



# Limits on oscillating fundamental constants from laser spectroscopy of molecular ensembles

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# DM and oscillating fundamental constants

- DM may consist of light bosons. These form a classical field  $\Phi$ , which coherently oscillates at their Compton frequency  $f_C = m_\Phi c^2/h$   $\langle \Phi \rangle = \frac{\sqrt{2\rho_{DM}}}{m_\Phi} \cos 2\pi f_C t$
- $\Phi$  may have scalar interactions with the SM fields
- The fundamental constants (FC) may be expectation values of SM fields
- The coupling of  $\Phi$  to SM fields may lead to oscillating fundamental constants

Low-energy effective lagrangian with linear coupling:

$$\mathcal{L}_{\text{eff}} \supset \frac{\phi}{M_{\text{Pl}}} \left( \sum_{f=e,u,d,s} d_{m_f} m_f \bar{f} f + \frac{d_\alpha \alpha}{4} F F + \frac{d_g \beta(g_s)}{2g_s} G G \right)$$

$F$ : electromagnetic field tensor

$G$ : gluon field tensor

$f$ : fermion fields (electron, up-quark, down-quark, strange quark)

$d$ : dimensionless coupling constants to the DM field  $\Phi$

$\alpha$ : fine-structure constant

$\beta$ : beta function, describes the running of the coupling constant with energy

$\alpha_s = g_s^2/4\pi$ , strong force coupling constant; for 3 massless quarks,  $\beta/2g_s = -9\alpha_s/8\pi$

# DM and oscillating fundamental constants

$$m_f(\phi) = m_f \left[ 1 + d_{m_f} \frac{\phi}{M_{\text{Pl}}} \right], \quad \alpha(\phi) \simeq \alpha \left[ 1 - d_\alpha \alpha \frac{\phi}{M_{\text{Pl}}} \right], \quad \alpha_s(\phi) \simeq \alpha_s \left[ 1 - \frac{2d_g \beta(g_s)}{g_s} \frac{\phi}{M_{\text{Pl}}} \right]$$

$$\frac{\partial \ln \Lambda_{\text{QCD}}}{\partial \phi} = -\frac{g_s}{2\beta(g_s)} \frac{\partial \ln \alpha_s}{\partial \phi} = \frac{d_g}{M_{\text{Pl}}} \quad \frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}} = \frac{\delta \Lambda_{\text{QCD}}}{\Lambda_{\text{QCD}}} + 0.093 \frac{\delta \hat{m}}{\hat{m}} + 0.043 \frac{\delta m_s}{m_s},$$
$$\hat{m} = (m_u + m_d)/2$$

- Observables of light scalar DM may come from FC oscillations
- A series of experiments on oscillating FC has already been performed [1]
- Also: Equivalence-Principle (EP)-violating/5th force accelerations searches for non-SM fields. Experiments have already set tight bounds [2]

[1]

For references see: D. Antypas et al.  
Qu. Sci. and Techn. 6, 034001 (2021)  
for a list of references

see also M. Tobar, this workshop

[2]

P. W. Graham, et al., Phys. Rev. D 93, 075029 (2016).  
A. Hees, et al., Phys. Rev. D 98, 064051 (2018).  
G. L. Smith, et al., Phys. Rev. D 61, 022001 (1999).  
S. Schlamminger, et al., Phys. Rev. Lett. 100, 041101 (2008).  
P. Touboul et al., Phys. Rev. Lett. 119, 231101 (2017).  
J. Bergé, et al., Phys. Rev. Lett. 120, 141101 (2018).

# An open search

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Which fundamental constants?

All?

Magnitude?

Period?

*Make an encompassing search*

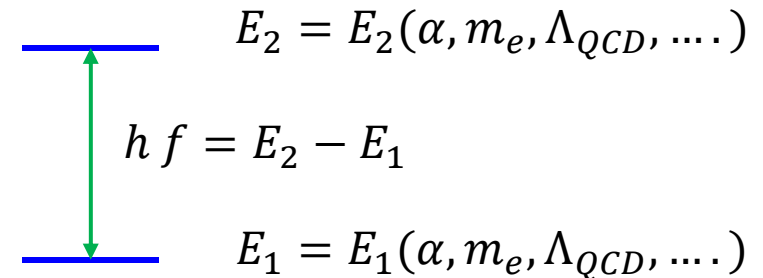
→ All constants

→ All frequencies

# Approach

## Frequency metrology

- Transition frequency  $f$  between internal levels of a quantum system



- Transition frequency between levels of a spin in an external magnetic field
- Mode frequency of an electromagnetic resonator
- Mode frequency of a mechanical resonator

The frequency ratio of two dissimilar oscillators is measured as a function of time

*High quality factor of the transitions/modes lead to:*

*high sensitivity*

*but slow reaction – low bandwidth*



Trade-off

# Some dependencies

- Optical transition frequency

$$f \propto m_e c^2 \alpha^2 H(\alpha)$$

- Hyperfine transition frequency

$$f \propto m_e c^2 \alpha^4 F(\alpha) \left(\frac{m_e}{m_p}\right) \mu_{nuc}$$

- Molecular vibrational transition frequency

$$f \propto m_e c^2 \alpha^2 \left(\frac{m_e}{M_{nuc}}\right)^{\frac{1}{2}} G\left(\frac{m_e}{M_{nuc}}\right)$$

- Electromagnetic cavity mode frequency (empty cavity)

$$f \propto m_e c^2 \alpha$$

- Mechanical mode frequency

$$f \propto m_e c^2 \alpha^2 \left(\frac{m_e}{M_{nuc}}\right)^{\frac{1}{2}}$$

$$\frac{\delta M_{nuc}}{M_{nuc}} = \frac{\delta \Lambda_{QCD}}{\Lambda_{QCD}} + 0.093 \frac{\delta \hat{m}}{\hat{m}} + 0.043 \frac{\delta m_s}{m_s}, \quad \hat{m} = (m_u + m_d)/2$$

$\mu_{nuc}$  has a small/modest dependence on  $m_s$  ( $\sim 0.01$ ) and on  $\hat{m}$  ( $\sim 0.1$ )

# Classes of systems and their performance

	Recent experiments	frac. accuracy/stability	$f_{\max}$ (Hz)
<b>Atomic clocks</b>	<i>optical transitions (electronic)</i>	$10^{-18}/10^{-18}$	1
	<i>microwave transitions (hyperfine)</i>	$10^{-16}/10^{-16}$	1
<b>Atomic spectroscopy</b>	<i>optical transitions (electronic)</i>	$10^{-14}/10^{-14}$	$10^7$
	<i>microwave (hyperfine)</i>	$10^{-14}/10^{-14}$	$10^3$
<b>Molecular standards/spectroscopy</b>	<i>optical transitions (electronic)</i>	$10^{-14}/10^{-15}$	$10^7$
	<i>mid-infrared transitions (vibrational)</i>	$10^{-15}/10^{-15}$	$10^6$
	<i>THz transitions (rotational)</i>	$10^{-11}/10^{-11}$	$10^4$
<b>Other</b>	<i>Mass spectrometers (mass ratios)</i>	$10^{-11}$	0.1
	<i>Atom interferometers (<math>h/mass</math>)</i>	$10^{-10}$	1
	<i>g-factors of electron, positron, nuclei</i>	$10^{-12}$	0.1
<b>Electromagnetic resonators</b>	<i>Microwave resonators</i>	--/ $10^{-16}$	$10^4$
	<i>Optical resonators</i>	--/ $10^{-17}$	$10^4$
<b>Mechanical resonators</b>	<i>quartz crystals</i>	--/ $10^{-13}$	$10^3$

Note: rough estimates

# Opportunity

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*Experiments so far have not addressed:*

Oscillations of nuclear mass with frequencies  $> 1$  Hz

Optical spectroscopy is well suited for this purpose!

*Proposal:*

D. Antypas et al.

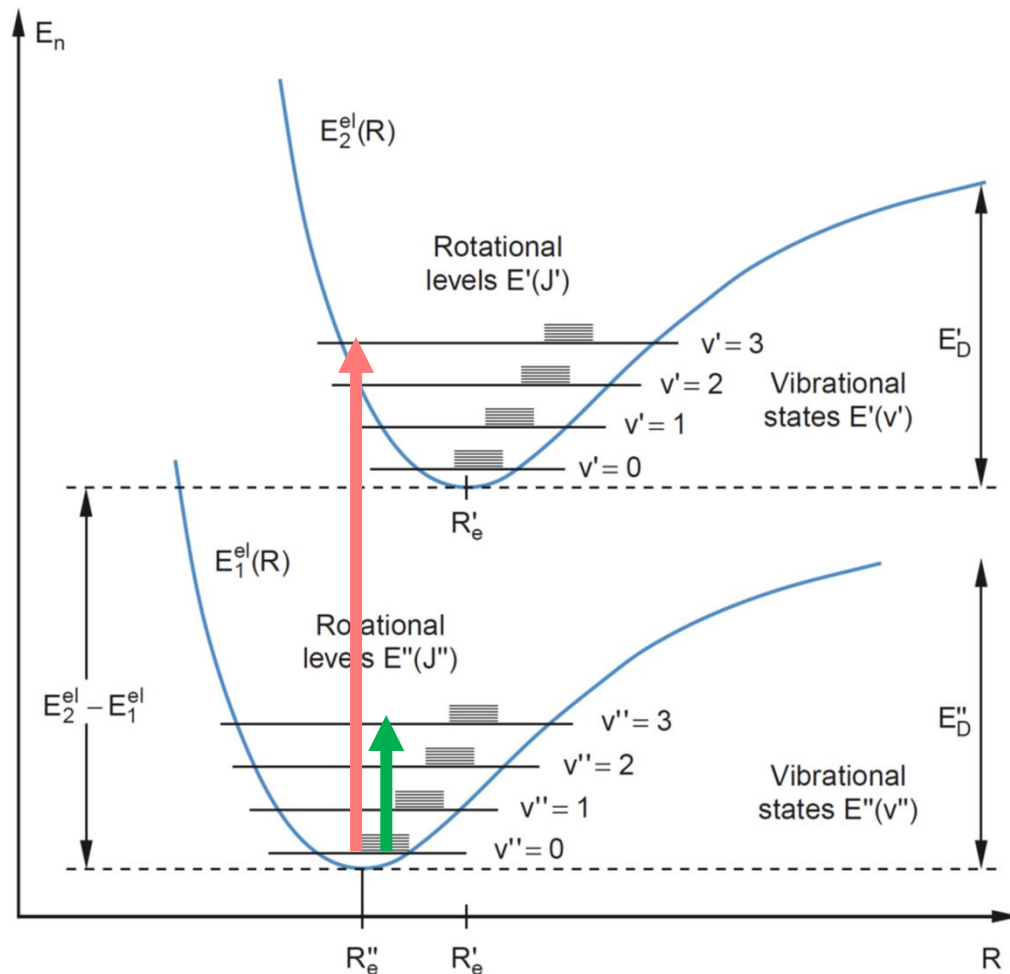
*„Probing fast oscillating scalar dark matter with atoms and molecules”*

Quantum Science and Technology 6, 034001 (2021)



# Optical transitions in molecules

Rotational and vibrational levels  
in two different electronic states of a diatomic molecule



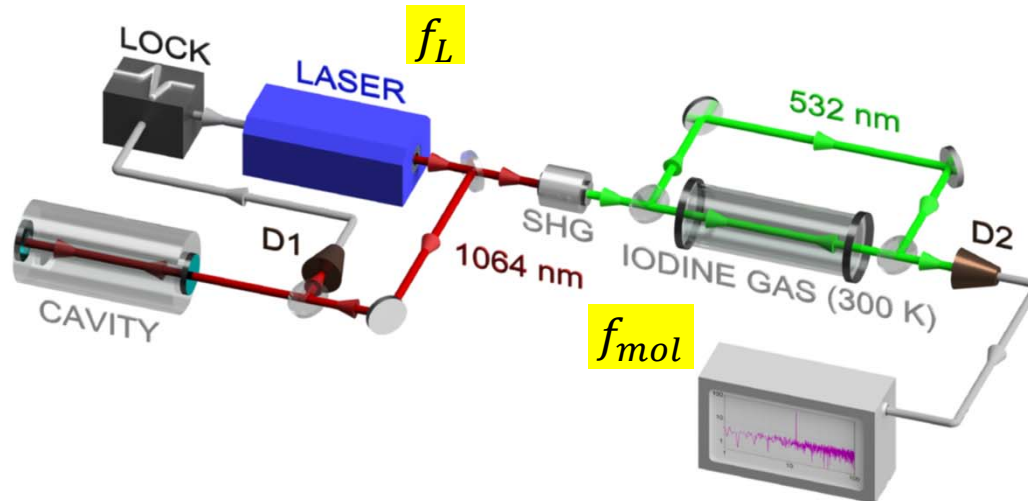
**Electronic transition:**  
contribution from vibration to  
transition frequency is small ( $\sim 0.1$ )

**Vibrational transition:**  
full contribution from vibration

From: W. Demtröder  
*Atoms, Molecules and Photons*

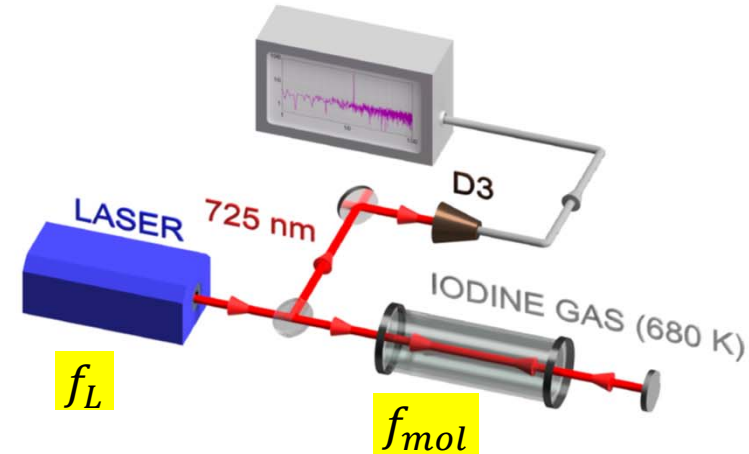
# Experimental setups

**Experiment A** Saturation spectroscopy



$100 \text{ Hz} < f < 0.1 \text{ MHz}$

**Experiment B** Absorption spectroscopy

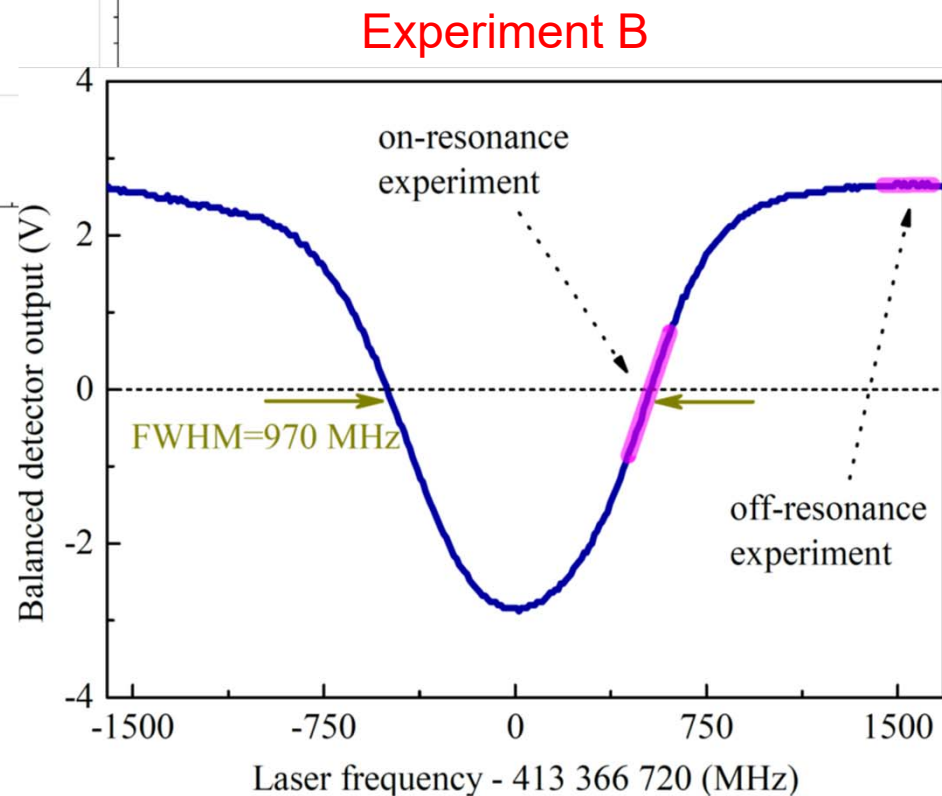
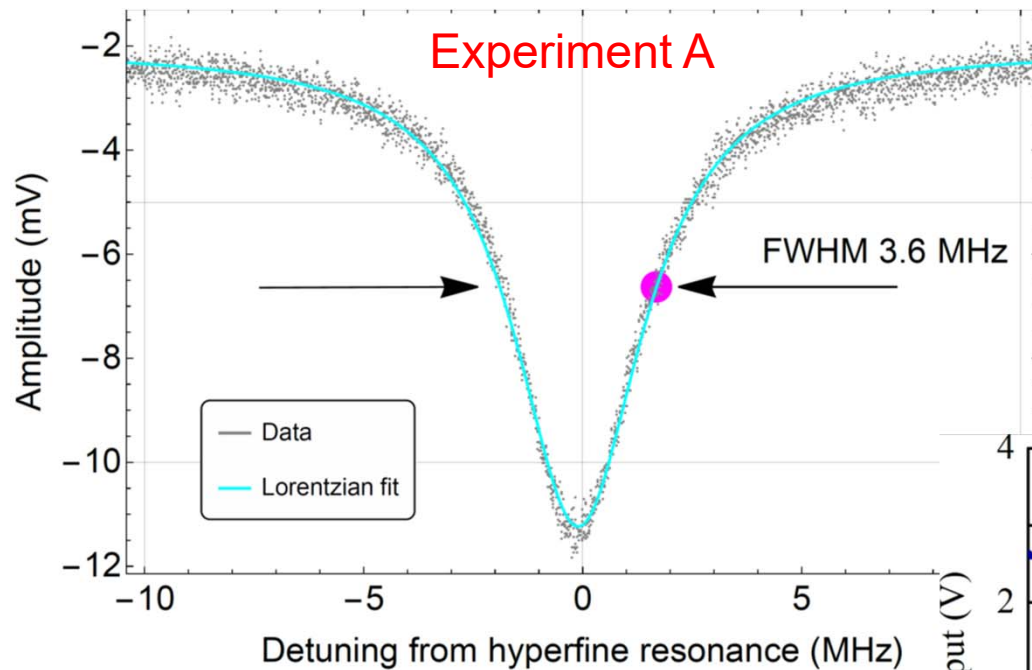


$0.1 \text{ MHz} < f < 100 \text{ MHz}$

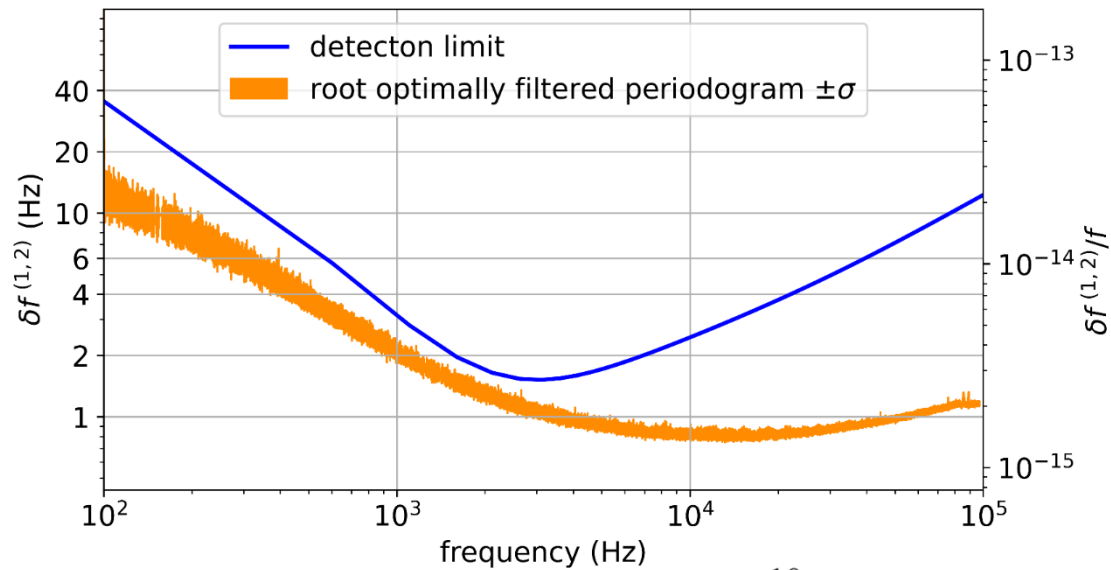
$$\frac{\delta(f_{\text{mol}} - f_L)}{f_{\text{mol}}} = \begin{cases} \frac{\delta\alpha}{\alpha} + \frac{f_{\text{vib}}}{2f_{\text{mol}}} \frac{\delta m_e}{m_e} - \frac{f_{\text{vib}}}{2f_{\text{mol}}} \frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}}, & f \leq f_{\text{cut}} \\ 2\frac{\delta\alpha}{\alpha} + \left(1 + \frac{f_{\text{vib}}}{2f_{\text{mol}}}\right) \frac{\delta m_e}{m_e} - \frac{f_{\text{vib}}}{2f_{\text{mol}}} \frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}}, & f \geq f_{\text{cut}} \end{cases}$$

Experiment is sensitive to 6 fundamental constants!

# Iodine transition lines



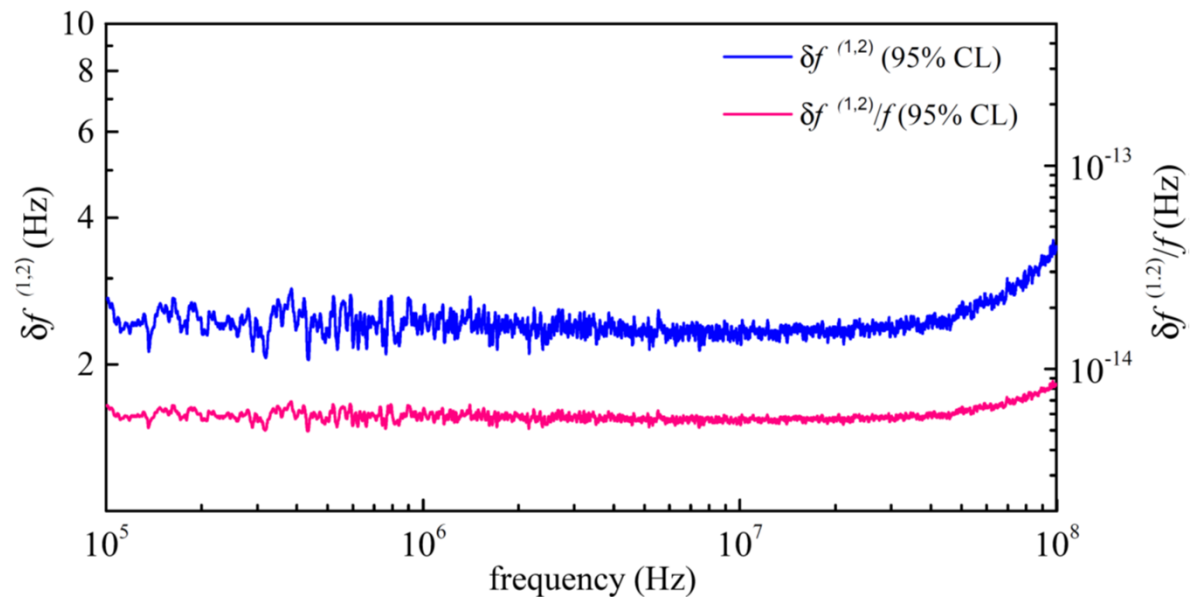
# Experimental limits



Experiment A  
19 h of data

Experiment B 60 h of data

- No evidence for oscillations
- Analysis takes axion lineshape into account
- Results are limited by technical noise



# Analysis

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effectively:

$$\frac{\delta|f_{\text{mol}} - f_L|}{f_{\text{mol}}} = \left| \frac{f_{\text{vib}}}{2f_{\text{mol}}} \right| \frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}}.$$

$$\frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}} = \frac{\delta \Lambda_{\text{QCD}}}{\Lambda_{\text{QCD}}} + 0.093 \frac{\delta \hat{m}}{\hat{m}} + 0.043 \frac{\delta m_s}{m_s},$$

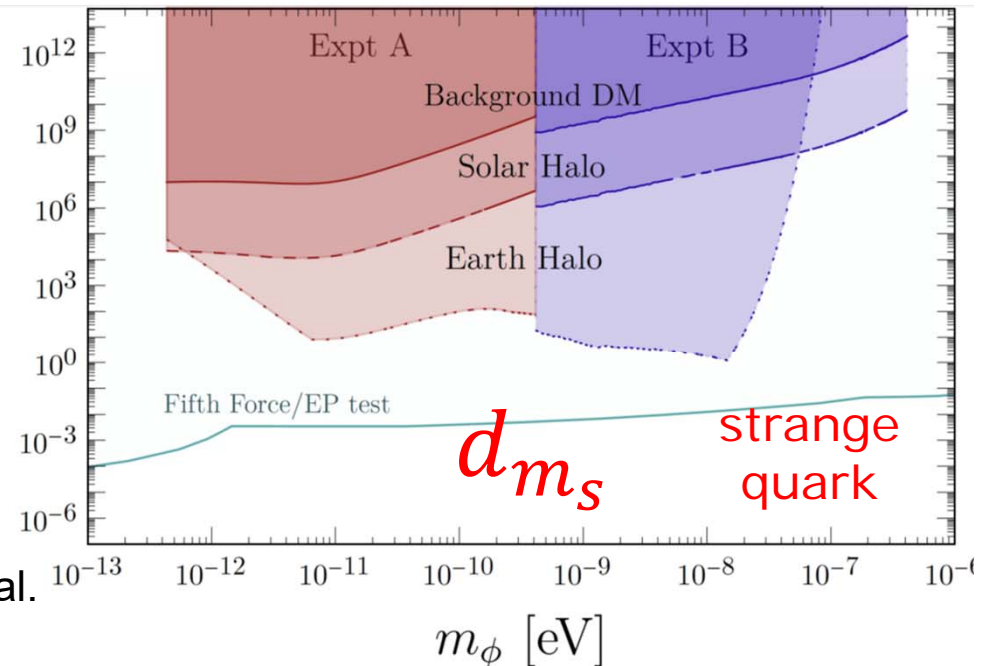
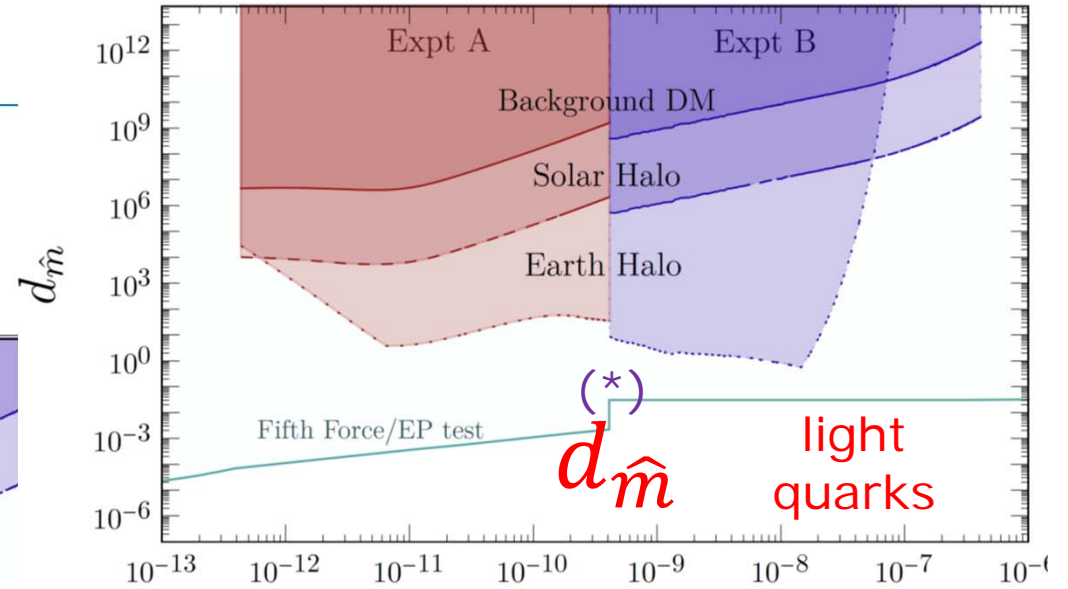
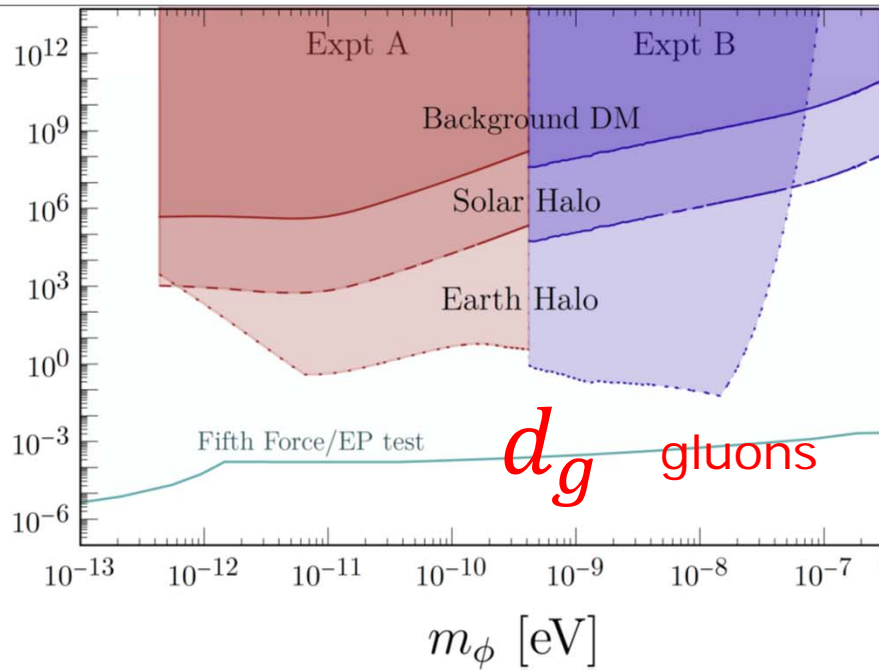
Dependence of nuclear mass on the DM field:

$$\frac{\delta M_{\text{nuc}}}{M_{\text{nuc}}} = (d_g + 0.093 d_{\hat{m}} + 0.043 d_{m_s}) \frac{\langle \Phi \rangle}{M_{Pl}}$$

$$\langle \Phi \rangle = \frac{\sqrt{2\rho_{DM}}}{m_\Phi} \cos 2\pi f_C t$$

# Bounds

preliminary



Earth halo limits: based on model in Banerjee et al.

Comm. Physics 3, 1 (2020)

Solar halo limits: based on model 2007.11016; extended

(\*) accidental

# Summary and conclusion

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- A search for oscillating nuclear mass is feasible in a wide spectrum using standard molecular spectroscopy of gas: sub-kHz to 100 MHz
- Experimental bounds on modulation of molecular transition frequency are at several  $\times 10^{-15}$
- We set bounds on the coupling of light DM scalar field to gluons and quark masses at level  $10^6$  (Galactic halo model)
- Improvement of bounds by several orders appears feasible, with effort





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1-loop result

$$\Lambda_{QCD} \sim \mu e^{2\pi/(\beta_0 \alpha_s(\mu^2, \Phi))}$$

$\mu$ : renormalization-running-energy scale

$\beta_0$ : leading-order beta function

$\alpha_s$ : strong force coupling constant;

here it also depends on  $\Phi$  due to the inner coupling added to the  $GG$  term

# EP-violation/5th force experiments

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- These do not depend on  $\Phi$  having a background value or being DM.
- Because  $\Phi$  is light it mediates a long-range force between two SM objects that couple to  $\Phi$
- the force can be tested via 5th-force-EP-type experiments.
- Example:  $m_\Phi = 1 \text{ neV} \rightarrow f = 240 \text{ kHz}, \Lambda = 1.2 \text{ km}$

$$\mathcal{L} \subset y_{nuc} \frac{\Phi}{M_{Pl}} m_{nuc} \bar{N} N$$

$y_{nuc}$ : Effective coupling constant  
 $N$ : nucleon field

$$F(r) = y_{nuc}^2 \frac{m_{nuc}^2}{M_{Pl}^2} \frac{e^{-r/\Lambda}}{4\pi r}$$

$\Lambda$ : Compton wavelength  $h/m_\Phi c$   
 $F$ : nucleon-nucleon 5th force

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## Models of ultra-light DM:

- [1405.2925](#) the DM is the dilaton (the Goldstone boson of scale invariance)
- [1810.01889](#) (the DM is ALP and due to spontaneous breaking of CP mixes with the Higgs)