Finding hints of New Physics in Tritium molecular spectra?

Wolfgang Gregor Hollik

Karlsruhe Institute of Technology

IKP & TTP @ KIT
KIT-Centrum für Elementarteilchen- und Astroteilchenphysik (KCETA)

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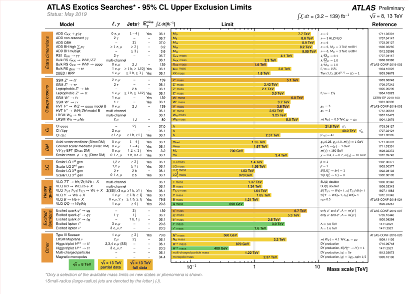
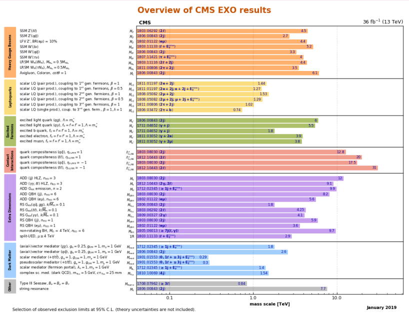
No New Physics at high energies?





[ATLAS Collaboration] [CMS Collaboration]

High Energy vs. Precision Frontier

- New machines, higher collider energies, more luminosity
- Lower energies, more precision, cheaper experiments

Room for New Physics in T_2?

Fundamental vibrational splittings: $Q(J)$ in T_2 [cm^{-1}]

Line	experiment	theory (nonrel)	diff.
$Q(0)$	2 464.5052 (4)	2 464.5021	0.0031 (4)
$Q(1)$	2 463.3494 (3)	2 463.3463	0.0031 (3)
$Q(2)$	2 461.0388 (4)	2 461.0372	0.0016 (4)
$Q(3)$	2 457.5803 (4)	2 457.5795	0.0008 (4)
$Q(4)$	2 452.9817 (4)	2 452.9803	0.0014 (4)
$Q(5)$	2 447.2510 (4)	2 447.2492	0.0017 (4)

exp.: [Trivikram, Schlösser, Ubachs, Salumbides: PRL**120**, 163002 (2018)]

theo.: [Pachucki, Komasa: JCP**143**, 034111 (2015)]

"Interpretation" by the experimentalists [PRL120,163002]

- missing QED and relativistic corrections for T_2
- "measured" contribs. larger than known ones for H_2 and D_2

Light particles and ultraweak forces

- absence of New Physics in High Energy Physics experiments
- quest for open problems and issues in the Standard Model
  - A possible solution to the strong CP problem: Axions?
  - A hidden sector: dark photons and kinetic mixing?
  - Small neutrino masses and sterile neutrinos?
- ultralight and ultraweakly coupled particles still weakly constrained
- e. g. modifcation of coulomb interaction by a light scalar ϕ (*axion*)

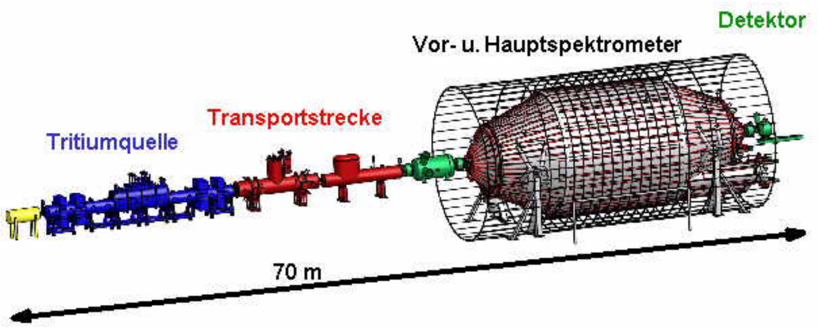
$$V_{AB}(\vec{r}) = -g_A g_B \, \frac{e^{-m_\phi r}}{4\pi r}$$

  - length scale–mass relation: $1\,\text{Å} \mathrel{\hat{\approx}} 1\,\text{keV}$, where molecular bond lengths are $\mathcal{O}(\text{Å})$
  - sensitivity up to several keV massive particles possible with large couplings
- Neutrino pair exchange: Dirac vs. Majorana?
- Modification of Coulomb force by Dark Photons?

Why Tritium!?

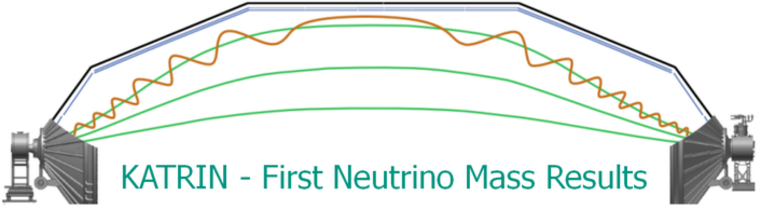
[stern.de]

Tritium spectroscopy



[www.itep.kit.edu]

[KIT KATRIN]



KATRIN - First Neutrino Mass Results

We are pleased to inform you about the results of our first neutrino mass measurement. The KATRIN experiment and its collaborators provide high quality data of beta decay electrons from molecular tritium. The first neutrino mass measurement campaign, which took place this year in spring, made a significant scientific contribution by setting a new upper limit for the absolute mass scale of neutrinos.

We derive a upper limit of **1.1 eV (90% confidence level)** on the absolute neutrino mass scale from our first 28 data days. In the coming years we will collect more data on the decay of molecular tritium and further reduce this limit.

[www.katrin.kit.edu]

13th / 16th September 2019

Toyama / Eggenstein-Leopoldshafen *(near Karlsruhe)*

[arXiv:1909.06048]

Tritium Laboratory Karlsruhe

- world's largest amount of civil tritium
- 25 g of gaseous tritium (T_2)
- research on nuclear fusion (discontinued) and β-decay

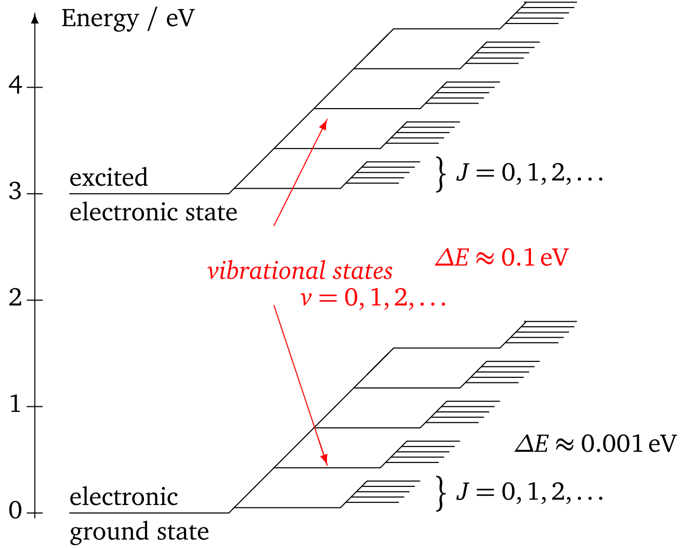
Hydrogen spectroscopy

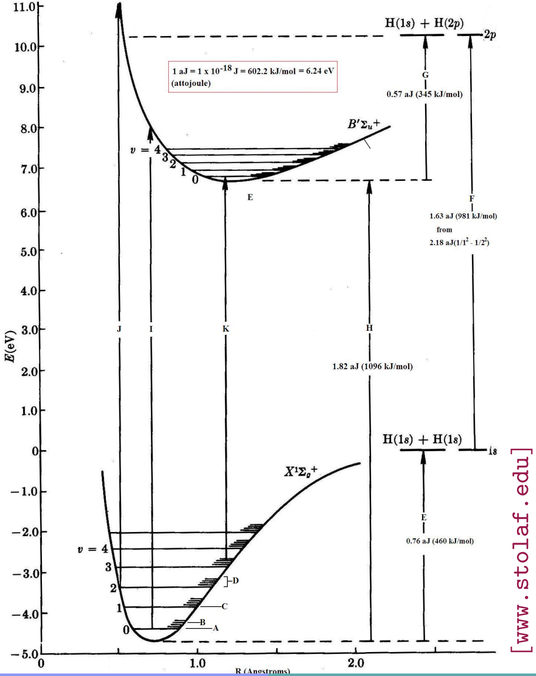
- very precise measurements
- Laser–Raman spectroscopy
- H_2 molecule and variants: clean theoretical prediction (?)

New Physics in molecules?

- test of different length and mass (energy) scales than atoms
- isotopic effects: T_2, D_2, H_2, DT, HT, HD

Schematic molecular energy level diagram (not to scale)




H(1s) + H(2p)
2p
1 aJ = 1 x 10-18 J = 602.2 kJ/mol = 6.24 eV
(attojoule)
G
0.57 aJ (345 kJ/mol)
B′Σu+
v = 4
3
2
1
0
E
F
1.63 aJ (981 kJ/mol)
from
2.18 aJ(1/1² - 1/2²)
J
I
K
H
1.82 aJ (1096 kJ/mol)
E(eV)
H(1s) + H(1s)
1s
X¹Σg+
E
0.76 aJ (460 kJ/mol)
v = 4
3
2
1
0
D
C
B
A
R (Angstroms)
11.0
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
−1.0
−2.0
−3.0
−4.0
−5.0
0
1.0
2.0
[www.stolaf.edu]

Atomic Units *(atomic and molecular physics)*

Hartree: $e = m_e = \hbar = k_e = k_B = 1$ $\quad c = \frac{1}{\alpha}$ *(speed of light)*

Coulomb's constant: $k_e = \frac{1}{4\pi\varepsilon_0}$

$$\text{length} \quad \ell_A = \frac{\hbar^2\, 4\pi\varepsilon_0}{m_e e^2} \mathrel{\hat{\approx}} 5.292 \cdot 10^{-11}\text{m} \approx 0.5\ \text{Å} \quad \textit{(Bohr radius)}$$

Natural Units *(particle physics)*

$$\hbar = c = 1 \;\; \text{e. g. } m_e = 511\,\text{keV},\; m_P \sim 1\,\text{GeV}$$

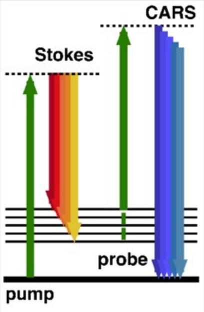
typical bond length $H_2 \sim 0.75\,$Å $\quad 1\,\text{Å} = 10^{-10}\,\text{m}$

Mass–energy–length relation: keV!

$$1\ \underbrace{\text{eV}^{-1}}_{10^3\,\text{keV}} = 1.97 \cdot \underbrace{10^{-7}\text{m}}_{10^3\,\text{Å}} \equiv \frac{\hbar c}{\text{eV}}$$

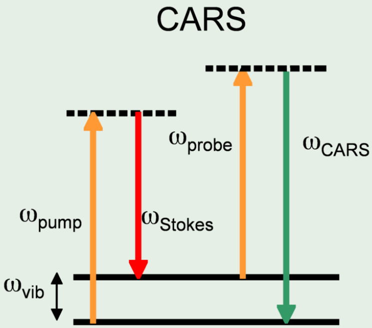
Coherent Anti-Stokes Raman Scattering

"CARS is a nonlinear variant of normal Raman vibrational spectroscopy that allows label-free chemical imaging at (sub)micron resolution. Owing to the inherently small CARS volume, depth profiling of a sample is possible without the need for a confocal set-up. The high sensitivity of the technique due to coherence effects overcomes the difficulty of low Raman scattering cross sections, making CARS three to eight orders of magnitude more sensitive than normal Raman scattering. Thus, vibrational fingerprints of the sample are acquired extremely fast with typical integration times between 20 and 100 ms per voxel."



[Max-Planck-Institut für Polymerforschung, Mainz]

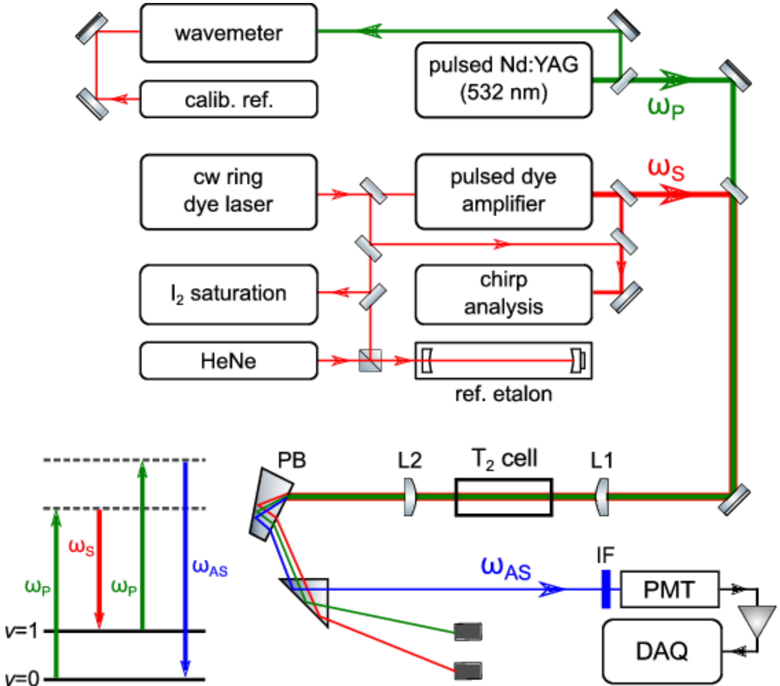
CARS principle and advantage



[wikipedia.org]

- quantum mechanical (an)harmonic oscillator: ω_{vib}, $\nu = 0 \to 1$
- two laser beams:
  - pump ω_{pump}
  - Stokes ω_{Stokes}
- difference frequency $\omega_{\mathrm{pump}} - \omega_{\mathrm{Stokes}} \to \omega_{\mathrm{vib}}$ resonance
- coherent superposition of $|0\rangle$ and $|1\rangle$

Measurements with Tritium (T_2, DT, HT) done by the molecular physics group and LaserLaB, Vrije Universiteit Amsterdam [W. Ubachs et al.]
Tritium source by KIT-TLK [M. Schlösser at al.]



[Madhu Trivikram, Schlösser, Ubachs, Salumbides: PRL **120**, 163002]

Is it feasible to search for New Physics effects in molecules?

Theory: constrain New Physics models

- **Matthias Linster**, KIT-TTP
- **Mustafa Tabet**, KIT-TTP
- Sonia Rani, IIT Bombay
- Aman Sardwal, IIT Bombay
- Wolfgang G. Hollik, KIT-IKP/TTP
- Ulrich Nierste, TTP-KIT

- light new particles: **ALPs, dark photons, sterile neutrinos**?

Experiment: measure precise spectra

- Magnus Schlösser, KIT-TLK
- Edcel Salumbides, VU Amsterdam
- Wim Ubachs, VU Amsterdam

Rotational Energies

- q. m. rotator: two masses $m_{1,2}$, distance $r = r_1 + r_2$
- center or mass: $m_1 r_1 = m_2 r_2$
- moment of inertia: $I = m_1 r_1^2 + m_2 r_2^2 = \frac{m_1+m_2}{m_1 m_2}(r_1^2 + r_2^2) = \mu r^2$

$$\left.\begin{array}{rl} \text{angular momentum} & \vec{L} = I\vec{\omega}/\hbar \\ \text{energy} & E = I\omega^2/2 \end{array}\right\} \quad E = \frac{\vec{L}^2\hbar^2}{2I}$$

$|\vec{L}| = \hbar\sqrt{J(J+1)}$, spinless masses: $J = 0, 1, 2, \ldots$

- quantized energy: $E_J = J(J+1)\,\hbar^2/(2I) \equiv B\,J(J+1)$
- energy separation between rotational levels $\sim 10^{-3}$ eV (mw)

Vibrational Modes: level separation $\sim 0.1\,$eV (IR)

anharmonic oscillator: $E_v = (v + \frac{1}{2})\,\hbar\omega - (v + \frac{1}{2})^2\, x_e\,\hbar\omega$

Q-band spectroscopy: $\Delta J = 0$, $v = 0 \rightarrow 1$ CARS technique

Hydrogen-like molecules: 2 nuclei A and B, 2 electrons

$$H = \underbrace{\frac{\vec{P}_1^2}{2m} + \frac{\vec{P}_2^2}{2m}}_{\text{electrons}} + \underbrace{\frac{\vec{P}_A^2}{2M_A} + \frac{\vec{P}_B^2}{2M_B}}_{\text{nuclei } A+B}$$

$$+ \alpha \left\{ -\frac{1}{r_{1A}} - \frac{1}{r_{2B}} + \left(\frac{1}{r_{12}} + \frac{1}{R_{AB}} - \frac{1}{r_{1B}} - \frac{1}{r_{2A}} \right) \right\}$$

coulombic terms:

- attractive: electron$_{1/2,2/1}$ at nucleus$_{A/B,B/A}$ $\sim \frac{1}{r_{1A/2B,1B,2A}}$
- repulsive: electron-electron $\sim \frac{1}{r_{12}}$, nucleus-nucleus $\sim \frac{1}{R_{AB}}$

Heitler–London approach

[Heitler/London 1927; Born/Oppenheimer 1927]

- neglection motion of nuclei (heavy wrt electrons)
- calculate electronic wave function
  $\hookrightarrow$ effective potential between nuclei

Electronic wave functions, nuclei at fixed positions

- $\psi_X(\vec{r}_j)$: electron j at nucleus X; two solutions: gerade, ungerade

$$\Phi_\pm(\vec{r}_1,\vec{r}_2) = \frac{\psi_A(\vec{r}_1)\psi_B(\vec{r}_2) \pm \psi_B(\vec{r}_1)\psi_A(\vec{r}_2)}{\sqrt{2(1\pm|S_{AB}|^2)}}$$

- overlap $S_{AB} = \int \mathrm{d}^3 r_1 \psi_A^*(\vec{r}_1)\psi_B(\vec{r}_1)$

Perturbation: "wrong" Hamiltonian ↪ energy shift

$$E_{11} = \int \mathrm{d}r_1\,\mathrm{d}r_2 \Bigg\{ \left(\frac{\alpha}{r_{12}} + \frac{\alpha}{R}\right) \frac{u_A(r_1)^2 u_B(r_2)^2 + u_A(r_2)^2 u_B(r_1)^2}{2} - \left(\frac{\alpha}{r_{1A}} + \frac{\alpha}{r_{2B}}\right)\frac{u_A(r_2)^2 u_B(r_1)^2}{2} - \left(\frac{\alpha}{r_{2A}} + \frac{\alpha}{r_{1B}}\right)\frac{u_A(r_1)^2 u_B(r_2)^2}{2} \Bigg\}$$

$$E_{12} = \int \mathrm{d}r_1\,\mathrm{d}r_2 \left(\frac{2\alpha}{r_{12}} + \frac{2\alpha}{R} - \sum_{x=1A,2A,1B,2B} \frac{\alpha}{r_x}\right) \frac{u_A(r_1)u_A(r_2)u_B(r_1)u_A(r_2)}{2}$$

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Perturbation: "wrong" Hamiltonian $\hookrightarrow$ energy shift

$$E_\pm = \frac{E_{11} \pm E_{12}}{1 \pm S_{AB}}$$

- Coulomb integral: E_{11}
- exchange integral: E_{12}
- potential energy curve: Morse potential $V(r) = D_0\left(1 - e^{-\alpha(r-r_0)}\right)^2$

Electronic wave functions, nuclei at fixed positions

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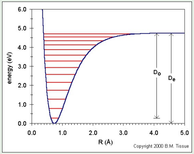

- Coulomb integral: E_{11}
- exchange integral: E_{12}
- potential energy curve: Morse potential $V(r) = D_0\left(1 - e^{-\alpha(r-r_0)}\right)^2$

Full Hamiltonian

$$H = \sum_{k=1}^{2} \frac{P_k^2}{2M_k} + \frac{1}{2} \sum_{i=1}^{2} \frac{p_i^2}{2m} + V_C(r,R) = \sum_{k=1}^{2} \frac{P_k^2}{2M_k} + H^0$$

- electronic problem solved
- eigenvalues $E_n^0(R)$ and eigenstates $\varphi_n(r,R)$ known:

$$\left(H^0 - E_n^0(R)\right) \varphi_n(r,R) = 0$$

Nuclear wave function

semi-separation ansatz: nuclear wavefunction $\psi_n(R)$

$$\Psi(r,R) = \sum_n \psi_n(R) \varphi_n(r,R)$$

solve

$$(H - E)\Psi(r,R) = 0$$

Integrating electronic coordinates $\int \mathrm{d}r \varphi^*_{n'}(r,R)(H-E)\Psi(r,R)$

$$\int \mathrm{d}r\, \varphi^*_{n'}(r,R) \left(\sum_{k=1}^{2} \frac{P_k^2}{2M_k} + H^0 - E \right) \sum_n \psi_n(R) \varphi_n(r,R) = 0\,,$$

$$\int \mathrm{d}r\, \varphi^*_{n'}(r,R) \left(\sum_{k=1}^{2} \frac{P_k^2}{2M_k} + E^0_n(R) - E \right) \sum_n \psi_n(R) \varphi_n(r,R) = 0\,,$$

$$\left(\sum_{k=1}^{2} \frac{P_k^2}{2M_k} + E^0_{n'}(R) - E \right) \psi_{n'}(R) - \sum_n C_{n'n} \psi_n(R) = 0\,,$$

Nuclear distance R not constant anymore

$$A^{(k)}_{n'n} = \int \mathrm{d}r\, \varphi^*_{n'}(r,R) P_k\, \varphi_n(r,R) \qquad B^{(k)}_{n'n} = \frac{1}{2} \int \mathrm{d}r\, \varphi^*_{n'}(r,R) P_k^2\, \varphi_n(r,R)$$

Schrödinger equation for the nuclei $\psi(R)$ $\qquad C_{n'n} = \sum_k \frac{1}{M_k} \left(A^{(k)}_{n'n} P_k + B^{(k)}_{n'n} \right)$

$$\left(\sum_{k=1}^{2} \frac{P_k^2}{2M_k} + U_n(X) - E \right) \psi_n + \sum_{n' \neq n} C_{nn'} \psi_{n'}(R) = 0$$

Separating electronic and nuclear motion

$$H_0 = -\frac{1}{2}\left(\Delta_{r_1} + \Delta_{r_2}\right) + V\,,$$
$$H_1 = -\frac{m}{8\mu}\left(\vec{\nabla}_{r_1} + \vec{\nabla}_{r_2}\right)^2\,,$$
$$H_2 = -\frac{m}{2\mu}\,\langle 0|\,\Delta_R\,|0\rangle\,,$$
$$H_3 = -\frac{m}{2\tilde{\mu}}\,\langle 0|\,\vec{\nabla}_R\left(\vec{\nabla}_{r_1} + \vec{\nabla}_{r_2}\right)|0\rangle\,,$$

- reduced mass $\mu = M_a M_b/(M_a + M_b)$ and $\tilde{\mu} = M_a M_b/(M_a - M_b)$
- electronic ground state $|0\rangle$ i. e. two 1s H atoms

elliptic coordinates [Hylleraas 1931]

$$\Psi = \frac{1}{2\pi}\sum_{i,n} c_{i,n}\psi_i(\xi_1, \eta_1, \xi_2, \eta_2, \phi)h_n(R)$$

with

$$\xi_i = \frac{r_{ai} + r_{bi}}{R} \quad \text{and} \quad \eta_i = \frac{r_{ai} - r_{bi}}{R}$$

Separating electronic and nuclear motion

$$H_0 = -\frac{1}{2}\left(\Delta_{r_1} + \Delta_{r_2}\right) + V\,,$$
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non-adiabatic theory: dissociation energy [Kołos, Wolniewicz 1963 ff]

variational method: $\delta \int \mathrm{d}\,\tau_1\,\mathrm{d}\,\tau_2\,\mathrm{d}^3 R\,\Psi^* H \Psi = 0$

"trial" wave functions

$$\psi_i = \exp\left[-\alpha\left(\xi_1 + \xi_2\right)\right]\left(\xi_1^{p_i}\eta_1^{q_i}\xi_2^{r_i}\eta_2^{s_i} + \xi_1^{r_i}\eta_1^{s_i}\xi_2^{p_i}\eta_2^{q_i}\right)\left(\frac{2r_{12}}{R}\right)^{m_i}$$

A short wrap-up [source: Paweł Czachorowski PhD 2019 Warsaw]

- *ab initio* calculations
- large sets of trial wave functions: higher precision, more CPU
- non-relativistic QED [Pachucki 2004, 2005]

$$E(\alpha) = \alpha^2 E^{(2)} + \alpha^4 E^{(4)} + \alpha^5 E^{(5)} + \alpha^6 E^{(6)} + \alpha^7 E^{(7)} + \dots$$

- leading relativistic correction: Breit–Pauli [Puchalski et al. 2018]
- non-adiabatic perturbation theory
- nuclear nuclear size correction: accuracy of charge radii
- relativistic non-adiabatic [Czarochowski et al. 2018]
- explicitly correlated exponentials vs. correlated Gaussian
- usable computer code: `H2spectre` [`www.fuw.edu.pl/~kpl`]

Total theoretical error budget $D_0^{H_2} = 36118.06945(53)\,\mathrm{cm}^{-1}$

e. g. dissociation energy H_2: relative error $\simeq 1.46 \times 10^{-8}$

Dissociation energies

$\frac{D_0}{\mathrm{cm}^{-1}}$	H_2	HD	D_2
Th.	36 118.069 636(26)	36 405.782 39(19)	36 748.362 28(15)
E. 1	36 118.069 62(37)	36 405.783 66(36)	36 748.362 86(68)
Δ 1	+0.000 02(37)	−0.001 27(41)	−0.000 58(70)
E. 2	36 118.069 45(31)		
Δ 2	+0.000 19(31)		

[Czachorowski PhD 2019 and references therein!]

- theoretical error estimates?
- missing non-adiabatic QED $E^{(5,1)}$ corrections?
- full nonadiabatic $E^{(2)}$ energies for rovibrational levels
- long-term perspective: $E^{(7)}$ and $E^{(6,1)}$ contributions
- tackling the proton charge radius puzzle: independent determination of r_p from Hydrogen-isotoper spectroscopy

[Czachorowski PhD 2019]

Simplest scenario: Yukawa potential

$$V_{\mathrm{Yuk}} = \frac{g_{\mathrm{NP}}}{2\pi}\frac{e^{-mr}}{r}$$

- massive particle with mass m, New Physics coupling g_{NP}
- exponential drop-off
- e. g. dark photon [Jaeckel, Roy 2010]
- e. g. "fifth force" ($m \to 1/\lambda$ length) [Salumbides et al. 2013]
- axion-like particles (small couplings) [Jaeckel, Ringwald 2010]
- constraints on electron-electron, nucleon-nucleon, electron-nucleon generic couplings [WGH et al.]

Simple task(?)

Yukawa potential as perturbation to Coulomb potentials

$$\langle \Delta E_{\mathrm{NP}} \rangle = \frac{g_{\mathrm{NP}}}{2\pi}\left[\left\langle \Psi_{v_f,J_f} \middle| \frac{e^{-mr_x}}{r_x} \middle| \Psi_{v_f,J_f} \right\rangle - \left\langle \Psi_{v_i,J_i} \middle| \frac{e^{-mr_x}}{r_x} \middle| \Psi_{v_i,J_i} \right\rangle\right]$$

Simple task(?)

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wave functions from `H2spectre` [Pachucki et al.]

Potential for New Physics

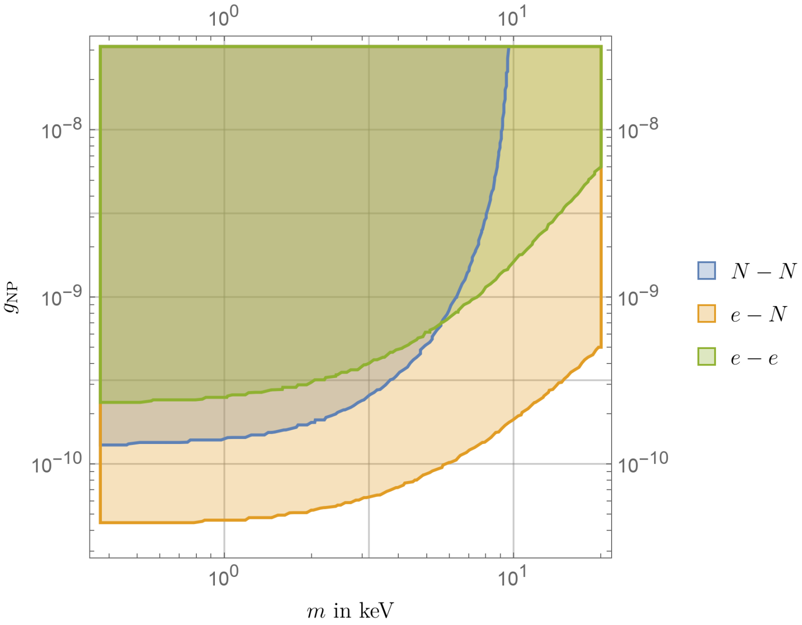
compare “old“ and new physics with experiment

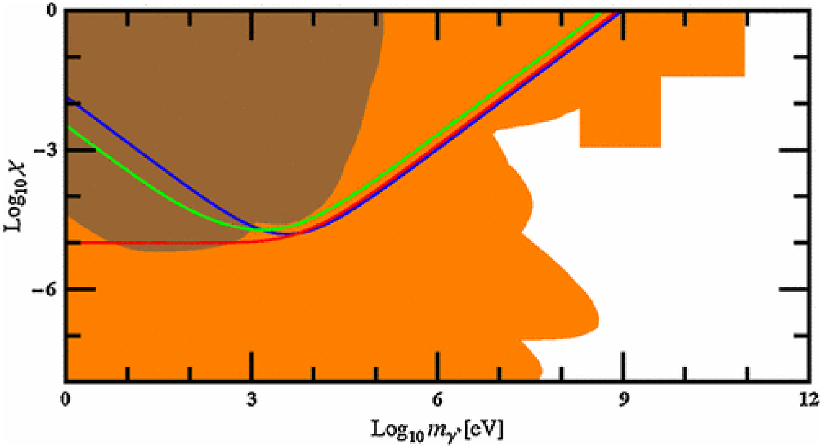
$$E_{\mathrm{exp}} + \delta E_{\mathrm{exp}} \quad \text{vs.} \quad E_{\mathrm{theo}} + \delta E_{\mathrm{theo}} + \Delta E_{\mathrm{NP}}$$

constraints on mass vs. coupling

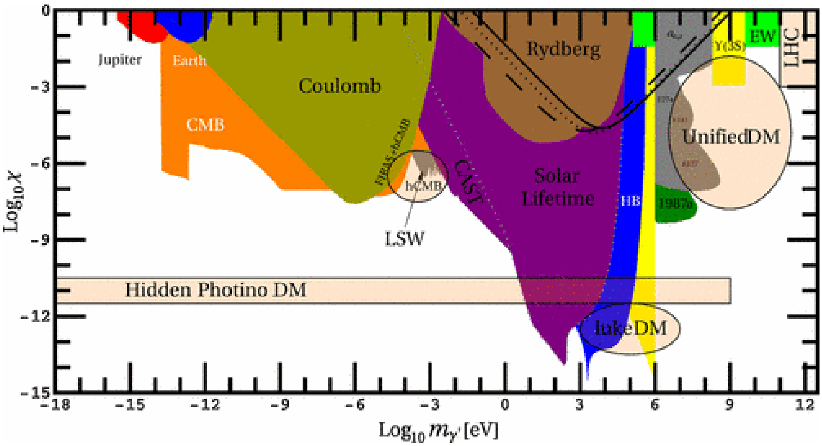
Results, exclusion limits (preliminary)

$$V_{\mathrm{NP}} = \frac{g_{\mathrm{NP}}}{2\pi}\frac{\exp(-mr)}{r}$$





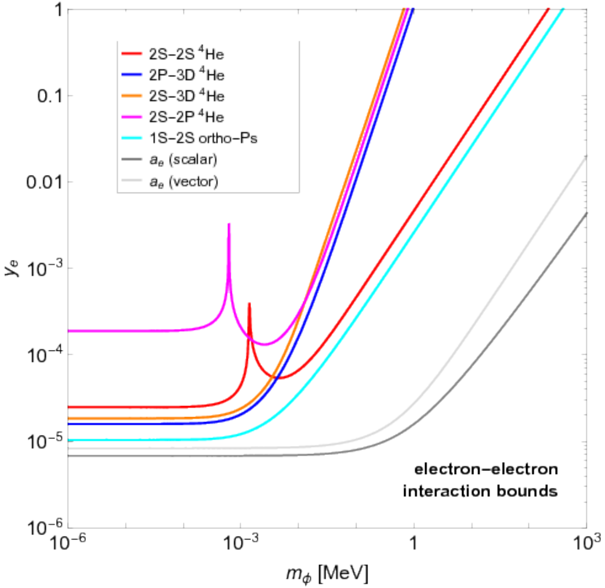
[Jaeckel, Roy: "Spectroscopy as a test of Coulomb's law: A probe of the hidden sector", PRD**82**,125020(2010)]



[Jaeckel, Roy: "Spectroscopy as a test of Coulomb's law: A probe of the hidden sector", PRD**82**,125020(2010)]

Atom spectroscopy with few electrons

[Delaunay, Frugiuele, Fuchs, Soreq 2017]



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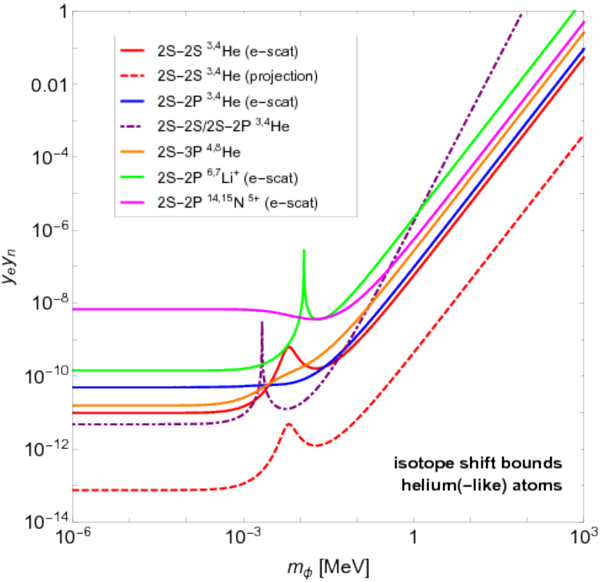

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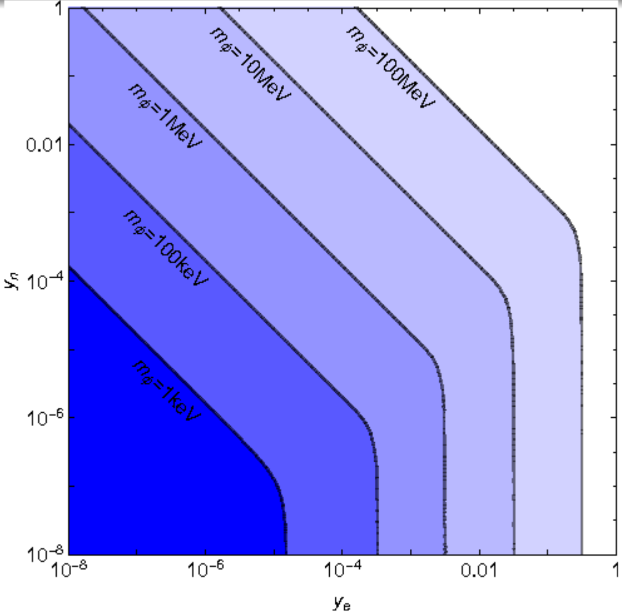

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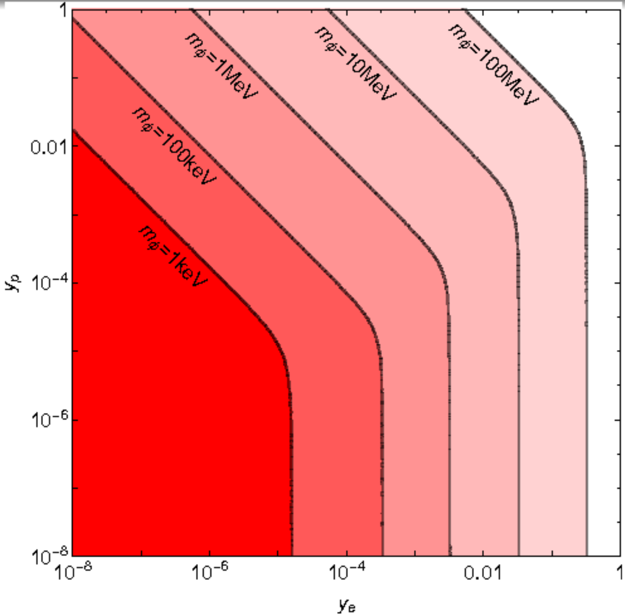
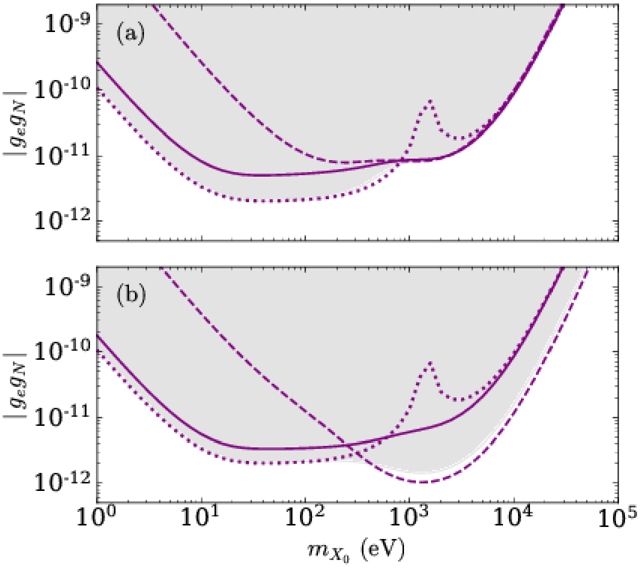


Rydberg states of atomic hydrogen

[Jones, Potvliege, Spannowsky arXiv:1909.09194]



Summary and Conclusions

- rovibrational spectroscopy as a probe of New Physics
- Hydrogen isotopomers: H_2, D_2, T_2, HD, HT, DT
- first step: constrain simple New Physics as Yukawa potential

$$V_{\mathrm{NP}} = \frac{g_{\mathrm{NP}}}{2\pi} \frac{e^{-mr}}{r}$$

Outlook

- isotopes for correlations [WGH et al.]
- improvement in theoretical *ab initio* calculations expected
- experimental refinements
- measurement of DT and HT to be published
- test of different potential types (also with spin) [WGH et al.]