza-Klein models, String theories 🛄 J.-P. Uzan, Rev. Mod. Phys. 75, 403 (2003)

Inflation Model S.G. Karshenboim, Can. J. Phys. 83, 767 (2005) G. Börner, The Early Universe (Springer-Verlag, 1993)



Identifying the *Ingredients*: Outline

Cold and slow molecules (enhancing the interrogation time): *beams, fountains, traps*

High-resolution + high-sensitivity:

sub-Doppler saturation spectroscopy, Doppler-free polarization spectroscopy, twophoton spectroscopy, Ramsey fringes + cavity-enhanced absorption spectroscopy

Long-term frequency stability & absolute frequency calibration: optical frequency combs referenced to the Cs primary standard

Narrow emission linewidth:

<u>semiconductor lasers</u>: diode lasers, quantum and interband cascade lasers <u>nonlinear sources</u>: difference frequency generators, optical parametric oscillators

THE NIR/MID-IR: A SOLID FOUNDATION

Strong vibrational transitions (up to 10⁻¹⁸ cm/molec) with reduced natural linewidths (down to a few Hz) + valuable laser sources and detectors

P. Maddaloni et al., Laser-based measurements for time and frequency domain applications, Taylor&Francis (2013)

Improved samples

Interrogation techniques

> Radiation Sources

Cw Laser Sources and Optical Frequency Combs

LINKING ULTRA-NARROW-LINEWIDTH NIR/MIR PROBE LASER SOURCES TO THE CESIUM PRIMARY STANDARD







The second is defined as the duration of 9,192,631,770 periods of the radiation corresponding to the transition between two hyperfine levels $|F = 4, m_F = 0\rangle \leftrightarrow |F = 3, m_F = 0\rangle$ in the ground state ${}^2S_{1/2}$ of the ${}^{133}Cs$ atom

P. Maddaloni et al., Meas. Sci. Technol. **20**, 052001 (2009)

I. Ricciardi et al., Optics Express B **20**, 9178 (2012)

Comb-assisted laser spectroscopy

FC1500-250-WG Synthesizer Er fiber oscillator 1050-2100 nm



FC8004 Synthesizer Ti:Sa oscillator 500-1100 nm Still limited to visible/nearinfrared spectral region

(special sources and fibers required)



P. Maddaloni et al., New Journal of Physics 8, 262 (2006)

P. Malara et al. Opt. Express 16, 8056 (2008)



P. Cancio et al., Appl. Phys B **102**, 255 (2011)

Comb referencing of DFG/OPO sources **YDFA** ECDL2 pump **PPLN** idler Ge ouartz+Rb+GPS signal to wavemeter BS PZT F LO BU PPLN OFCS BS M DFG beam $f_{\rm loi}$ OFC BS LO BU BS to wavemeter Servo 1 ervo 2 ► EOM Signal PZT V 10 W SL PZT Yb-doped Yb-doped OPO BS ibre Lase ibre Amplifier ECDL1 **EDFA** Idler $f_n = n_n f_r + f_0 + f_{hast}^p$

$$f_s = n_s f_r + f_0 + f_{beat}^s$$

$$F_{DFG} \equiv f_p - f_s = (n_p - n_s)f_r + (f_{beat}^p - f_{beat}^s)$$

$$f_p - f_{EOM} = f_0 + n_p f_r + f_{beat}^p$$
$$f_s = f_0 + n_s f_r + f_{beat}^s$$

$$f_{OPO} = f_p - f_s = (n_p - n_s)f_r + (f_{beat}^p - f_{beat}^s) + f_{EOM}$$

Direct cooling of stable molecules

ACCESS TO HYDRIDES, NITRIDES, OXIDES, FLUORIDES,...

AS OPPOSED TO CREATION OF DIMERS VIA MAGNETO (PHOTO)-ASSOCIATION OF ULTRACOLD ALKALI ATOMS

Stark Deceleration



Stark effect $H = H_0 - \vec{\mu} \cdot \vec{\mathcal{E}}$

□ H.L. Bethlem et al., Phys. Rev. Lett. **83**, 1558 (1999)

Hexapole lens



In linear regime, the hexapole provides a radial (velocity) confinement due to a quadratic potential

 $F_r = -k_{el}r$

-5 kV -3.5 kV 0 kV 3.5 kV 5 kV 3.5 kV 0 kV -3.5 kV -5 kV

Hundreds of ring-shaped tantalum electrodes are connected in sets of 8. Oscillating high voltages are applied to these sets with a phase difference of $2\pi/8$, thereby creating a series of electric field minima, which are true 3D traps for molecules in a low field seeking state.

$$V_n(t) = V_0 \sin\left[-\phi(t) + \frac{2\pi n}{8}\right]$$
$$v_z(t) = f(t)L = \frac{1}{2\pi} \frac{d\phi}{dt}L$$

The frequency of the voltages determines the velocity of the moving traps. Initially, as the molecules enter the decelerator, the traps are set to move at the same speed as the molecules. Then, gradually, the oscillation frequency of the voltages is swept down resulting in the deceleration and ultimately stopping of the traps, with the molecules remaining in the traps.

A. Osterwalder et al., Phys. Rev. A **81**, 051401(R) (2010)



3D MOT of strontium monofluoride at a temperature of approximately 2.5 mK, loaded with pulses of SrF from a cryogenic buffer gas beam source that have been slowed using radiation pressure

factor of 29.

centre in a more weakly trapped state. Combined with the

unidirectionality of the optical pumping back to the strongly

trapped states, this cools about one million of CH_3F molecules by a factor of 13.5, with the phase space density increased by a



Microwave-forced evaporative cooling of neutral hydroxyl molecules loaded from a Stark-decelerated beam into a high-gradient magnetic quadrupole trap → cooling by one order of magnitude in temperature (correspondingly phase-space density increases by 3 orders of magnitude)

Symphatetic cooling

W.G. Rellergert et al., Nature **495**, 490 (2013)



The vibrational motion of trapped BaCl⁺ molecules is quenched by collisions with ultracold calcium atoms at a rate that is over four orders of magnitude more efficient than traditional sympathetic cooling schemes. The high cooling rate, a consequence of a strong interaction potential (due to the high polarizability of calcium), along with the low collision energies involved, leads to molecular samples with a vibrational ground-state occupancy of at least 90%. Because sympathetic cooling of molecular rotational motion is much more efficient than vibrational cooling in traditional systems, the method should also allow efficient cooling of the rotational motion of the molecules Sounding the time Unwinding of the PRoton-to-Electron Mass ratiO - SUPREMO

INFN EXPERIMENT - CSN II

https://web.infn.it/supremo



Main goal

Constrain over a-few-year timescale the fractional temporal variation of the proton-to-electron mass ratio at a level of <u>10⁻¹⁵/yr</u> by means of a spectroscopic frequency measurement on a beam of cold and slow molecules

$$\beta = \frac{m_p}{m_e} \qquad \qquad \frac{1}{\beta} \frac{d\beta}{dt} < 10^{-15} \,\mathrm{yr}^{-1}$$

Possible validation of multidimensional theories seeking to unify the three distinct gauges of the Standard Model towards a GUT (Lie groups,...)

🚇 H. Fritzsch, Lect. Notes Phys. 648, 107 (2004)

Testing TOE-aspirant theoretical frameworks (String/M-theories, Loop quantum gravity,...) attempting to reconcile Standard Model and General Relativity
 C. Kiefer, Lect. Notes Phys. 648, 115 (2004)

State of the art

Hybrid Cosmological Approach

look-back time of several Gyr

poor control on data

Compare wavelengths of atomic/molecular lines on Earth (at present epoch) and from astronomical objects at high redshifts (up to z≈3)

P. Molaro, Highlights of Astronomy **15**, 326 (2010)

Transition		Energy scaling
Atomic	Gross structure Fine structure Hyperfine structure	$Ry lpha^2 Ry lpha^2 (\mu/\mu_B) Ry$
Molecular	Electronic structure Vibrational structure Rotational structure	$Ry \\ \beta^{-1/2} Ry \\ \beta^{-1} Ry$
Relativistic corrections		Function of α



All-Lab Approach

reduced look-back time

accuracy, and reproducibility

High-precision comparisons between transitions with different sensitivities to β

S.N. Lea, Rep. Prog. Phys. **70**, 1473 (2007)

 $\Delta\beta/\beta = (0.0 \pm 1.0) \cdot 10^{-7}$ z=0.89 (7 Gyr)

🛄 J. Bagdonaite et al., Science **339**, 46 (2013)

 $\dot{\beta}/\beta = (-3.8 \pm 5.6) \cdot 10^{-14} \text{ yr}^{-1}$

A. Shelkovnikov et al., Phys. Rev. Lett. **100**, 150801 (2008)



L. Santamaria et al., J. Mol. Spectrosc. **300**, 116 (2014)



Buffer Gas Cooling



Both translational and rotational (internal) degrees of freedom of the desired molecular species are cooled via collisions with a thermal bath of helium in a cryogenic cell
S.E. Maxwell et al., Phys. Rev. Lett. 95, 173201 (2005)
N.R. Hutzler et al., Chem. Rev. 112, 4803 (2012)



Molecular Beam Parameters



Reduced mean forward velocity (*u*) and temperature (i.e. spread around $u, \Delta u$)

Effusive beam	<i>Re</i> < 1	$\Delta \theta \approx 120^{\circ}$
Partially hydrodynamic	1 < Re < 100	lower divergence
Supersonic beam	<i>Re</i> > 100	$\Delta\theta \approx 78^\circ$

$$Re \equiv \frac{F_{inert}}{F_{viscous}} \simeq \frac{8\sqrt{2} f_b \sigma_{b-b}}{\sqrt{\frac{8k_B T_b}{\pi m_b}} r} \qquad \sigma_{b-b} \simeq 3 \cdot 10^{-19} \text{ m}^2$$

 $1 \leq Re \leq 10$

$$\begin{split} u &\simeq \sqrt{\frac{8k_B T_b}{\pi m}} \left(1.2 + 0.6 \, Re \sqrt{\frac{m_b}{m}} \right) \\ \Delta u &\simeq \Delta v_\perp \simeq 1.5 \, \sqrt{\frac{8k_B T_b}{\pi m}} \\ \Delta \theta &= 2 \arctan\left(\frac{\Delta v_\perp}{u}\right) \simeq 2 \arctan\left(\frac{1.5}{1.2 + 0.6 \, Re \sqrt{\frac{m_b}{m}}}\right) \end{split}$$

Characterizing the collisional cooling process Translational temperatures are Laser-absorption ro-vibrational spectroscopy extracted by Doppler thermometry of the ${}^{12}C_{2}H_{2}(v_{1}+v_{3})$ band is performed 1.00 $T_{trans} = 115 \text{ K}$ Spectroscopic absorption Pulse $T_{trans} = 15 \text{ K}$ Tube 0.75 signal (arb. unit) cell activated 0.50 acetylene charcoal line Shields 0.25 Laser Reference cell 0.00 250 -750 -500 -250 0 500 750 Frequency detuning (MHz) BS $-\frac{4 \ln 2 (\nu - \nu_0)^2}{\sigma^2}$ $G(\nu) = G_0 \exp$ Due to a non perfect thermal exchange between the copper pipe and the two PT plates, a temperature of about 15 K is measured on the He line just before the entrance into the BGC cell (at 4.2 K); to bridge this gap, $/8 \ln 2 k_B T_{trans}$ $\sigma_D = \frac{\nu_0}{\cdot}$ an improved setup for better cooling of the He line is under construction





The diffusion time of ${}^{12}C_{2}H_{2}$ in the BGC cell is measured against the ⁴He flux both at two separate translational temperatures: 100 and 25 K. The observed behavior is compared with that predicted by a Monte Carlo simulation to provide an estimate for the respective total elastic cross sections:

$$\sigma_{el} = (100 \text{ K}) = (4 \pm 1) \cdot 10^{-20} \text{ m}^2$$
$$\sigma_{el} = (25 \text{ K}) = (7 \pm 2) \cdot 10^{-20} \text{ m}^2$$

Future

In general, unlike what happens to translational states, even if the initial distribution over the rotational states is Boltzmannian, it will relax without preserving the canonical invariance, and it will not be possible to define a rotational temperature. To address this issue in our case, a normalized linestrength is measured for several ro-vibrational lines at a given translational temperature. The acquired behavior is then compared with the theoretical line dictated by the Boltzmann law. In both cases, the correlation coefficient is $\chi = 0.98$, consistent with the hypothesis of canonical invariance.



Characterization of the beam parameters (flux and velocities) by cavity ring-down spectroscopy





Pushing the ultimate resolution in the spectroscopic frequency measurement down to 10⁻¹⁸ by stabilizing the frequency comb, to which the probe laser is referenced, against an optical atomic standard

 \rightarrow Yb lattice clock under construction at INRIM

 $iggrad rac{1}{rac{
u_{vib}(M)}{
u_{el}(Yb)}}rac{\partial\left[rac{
u_{vib}(M)}{
u_{el}(Yb)}
ight]}{\partial t}=-0.5rac{\dot{eta}}{eta}-N_{Yb}rac{\dot{lpha}}{lpha}$

Team and Collaborations

FIRENZE (INO & INFN) Stark decelerated beam and REMPI spectroscopy

Giacomo Insero Gabriele Santambrogio Simone Borri Paolo De Natale

BOLOGNA (UNIBO) Molecular theory

Cristina Puzzarini

NAPOLI (INO & INFN) Precision spectroscopy and buffer gas cooling

Luigi Santamaria Valentina Di Sarno Gianpiero Mangano Iolanda Ricciardi Simona Mosca Maria Parisi Maurizio De Rosa

TORINO (INRIM) National optical fiber link

Davide Calonico Filippo Levi Massimo Inguscio

WASHINGTON, DC (NRL) Interband cascade lasers

Jerry Meyer