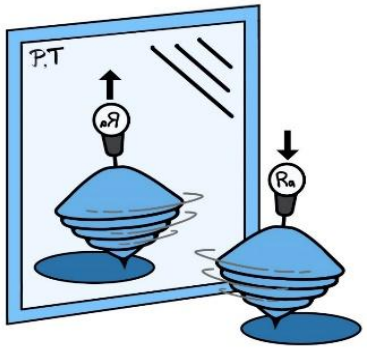

St Petersburg University

Search for the Dark Sector particles with molecules

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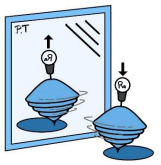
THE AXION QUEST 2024

Intro



- **Experiments with molecules** are a successful platform for searching for P,T-violating processes

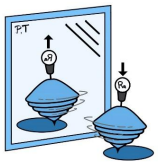
- Experiments on molecules make it possible to achieve **high precision** in measuring these effects.

- Polyatomic molecules with heavy atoms are promising because these molecules have high intramolecular electric fields, can be laser cooled, and have similar **levels of opposite parity** in their vibrational spectrum, which makes it possible to suppress many systematic effects

Current limit on the eEDM: $|de| < 4.1 \times 10^{-30}\ e \cdot cm$

- Is it possible to see in these experiments the interaction of known particles with cosmic fields of the Dark Sector, such as pseudoscalar fields of axions and vector fields of Dark photons?

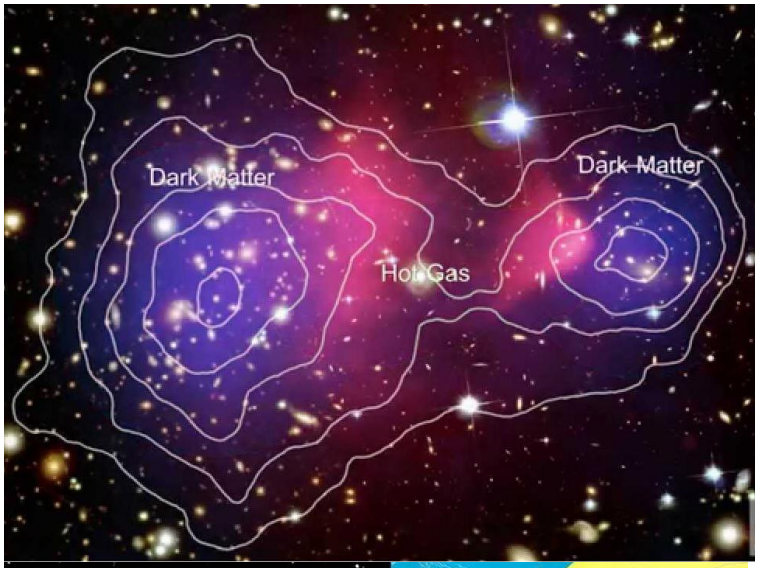
The nature of dark matter

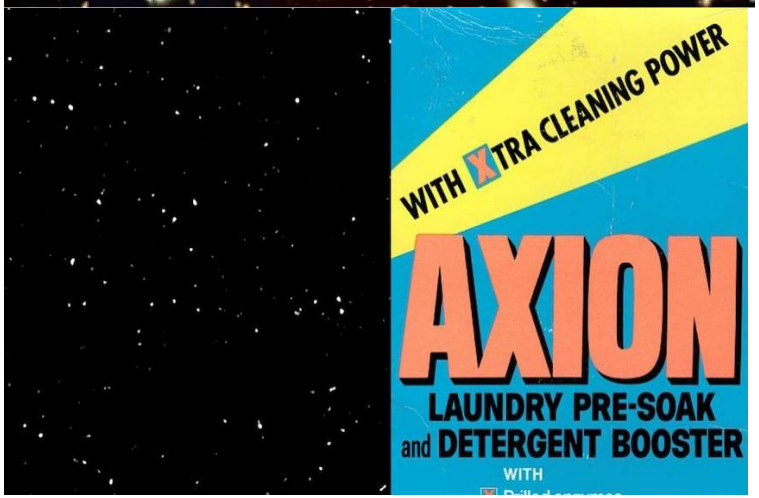


- Its contribution to the energy density in the Universe is several times greater than the contribution of all known particles.

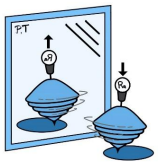
- Most observations are best described by the concept of **cold dark matter (CDM):** in which dark matter consists of weakly interacting, long-lived heavy particles.

- The CDM concept faces a number of difficulties, so other options for the structure of the dark sector are being considered.





Axions and dark photons

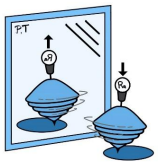


In CDM, Dark Matter behaves like a gas of particles. Scenarios in which it is in fairly coherent states and behaves more like classical field configurations are also popular

- One popular candidate for the role of dark matter is an oscillating condensate of **ultralight pseudoscalar particles - axions**.

- The proposed extension of the Standard model to include the Dark Sector also often assumes a more complex structure. It consists not only of scalar fields, but also of **vector fields known as Dark Photons**. In such a scenario, the Solar System is in the external scalar and vector fields of the Dark Sector.

Interaction of leptons with dark matter fields



Proposed new interactions

$$\mathcal{L}^{a}_{\not P\not T} = -\frac{g_{a\bar{e}e}}{m_e}\left(\partial_\mu a\right)\bar{\psi}_{\rm e}\gamma^\mu\gamma^5\psi_{\rm e},\quad \mathcal{L}^{\tilde{A}}_{\not P\not T} = -\frac{g_{\tilde{A}\bar{e}e}}{m_e}\tilde{A}^\mu\bar{\psi}_{\rm e}\gamma^\mu\gamma^5\psi_{\rm e}$$

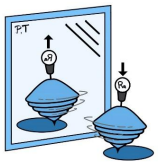
Let the DM halo be a condensate of axions, as well as possible galactic dark vector fields. Solar system speed ~10^{-3} c

$$a(t) = a_0\cos m_a t \qquad \tilde{A}^\mu(t) = \tilde{A}\delta^\mu_0\cos m_{\tilde{A}} t$$

Effective interactions with the electron shell can be considered as small P, T-odd additions to the Hamiltonian of the molecule

$$\hat{H}^{a}_{eff} = \frac{g_{a\bar{e}e}}{m_e} m_a a_0\gamma^5\cos m_a t, \qquad \hat{H}^{\tilde{A}}_{eff} = \frac{g_{\tilde{A}\bar{e}e}}{m_e}\tilde{A}\gamma^5\cos m_{\tilde{A}} t$$

Violation of C- and P-parities

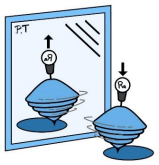


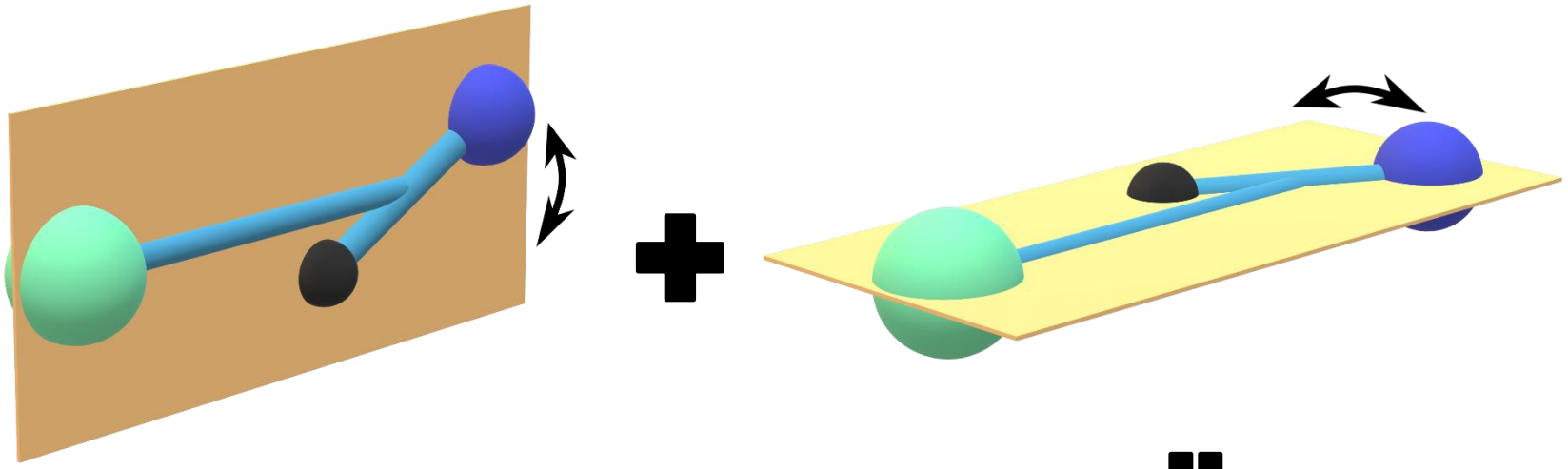
- At the moment, **experiments to search for fundamental parity violation using molecules are leaders in the accuracy** of restrictions on such effects as the electric dipole moment of the electron and the parity-violating interaction of electrons with the nucleus.

- Previous experiments looking for odd effects such as electron EDM have been done with diatomic molecules, but experiments are now being planned with polyatomic molecules such as the triatomic molecules RaOH and YbOH, as well as hexatomic molecules like the symmetrical top $YbOCH_3$ and $RaOCH_3$

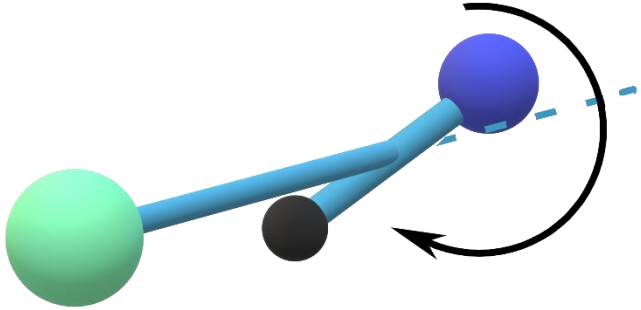
- **Can the interactions under consideration affect the spectrum of such molecules?**

Transverse vibrations



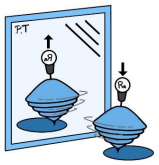


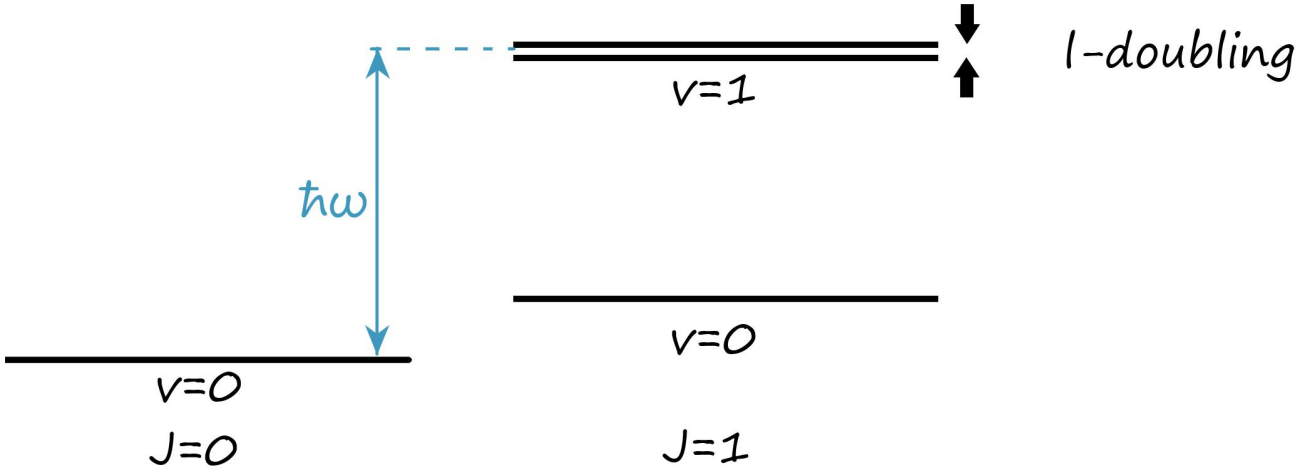
- Transverse vibrations of the ligand (not necessarily linear) can occur in two transverse planes. To a first approximation, we can describe them as two **uncoupled harmonic oscillators** with quantum numbers v=v1+v2.



- **The superposition of vibrations in two planes can be considered as rotation** around the axis of the molecule. Therefore, we can attribute to them the vibrational angular momentum l, which contributes to the total angular momentum of the molecule J.

Rovibrational spectrum





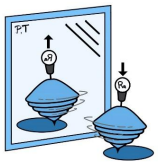
v=1 corresponds to two approximately degenerate levels

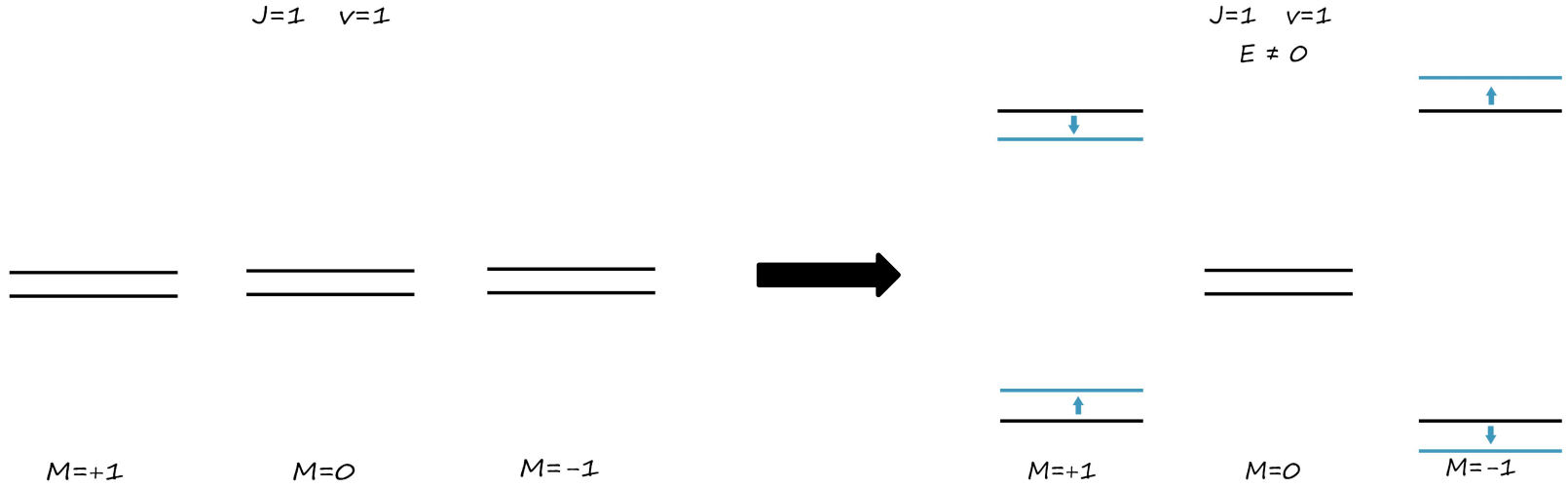
The degeneracy is broken by the Coriolis force and the interaction between longitudinal and transverse vibrations due to potential anharmonicities. The result is two **closely spaced levels of opposite parity**.

$$|\pm\rangle = \frac{1}{\sqrt{2}}|+l\rangle \pm \frac{1}{\sqrt{2}}|-l\rangle$$

Level splitting is called **l-doubling.**

Spectrum in the external field with P,T-violating effects



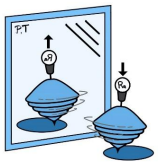


Without an external field, states with different projections of the total angular momentum onto the z axis of the laboratory coordinate system are **degenerate**.

An external electric field leads to a shift of levels from M=±1. **P, T – odd effects lead to different shifts** for opposite M (blue lines). Measuring the difference in shifts for different l-doubled levels allows us to eliminate many systematic effects.

The effect is maximum at a value of the field E when the shifted levels become eigenstates l. **E depends on the magnitude of l-doubling.**

Impact of the halo on the spectrum



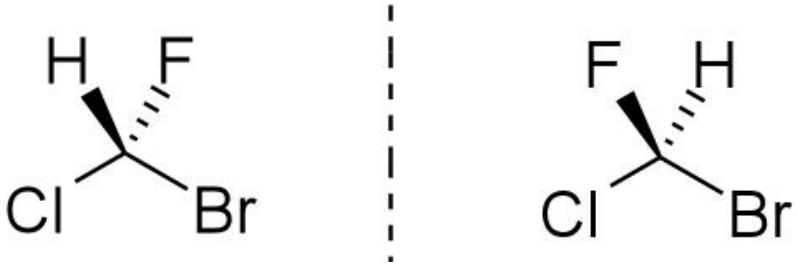
- We consider new interactions as a **perturbation of the P, T-even Hamiltonian**. The influence of the halo is determined by the average values of the effective Hamiltonian of interactions on solutions of the P, T-even Hamiltonian.

- **Problem:** if a molecule upon reflection can be returned to its original configuration by rotation, then the electronic states have a certain parity with respect to this symmetry transformation.

- Y5 will be odd, which means its average value = 0.

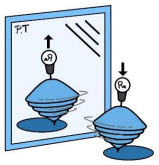
$$\langle\psi|\gamma_5|\psi\rangle \mapsto -P_\psi^2\langle\psi|\gamma_5|\psi\rangle = -\langle\psi|\gamma_5|\psi\rangle$$

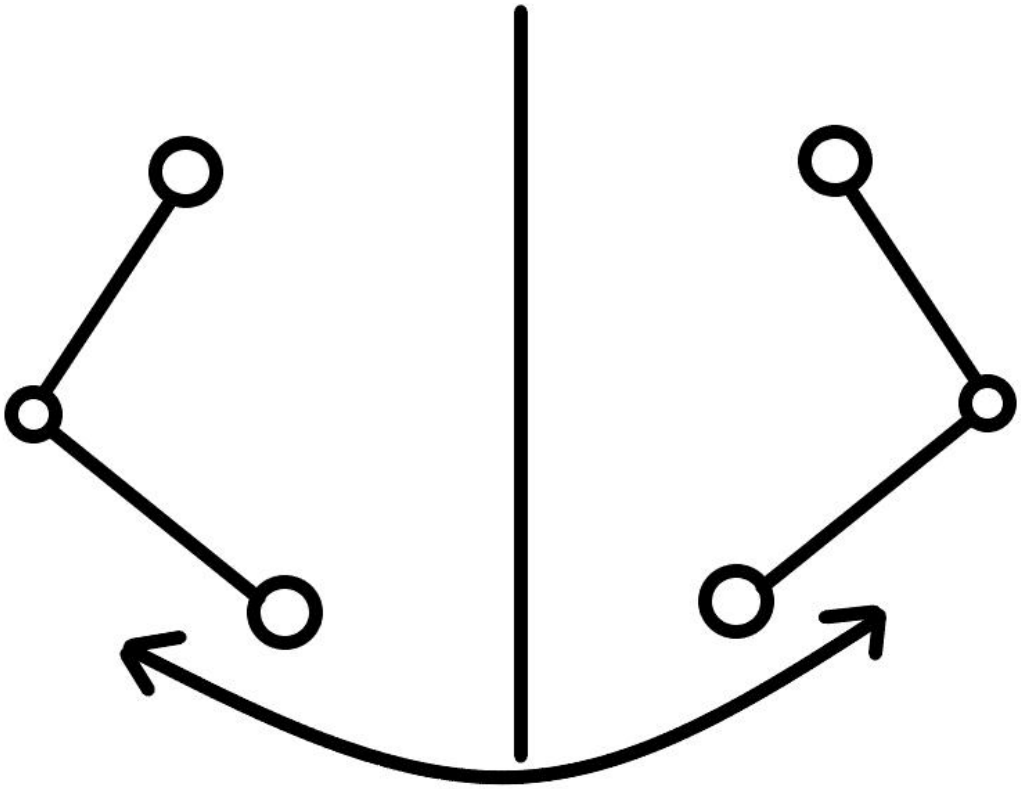
- **Chiral molecules** [K.Gaul, M.G.Kozlov, T.A.Isaev, R.Berger, 2020]



- **But molecules vibrate and can become sensitive in non-equilibrium configurations!**

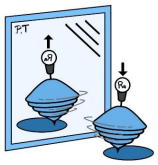
Triatomic molecule



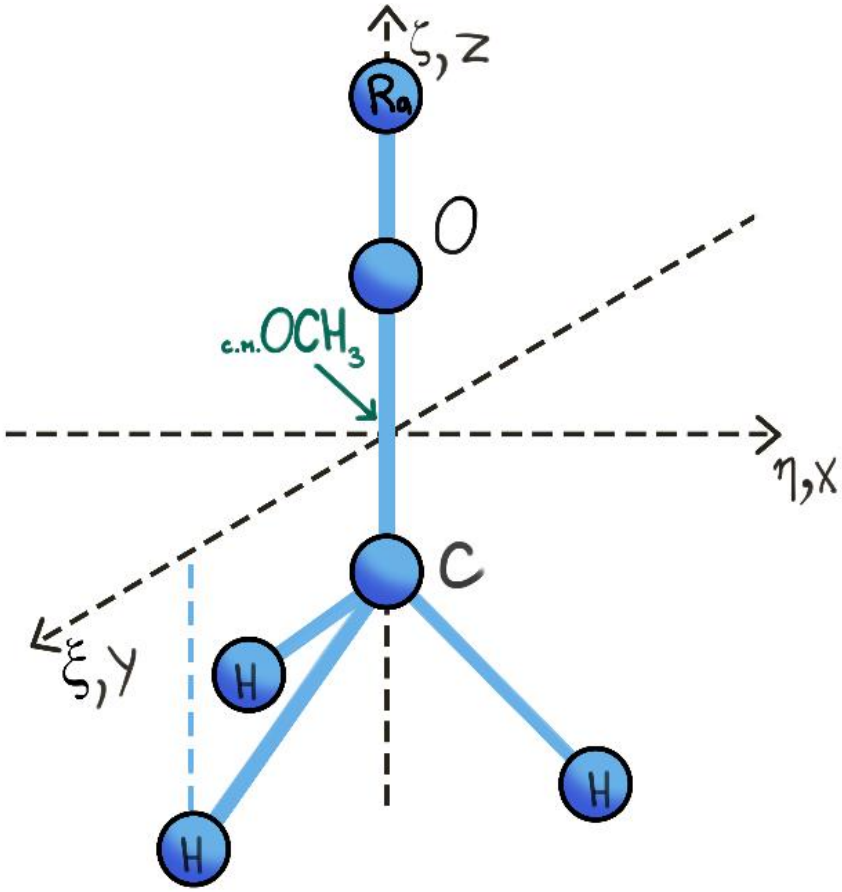


- **Triatomic molecules are not suitable**, since a bent molecule can always be rotated to its original configuration

Symmetric top



RaOCH3 and YbOCH$_3$

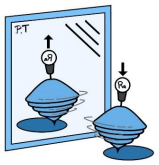


eEDM and SP-S electron-nucleon interaction

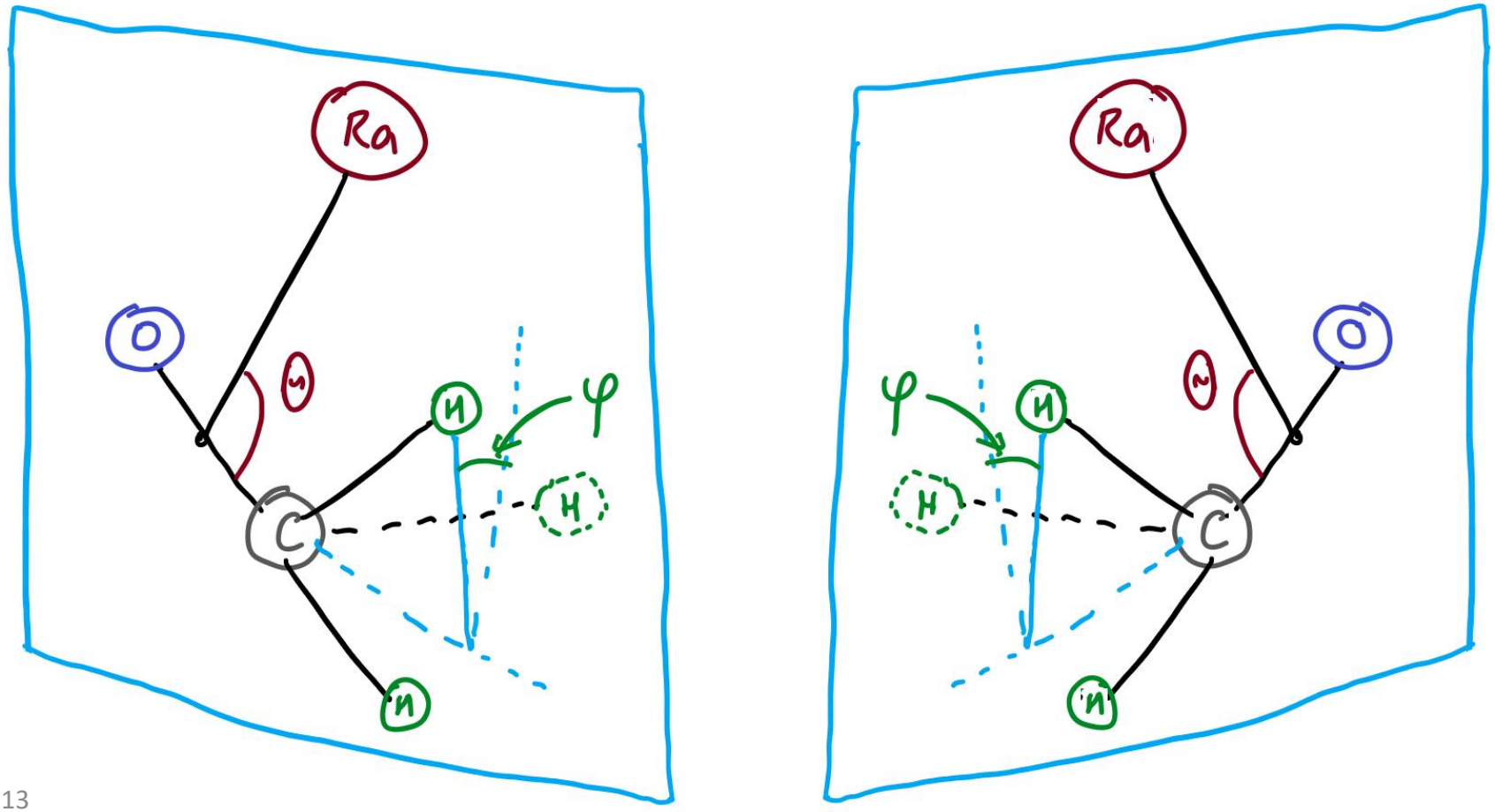
1) Zakharova, A. (2022). Rotating and vibrating symmetric-top molecule RaOCH$_3$ in fundamental P, T-violation searches. Phys.Rev. A

2) Zakharova, A. (2024). Symmetric top molecule YbOCH3 in the fundamental P, T-violation searches. arXiv preprint arXiv:2404.07629. Chem. Phys. Letters (accepted)

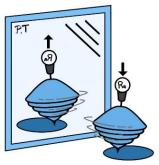
Symmetric top



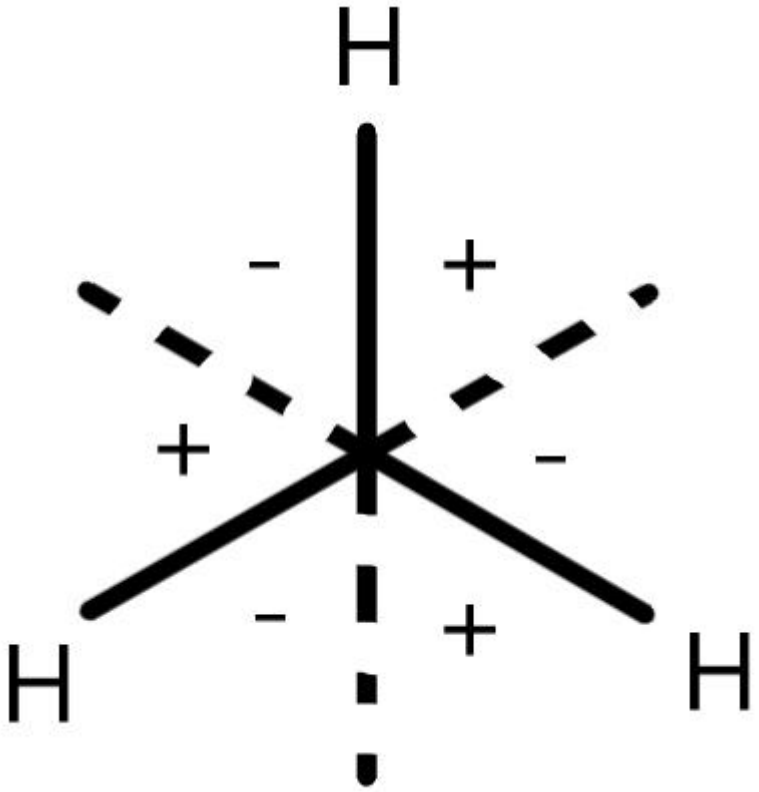
- A **curved symmetrical top**, for example $RaOCH_3$, can be found in chiral configurations
- If the angle **ϕ < 60 degrees**, then no rotation can transform the mirrored molecule into the original one



Symmetries of the $RaOCH_3$ molecule



- The symmetries of the molecule and the oddness of the interaction operator make it possible to determine the nature of the dependence of the average value on the electron wave function on the direction of bending of the molecule



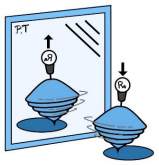
1). Antisymmetry

2). Periodicity 2pi/3

Then the dependence can be represented in terms of the Fourier series

$$E_{a,\tilde{A}} = \sum_{n=1}^{+\infty} E_{a,\tilde{A}}^{(n)} \sin 3n\phi$$

Sensitivity of vibrational states



- Although in RaOCH3 the complete rotational symmetry around the molecular axis is broken (this will play an additional role in removing the degeneracy of the l-doublet), the potential only weakly depends on ϕ

- As a first approximation – a 2-dimensional harmonic oscillator of transverse oscillations

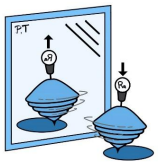
$$|v,l\rangle \mapsto \exp(il\phi)\Phi_{v,l}(\theta), \quad l = -v, -v+2, \ldots + v$$

- The sines in $E_{a,\tilde{A}}$ contain exponents ϕ, multiplying which by these wave functions we shift the oscillatory angular momentum l

- We start with sin 6ϕ - the lowest vibrational states, giving non-trivial matrix elements |3,3> and |3,-3>

$$\hat{E}_{a,\tilde{A}} \to W_{\text{halo}} \left(\frac{1}{2i} \Big| 3,3 \Big\rangle \Big\langle 3,-3 \Big| - \frac{1}{2i} \Big| -3,3 \Big\rangle \Big\langle 3,3 \Big| \right)$$

Influence of the halo on the doublet in an external field



- Consider|3,3> , |3,-3> l-doublet. **Restricted on this subspace the Hamiltonian becomes the 2x2 matrix**

$$H_{v=3,|l|=3} = \begin{pmatrix} 0 & \frac{\Delta E}{2} \\ \frac{\Delta E}{2} & 0 \end{pmatrix} + \begin{pmatrix} d_e\mathcal{E} & 0 \\ 0 & -d_e\mathcal{E} \end{pmatrix} + \begin{pmatrix} 0 & -\frac{i}{2}W_{\text{halo}} \\ \frac{i}{2}W_{\text{halo}} & 0 \end{pmatrix}$$

even Hamiltonian (doublet splitting)

external electric field interacting with the dipole moment of the molecule

interaction with dark matter halo

- It is **hard to extract the effect of interest** as in this case it is quadratic in W_{halo}

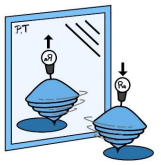
$$\hat{H} = \sum_k h_k\sigma_k \mapsto E_\pm = \pm\sqrt{\sum_k h_k^2} \qquad \delta E \simeq \frac{W_{halo}^2}{4d_e\mathcal{E}}$$

- **In [Norrgard et al, 2018]** P, T-odd interaction of nuclear spin with magnetic field – **similar problem**

Use $\mathcal{E} = \mathcal{E}_0 \cos\Omega t$ to induce transitions between doublet states. Get effect of interest from probability difference for the same state and opposite phases of external field

$$|\Psi\rangle\Big|_{t=0} = |-\rangle, \qquad \frac{P_{|+\rangle}(+\mathcal{E}_0) - P_{|+\rangle}(-\mathcal{E}_0)}{P_{|+\rangle}(+\mathcal{E}_0) + P_{|+\rangle}(-\mathcal{E}_0)} \simeq W_{\text{halo}}\frac{\Omega}{d_e\mathcal{E}_0\Delta E}$$

Molecular computation



Born-Oppenheimer approximation $\Psi \simeq \Psi_{\mathrm{nuc}}(\vec{R}, \hat{r}, \gamma)\psi_{\mathrm{elec}}(\{\vec{r}_i\}, \vec{R}, \hat{r}, \gamma)$

- Start with **SCF** approximation for the electronic wavefunction

$$\Psi^{SCF}_{\alpha_1 \cdots \alpha_N}(x_1, \cdots x_N) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \Psi^1_{\alpha_1}(x_1) & \cdots & \Psi^N_{\alpha_1}(x_1) \\ \vdots & \ddots & \vdots \\ \Psi^1_{\alpha_N}(x_N) & \cdots & \Psi^N_{\alpha_N}(x_N) \end{vmatrix}$$

- **CCSD method** $\Psi = \exp(T)\Psi^{SCF}$ with single and double excitations in T to take into account electronic correlation
- **To reduce the computation** with heavy nuclei the core electrons of the Ra atom are described with help of the **generalized effective core potential (GRECP)**
- **One-center restoration technique:** produce a large basis of solutions of GRECP and full-electron atomic problems with same quantum numbers

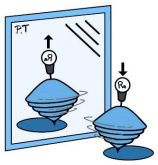
Decompose 2-component solution of the molecular problem using

$$\tilde{f}_{nlj}(r)\chi_{ljm}$$

Get 4-component wavefunction using same coefficients with

$$\begin{pmatrix} f_{nlj}(r)\chi_{ljm} \\ g_{nlj}(r)\chi_{2j-l,jm} \end{pmatrix}$$

Preliminary results



Based on R=5.7 a.u. data and θ bending angle corresponding to the peak of v=3 l=3 rovibrational wavefunction

If similar sensitivities can be achieved with oscillating field as for the current eEDM searches, then **for the dark matter density in our galaxy we could sense**

$$g_{a\bar{e}e} \sim 10^{-15}$$

The results strongly depend on the electronic correlation. These results must be checked for numerical stability.

Thank you for your attention!

https://arxiv.org/a/zakharova_a_1.html

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