Molecular Spectroscopy Experiments for Direct Detection of Dark Matter

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• Outline

1) New physical phenomena from wavelike dark matter

2) Dark matter detection by laser spectroscopy of molecular transitions

3) Potential from quantum-enhanced spectroscopy

4) Conclusion

1) New physical phenomena from wavelike dark matter

• Standard Model and Dark Matter

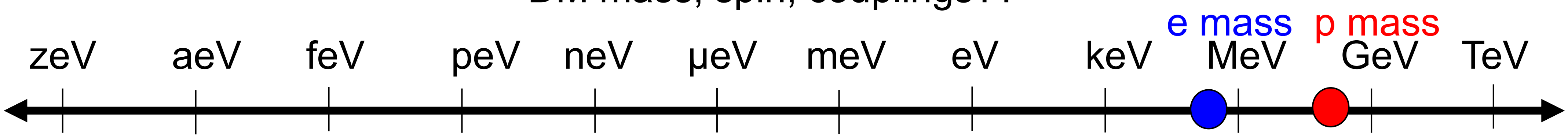
- The Standard Model (SM) and the General Relativity (GR) theories

=> fundamental constants (FC) are free parameters of the theory

Uzan, C. R. Physique 16, 576 (2015)

=> Failure of SM&GR to explain observational data on mass distribution in the Universe

DM mass, spin, couplings??



- BSM theories: DM is sub-eV bosonic field $\phi(t) = \phi_0 \cos\left(\omega_\phi t\right)$; $\omega_\phi \cong \frac{m_\phi c^2}{\hbar}$; $\phi_0 = \sqrt{\frac{4\pi G \rho_{DM}}{\omega_\phi^2 c^2}}$

Arvanitaki *et al*, PRD 91, 015015 (2015)

=> Introducing galactic DM : $\rho_{DM} = 0.4\ \mathrm{GeV/cm^{-3}}$; $v \cong 230\ \frac{\mathrm{km}}{\mathrm{s}}$; $\sigma_v \cong 10^{-3}c$; $\tau_c = 10^{-4}/\omega_\phi$

- UDM coupling to the Standard Model bring new physical phenomena:

=>precession of nuclear or electron spins

=>currents in electromagnetic systems

=>photon generation

=>induce accelerations of matter

=>variations of fundamental constants

• Variations of fundamental constants and AMO physics

- fermion X=(e,u,d,s) masses : $m_X(\phi) = m_X\left(1 + d_{m_X}\cdot\phi/\sqrt{c\hbar/8\pi G}\right)$

- fine structure constant : $\alpha(\phi) = \alpha\left(1 + d_\alpha\cdot\phi/\sqrt{c\hbar/8\pi G}\right)$

- QCD scale parameter : $\Lambda_{QCD}(\phi) = \Lambda_{QCD}\left(1 + d_g\cdot\phi/\sqrt{c\hbar/8\pi G}\right)$

- $\mu(m_e, \Lambda_{QCD}, m_X) = m_p/m_e$: $\mu(\phi) = \mu\left(1 - \left(d_{m_e} + d_g + 0.036 d_{\hat{m}}\right)\cdot\phi/\sqrt{c\hbar/8\pi G}\right)$

Damour and Donoghue, PRD 82, 084033 (2010)

=> Atomic, molecular and optical cavity resonance frequencies depend on FC:

- atomic optical transition $f_{A,opt} = C_{A,opt}\dfrac{\alpha^2 m_e c^2}{4\pi\hbar}\cdot F_{opt}(\alpha)$
- atomic hyperfine transition $f_{A,HF} = C_{A,HF}\dfrac{\alpha^2 m_e c^2}{4\pi\hbar}\left(g_i\left(m_q/\Lambda_{QCD}\right)\times m_e/m_p\right)\cdot F_{HF}(\alpha)$
- vibrational molecular transition : $f_{vib} = C_{vib}\dfrac{\alpha^2 m_e c^2}{4\pi\hbar}\sqrt{m_e/m_p}$
- rotational molecular transition : $f_{rot} = C_{rot}\dfrac{\alpha^2 m_e c^2}{4\pi\hbar} m_e/m_p$
- optical cavity resonance : $f_C = C_C\dfrac{\alpha m_e c^2}{\hbar}\cdot F_C\left(\alpha, m_X, \Lambda_{QCD}\right)$

=> Parametrization in terms of $\left(\alpha, \mu, \hat{m}/\Lambda_{QCD}\right)$ with appropriate sensitivity coefficients:

$$\frac{\Delta f_{C,A,M}}{f_{C,A,M}} = Q_\alpha^{C,A,M}\frac{\Delta\alpha}{\alpha} + Q_\mu^{C,A,M}\frac{\Delta\mu}{\mu} + Q_q^{C,A,M}\frac{\Delta\left(\hat{m}/\Lambda_{QCD}\right)}{\hat{m}/\Lambda_{QCD}}$$

• Searches for UDM and quantum sensing

- **Snowmass 2021 UDM whitepaper**

Antypas *et al*, arXiv 2203.14915

- **Approaches**

=> Atomic clock comparisons

Hees *et al*, PRL 117, 061301 (2016)

Kennedy *et al*, PRL 125, 201302 (2020)

=> Molecular spectroscopy

Oswald *et al*, PRL 129, 031302 (2022)

Constantin, Proc. IFCS-EFTF 2023

- **Quantum-enhancement**

-entanglement

Colombo *et al*, APL 121, 210502 (2022)

Shi *et al*, PRL 125, 180502 (2020)

-squeezing

Schulte et al, Nat Commun 11, 5955 (2020)

Heng *et al*, Appl. Sci. 15, 10179 (2025)

=>enhanced GW-detection

Tse *et al*, PRL 123, 231107 (2019)

- **Networks for quantum sensing**

-optical clock networks

Riehle, Nature Photon. 11, 25 (2017)

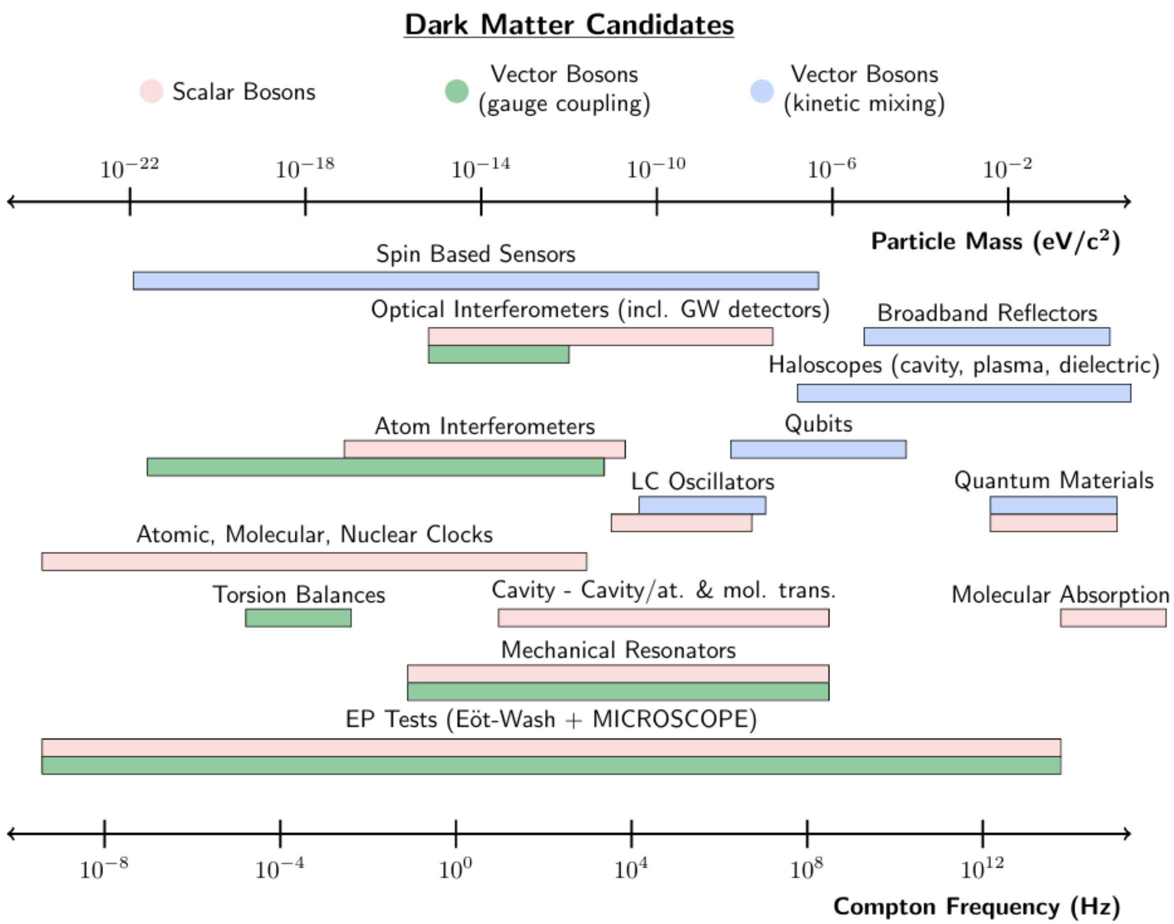
-entangled atomic clocks

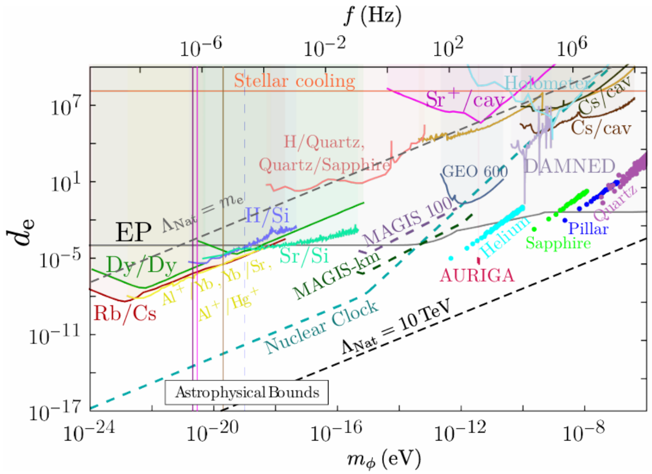
Komar *et al*, Nat. Phys. 10, 582 (2014)

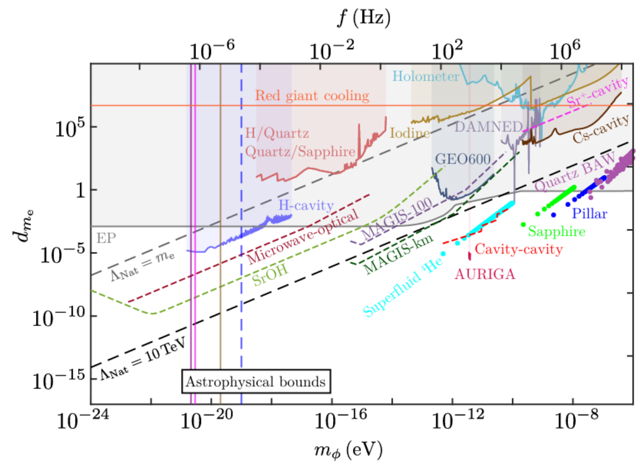
Polzik and Ye, PRA 93, 021404(R) (2016)

-CV entanglement in optical networks

Guo *et al*, Nat. Phys. 16, 281–284 (2020)







2) Dark matter detection by laser spectroscopy of molecular transitions

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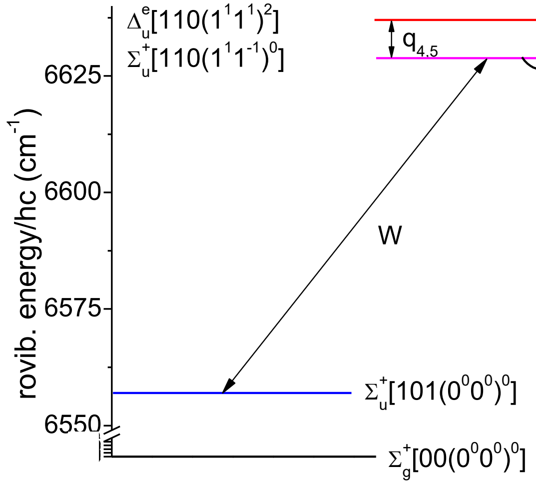
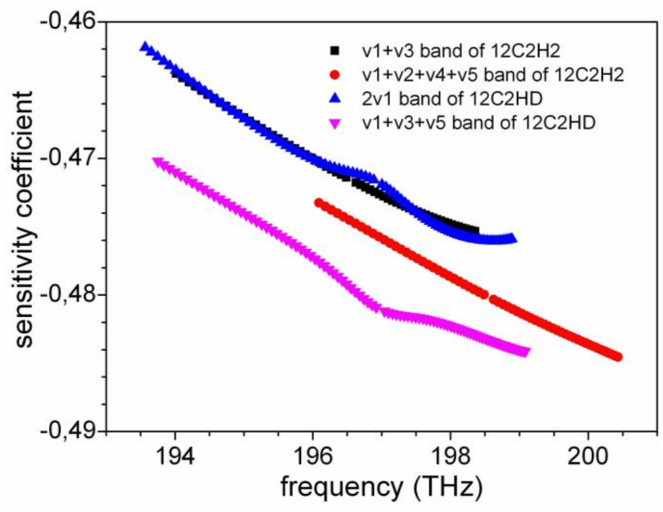
Acetylene rovibrational transitions at 1.5 µm

- Effective Hamiltonian models for rovibrational energy levels

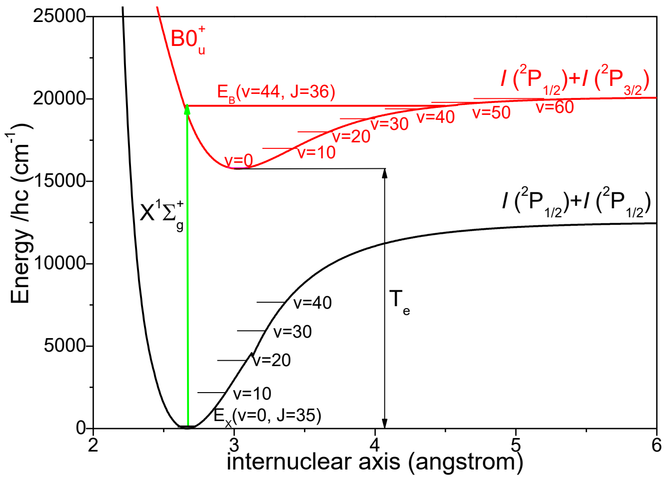


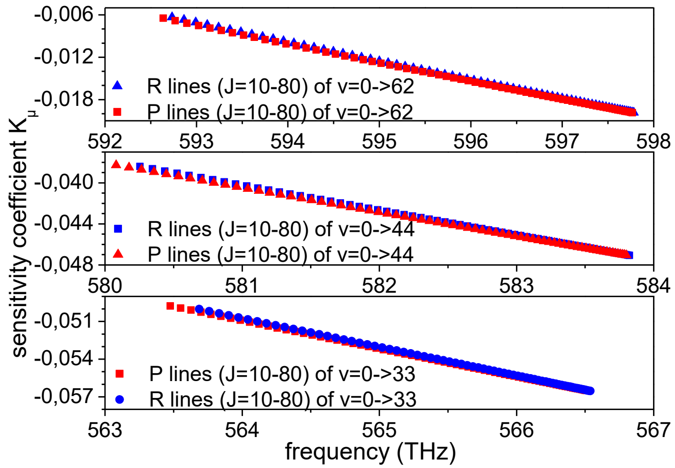


Constantin, Vibrational Spectr. 85, 228 (2016)

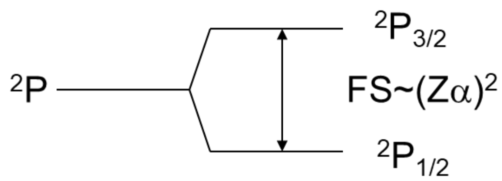
Molecular iodine rovibronic transitions at 0.5 µm

- Dunham expansion of energy levels





Sensitivity coefficient $K_\alpha \sim 2$



Constantin, Proc. IFCS, 9234878 (2020)

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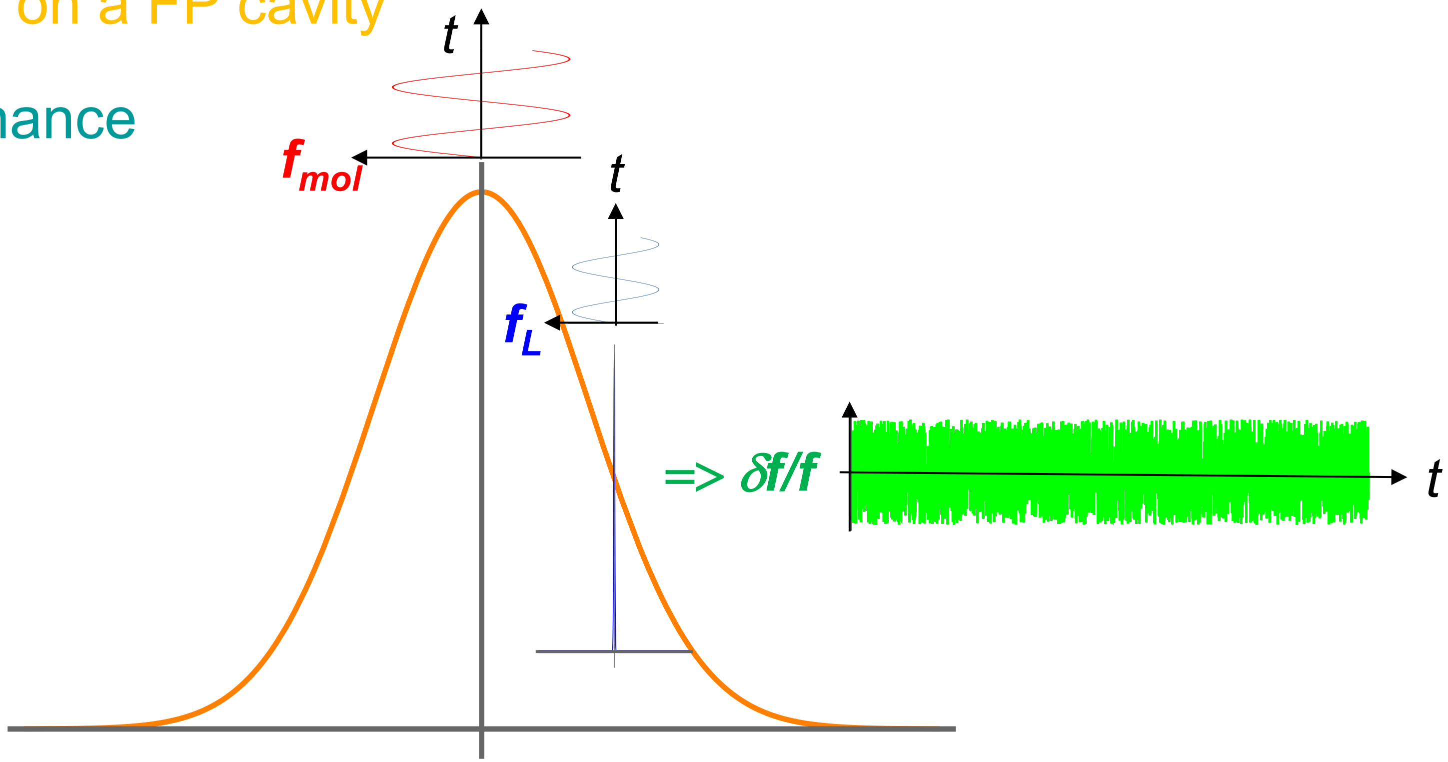
- Laser stabilized on a FP cavity
- Molecular resonance



$$\frac{f_L(t)-f_{mol}(t)}{f_L(t)} = \left(Q_\alpha^L h^L(f_\phi) - Q_\alpha^{mol} h^{mol}(f_\phi)\right)\frac{\Delta\alpha(t)}{\alpha} + \left(Q_\mu^L h^L(f_\phi) - Q_\mu^{mol} h^{mol}(f_\phi)\right)\frac{\Delta\mu(t)}{\mu}$$

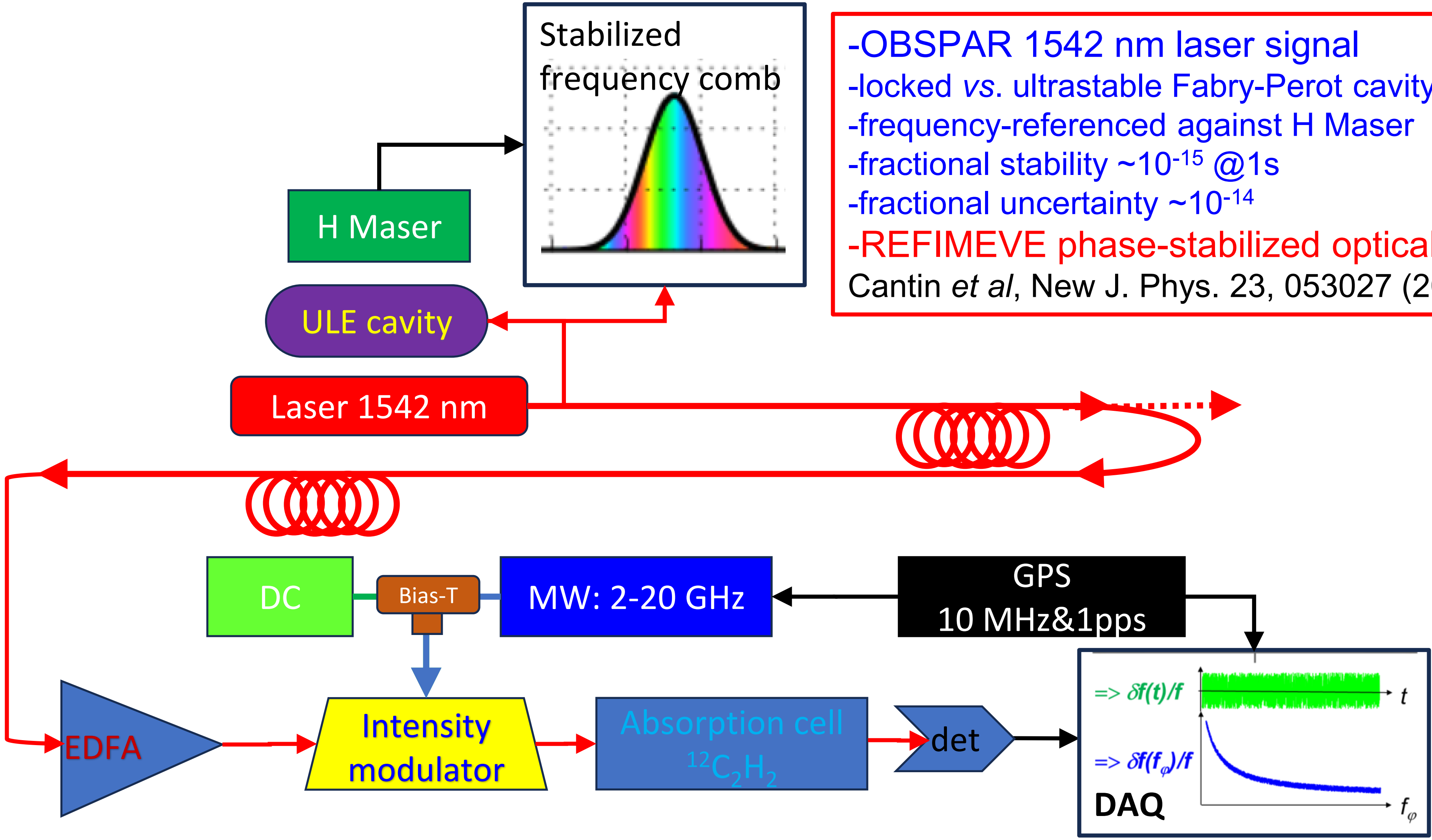
- Sensitivity coefficients: $Q_\alpha^L \cong 1; Q_\mu^L \cong 0;\ Q_\alpha^{mol} = 0 \ldots 2;\ Q_\mu^{mol} = -0.01 \ldots - 0.5$

Pašteka *et al*, PRL 122, 160801 (2019); Constantin, Vibrational Spectr. 85, 228 (2016) & Proc IFCS 2020

- Response functions $h^L,\ h^{mol}$ with high-frequency cutoffs

-frequency cutoffs : linewidth of the molecular line, delay in propagation of sound in the ULE spacer, …

• Probing C_2H_2 absorption by optical network spectroscopy



-OBSPAR 1542 nm laser signal
-locked *vs.* ultrastable Fabry-Perot cavity
-frequency-referenced against H Maser
-fractional stability ~10^{-15} @1s
-fractional uncertainty ~10^{-14}
-REFIMEVE phase-stabilized optical link
Cantin *et al*, New J. Phys. 23, 053027 (2021)

- EDFA amplifier & fiber-coupled LN intensity modulator (MW+DC)
- Absorption cell with 5 cm pathlength filled with $^{12}C_2H_2$ at 50 Torr
- Photodetector with 1 GHz BW & 14 bits DAQ system sampling up to GSPS
- GPS receiver: timestamps and 10 MHz frequency reference

• First experimental data recording and exploitation

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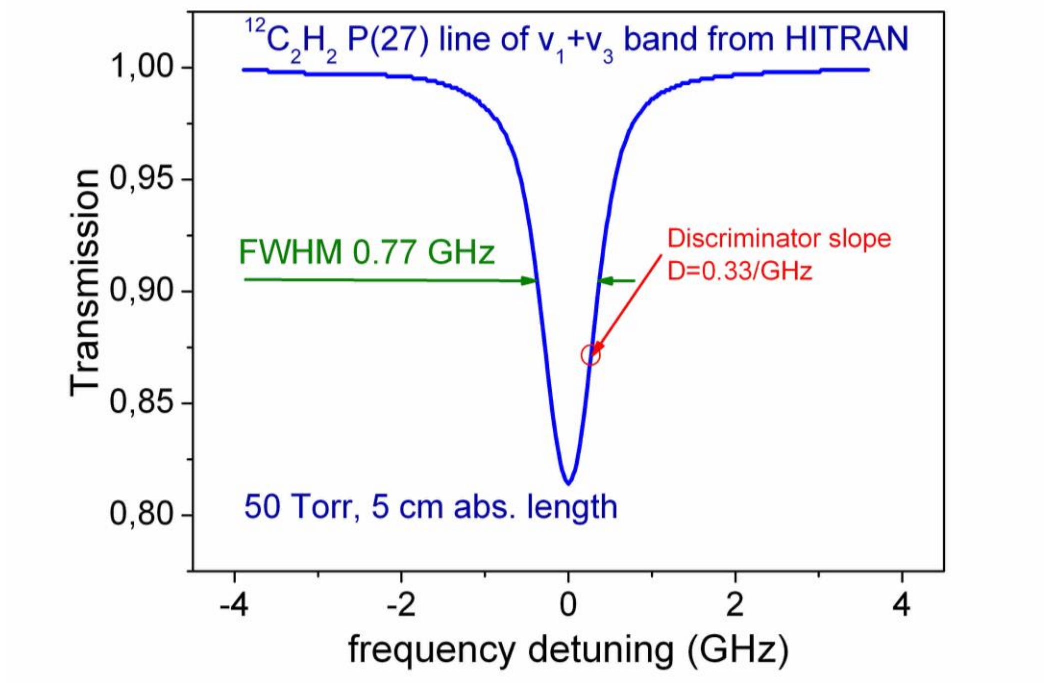
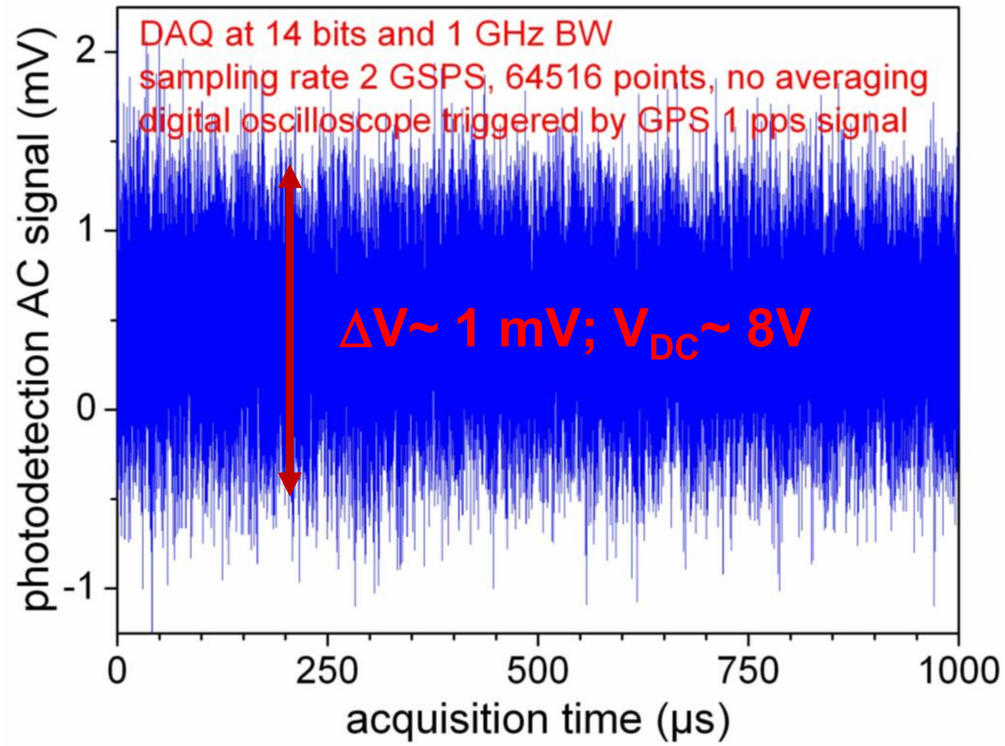




- Simulated experimental transfer function

-conversion of fluctuations Δf <-> ΔV via slope

- Fast data acquisition

-sampling oscilloscope at 2 GSPS

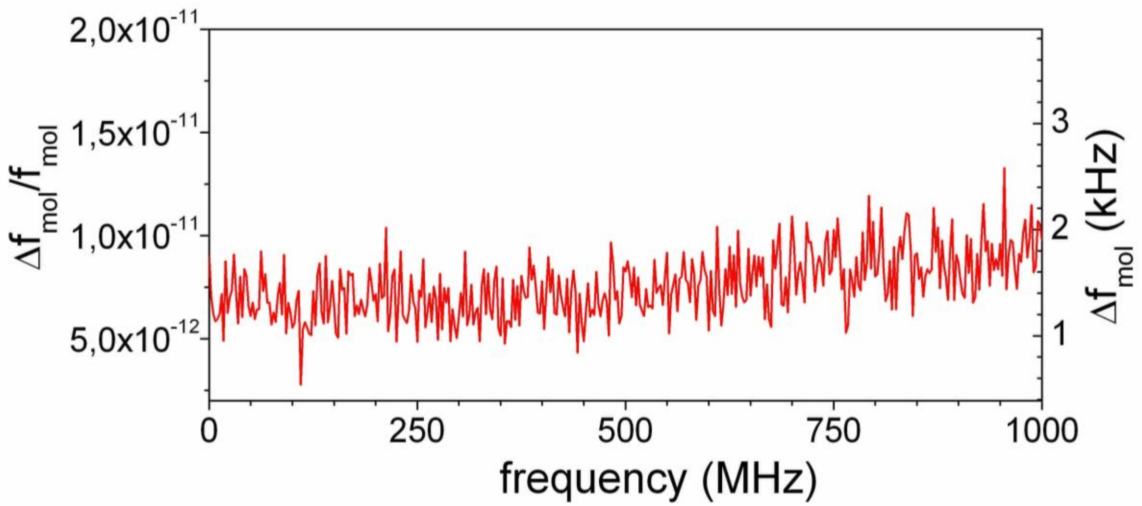
-MW spectrum analyser with BW>1 GHz

- Conversion of the photodetection signal in kHz-level frequency excursions

=> Spectroscopy with 10^{-12} precision, in line with previous estimations

=> Improvement by differential detection

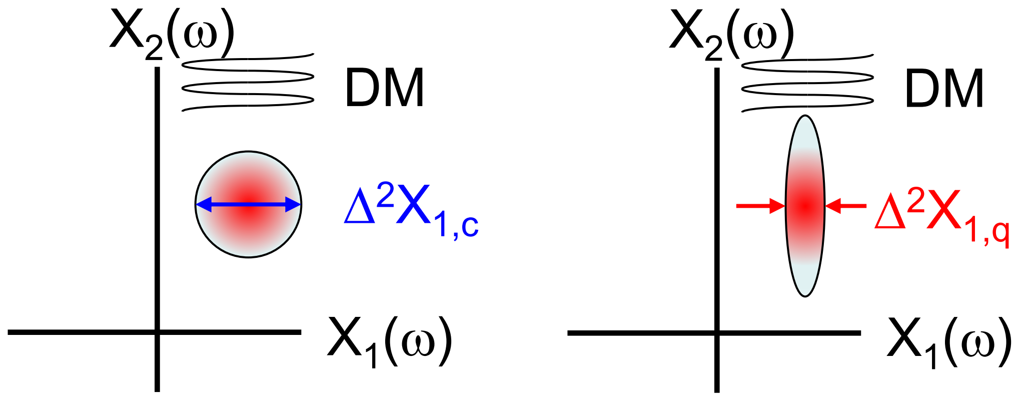
Constantin, Proc. IFCS-EFTF 2023



10 kHz RBW, 400 points, 0.6 s averaging time/point

3) Potential from quantum-enhanced spectroscopy

-Use amplitude squeezed light to push noise out of measurement:

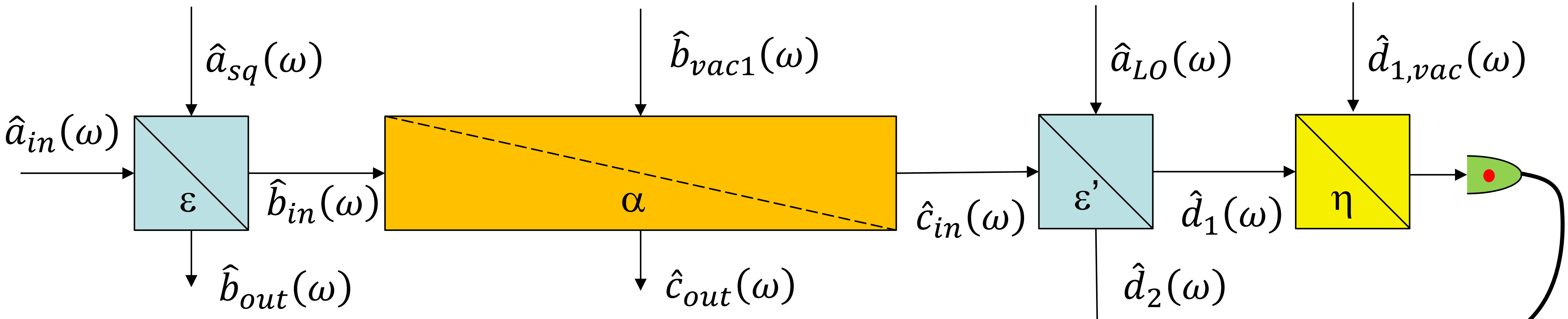


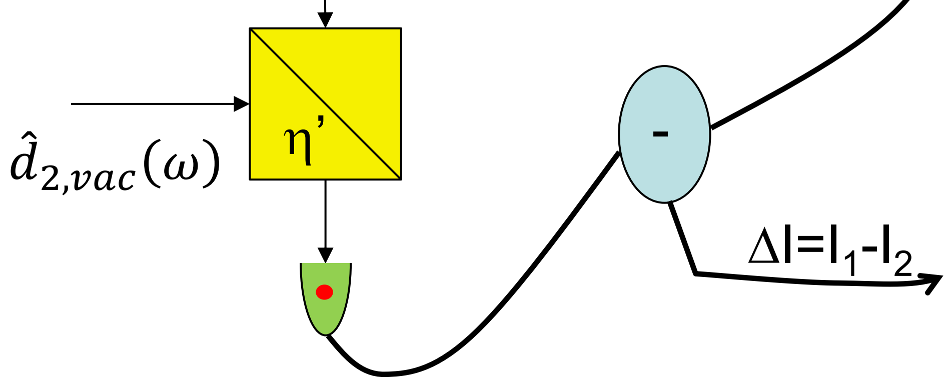
$$X(\theta) = X_1 \cos\theta + X_2 \sin\theta$$

$$Squeezing(dB) = -10\, log_{10}\left(\frac{\Delta^2 X_{1,q}}{\Delta^2 X_{1,c}}\right)$$

=>bright, tunable, cw squeezed radiation generation

=>probing molecular absorption

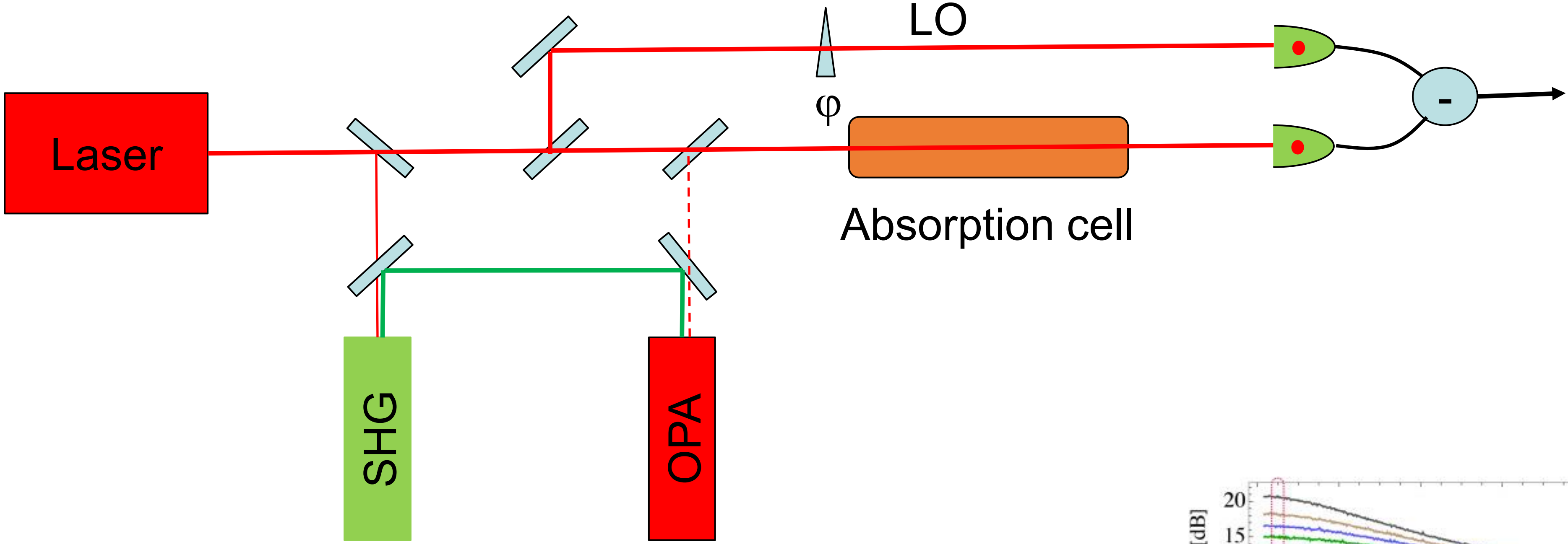


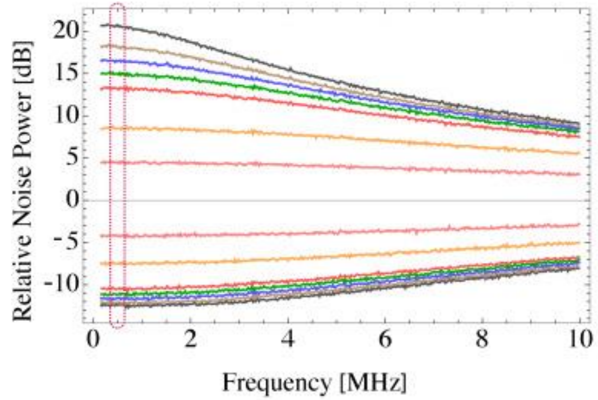


=>BHD amplification and quadrature detection with LO:

$$\Delta I(t) = K\langle \hat{a}_{LO}\rangle X_{1,c_{in}}(t) + \cdots$$

• Instrumentation for spectroscopy with squeezed light





- Cw laser, SHG, sub-thresold pumped OPA => squeezed vacuum
- Superposition with coherent radiation
- Probing molecular absorption
- Balanced homodyne detection

=> Molecular frequency references: C_2H_2 at 1.5 µm and I_2 at 0.5 µm

CCL BIPM report (2007), Riehle *et al*, Metrologia 55, 188 (2018)

=> Strong squeezing at 1550 nm up to 15 dB

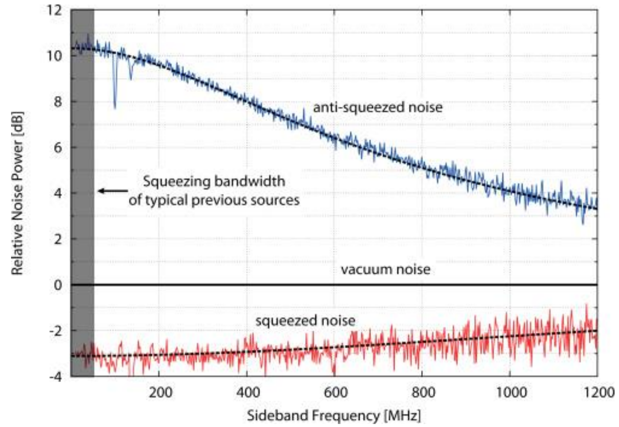
Shajilal *et al*, Opt. Express 30, 37213 (2022)

=>High-bandwidth squeezing at 1550 nm up to GHz-level

Ast *et al*, Opt. Express 21, 13572 (2013)

=>Strong squeezing at 532 nm by upconversion

Baune *et al*, Opt. Express 23, 16035 (2015)



• Constraints from time-synchronized laser spectroscopy

Exp. A : Doppler-free acetylene spectroscopy with $\frac{\delta f}{f} = 5 \times 10^{-13}/\sqrt{\tau}$

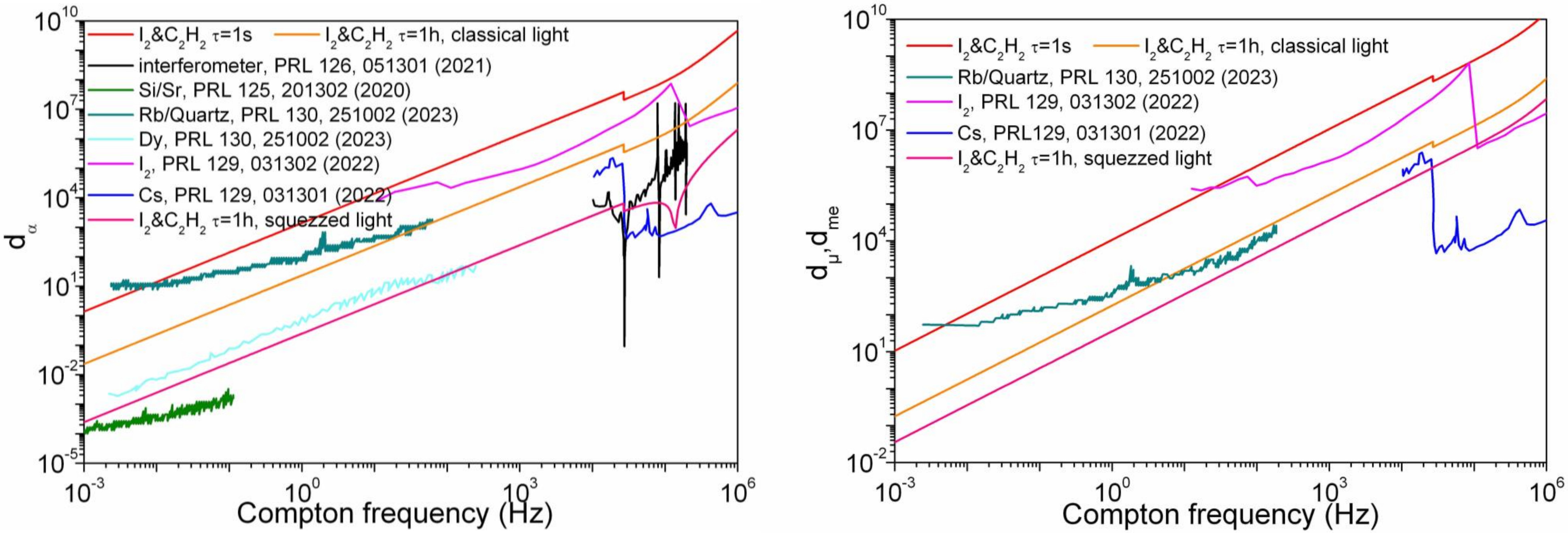
Hald *et al,* Opt. Express 19, 2052 (2011)

Exp. B : Doppler-free molecular iodine spectroscopy with $\frac{\delta f}{f} = 3 \times 10^{-14}/\sqrt{\tau}$

Barbarat *et al*, Proc. SPIE 11180, 111800T (2018)

=> Improved spectroscopy with squeezed light: $\left(\frac{\delta f}{f}\right)_{C_2H_2} = 3 \times 10^{-14}/\sqrt{\tau}$ and $\left(\frac{\delta f}{f}\right)_{I_2} = 8 \times 10^{-15}/\sqrt{\tau}$

(in case of negligible losses)



=> order-of-magnitude sensitivity improvement by using spectroscopy with squeezed light

=> constraints on α- and μ-variations conjointly

=> address Compton frequency domain complementary to that explored with atomic clocks

=> Improved constraints in the kHz range, useful probe for DM couplings to the nuclear interaction

➢ Sensitivity to μ- and α-variation for C_2H_2 and I_2 reference transitions

➢ Laser spectroscopy experimental setup at 194 THz with precision at 10^{-12} level that exploits the REFIMEVE network signal

- Compact and robust molecular spectrometer
- Fiber-coupled components enabling easy integration into optical fiber networks

➢ Proposal for enhanced spectroscopy with squeezed light

➢ Evaluation of constraints from precision spectroscopy measurements

Thank you for your attention !

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FIRST TF

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Refimeve+ — Réseau fibré métrologique à vocation européenne