Novel Mechanisms of Electric Dipole Moments in Atoms and Molecules

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Conventional Wisdom in the Classification of Atomic/Molecular EDM Experiments

Diamagnetic systems (contain *no* unpaired electrons) are mainly sensitive to **hadronic** sources of CP violation – e.g., **Hg**, **Xe**, **n**

Paramagnetic systems (contain *one or more* unpaired electrons) are mainly sensitive to **leptonic** sources of CP violation – e.g., **ThO**, **HfF^+**, **YbF**, **Tl, Cs**

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For **semi-leptonic** sources of CP violation, the story is more complicated – the "classification" generally depends on whether the interactions involve mainly electron spin or nuclear spin

Leptonic CP Violation in Paramagnetic Molecules

Over the past decade, molecular experiments have improved the sensitivity to electron EDM d_e by more than 100-fold:

^{232}ThO bound: $|d_e| < 10^{-29}\ e\ \mathrm{cm}$

[Andreev *et al*. (ACME collaboration), *Nature* **562**, 355 (2018)]

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What about sensitivity of paramagnetic systems to hadronic CP violation?

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Hadronic CP Violation in Diamagnetic Atoms

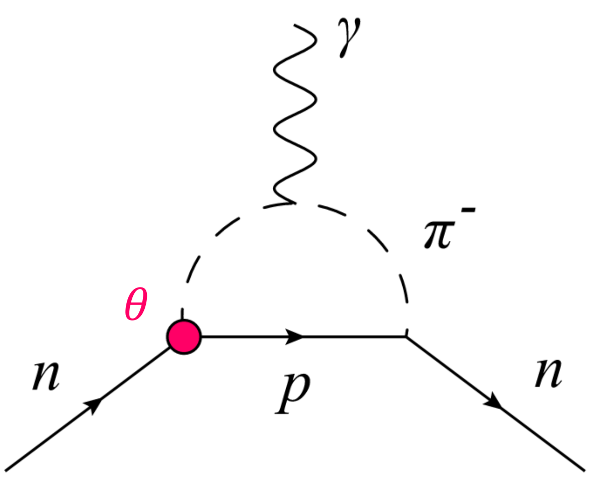
Nucleon EDMs: [Crewther, Di Vecchia, Veneziano, Witten, *PLB* **88**, 123 (1979)]

Intranuclear forces: [Haxton, Henley, *PRL* **51**, 1937 (1983)],

[O. Sushkov, Flambaum, Khriplovich, *JETP* **60**, 873 (1984)]

Illustrative example: $\mathcal{L} = \theta \frac{g_s^2}{32\pi^2} G\tilde{G}$

Nucleon EDMs



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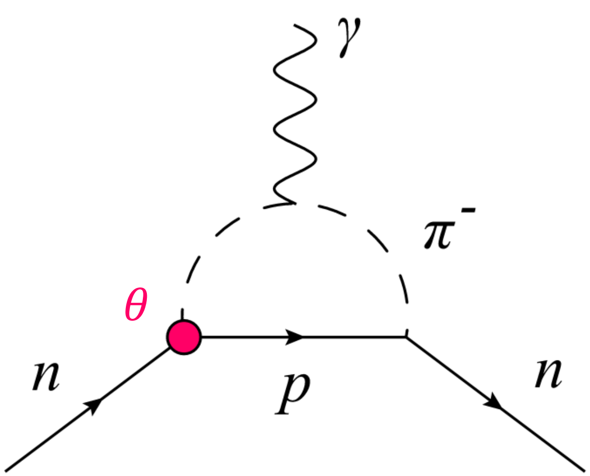
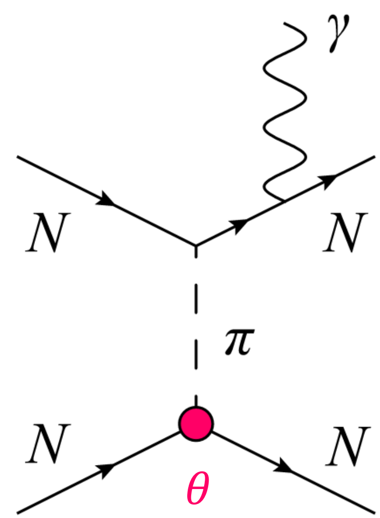


CP-violating intranuclear forces

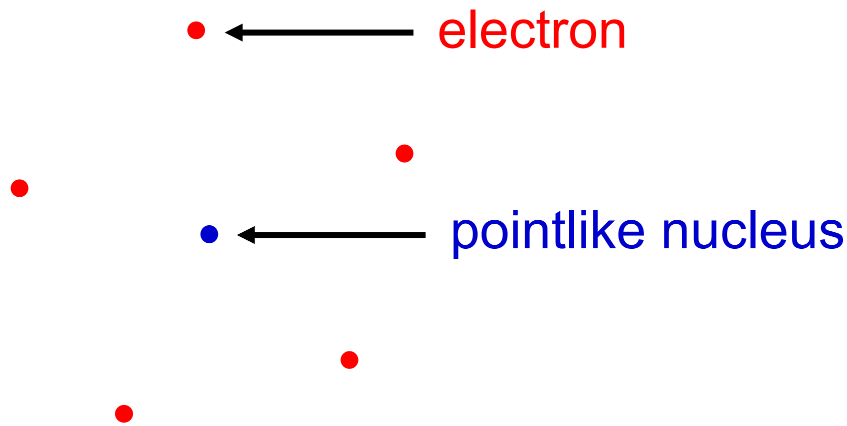


In nuclei, **tree-level** CP-violating intranuclear forces dominate over **loop-induced** nucleon EDMs [loop factor $= 1/(8\pi^2)$].

Schiff's Theorem

[Schiff, *Phys. Rev.* **132**, 2194 (1963)]

Schiff's Theorem: "In a neutral atom made up of point-like non-relativistic charged particles (interacting only electrostatically), the constituent EDMs are screened from an external electric field."



Classical explanation for nuclear EDM: A neutral atom does not accelerate in an external electric field!

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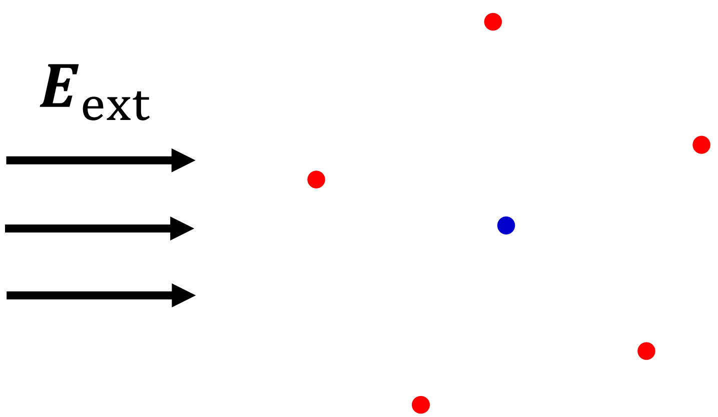

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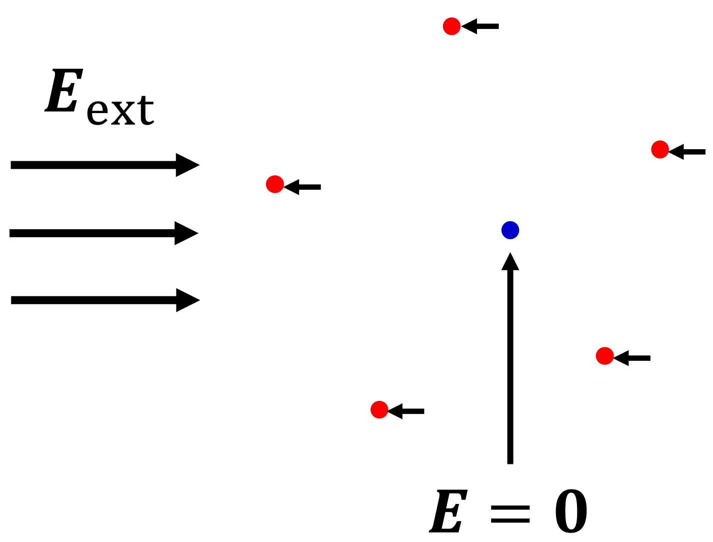

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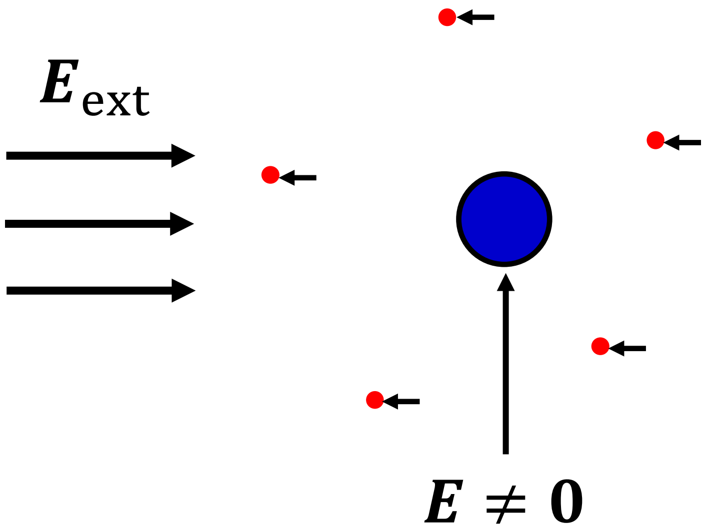
Lifting of Schiff's Theorem

[Sandars, *PRL* **19**, 1396 (1967)],

[O. Sushkov, Flambaum, Khriplovich, *JETP* **60**, 873 (1984)]

In real (heavy) atoms: Incomplete screening of external electric field due to finite nuclear size, parametrised by **nuclear Schiff moment**

("screened nuclear EDM")

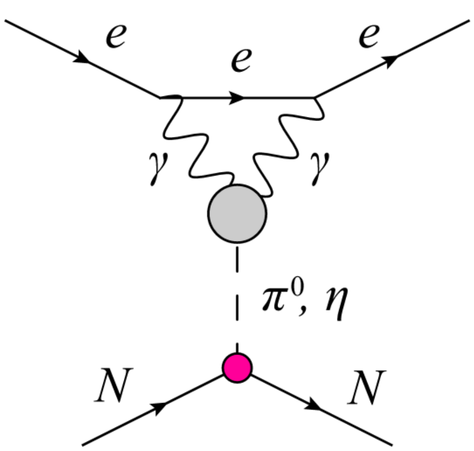


Hadronic CP Violation in Paramagnetic Molecules

[Flambaum, Pospelov, Ritz, Stadnik, *PRD* **102**, 035001 (2020)]

- Hadronic CP-violating effects arise via 2γ-exchange starting at **2-loop level**

LO: $\mathcal{O}\left(m_\pi^{-2}\right)$



CP-odd nucleon polarisabilities ($\propto \boldsymbol{E}\cdot\boldsymbol{B}$)

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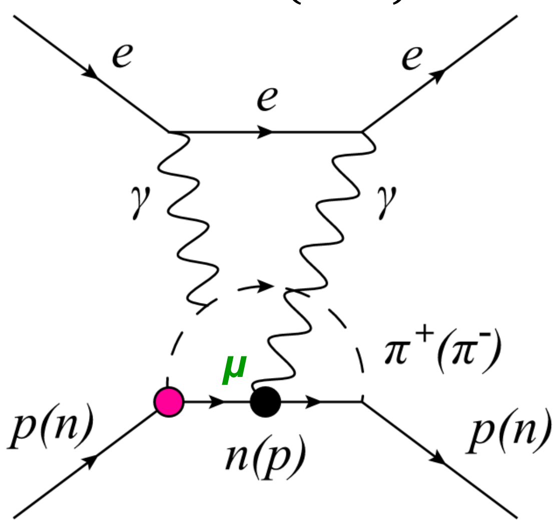

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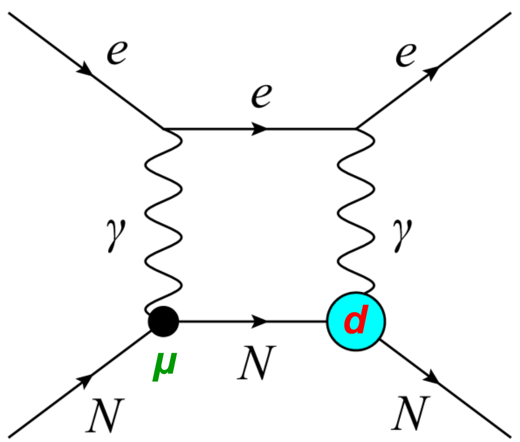
$(\boldsymbol{\mu}-\boldsymbol{d})$**:** $\mathcal{O}[\ln(A)/p_{\mathrm{F}}]$

[Fermi-gas model]



Internal nuclear excitations

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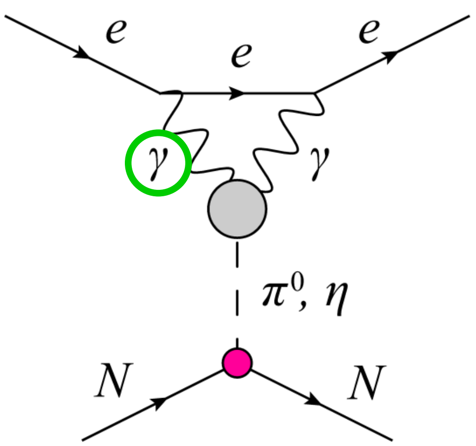

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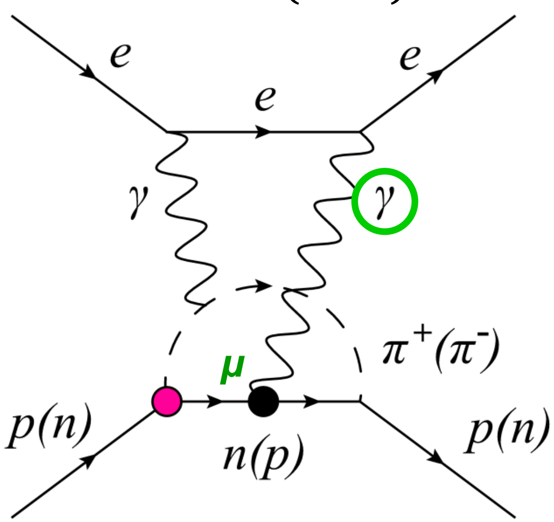

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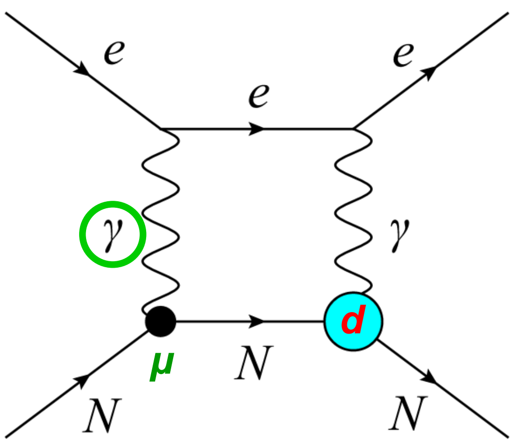

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  - **$\mathcal{O}(A)$-enhanced** CP-odd nuclear *scalar* polarisability

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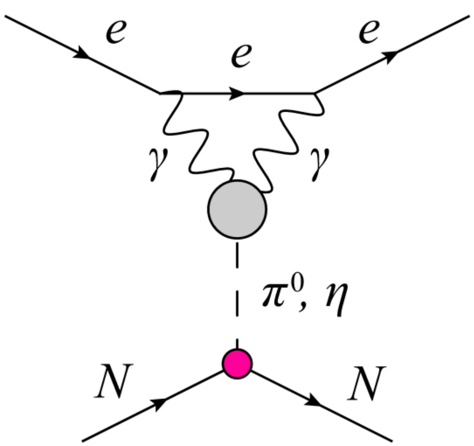

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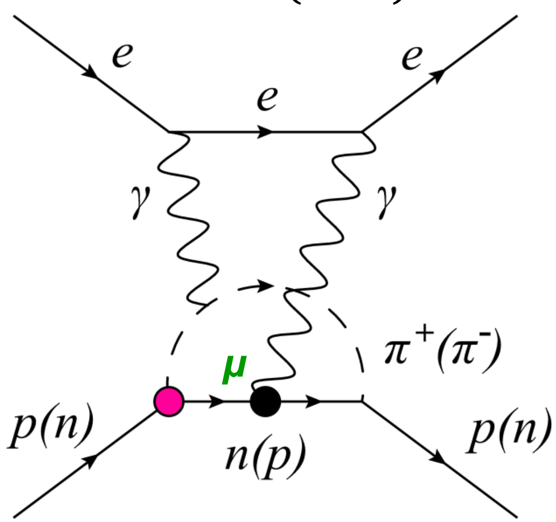

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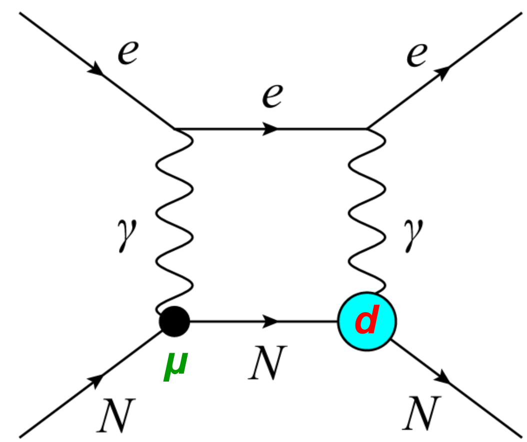

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  - $\boldsymbol{\mathcal{O}(A)}$**-enhanced** CP-odd nuclear *scalar* polarisability
  - Operative even in *spinless* nuclei (e.g., ^{232}ThO, ^{180}HfF^{+})

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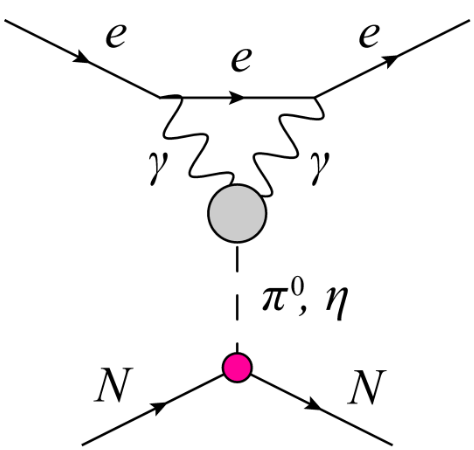

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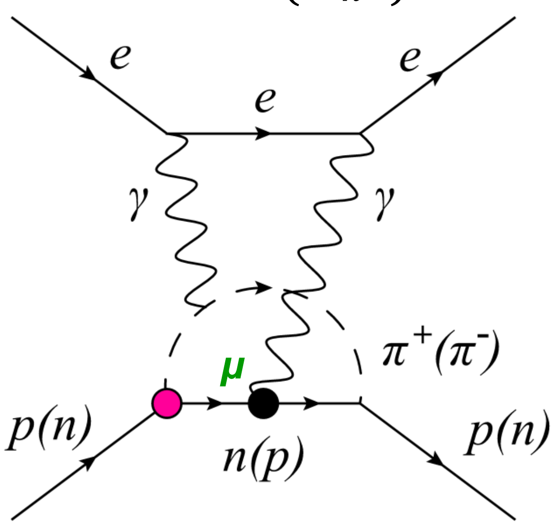

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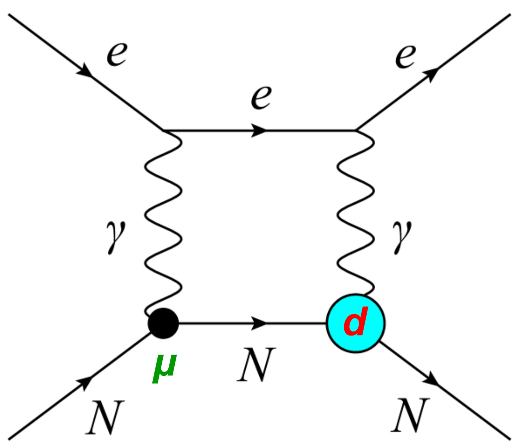

Internal nuclear excitations

Isoscalar CP-Odd π-N Coupling $\mathcal{L} = \bar{g}^{(1)}_{\pi NN}\pi^0\bar{N}N$

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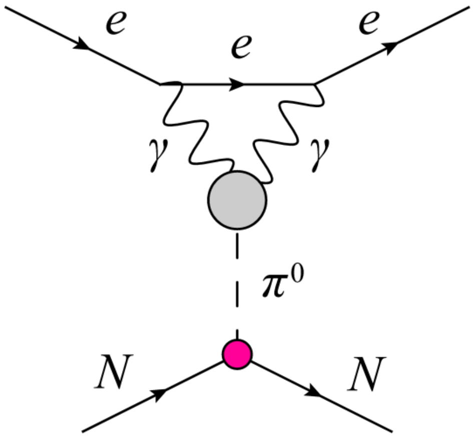

π^0 exchange

In molecules with *spinless* nuclei (e.g., ^{232}ThO, ^{180}HfF$^+$), effect dominated by a **"bulk"** property of the nucleus that grows with A in a regular manner, with *no contribution* from the nuclear Schiff moment mechanism (needs $I \neq 0$)

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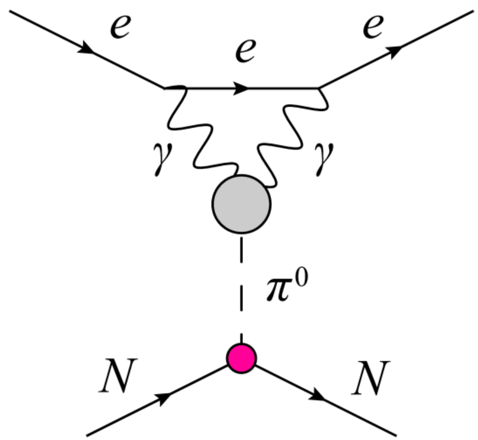

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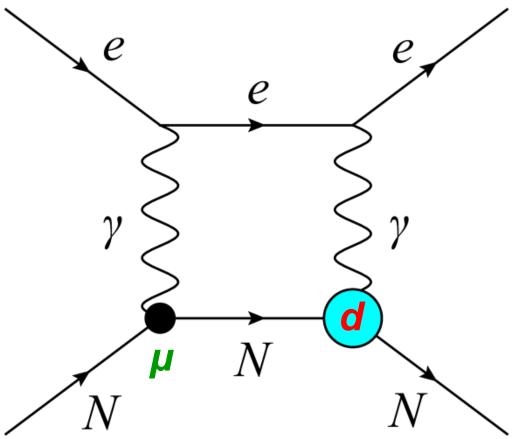
=> Clean bounds, since less sensitivity to details of nuclear structure

(cf. strong sensitivity of ^{199}Hg Schiff moment to assumptions about underlying nuclear structure – different models give different signs for sensitivity coefficient!)

Nucleon EDMs $\mathcal{L} = -\frac{i}{2} d_N \bar{N} F_{\mu\nu} \sigma^{\mu\nu} \gamma_5 N$

[Flambaum, Pospelov, Ritz, Stadnik, *PRD* **102**, 035001 (2020)]



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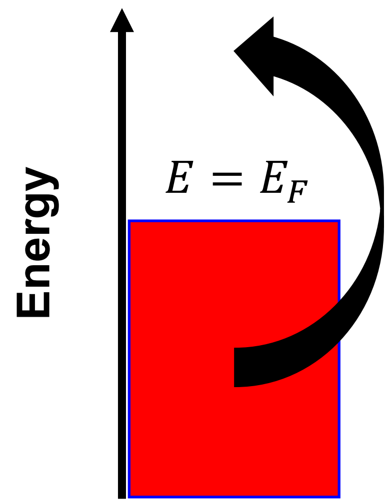

Continuum

Excitations to continuum above Fermi surface: $\sim \ln(A)/p_{\mathrm{F}}$ [Fermi-gas model]

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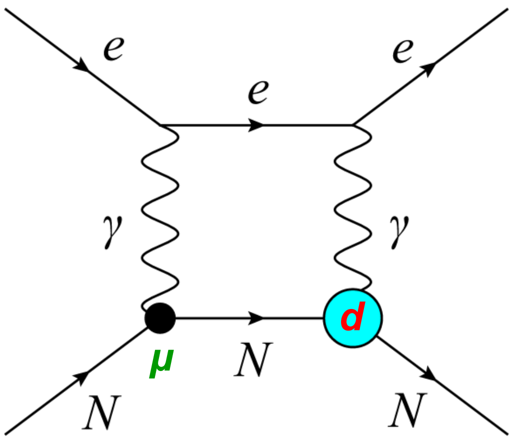

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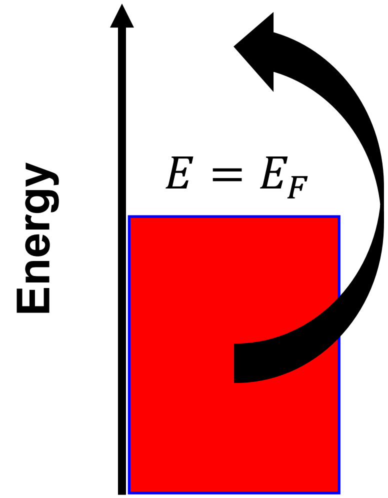

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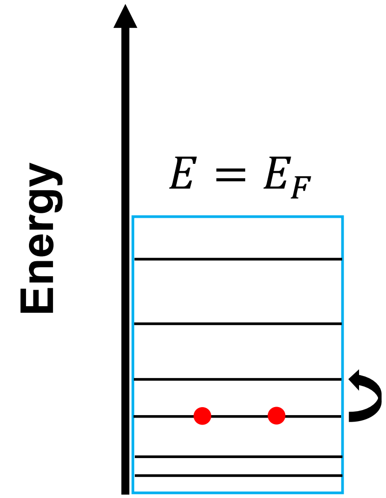

Discrete

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Discrete transitions between L-S doublets: $\sim[\mathcal{O}(10)/A] \times (1/\Delta E_{\mathrm{nucl}})$

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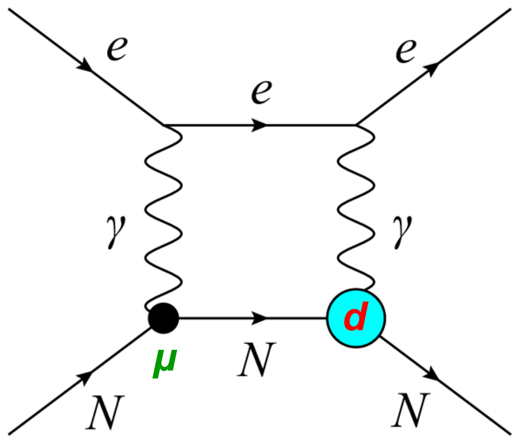

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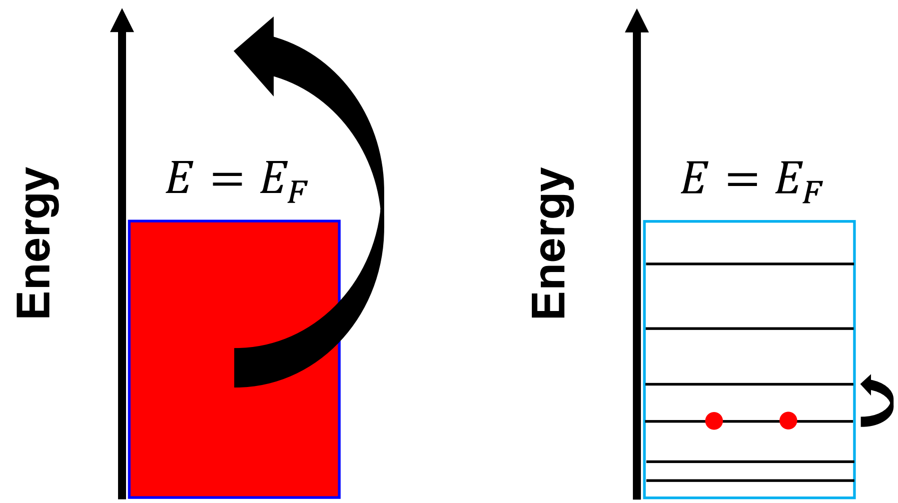

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For $A \sim 200$ and $\Delta E_{\mathrm{nucl}} \sim \mathrm{several\ MeV}$, the two contributions are comparable in size (and of the same sign)

QCD Vacuum Angle $\mathcal{L} = \theta \frac{g_s^2}{32\pi^2} G\tilde{G}$

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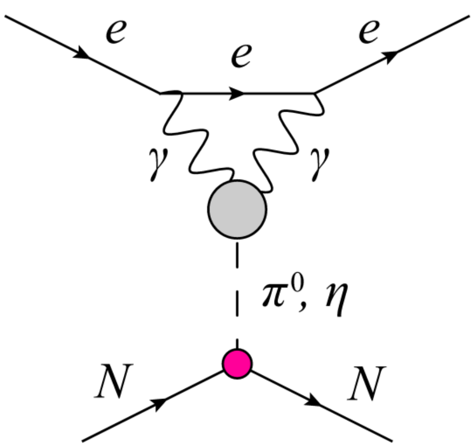

π^0, η contributions: ***opposite*** sign (near-cancellation in heavy nuclei)

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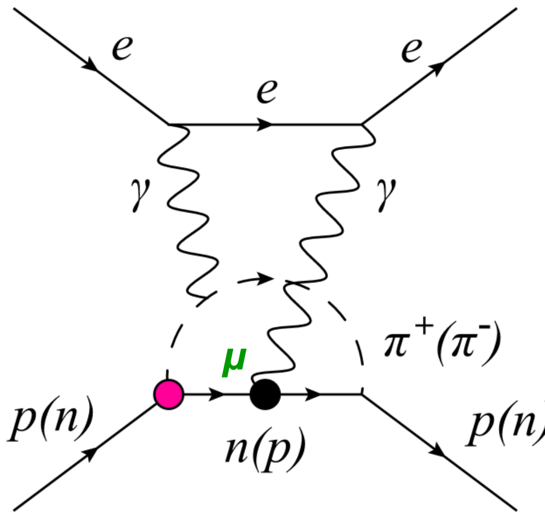

+

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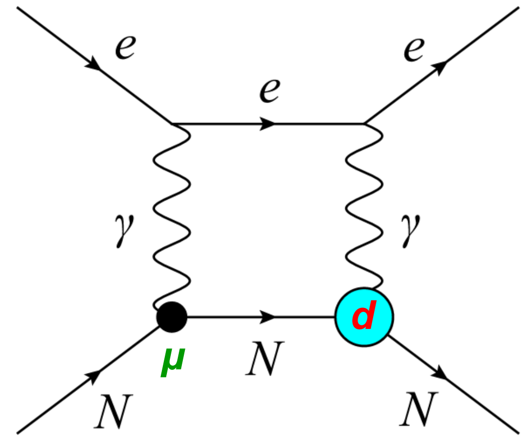

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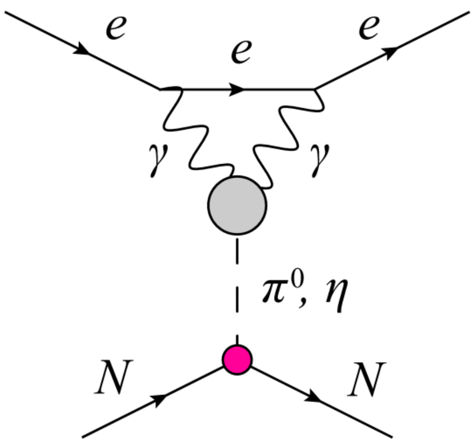

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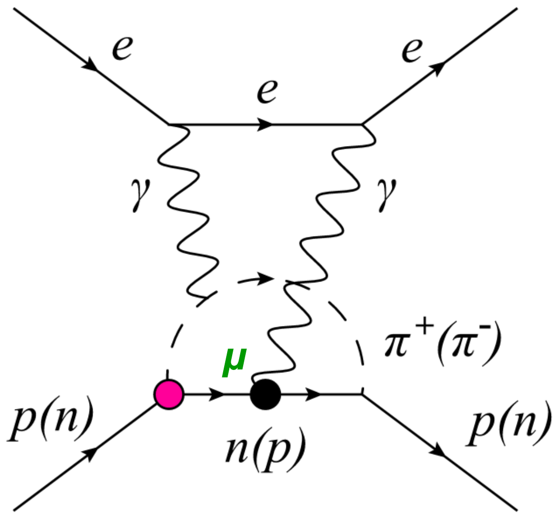

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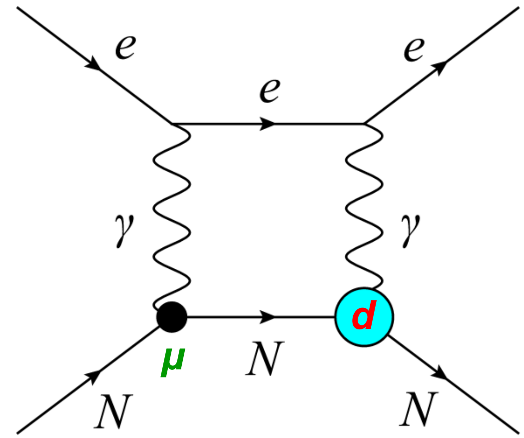

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For $Z \sim 80$ & $A \sim 200$: $C_\mathrm{SP}(\theta) \approx \left[0.1_\mathrm{LO} + 1.0_\mathrm{NLO} + 1.7_{(\mu-d)}\right] \times 10^{-2}\theta \approx 0.03\theta$

$$\mathcal{L}_\mathrm{contact} = -\frac{G_F C_\mathrm{SP} \bar{N} N \bar{e} i \gamma_5 e}{\sqrt{2}}$$

Bounds on Hadronic CP Violation Parameters

ThO bounds: [Flambaum, Pospelov, Ritz, Stadnik, *PRD* **102**, 035001 (2020)]

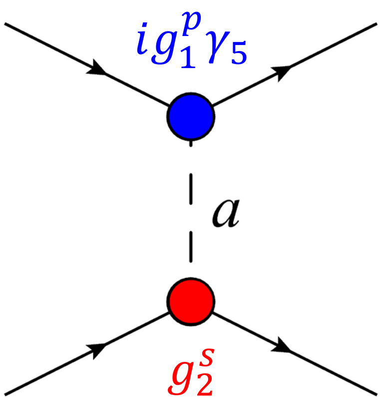
System	$\left\|\bar{g}_{\pi NN}^{(1)}\right\|$	$\left\|\tilde{d}_u - \tilde{d}_d\right\|$ (cm)	$\left\|d_p\right\|$ (e cm)	$\|\theta\|$
ThO	$\mathbf{4 \times 10^{-10}}$	$\mathbf{2 \times 10^{-24}}$	$\mathbf{2 \times 10^{-23}}$	$\mathbf{3 \times 10^{-8}}$
n	1.1×10^{-10}	5×10^{-25}	—	2.0×10^{-10}
Hg	1×10^{-12}	5×10^{-27}	2.0×10^{-25}	1.5×10^{-10}
Xe	6.7×10^{-8}	3×10^{-22}	3.2×10^{-22}	3.2×10^{-6}

* These limits can formally be null within nuclear uncertainties

Current bounds from paramagnetic molecules are $\sim 10 - 100$ times weaker than from diamagnetic Hg & n, but are $\sim 10 - 100$ times stronger than bounds from diamagnetic Xe

P,T-Violating Forces Mediated by Dark Bosons

[Stadnik, Dzuba, Flambaum, *PRL* **120**, 013202 (2018)],
[Dzuba, Flambaum, Samsonov, Stadnik, *PRD* **98**, 035048 (2018)]



$$\mathcal{L}_{\mathrm{int}} = a\bar{f}\left(g_f^s + ig_f^p\gamma_5\right)f$$

$$V(r) \approx \frac{g_1^p g_2^s}{8\pi m_1}\boldsymbol{\sigma}_1 \cdot \hat{\boldsymbol{r}}\left(\frac{m_a}{r} + \frac{1}{r^2}\right)e^{-m_a r}$$

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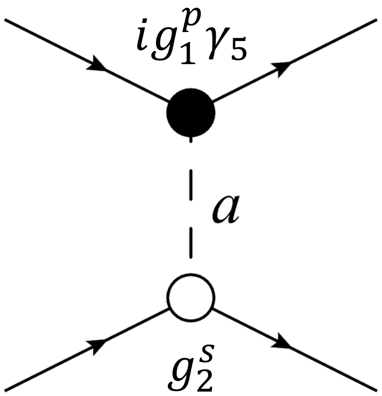


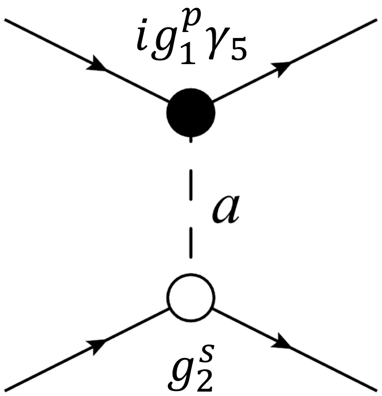
$$\mathcal{L}_{\text{int}} = a\bar{f}\left(g_f^s + ig_f^p\gamma_5\right)f$$

$$V(r) \approx \frac{g_1^p g_2^s}{8\pi m_1}\boldsymbol{\sigma}_1 \cdot \hat{\boldsymbol{r}}\left(\frac{m_a}{r} + \frac{1}{r^2}\right)e^{-m_a r}$$

P,T-violating forces => Atomic and Molecular EDMs

P,T-Violating Forces Mediated by Dark Bosons

[Stadnik, Dzuba, Flambaum, *PRL* **120**, 013202 (2018)],
[Dzuba, Flambaum, Samsonov, Stadnik, *PRD* **98**, 035048 (2018)]



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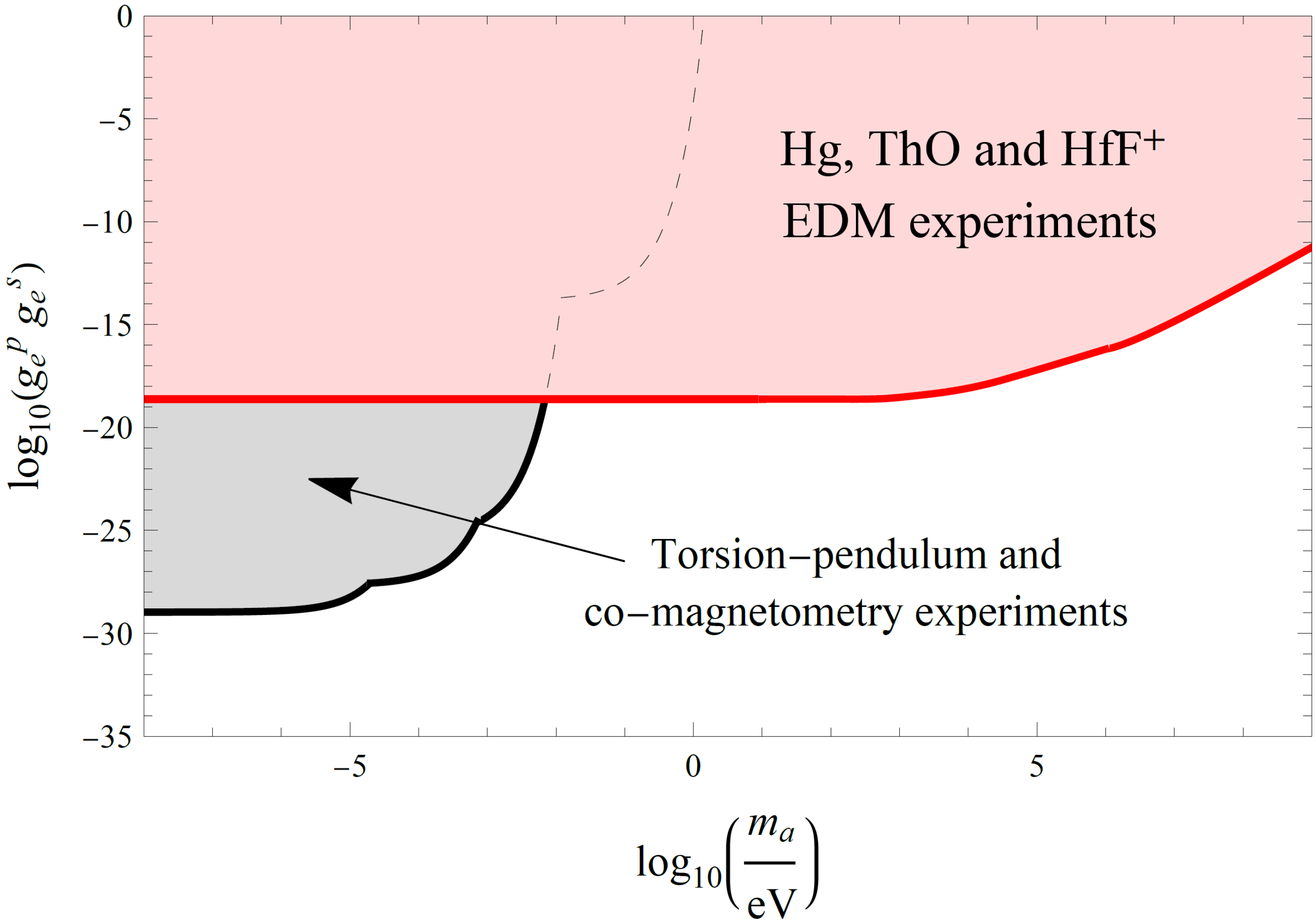
P,T-violating forces => Atomic and Molecular EDMs

If exchanged boson is sufficiently low-mass, then P,T-violating forces are long-range on the scale of atom/molecule, and the non-vanishing contribution arises from the Thomas-Fermi length scale $r \sim a_\mathrm{B}/Z^{1/3}$

Constraints on Scalar-Pseudoscalar Electron-Electron Interaction

EDM constraints: [Stadnik, Dzuba, Flambaum, *PRL* **120**, 013202 (2018)]

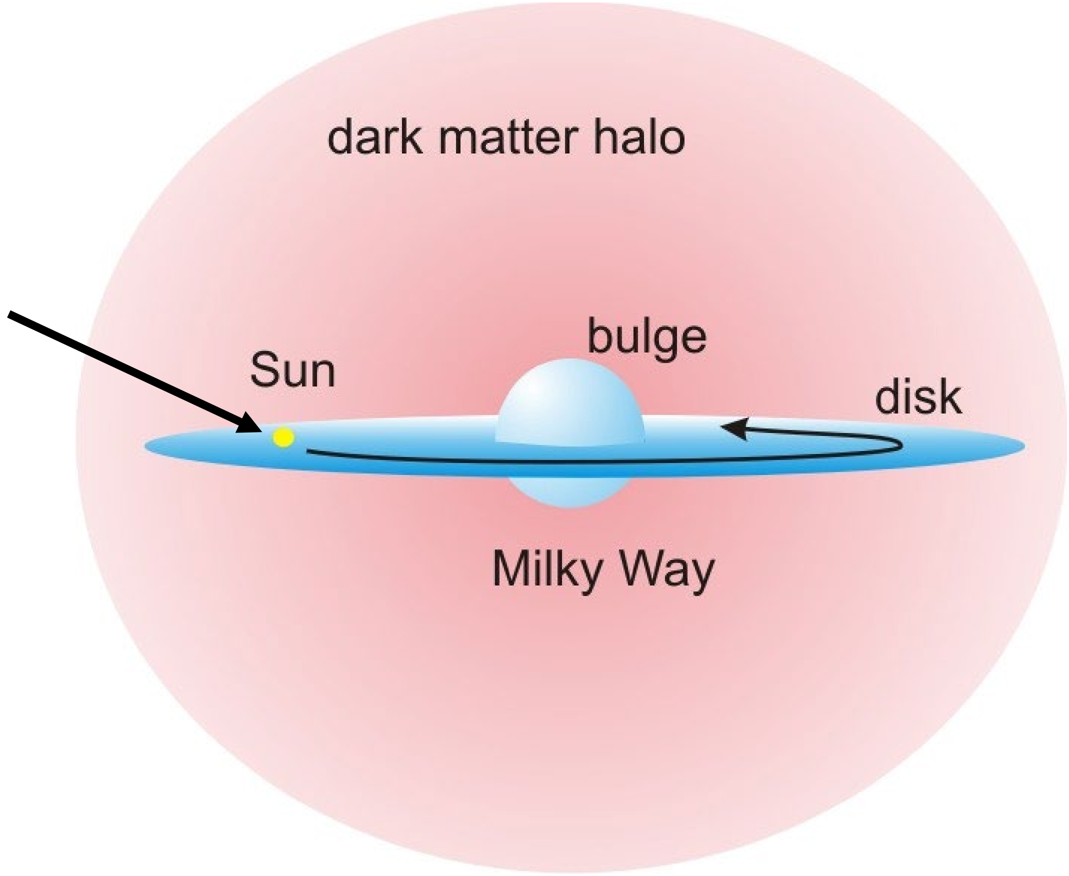


Dark Matter

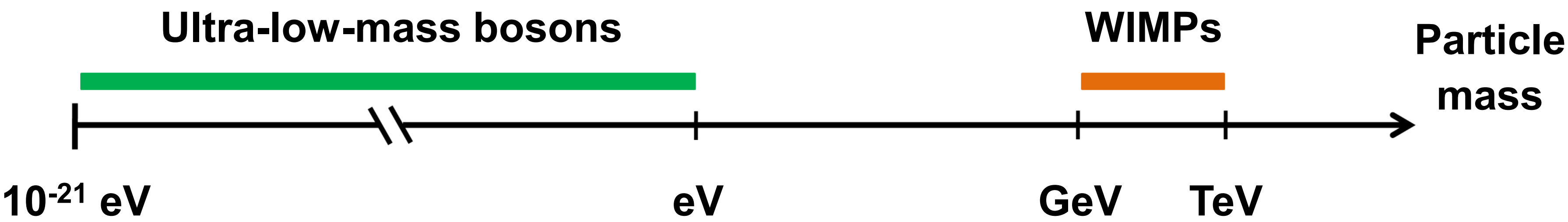
Strong astrophysical evidence for existence of **dark matter** (~5 times more dark matter than ordinary matter)

$$\rho_{\mathrm{DM}} \approx 0.4\ \mathrm{GeV/cm^3}$$

$$v_{\mathrm{DM}} \sim 300\ \mathrm{km/s}$$



Dark Matter



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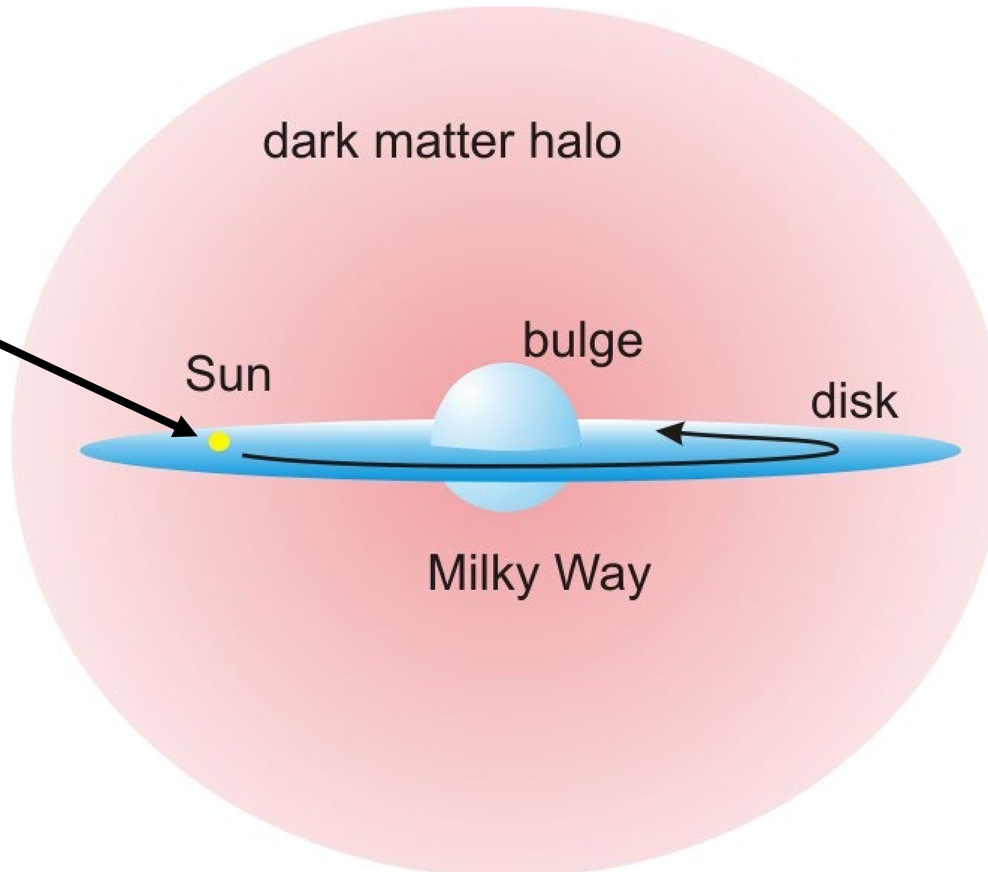

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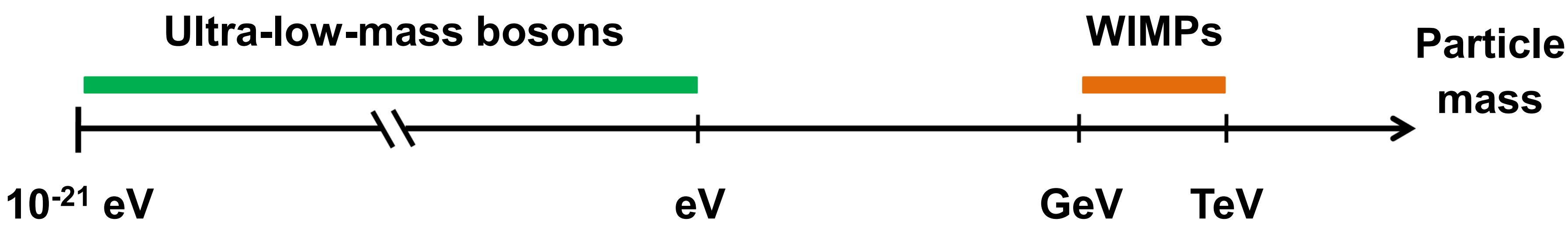
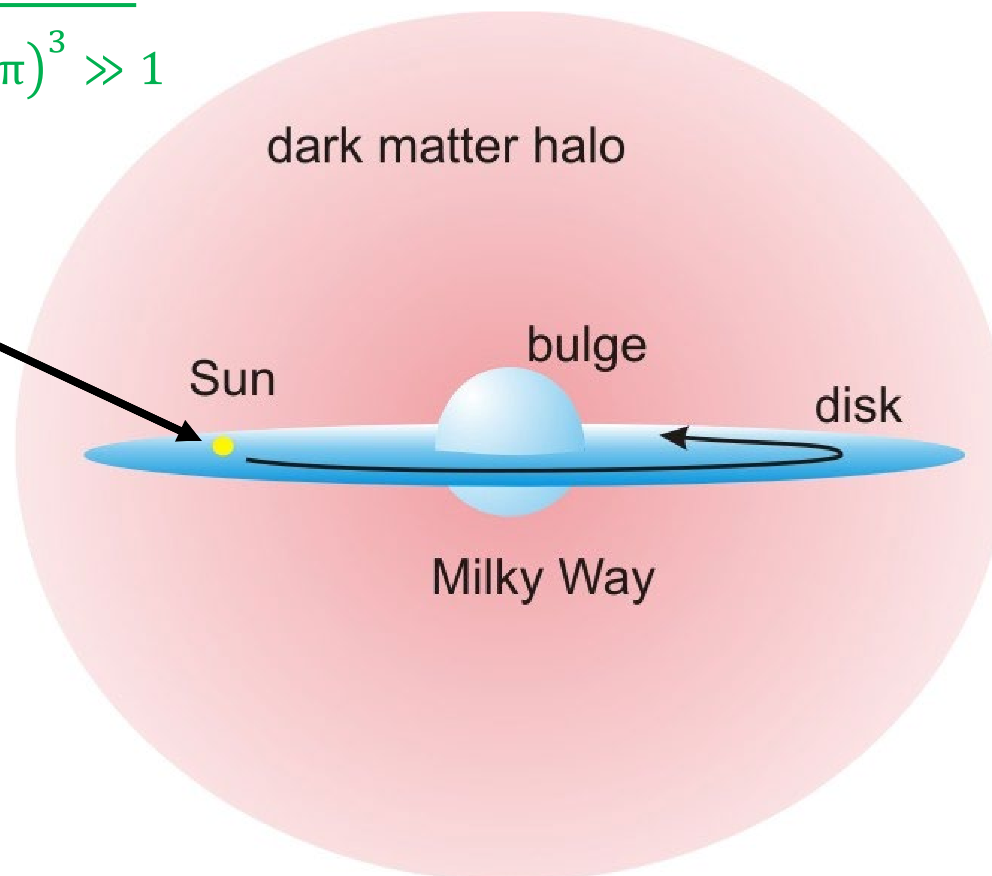


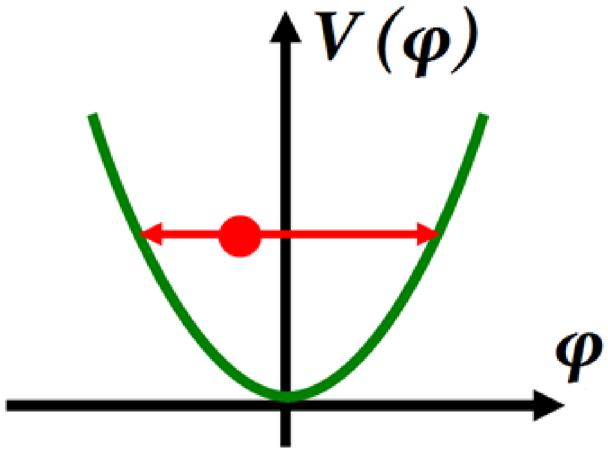
Classical field: $n_{\mathrm{DM}}\left(\lambda_{\mathrm{dB,DM}}/2\pi\right)^3 \gg 1$

$\rho_{\mathrm{DM}} \approx 0.4\ \mathrm{GeV/cm^3}$

$v_{\mathrm{DM}} \sim 300\ \mathrm{km/s}$



Oscillating Electric Dipole Moments



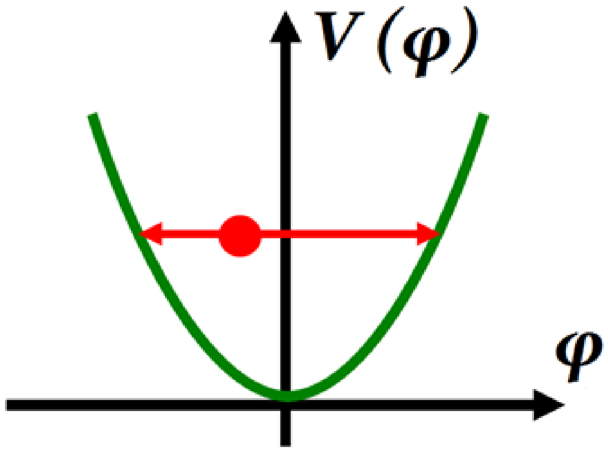
$$\varphi(t) = \varphi_0 \cos\left(m_\varphi c^2 t/\hbar\right),$$

$$\langle \rho_\varphi \rangle \approx m_\varphi^2 \varphi_0^2/2$$

Oscillating Electric Dipole Moments

Nucleons: [Graham, Rajendran, *PRD* **84**, 055013 (2011)]

Atoms and molecules: [Stadnik, Flambaum, *PRD* **89**, 043522 (2014)]; [Flambaum, Pospelov, Ritz, Stadnik, *PRD* **102**, 035001 (2020)]



$$\varphi(t) = \varphi_0 \cos\left(m_\varphi c^2 t/\hbar\right),$$

$$\langle \rho_\varphi \rangle \approx m_\varphi^2 \varphi_0^2 / 2$$

$$\mathcal{L} = \frac{g_s^2}{32\pi^2} \frac{C_G}{f_a} \varphi_0 \cos\left(m_\varphi t\right) G\tilde{G} \quad \Rightarrow \quad \boldsymbol{d}(t) \propto \boldsymbol{J} \cos\left(m_\varphi t\right), \quad H_{\mathrm{EDM}}(t) = \boldsymbol{d}(t) \cdot \boldsymbol{E}$$

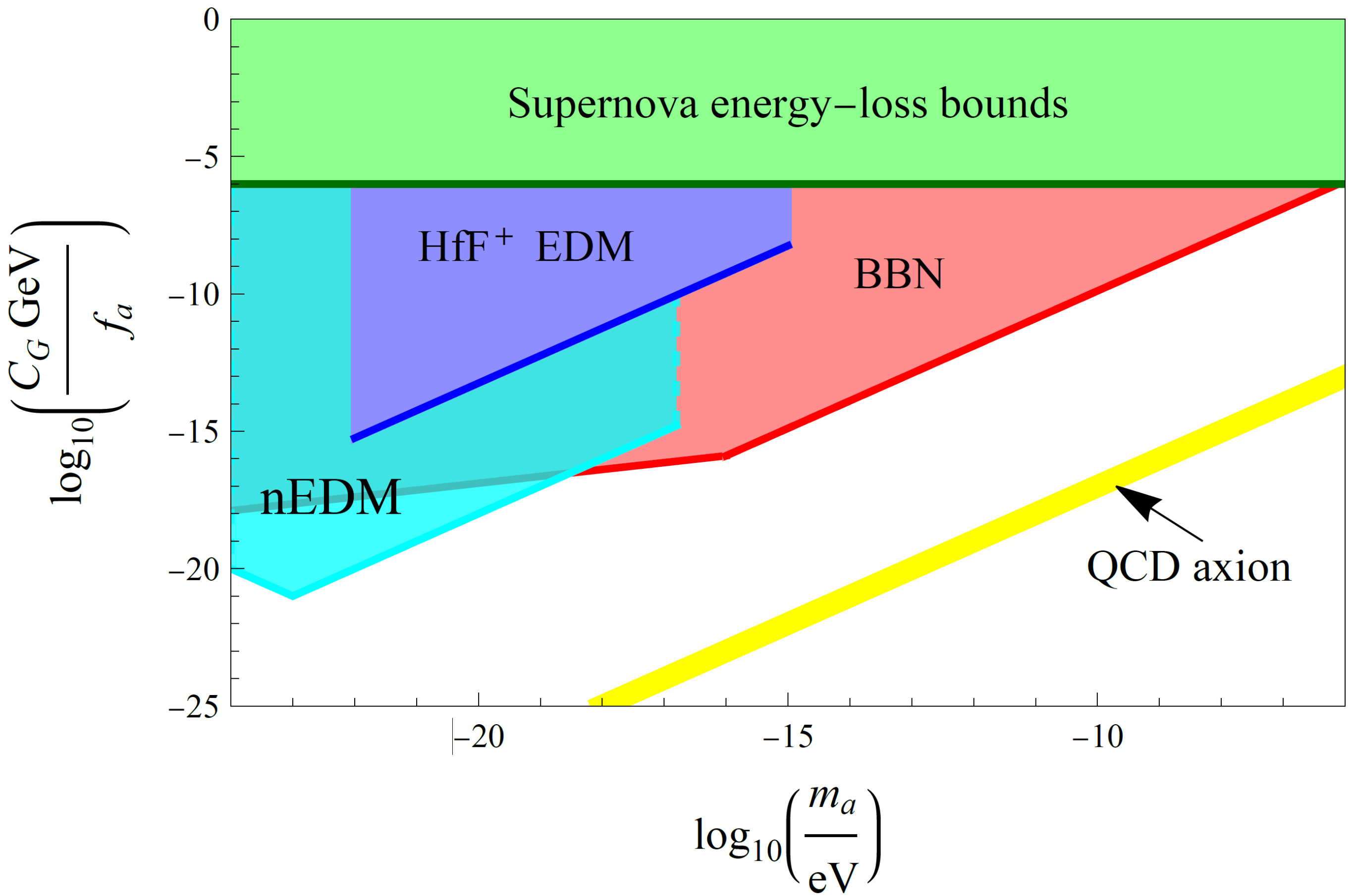
$$\text{cf. } \mathcal{L} = \frac{g_s^2}{32\pi^2} \theta G\tilde{G} \quad \Rightarrow \quad \theta \leftrightarrow \frac{C_G}{f_a} \varphi_0 \cos\left(m_\varphi t\right)$$

Constraints on Interaction of Axion Dark Matter with Gluons

nEDM constraints: [nEDM collaboration, *PRX* **7**, 041034 (2017)]

HfF⁺ EDM constraints: [Roussy *et al.*, *PRL* **126**, 171301 (2021)]

3 orders of magnitude improvement!



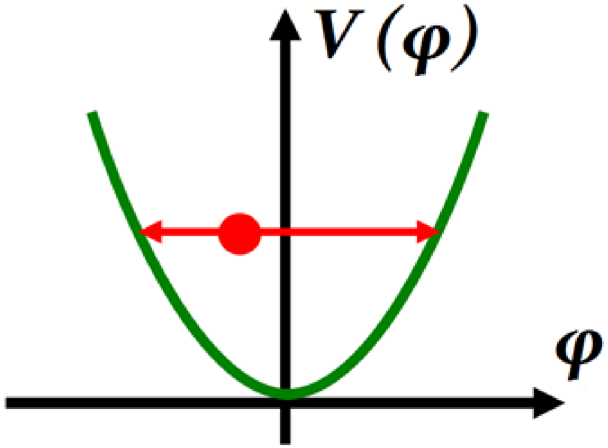
Summary

- Paramagnetic atoms and molecules are sensitive to hadronic sources of CP violation via two-photon-exchange processes, leading to novel and independent constraints using data from ThO (ACME experiment)
- Exchange of low-mass dark bosons within atoms/molecules can induce "long-range" P,T-violating forces, generating atomic/molecular EDMs – limits from ThO, HfF^+, Hg and other EDM experiments
- Ultra-low-mass bosonic dark matter in the form of an oscillating classical field can induce oscillating-in-time EDMs – limits from nEDM and HfF^+

Back-Up Slides

Low-mass Spin-0 Dark Matter

- Low-mass spin-0 particles form a coherently oscillating classical field $\varphi(t) = \varphi_0 \cos\left(m_\varphi c^2 t/\hbar\right)$, with energy density $\langle \rho_\varphi \rangle \approx m_\varphi^2 \varphi_0^2/2 \ \ (\rho_{\mathrm{DM,local}} \approx 0.4\ \mathrm{GeV/cm^3})$



$$V(\varphi) = \frac{m_\varphi^2 \varphi^2}{2}$$

$$\ddot{\varphi} + m_\varphi^2 \varphi \approx 0$$

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- *Coherently* oscillating field, since *cold* $(E_\varphi \approx m_\varphi c^2)$
- $\Delta E_\varphi / E_\varphi \sim \langle v_\varphi^2 \rangle / c^2 \sim 10^{-6} \;\; \Rightarrow \;\; \tau_{\mathrm{coh}} \sim 2\pi/\Delta E_\varphi \sim 10^6 T_{\mathrm{osc}}$

$\uparrow$

$v_{\mathrm{DM}} \sim 300\ \mathrm{km/s}$

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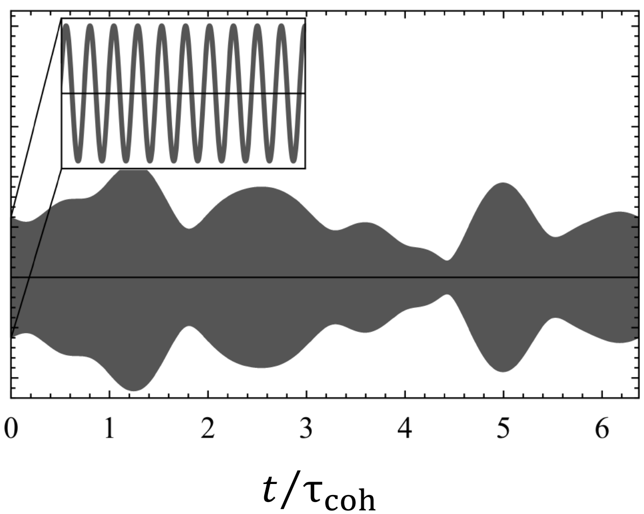
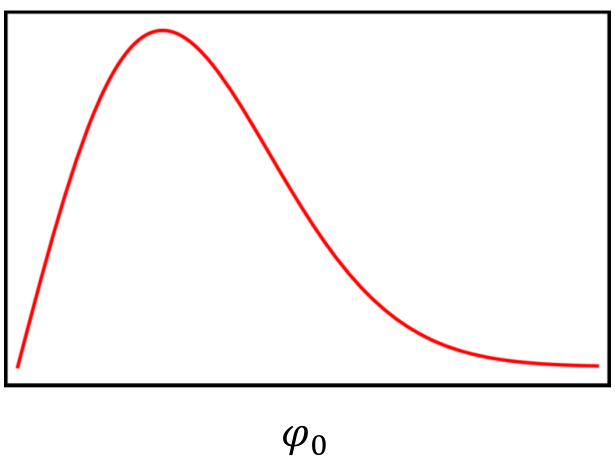
Evolution of φ_0 with time



Probability distribution function of φ_0 (e.g., Rayleigh distribution)



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- $10^{-21}\ \mathrm{eV} \lesssim m_\varphi \lesssim 1\ \mathrm{eV} \;\; \Leftrightarrow \;\; 10^{-7}\ \mathrm{Hz} \lesssim f_{\mathrm{DM}} \lesssim 10^{14}\ \mathrm{eV}$

$T_{\mathrm{osc}} \sim 1\ \mathrm{month}$

IR frequencies

Lyman-α forest measurements [suppression of structures for $L \lesssim \mathcal{O}(\lambda_{\mathrm{dB},\varphi})$]

[Related figure-of-merit: $\lambda_{\mathrm{dB},\varphi}/2\pi \leq L_{\mathrm{dwarf\ galaxy}} \sim 100\ \mathrm{pc} \;\; \Rightarrow \;\; m_\varphi \gtrsim 10^{-21}\ \mathrm{eV}$]

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↑

Lyman-α forest measurements [suppression of structures for $L \lesssim \mathcal{O}(\lambda_{\mathrm{dB},\varphi})$]

- *Wave-like* signatures [cf. *particle-like* signatures of WIMP DM]

Low-mass Spin-0 Dark Matter

Dark Matter →

Pseudoscalars (Axions):

$\varphi \xrightarrow{P} -\varphi$

More traditional axion detection methods tend to focus on the ***electromagnetic*** *coupling*

Here I focus on relatively new detection methods based on ***non-electromagnetic*** *couplings* →

Time-varying spin-dependent effects

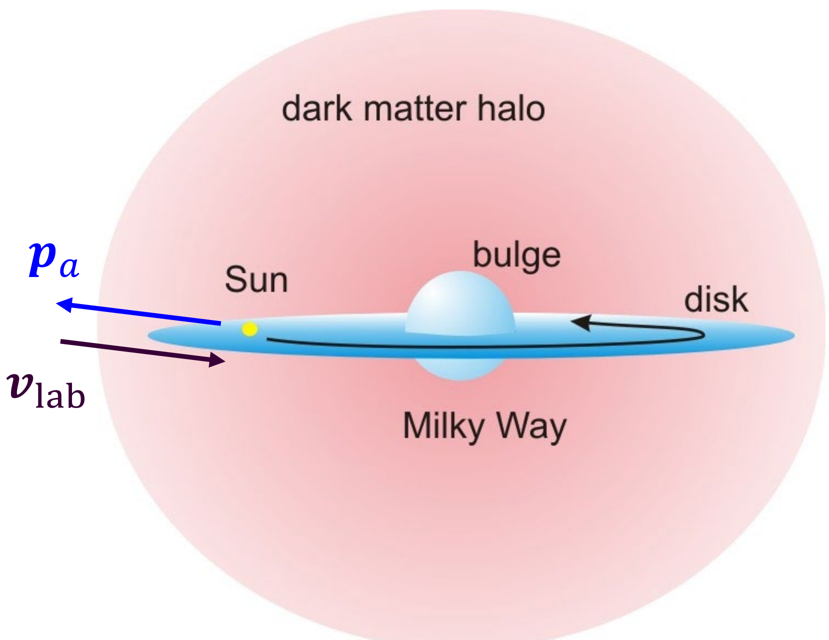
- Co-magnetometers
- Particle g-factors
- Spin-polarised torsion pendula
- Spin resonance (NMR, ESR)

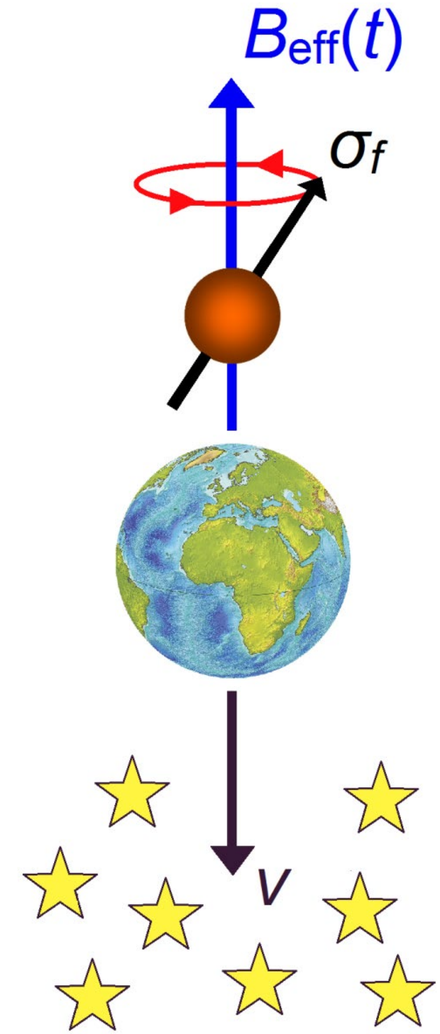
"Axion Wind" Spin-Precession Effect

[Flambaum, talk at *Patras Workshop*, 2013], [Stadnik, Flambaum, *PRD* **89**, 043522 (2014)]

$$\mathcal{L}_f = -\frac{C_f}{2f_a}\partial_i[a_0\cos(m_a t - \boldsymbol{p}_a \cdot \boldsymbol{x})]\bar{f}\gamma^i\gamma^5 f$$

$$\Rightarrow \quad H_{\mathrm{wind}}(t) = \boldsymbol{\sigma}_f \cdot \boldsymbol{B}_{\mathrm{eff}}(t) \propto \boldsymbol{\sigma}_f \cdot \boldsymbol{p}_a \sin(m_a t)$$



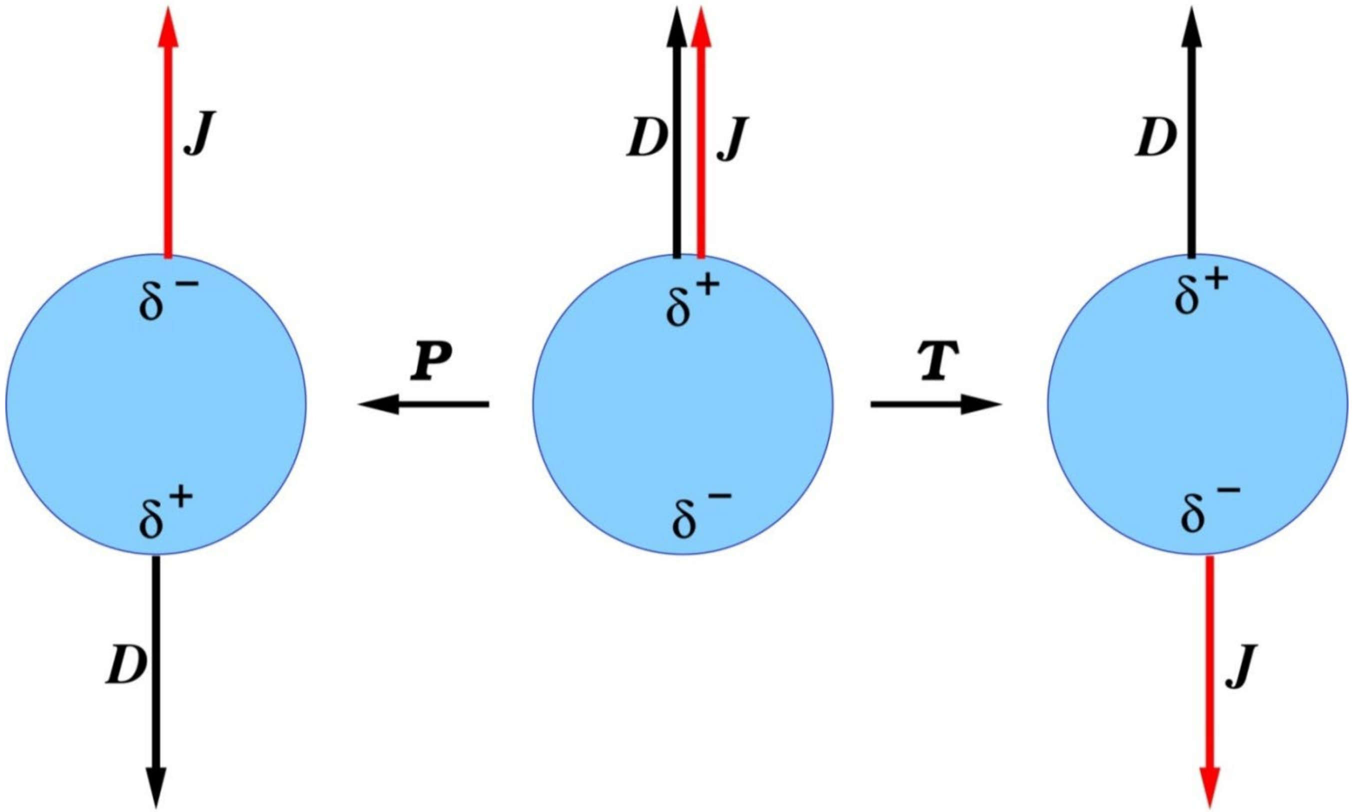


Oscillating Electric Dipole Moments

Nucleons: [Graham, Rajendran, *PRD* **84**, 055013 (2011)]

Atoms and molecules: [Stadnik, Flambaum, *PRD* **89**, 043522 (2014)]

Electric Dipole Moment (EDM) = parity (P) and time-reversal-invariance (T) violating electric moment



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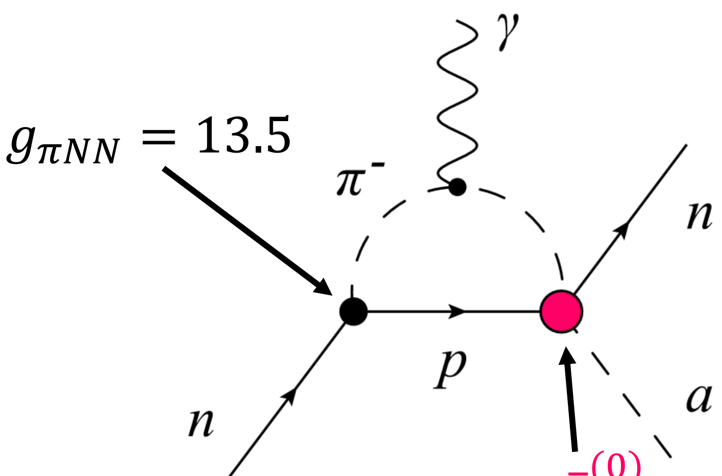
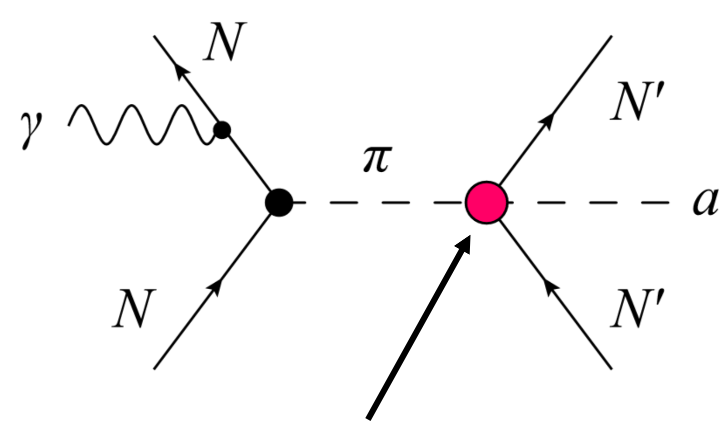
$$\mathcal{L}_G = \frac{C_G g^2}{32\pi^2 f_a} a_0 \cos(m_a t) G\tilde{G} \quad \Rightarrow \quad H_{\mathrm{EDM}}(t) = \boldsymbol{d}(t) \cdot \boldsymbol{E}, \quad \boldsymbol{d}(t) \propto \boldsymbol{J} \cos(m_a t)$$

Nucleon EDMs

CP-violating intranuclear forces





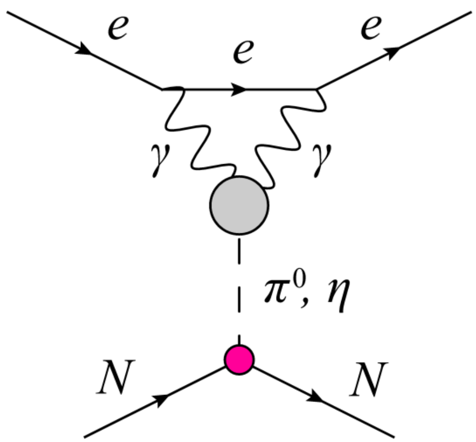
$\bar{g}_{\pi NN}^{(0)} \approx 0.016\, C_G a_0 \cos(m_a t)/f_a$

In nuclei, ***tree-level*** *CP*-violating intranuclear forces dominate over ***loop-induced*** nucleon EDMs [loop factor = $1/(8\pi^2)$].

QCD Vacuum Angle $\mathcal{L} = \theta \frac{g_s^2}{32\pi^2} G\tilde{G}$

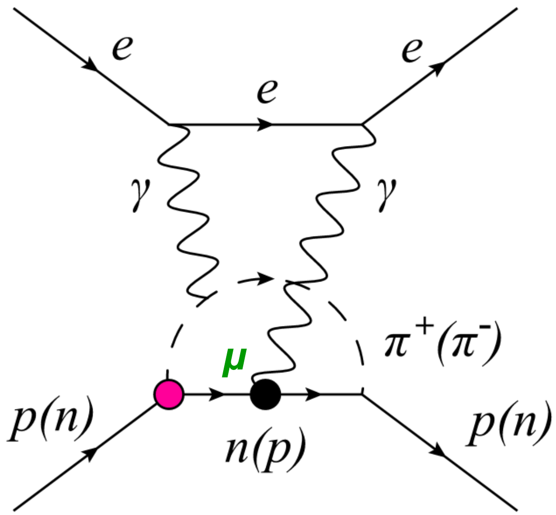
[Flambaum, Pospelov, Ritz, Stadnik, *PRD* **102**, 035001 (2020)]

LO: $\mathcal{O}(m_\pi^{-2})$



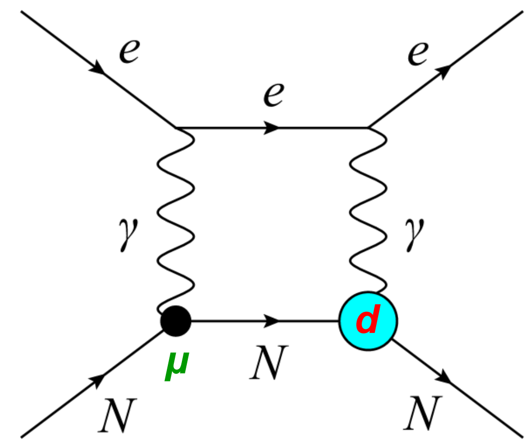
π^0, η contributions:
opposite sign

NLO: $\mathcal{O}(m_\pi^{-1})$



$(\boldsymbol{\mu} - \boldsymbol{d})$**:** $\mathcal{O}[\ln(A)/p_\mathrm{F}]$

[Fermi-gas model]



p, n contributions: ***same*** sign

For $Z \sim 80$ & $A \sim 200$: $C_\mathrm{SP}(\theta) \approx \left[0.1_\mathrm{LO} + 1.0_\mathrm{NLO} + 1.7_{(\mu-d)}\right] \times 10^{-2}\theta \approx 0.03\theta$

$$\mathcal{L}_\mathrm{contact} = -\frac{G_F C_\mathrm{SP} \bar{N} N \bar{e} i\gamma_5 e}{\sqrt{2}}$$

QCD Vacuum Angle $\mathcal{L} = \theta \frac{g_s^2}{32\pi^2} G\tilde{G}$

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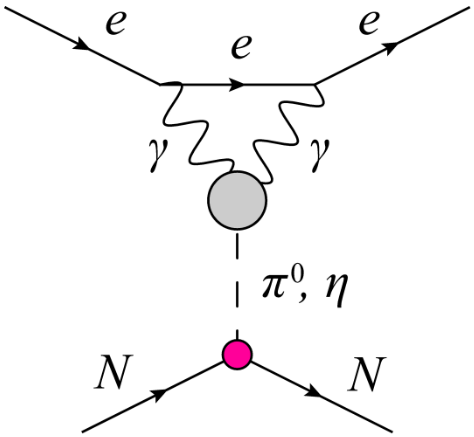

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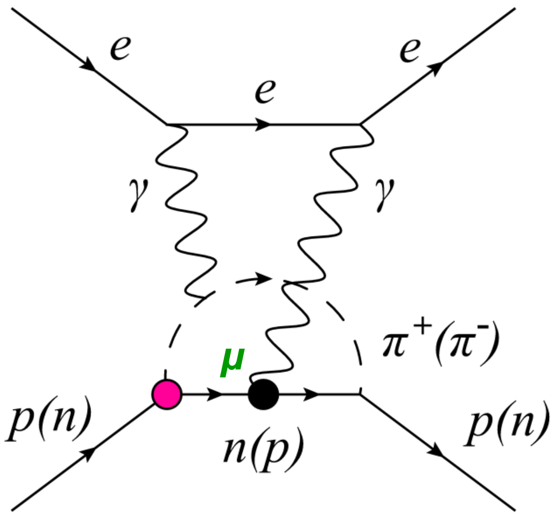

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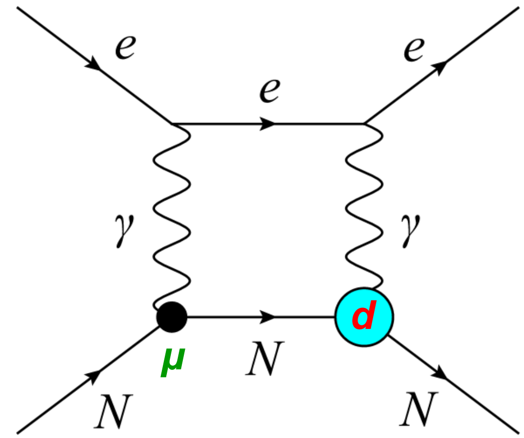

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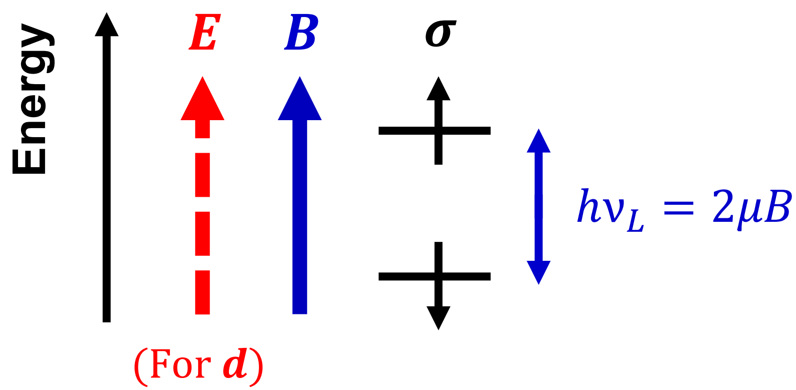
Future work: η' contribution and other N^{2}LO contributions, nuclear in-medium effects (NLO process), nuclear structure effects [$(\mu - d)$ process]

Searching for Spin-Dependent Effects

Proposals: [Flambaum, talk at *Patras Workshop*, 2013;
Stadnik, Flambaum, *PRD* **89**, 043522 (2014); Stadnik, thesis (Springer, 2017)]

Use spin-polarised sources: ***Atomic magnetometers***,
cold/ultracold particles, *torsion pendula*

Experiment (n/Hg): [nEDM collaboration, *PRX* **7**, 041034 (2017)]

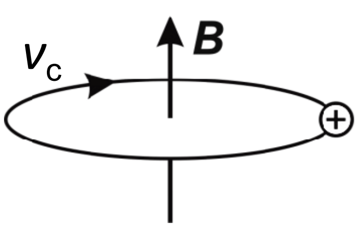


$$\frac{\nu_n}{\nu_{\mathrm{Hg}}} = \left|\frac{\mu_n \cancel{B}}{\mu_{\mathrm{Hg}} \cancel{B}}\right| + R(t)$$

***B*-field effect**

DM effect (*non-magnetic* coupling to $\boldsymbol{\sigma}$ or $\boldsymbol{d}$)

Proposal + Experiment ($\bar{p}$): [BASE collaboration, *Nature* **575**, 310 (2019)]



$$\left(\frac{\nu_L}{\nu_c}\right)_{\bar{p}} = \frac{|g_{\bar{p}}|}{2} + R(t)$$

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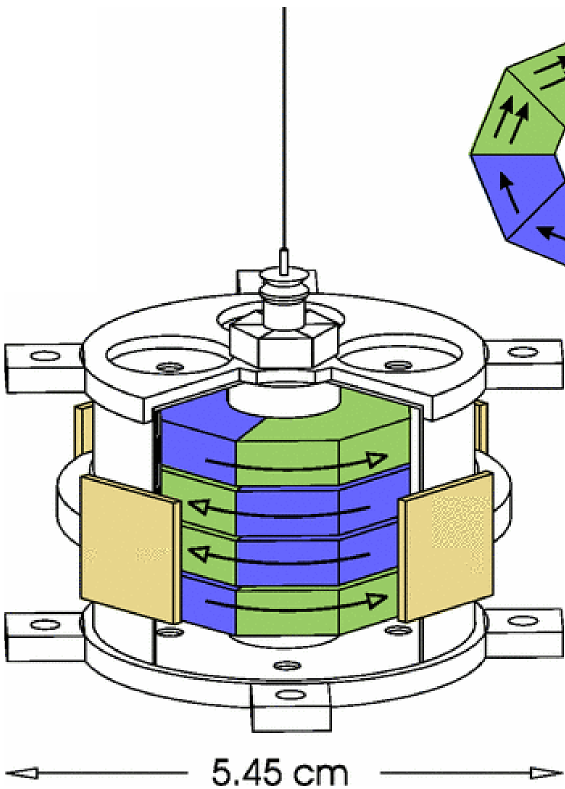
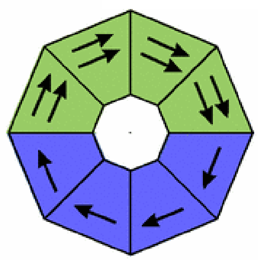
Stadnik, Flambaum, *PRD* **89**, 043522 (2014); Stadnik, thesis (Springer, 2017)]

Use spin-polarised sources: *Atomic magnetometers, cold/ultracold particles*, *torsion pendula*

Experiment (Alnico/$SmCo_5$): [Terrano *et al.*, *PRL* **122**, 231301 (2019)]





$\boldsymbol{\mu}_{\text{Alnico}}$ →

← $\boldsymbol{\mu}_{\text{SmCo}_5}$

$$\boldsymbol{\mu}_{\text{pendulum}} \approx \mathbf{0}$$

$(\boldsymbol{\sigma}_e)_{\text{Alnico}}$ →

← $(\boldsymbol{\sigma}_e)_{\text{SmCo}_5}$

$$(\boldsymbol{\sigma}_e)_{\text{pendulum}} \neq \mathbf{0}$$

$$\boldsymbol{\tau}(t) \propto (\boldsymbol{\sigma}_e)_{\text{pendulum}} \times \boldsymbol{B}_{\text{eff}}(t)$$