8 Luglio 2013

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ISTITUTO NAZIONALE DI OTTICA

Assessing the time constancy of the proton-to-electron mass ratio by precision ro-vibrational spectroscopy of cold fluoroform molecules

Pasquale Maddaloni

Proposal for a new INFN experiment

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MAIN GOAL

Constrain over a-few-years timescale

the fractional temporal variation of the proton-to-electron mass ratio

at a level of 10^{-15}/yr

by means of a spectroscopic frequency measurement

on a decelerated beam of cold molecules

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

- **Direct cooling of ground-state molecules as opposed to creation of dimers via magneto (photo)-association of ultracold alkali atoms → hydrides, nitrides, oxides, fluorides,...**

"CONSTANCY" OF CONSTANTS

- First conjecture: Dirac's big-numbers hypothesis

$$G \sim t^{-1}$$

P.A.M. Dirac, Nature **139**, 323 (1937)

- Generalized Kaluza-Klein models, String theories, other TOE candidate theories

$$R \sim l_P = \sqrt{\frac{\hbar G}{c^3}} \approx 1.62 \cdot 10^{-35}\ \mathrm{m}$$

$\dot{R} \neq 0$ → real changes in values of α, β,… take place

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)

…but Universe is still expanding and cooling → **continuous, slow (tiny) variation of fundamental constants over time**

ADDRESSING α AND β WITH ATOMIC/MOLECULAR SPECTROSCOPY

QED

$$\alpha = \frac{e^2}{4\pi\varepsilon_0\hbar c}$$

QCD

$$\alpha_s(r) = \frac{C}{\ln\left[\frac{r\Lambda_{QCD}}{\hbar c}\right]} \qquad \Lambda_{QCD} = 214^{+38}_{-35}\ \text{MeV} \Rightarrow m_p \propto \Lambda_{QCD}$$

β characterizes the strength of strong interactions in terms of the electro-weak

GUT prediction

X. Camet et al., Eur. Phys. J. C **24**, 639 (2002)

$$\frac{\dot\beta}{\beta} = R_C\frac{\dot\alpha}{\alpha} \qquad R_C = 20 \div 40$$

α and β appear prominently in atomic/molecular transitions

$$\alpha^{-1} = 137.035999074(44)$$

$$\beta = 1836.15267245(75)$$

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

ALL-LAB vs HYBRID COSMOLOGICAL APPROACH

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

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

+ look-back time of several Gyr

- poor control on data

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

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

Constraints on the fractional temporal variation of α (β) can be inferred from precise frequency comparisons of transitions with different sensitivities to α (β)

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

+ accuracy, reproducibility and unequivocal interpretation

β VARIATION – STATE OF THE ART

$$\frac{\dot{\alpha}}{\alpha} = (-1.6 \pm 2.3) \cdot 10^{-17}\ \text{yr}^{-1}$$

$$\frac{d}{dt} \ln \frac{\mu_{Cs}}{\mu_B} = (-1.9 \pm 4.0) \cdot 10^{-16}\ \text{yr}^{-1}$$

L. Lorini et al., Eur. Phys. J. Special Topics **163**, 19 (2008)

In principle, constraints on α, *Ry*, μ/μ_B (as inferred from atomic clocks) could be transferred to β by applying the nuclear Schmidt model; unfortunately, the uncertainty of the calculation within such a model is quite high (usually from 25% to 50%)

Atoms fail, but molecules come to the rescue!

Measure the frequency of a molecular vibrational transition relative to the hyperfine transition ($F = 4$, $m_F = 0$) ↔ ($F = 3$, $m_F = 0$) in the Cs electronic ground state

$$\frac{1}{\frac{\nu(S)}{\nu(Cs)}} \frac{\partial\left[\frac{\nu(S)}{\nu(Cs)}\right]}{\partial t} = -\frac{1}{2\beta}\frac{\partial\beta}{\partial t} - 2.83\frac{1}{\alpha}\frac{\partial\alpha}{\partial t} - \frac{1}{\frac{\mu_{Cs}}{\mu_B}} \frac{\partial\left(\frac{\mu_{Cs}}{\mu_B}\right)}{\partial t} \cong -\frac{1}{2\beta}\frac{\partial\beta}{\partial t}$$

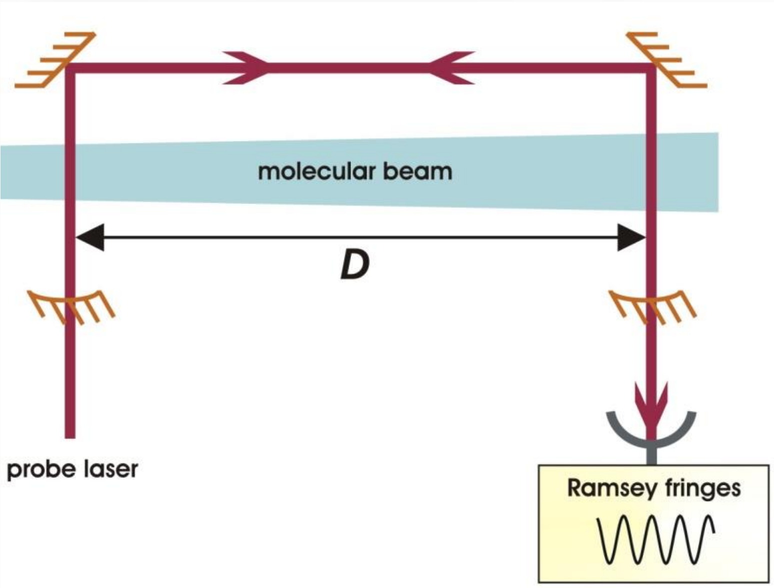
→ **increase the interrogation time *T* by adopting a (two-photon) Ramsey-fringes scheme**

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

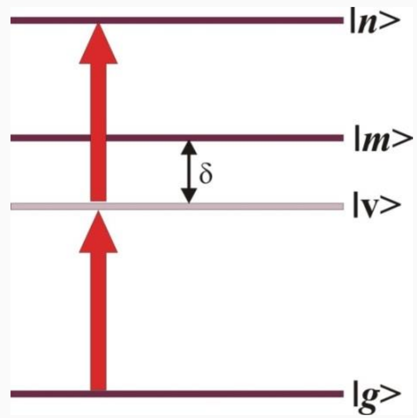
$$\frac{\dot{\beta}}{\beta} = (-3.8 \pm 5.6) \cdot 10^{-14}\ \text{yr}^{-1}$$

Conventional supersonic beam of SF_6 @ 10 μm

OUR PROPOSAL: KEY CONCEPTS



N.F. Ramsey, Phys. Rev. A **78**, 695 (1950)



$$\mathcal{B}(\omega) = \tau^2 \frac{|\mu_{nm}|^2 |\mu_{mg}|^2 |E|^4}{\hbar^4 (\omega_{mg} - \omega)^2}$$

$$\mathcal{A} = 1 + e^{-2\gamma T} \qquad \mathcal{C} = 2e^{-2\gamma T}$$

$$\mathcal{P} = \frac{1}{2T} = \frac{\mathrm{v}}{2D}$$

$$P_{g\to n}(\omega,\mathrm{v}) \cong \mathcal{B}(\omega)\left\{\mathcal{A} + \mathcal{C}\cos\left(\frac{\omega - \frac{\omega_{ng}}{2}}{\mathcal{P}}\right)\right\}$$

Decrease fringe period

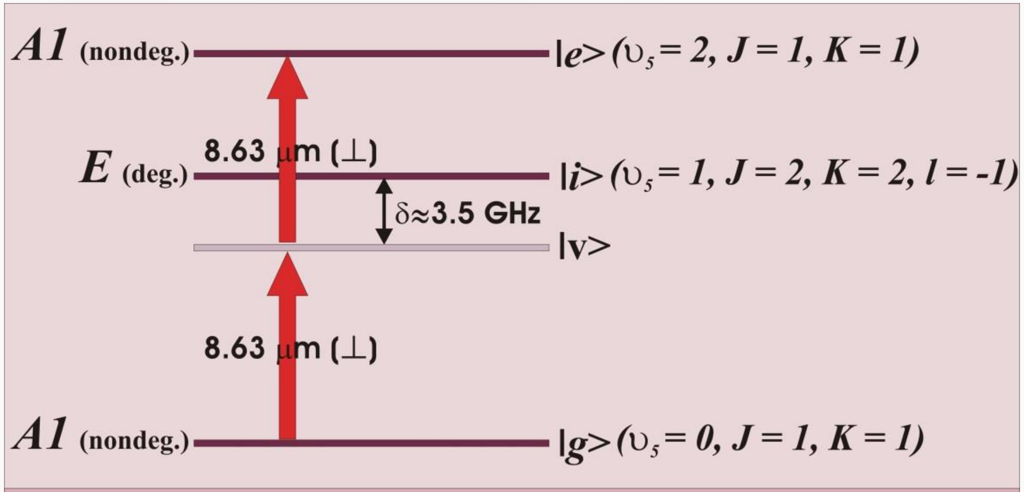
Increase signal amplitude

$$N_{JK} \sim e^{-\frac{E_{JK}}{k_B T_S}} \qquad (\Delta\mathrm{v})^{-1} \sim 1/\sqrt{T_s}$$

slow (decelerated) molecules

cold molecules

THE PROTAGONIST: CHF_3 MOLECULE



$-|J| \leq K \leq |J|$ $\quad$ l : $+\upsilon_5, \upsilon_5 - 2, \upsilon_5 - 4, \ldots, -\upsilon_5$ (only for deg. vib. levels)

⊥ = vibrational transition moment perpendicular to top axis
Selection rules :

$$\Delta K = K_i - K_f = \pm 1, \Delta J = 0, \pm 1$$

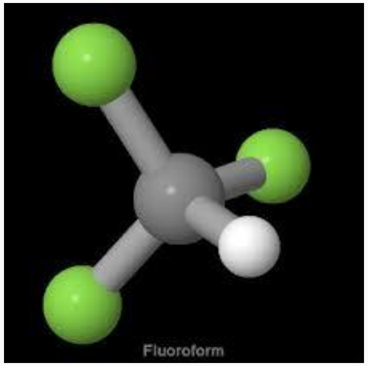
$A1 \longrightarrow E$:
$+l$ with $\Delta K = +1$; $-l$ with $\Delta K = -1$

$E \longrightarrow A1$:
$+l$ with $\Delta K = -1$; $-l$ with $\Delta K = +1$

$$E(J,K) = BJ(J+1) + (C-B)K^2 - D_J J^2 (J+1)^2 - D_{JK} J(J+1)K^2 - D_K K^4 - 2\xi K l$$

$[B = 10.425\text{ GHz}, C = 5.716\text{ GHz}]$

- ✓ **Strong fundamental ro-vibrational band at 8.63 micron (Band Intensity = 550 km mol^{-1}) → QCLs and OFCSs available**



- ✓ **Relatively high dipole moment (1.67 Debye) → manipulation with electric fields**
- ✓ **Fundamental ro-vibrational band at 3.30 micron (Band Intensity: 30 km mol^{-1}) → high power OPOs available in Naples → dipole force potential**
  S. Kuma et al., New J. Phys. **11**, 055023 (2009)
- ✓ **Favourable two-photon transition**

$$R^{(2)}_{g\to e} \propto \left(\frac{I_{laser}}{\delta}\right)^2 |\mu_{ei}|^2 |\mu_{ig}|^2$$

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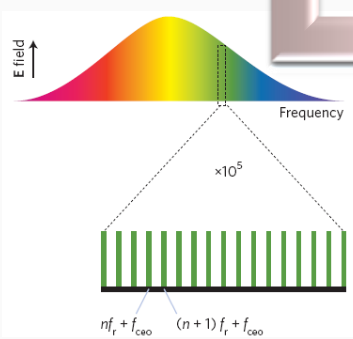
THE INGREDIENT LADDER

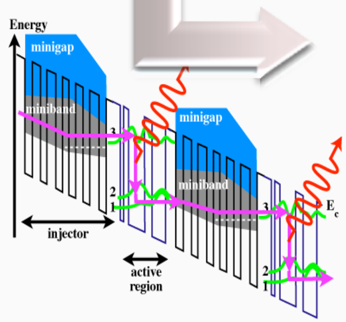
Optical fiber link

- In collaboration with INRIM (F. Levi) and INFN (N. Poli, G.M. Tino)

MIR optical frequency comb

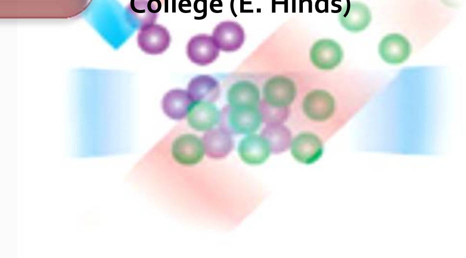


Quantum cascade laser spectrometer



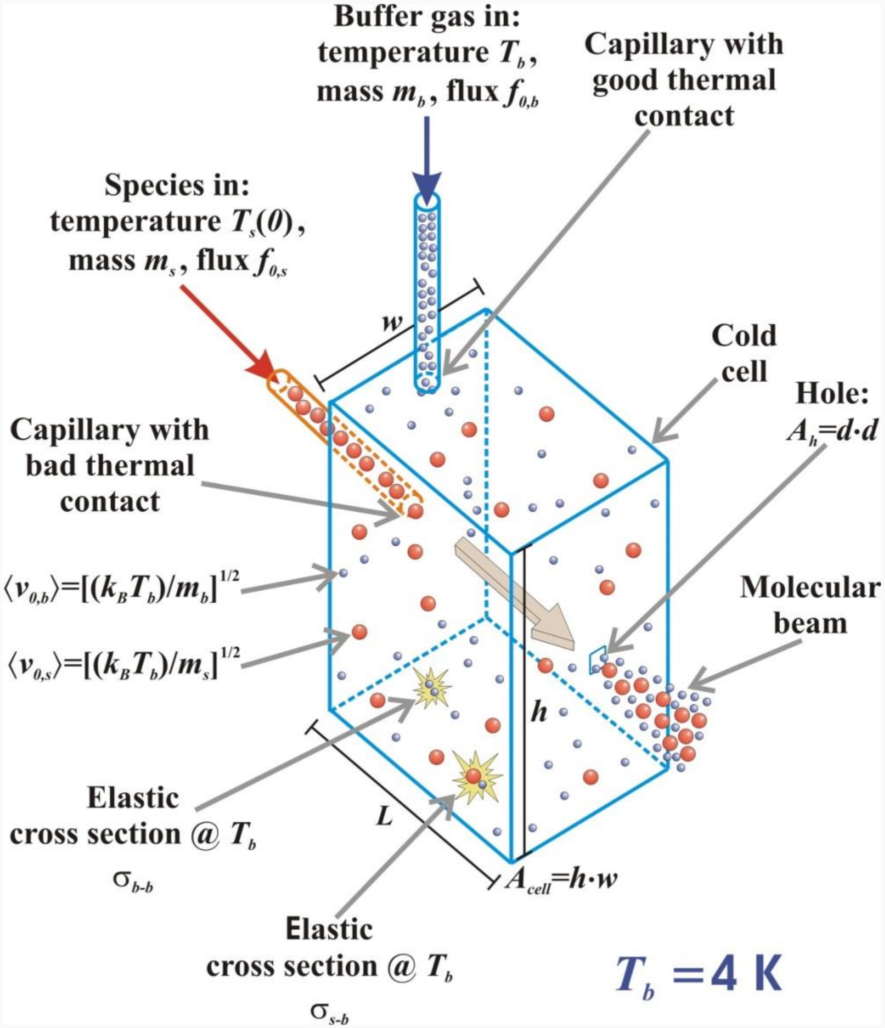
Beam of cold and slow molecules

- In collaboration with Max Planck Institut (G. Santambrogio) and Imperial College (E. Hinds)



$$\frac{1}{\beta}\frac{\partial \beta}{\partial t} = \frac{-2}{\frac{\nu(CF_3H)}{\nu(Cs)}} \frac{\partial \left[\frac{\nu(CF_3H)}{\nu(Cs)}\right]}{\partial t}$$

BUFFER GAS COOLING

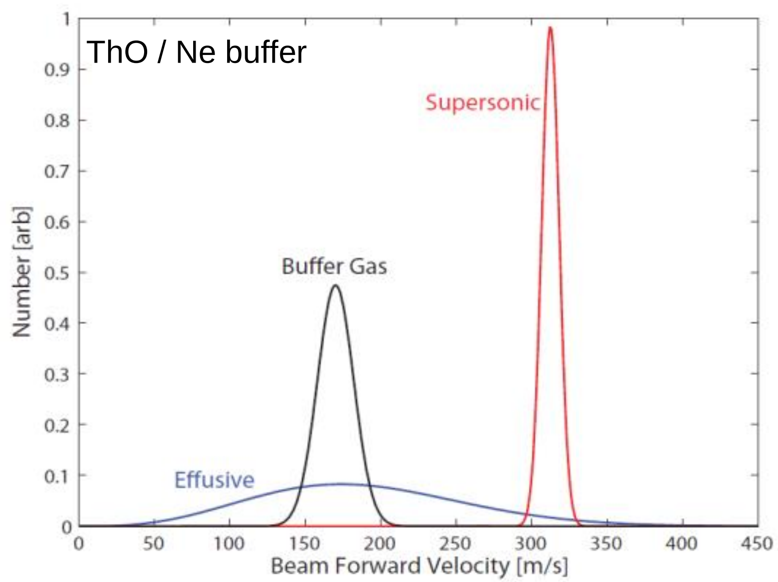


S.E. Maxwell et al., Phys. Rev. Lett. **95**, 173201 (2005)

N.R. Hutzler et al., Chem. Rev. **112**, 4803 (2012)

Both translational and rotational degrees of freedom of the desired molecular species are cooled via elastic collisions with a thermal bath of helium in a cryogenic cell

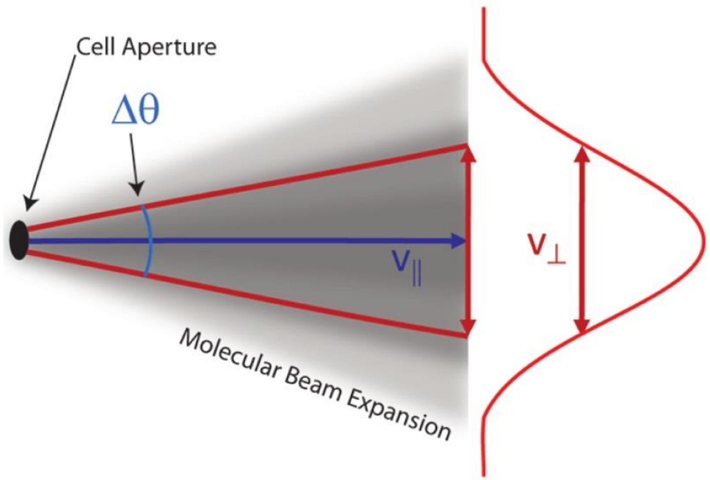
BEAM PARAMETERS



$$Re = \frac{F_{inert}}{F_{viscous}} \approx \frac{8\sqrt{2} f_{0,b} \sigma_{b-b}}{\langle v_{0,b} \rangle d}$$

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

Reduced mean forward velocity (MFV)
and temperature (i.e. spread around MFV)



Buffer-Gas-Cell parameters for fluoroform (CHF_3)		
$f_{0,b} = 1$ SCCM	$d = 2$ mm	$\boldsymbol{Re = 5}$
$f_{0,s} < 1$ SCCM	$h = w = 44$ mm	$\boldsymbol{\gamma \approx 0.1}$
$T_s(0) = 298$ K	$L = 20$ mm	$\boldsymbol{v_{\parallel,s} = 69}$ **m/s**
$N_{coll} \approx 75$	$\lambda_{s-b} = 0.08$ mm	$\boldsymbol{\Delta v_{\perp,s} = \Delta v_{\parallel,s} = 53}$ **m/s**
$\sigma_{b-b} \approx 3 \cdot 10^{-19}$ m^2	$\langle v_{0,b} \rangle = 150$ m/s	$\boldsymbol{\Delta\theta = 21^{\circ}}$
$\sigma_{s-b} \approx 10^{-18}$ m^2	$\langle v_{0,s} \rangle = 36$ m/s	$\boldsymbol{\eta_{extr} = 1\%}$

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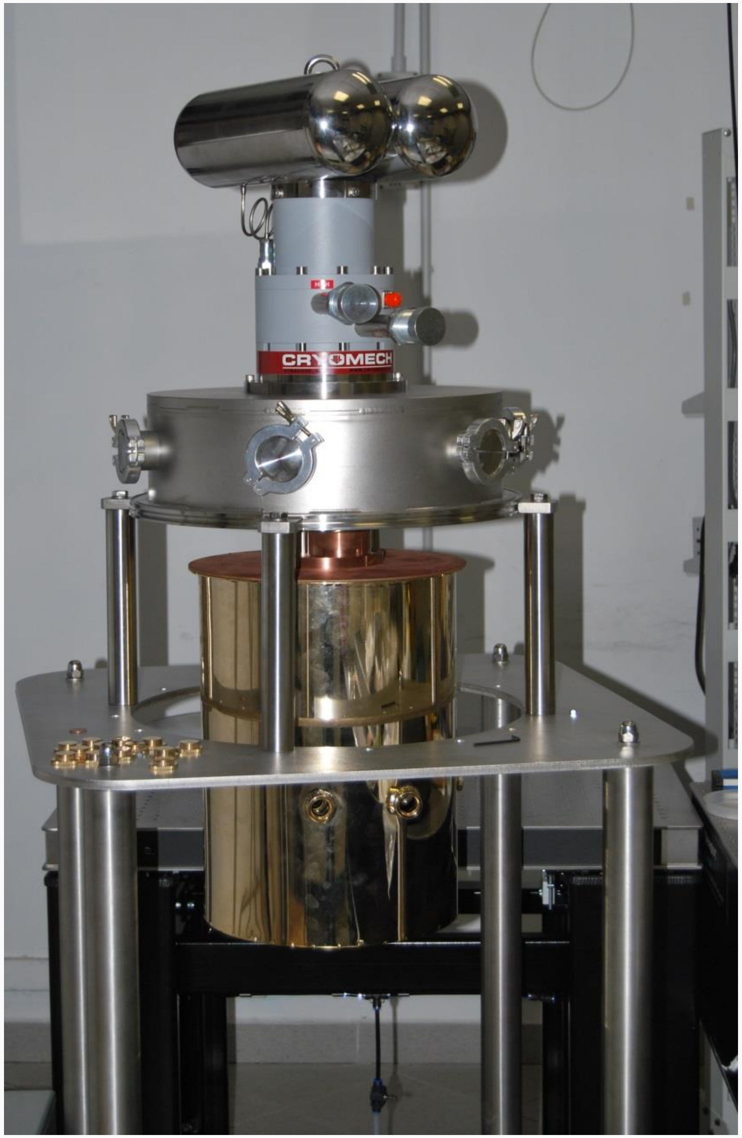
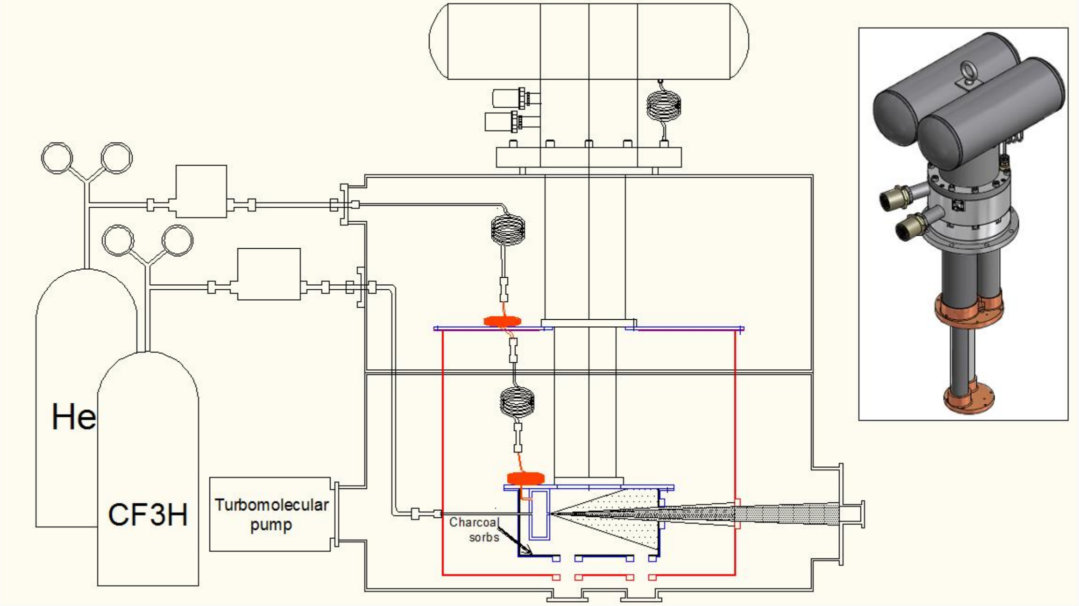
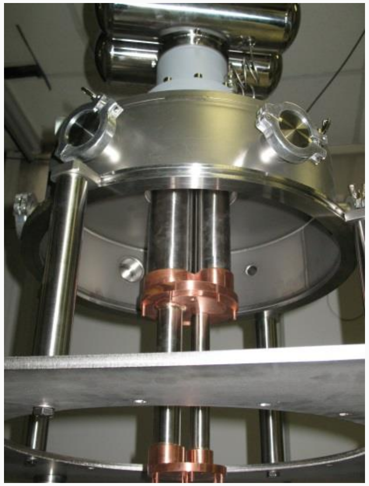
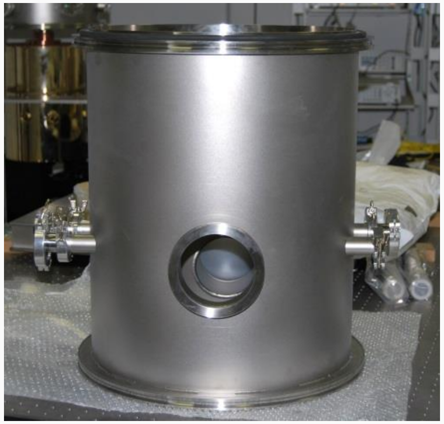
BUFFER-GAS-COOLING SETUP









TRAVELING-WAVE STARK DECELERATOR

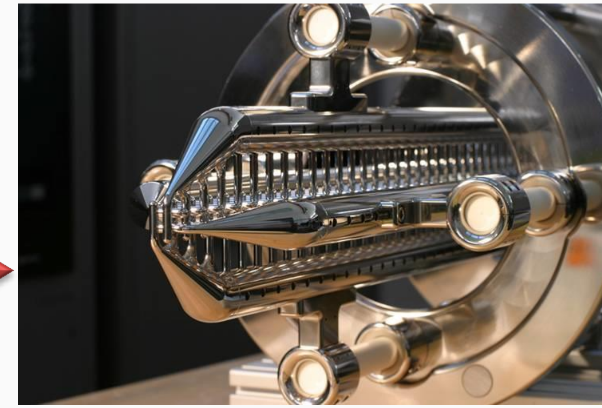
Hexapole lens

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

$$F_r = -k_{el} r$$

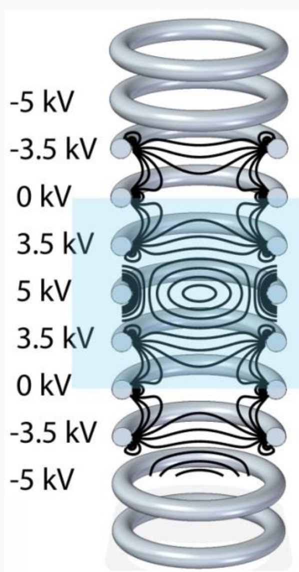
Stark effect

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



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

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



The frequency of the applied 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.

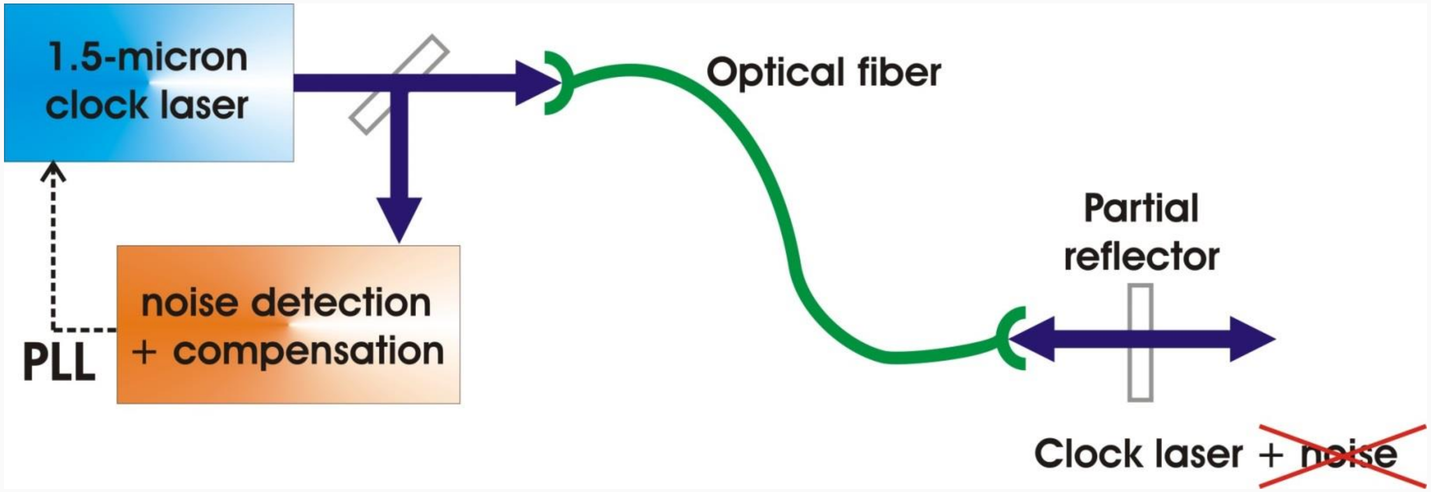
$$V_n(t) = V_0 \sin\left[-\phi(t) + \frac{2\pi n}{8}\right]$$

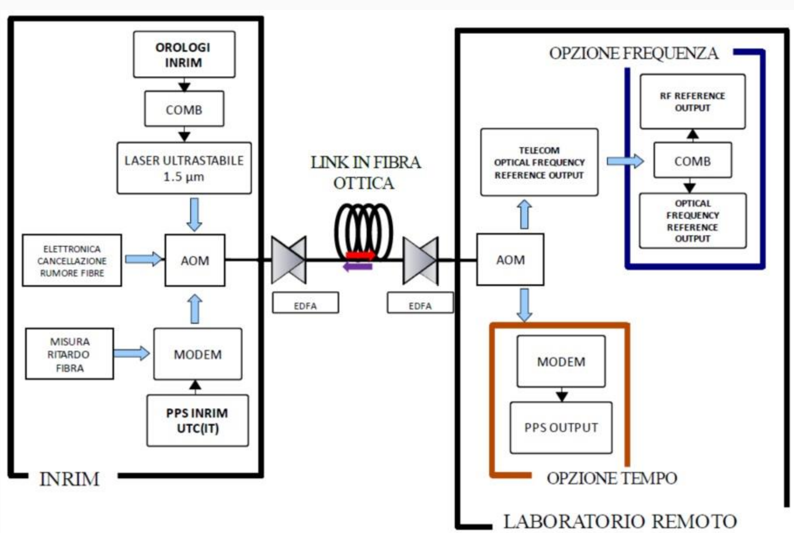
$$\mathrm{v}_z(t) = f(t)L = \frac{1}{2\pi}\frac{d\phi}{dt}L$$

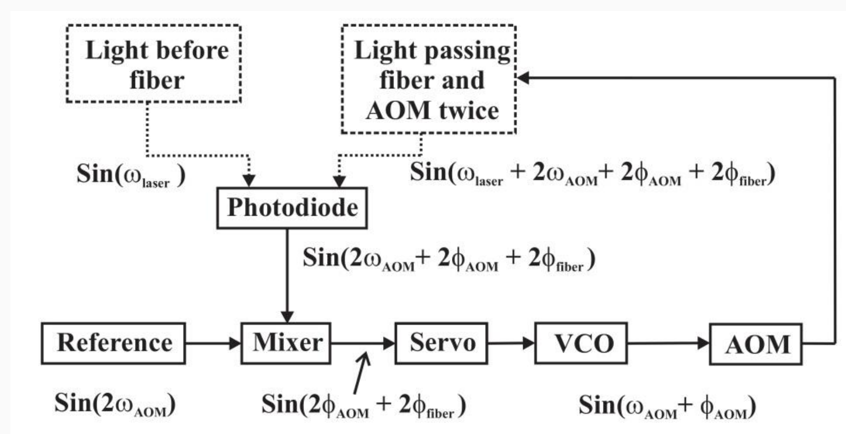
A. Osterwalder et al., Phys. Rev. A **81**, 051401(R) (2010)

THE NATIONAL OPTICAL FIBER LINK

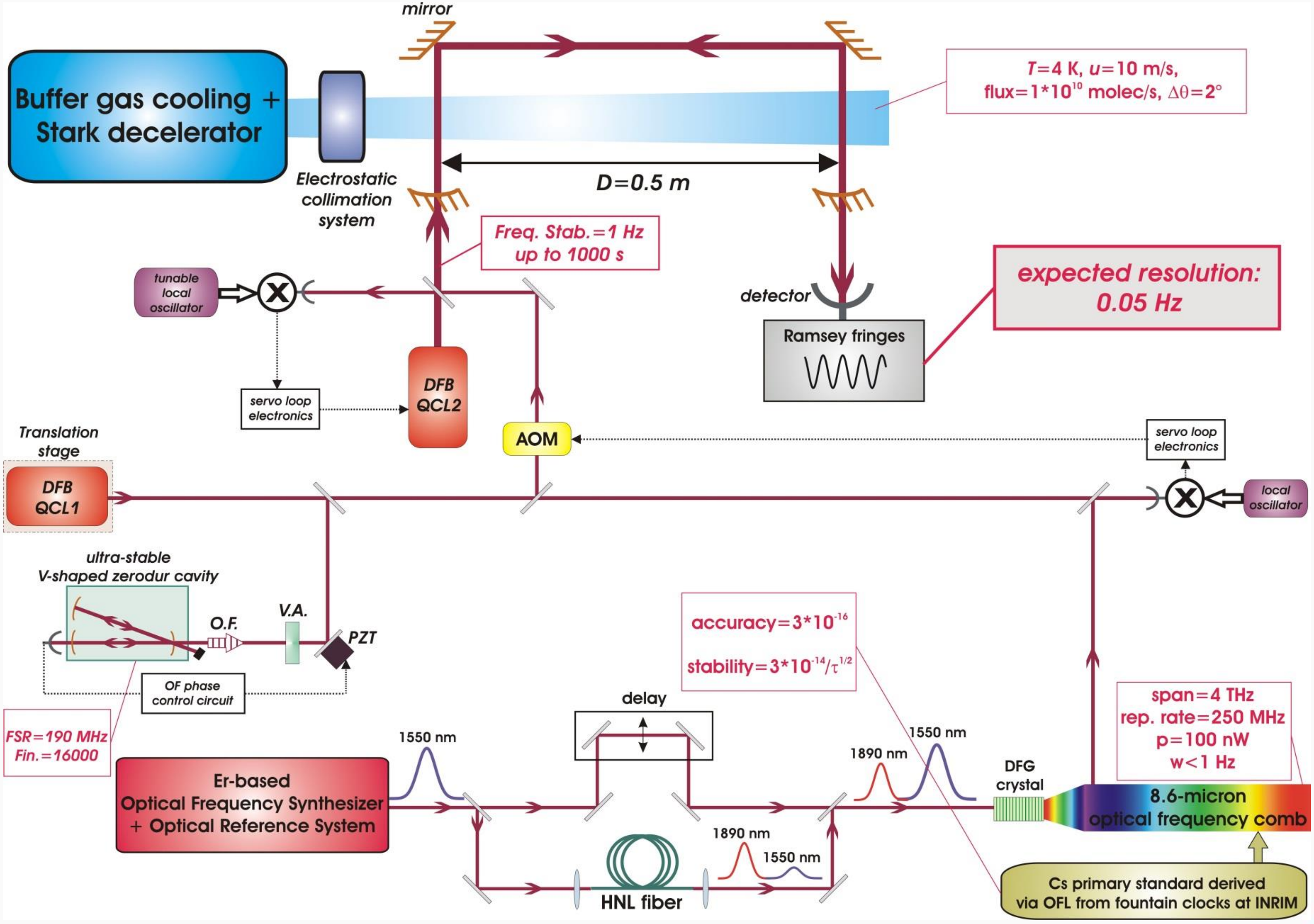
L.-S. Ma et al., Opt. Lett. **19**, 1777 (1994)







WHOLE EXPERIMENTAL APPARATUS



MAIN RESEARCH OUTCOME

- Possible validation of modern multi-D theories beyond the SM towards a GUT (Lie groups)

While the observation of the new particles predicted by candidate GUTs is far beyond the reach of currently foreseen collision experiments, other predictions, like *time variation of fundamental constants* or *the existence of EDMs of elementary particles* might be successfully addressed with precision spectroscopy of cold molecular samples

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

- Testing ToE-aspirant theoretical frameworks (String/M-theories, Loop Quantum Gravity, …) beyond SM and GR

C. Kiefer, Lect. Notes Phys. **648**, 115 (2004)

Pushing the ultimate resolution (accuracy) 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 (clock)

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PERSPECTIVES AND HORIZONS

Spectroscopic tests of fundamental simmetries	…towards and "beyond" quantum degeneracy
• **Spin-statistic relation (with three identical atoms)** G. Modugno et al., Phys. Rev. A **62**, 022115 (2000) • **Parity violation in chiral molecules** B. Darquie et al., Chirality **22**, 870 (2010) • **Electron EDM measurements** J.J. Hudson et al., Nature **473**, 493 (2011)	• **Opto-electrical cooling** M. Zeppenfeld et al., Nature **491**, 570 (2012) **or Cavity-assisted laser cooling** B.L. Lev et al., Phys. Rev. A **77**, 023402 (2008) • **Evaporative** B.K. Stuhl et al., Nature **492**, 396 (2012) **/ Symphatetic cooling** S.K Tokunaga et al., Eur. Phys. J. D **65**, 141 (2011) • **Study of electric dipole-dipole interaction in optical lattices: Quantum Computation and Exotic Quantum States** H.P. Büchler et al., Nature Physics **3**, 726 (2007)

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TIME SCHEDULE AND TOTAL BUDGET

3° year	• Integration of the molecular machine with the comb-referenced QCL interrogation source, construction of the Ramsey cavity, and first spectroscopic measurements	150 k€
2° year	• Generation/characterization of the SD beam • Implementation of the master-slave QCL source and linking to the MIR comb • Generation of the MIR comb	190 k€
1° year	• Realization/characterization of the BGC beam + hexapole lens • Completion of the National optical fiber link • Stabilization of the master QCL against the zerodur cavity	205 k€

PROPOSED ACRONYM

Sounding the time Unwinding of the PRoton-to-Electron Mass ratiO with COld Stable MOlecules

S.U.PR.E.M.O (CO.S.MO)

Thank you for your attention

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EC/EN6, EC/EN7

ANNO	missioni	consumo	inventario	TOTALE
2014	5 k€	95 k€	105 k€	205 k€
2015	7 k€	43 k€	140 k€	190 k€
2016	10 k€	55 k€	85 k€	150 k€
totali	22 k€	193 k€	330 k€	**545 k€**

Name	FTE
Maddaloni Pasquale (Resp. Naz.)	0,4
Pablo Cancio Pastor	0,4
Davide Mazzotti	0,4
Saverio Bartalini	0,4
Marco Barucci	0,3
Paolo De Natale (Resp. Loc.)	0,1
Massimo Inguscio	0,1