

Supplemental Material for Probing Fundamental Symmetries of Deformed Nuclei in Symmetric Top Molecules

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MOLECULAR STRUCTURE

As discussed in the main text, we numerically diagonalize the effective molecular Hamiltonian for the ground electronic state (\tilde{X}^1A_1) of $^{225}\text{RaOCH}_3^+$

$$H_{\text{total}} = H_{\text{rot}} + H_{\text{stark}} + H_{\text{zeeman}} + H_{\text{ss}} + H_{\text{nsr}} + H_{\text{sm}} \quad (\text{S1})$$

where we have included the rotational (rot), Stark, Zeeman, nuclear spin dipolar (ss), nuclear spin-rotation terms (nsr), and Schiff moment (sm) terms. No electron spin terms are included, as the molecule has a closed shell. For generality, however, the matrix elements are written in the fully decoupled basis including the electron spin S : $|N, K, S, J, m_J\rangle |I_M, m_{IM}\rangle |\Gamma, I_H, m_{IH}\rangle$.

Rovibrational Structure

RaOCH_3^+ is a prolate symmetric top with point group C_{3v} , corresponding to its three-fold cylindrical symmetry about the principal molecular axis (Z). The Hamiltonian that corresponds to the rotational energy for a prolate top is

$$H_{\text{rot}} = BN^2 + (A - B)\mathbf{N}_Z^2, \quad (\text{S2})$$

which has eigenenergies $BN(N+1) + (A-B)K^2$. A and B are rotational constants, while $\langle \mathbf{N} \rangle = N$ and $\langle \mathbf{N}_Z \rangle = K$ are the canonical rotational quantum numbers. A corresponds to the rotation of the molecule about the symmetry axis, while B corresponds to end-over-end rotation. For $|K| \neq 0$, each $|N, |K|\rangle$ -level in the rotational Hamiltonian has $2 \times (2N + 1)$ degeneracies, which are given by the $2N + 1$ lab-frame projections m_N and the two projections of $|K|$ onto the molecular axis.

The *ab initio* geometries, with corresponding rotational constants, for RaOCH_3^+ (see table I in main text) were optimized using the coupled cluster method with single, doubles, and perturbative triples [CCSD(T)] [1, 2] in CFOUR [3–5]. Relativistically contracted atomic natural orbital basis sets of polarized double, triple, and quadruple-zeta quality [ANO-RCC- $VnZP$ ($n=D, T, Q$)] are used [6–11]. Scalar relativistic effects are included via the one-electron variant of the spin-free exact two component theory (SFX2C-1e) [12–14].

There are eight vibrational modes for symmetric top molecules of the MOCH_3 form, four symmetric modes

Vibration	Energy [cm^{-1}]	Character
Ra-O-C bend	164.96/168.68	e
Ra-O stretch	390.78	a_1
CH_3 symmetric bend	1086.84	a_1
CH_3 rock	1170.41/1173.36	e
C-O stretch	1481.50	a_1
CH_3 asymmetric bend	1497.61/1498.56	e
CH_3 symmetric stretch	2993.19	a_1
CH_3 asymmetric stretch	3048.22/3059.15	e

Table S1. Vibrational energies, computed from B3LYP Hessians [15] at DFT optimized geometry, where each mode is classified with respect to its transformations under C_{3v} symmetry. In total, there are four symmetric a_1 states and four doubly-degenerate e states. The pair of frequencies for the degenerate vibrations is due to slight symmetry-breaking in the computed geometry.

of a_1 character and four asymmetric modes of e character. The vibrational energies and intensities of each mode are calculated via analytic B3LYP Hessians [15, 16] with ORCA [17]. For this calculation, we employ correlation-consistent basis sets at the quadruple-zeta level [18], with core-valence [19] and pseudopotential sets for the radium atom. The 78 core electrons of radium are modeled using the SK-MCDHF-RSC effective core potential (ECP) [20]. Energies and symmetry characters are listed in Table S1. While the proposed Schiff moment search would take place in the ground vibrational state, excited rovibrational states could be a valuable resource for state preparation or readout [21–25]. The energy difference between the nominally degenerate states of e character indicate that the accuracy of the energies is on the few percent level.

The vibrational states are also relevant as they will likely present a limitation on the coherence time [26]. Black-body excitation [27] of the Ra-O stretch mode (with the transition dipole moment calculated to be $\mu \sim 0.26$ D) is estimated to occur with a ~ 5 second time scale in a 300 K environment, though that can be reduced to 20 minutes in a 77 K environment. The radiative lifetime of the $N = |K| = 1$ states are much longer than one day due to their small energy spacing (a state with one atomic unit of transition dipole moment at this frequency would last around one hour) and the fact that they are spin-forbidden to decay to the ground rotational state, so black-body effects are likely to dominate over radiative

decay at room temperature.

Molecular Symmetries

Classification of Rovibronic and Nuclear Spin Wavefunctions

The C_{3v} point group (which is isomorphic to S_6) has three irreducible representations: A_1 , A_2 , and E . A_1 denotes the fully symmetric representation, and A_2 is the anti-symmetric representation. E denotes the degenerate representation, which splits into positive (E_+) and negative (E_-) components under the action of the cyclic group $C_3 \subset C_{3v}$. The character tables for C_{3v} and C_3 are written in Table S2 for reference.

	\hat{E}	$2\hat{C}_3(z)$	$3\sigma_v$		\hat{E}	\hat{C}_3	\hat{C}_3^2
A_1	+1	+1	+1	A	+1	+1	+1
A_2	+1	+1	-1	E_{\pm}	+1	$e^{\mp 2\pi i/3}$	$e^{\pm 2\pi i/3}$
E	+2	-1	0				

Table S2. Character Tables for C_{3v} (left) and C_3 (right)

We start by classifying the rovibronic states. In the electronic ground state and a vibrationally relaxed manifold ($|\Lambda = 0, \ell = 0\rangle$), the symmetry classification of the $|N, |K\rangle$ rovibronic states is dependent only on K . The $|N, K = 0\rangle$ state transforms as A_1 or A_2 (depending on N), while $|N, |K| = 3n\rangle$ states, where n is an integer and $K \neq 0$, transform as $A_1 \oplus A_2$. The remaining $|N, |K| \neq 3n\rangle$ states transform as the doubly degenerate E_{\pm} character, where the positive and negative components correspond to the positive and negative projections of $|K|$. (A generalized classification for non-zero orbital and vibrational angular momentum ($|\Lambda \neq 0, l \neq 0\rangle$))

can be found in [28].) The dependence of the molecular symmetries on K arises from the fact that the hydrogen atoms in the methyl group are indistinguishable and must obey the Pauli principle, as discussed in detail later.

All states where $|K| > 0$ are doubly degenerate and thus correspond to either the mixed E_{\pm} or $A_1 \oplus A_2$ symmetry characters. The splitting of K -doublets (and thus the breaking of C_{3v} symmetry) is naturally associated with the splitting of E_{\pm} or $A_1 \oplus A_2$ into distinct representations.

We discuss the specific mechanisms lifting the degeneracy below, but there is a simple and intuitive picture how this arises due to hyperfine couplings [29]. The K -doubled states $|\pm\rangle$ have a different distribution of proton spin about the azimuthal angle relative to the symmetry axis and therefore the anisotropic nature of the dipolar or nuclear spin-rotation interaction splits the two states. For $|K| = 1$ states, the hyperfine anisotropies directly couple states differing by $\Delta K = 2$ to produce a first-order K -splitting, but have progressively suppressed effects on the splitting for higher $|K| > 1$ [29, 30].

For states where $|K| = 3n$, the leading order source of K -doubling arises from a sextic centrifugal distortion term, which couples $\Delta K = 6$. This term appears as a correction to the rotational Hamiltonian $H_{\text{sextic}} = q_3(J_+^6 + J_-^6)/2$, where q_3 is the distortion constant [31]. In the CH_3 radical, the q_3 distortion constant has been measured to be 370 Hz [32]. At higher multiples of three, the K -splitting generated by q_3 is again suppressed. Using higher K states is therefore a general way to obtain even smaller K -doublets.

The composite nuclear spin states $|\Gamma, I_H, m_{IH}\rangle$ of the three hydrogen atoms, where Γ denotes the symmetry, are also classified into four states that transform as A_1 , and four states that transform as E_{\pm} :

$$\left|A, \frac{3}{2}, \pm \frac{3}{2}\right\rangle = \left|\pm \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2}\right\rangle \quad (\text{S3})$$

$$\left|A, \frac{3}{2}, \pm \frac{1}{2}\right\rangle = \frac{1}{\sqrt{3}} \left(\left|\pm \frac{1}{2}, \pm \frac{1}{2}, \mp \frac{1}{2}\right\rangle + \left|\pm \frac{1}{2}, \mp \frac{1}{2}, \pm \frac{1}{2}\right\rangle + \left|\mp \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2}\right\rangle \right) \quad (\text{S4})$$

$$\left|E_+, \frac{1}{2}, \pm \frac{1}{2}\right\rangle = \frac{1}{\sqrt{3}} \left(\left|\mp \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2}\right\rangle + e^{+2\pi i/3} \left|\pm \frac{1}{2}, \mp \frac{1}{2}, \pm \frac{1}{2}\right\rangle + e^{-2\pi i/3} \left|\pm \frac{1}{2}, \pm \frac{1}{2}, \mp \frac{1}{2}\right\rangle \right) \quad (\text{S5})$$

$$\left|E_-, \frac{1}{2}, \pm \frac{1}{2}\right\rangle = \frac{1}{\sqrt{3}} \left(\left|\mp \frac{1}{2}, \pm \frac{1}{2}, \pm \frac{1}{2}\right\rangle + e^{-2\pi i/3} \left|\pm \frac{1}{2}, \mp \frac{1}{2}, \pm \frac{1}{2}\right\rangle + e^{+2\pi i/3} \left|\pm \frac{1}{2}, \pm \frac{1}{2}, \mp \frac{1}{2}\right\rangle \right) \quad (\text{S6})$$

I_H refers to the total composite spin of the three protons ($\mathbf{I}_H = \mathbf{I}_{H1} + \mathbf{I}_{H2} + \mathbf{I}_{H3}$) and m_{IH} is the corresponding lab frame projection. Γ denotes the symmetry of the composite spin state.

To obey Fermi-Dirac statistics, the total nuclear-rovibronic wavefunction must transform as either A_1

(symmetric under inversion) or A_2 (antisymmetric under inversion) [28]. Therefore, the $K = 1$ and $K = 2$ (in the ground state) cases correspond to combined nuclear (${}^n\Gamma$) and rovibronic (${}^{evsr}\Gamma$) wavefunctions that have the

symmetries

$$|^{evsr}E_+\rangle|^nE_-\rangle \text{ or } |^{evsr}E_-\rangle|^nE_+\rangle, \quad (\text{S7})$$

whereas the fully symmetric $K = 0$ or $K = 3$ rotational states correspond to two possibilities

$$|^{evsr}A_1\rangle|^nA\rangle \text{ or } |^{evsr}A_2\rangle|^nA\rangle. \quad (\text{S8})$$

From eq. (S7), we can observe that any hyperfine interaction which couples $|^nE_+\rangle$ and $|^nE_-\rangle$ states also couples the $|^{evsr}E_+\rangle$ and $|^{evsr}E_-\rangle$ states, and therefore splits any residual K -degeneracy into the doublets,

$$\frac{1}{\sqrt{2}}(|^{evsr}E_+\rangle|^nE_-\rangle \pm |^{evsr}E_-\rangle|^nE_+\rangle). \quad (\text{S9})$$

This is the case for the K -doubling that originates from the nuclear dipolar spin couplings of RaOCH_3^+ , for instance.

The symmetry assignments in eqs. (S7) and (S8) also lead to additional selection rules for electric dipole transitions. For instance, let us consider transitions between different $|N, K\rangle$ manifolds in the electronic-vibrational ground state. Starting from the $K = 1$ manifold, the electric dipole operator only couples to $\Delta K = 0, \pm 1$ states. $|N, K = 2\rangle$ has the same nuclear symmetry Γ as $|N, K = 1\rangle$ and the transition is allowed. $|N, K = 0\rangle$ does not, and the transition is symmetry-forbidden (as well as spin-forbidden), which is why the radiative lifetime of $|N, K = 1\rangle$ is so long.

Hyperfine Structure

Parameterization of the Tensor Operators

In order to evaluate hyperfine structure of the hydrogen spins, it is necessary to sum over the interactions involving each individual hydrogen nucleus. A spin-spin interaction between the hydrogen spins I_i and an arbitrary spin \mathcal{S} , for instance, has the form

$$\sum_{i=1}^3 \mathcal{S} \cdot T_i \cdot I_i, \quad (\text{S10})$$

where T_i is the interaction tensor between the i th hydrogen and \mathcal{S} . In Cartesian form, T_1 can be written as

$$T_1 = \begin{pmatrix} T_{xx} & 0 & T_{xz} \\ 0 & T_{yy} & 0 \\ T_{zx} & 0 & T_{zz} \end{pmatrix}, \quad (\text{S11})$$

while T_2 and T_3 can be obtained with the appropriate rotations about the z axis [33].

The form of eq. (S10), however, is unwieldy for evaluating matrix elements over the symmetrized nuclear spin states derived in the previous section. A more intuitive

form can be obtained by parameterizing the T and I operators, as first formulated by Hougen in cartesian form [28] and Endo et al. in spherical tensor form [33, 34].

$$T_0 = (T_1 + T_2 + T_3)/3, \quad (\text{S12})$$

$$T_{\pm} = (T_1 + e^{\pm 2\pi i/3}T_2 + e^{\mp 2\pi i/3}T_3)/3, \quad (\text{S13})$$

$$I_0 = I_1 + I_2 + I_3, \quad (\text{S14})$$

$$I_{\pm} = I_1 + e^{\pm 2\pi i/3}I_2 + e^{\mp 2\pi i/3}I_3. \quad (\text{S15})$$

This allows us to rewrite eq. (S10) as

$$\sum_{\alpha=0,\pm} \mathcal{S} \cdot T_{\alpha} \cdot I_{-\alpha}. \quad (\text{S16})$$

Direct evaluation via the Wigner-Eckart Theorem yields the following reduced matrix elements for the parameterized nuclear spin operators over the symmetrized nuclear spin basis:

$$\langle \Gamma, I_0 | T^1(\mathbf{I}_0) | \Gamma, I_0 \rangle = \sqrt{I_0(I_0 + 1)(2I_0 + 1)}, \quad (\text{S17})$$

$$\langle E_{\pm}, I_0 | T^1(\mathbf{I}_{\pm}) | E_{\mp}, I_0 \rangle = -2\sqrt{I_0(I_0 + 1)(2I_0 + 1)}, \quad (\text{S18})$$

$$\langle E_{\pm}, I_0 = 1/2 | T^1(\mathbf{I}_{\pm}) | A_1, I_0 = 3/2 \rangle = \sqrt{6}. \quad (\text{S19})$$

Meanwhile, the parameterized T operators, in cartesian form, are:

$$T_0 = \begin{pmatrix} (T_{xx} + T_{yy})/2 & 0 & 0 \\ 0 & (T_{xx} + T_{yy})/2 & 0 \\ 0 & 0 & T_{zz} \end{pmatrix}, \quad (\text{S20})$$

$$T_{\pm} = \begin{pmatrix} (T_{xx} - T_{yy})/4 & \mp i(T_{xx} - T_{yy})/4 & T_{xz}/2 \\ \mp i(T_{xx} - T_{yy})/4 & -(T_{xx} - T_{yy})/4 & \pm i T_{zx}/2 \\ T_{zx}/2 & \pm i T_{zx}/2 & 0 \end{pmatrix}, \quad (\text{S21})$$

which have the corresponding non-zero rank-2 components in spherical tensor form $(T_{\alpha})_q^2$ [35]:

$$(T_0)_0^2 = (2T_{zz} - T_{xx} - T_{yy})/\sqrt{6}, \quad (\text{S22})$$

$$(T_{\pm})_{\mp}^2 = \mp(T_{xz} + T_{zx})/2, \quad (\text{S23})$$

$$(T_{\pm})_{\pm 2}^2 = (T_{xx} - T_{yy})/2. \quad (\text{S24})$$

By applying a Wigner rotation to the molecule-frame (q) spherical components, we obtain

$$\langle N'K' | \mathcal{D}_{pq}^2(\omega) (T_{\alpha})_q^2 | NK \rangle = \sum_q [(-1)^{N'-K'} \times \sqrt{(2N'+1)(2N+1)} \begin{pmatrix} N' & 2 & N \\ -K' & q & K \end{pmatrix} (T_{\alpha})_q^2]. \quad (\text{S25})$$

We can see that the anisotropic tensor component in eq. (S24) will couple states with $\Delta K = 2$. In the case of $|K| = 1$, this term would couple $K = +1$ and $K = -1$ terms, providing a first-order contribution to the K -doubling. Most of the sources of K -splitting in the hyperfine structure of symmetric tops arise in this manner.

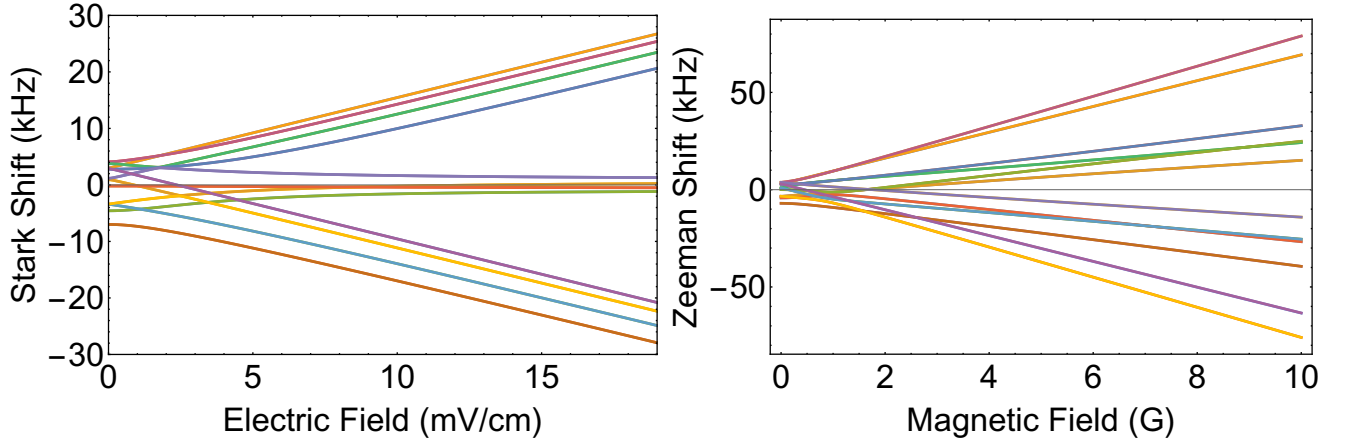


Figure S1. (left) Stark plot of hyperfine states in the $|N = 1, |K| = 1\rangle$ manifold, up to 15 mV cm^{-1} electric field. (right) Zeeman plot of hyperfine states in the $|N = 1, |K| = 1\rangle$ manifold, up to 10 G magnetic field.

Stark and Zeeman Matrix Elements

The Stark Hamiltonian is $\hat{H}_S = -T^1(\mathbf{d}) \cdot T^1(\mathbf{E})$, where \mathbf{d} is the molecule frame dipole moment and \mathbf{E}

is the electric field. Without loss of generality, we set the electric field along the \hat{z} direction in the lab frame. The matrix elements in the decoupled basis $|N, K, S, J, m_J\rangle |I_M, m_{IM}\rangle |\Gamma, I_H, m_{IH}\rangle$ are given by

$$\begin{aligned}
 & \langle N' K' S' J' m'_J; I_M m_{IM}; \Gamma I_H m_{IH} | \hat{H}_S | N K S J m_J; I_M m_{IM}; \Gamma I_H m_{IH} \rangle \\
 &= -E \langle N' K' S' J' m'_J | T_0^1(\mathbf{d}) | N K S J m_J \rangle \\
 &= -E d_0 (-1)^{J' - m'_J} \begin{pmatrix} J' & 1 & J \\ -m'_J & 0 & m_J \end{pmatrix} (-1)^{J + N' + 1 + S'} \sqrt{(2J + 1)(2J' + 1)} \begin{Bmatrix} N & J & S' \\ J' & N' & 1 \end{Bmatrix} \\
 & \quad \times \delta_{S, S'} (-1)^{N' - K'} \sqrt{(2N' + 1)(2N + 1)} \begin{pmatrix} N' & 1 & N \\ -K' & 0 & K \end{pmatrix}. \tag{S26}
 \end{aligned}$$

Note that the dipole moment is along the molecular axis such that $T_0^1(\mathbf{d}) = d_0$ and that the matrix element is diagonal with respect to the nuclear and electron spins.

The Zeeman Hamiltonian is written as $\hat{H}_z = \hat{H}_{z,I} + \hat{H}_{z,R}$, with two terms corresponding to the coupling of the nuclear spin and molecular rotation to the external magnetic field.

The nuclear spin term $\hat{H}_{z,I}$ is given by

$$\begin{aligned}
 \hat{H}_{z,I} &= -g_N \mu_N \sum_i T^1(\mathbf{I}_i) \cdot T^1(\mathbf{B}) \\
 &= -g_N \mu_N \sum_{i,p} (-1)^p T_p^1(\mathbf{I}_i) T_{-p}^1(\mathbf{B}) \tag{S27}
 \end{aligned}$$

where we note that we need to sum over all the hydrogen and radium nuclear spins. Evaluating the matrix element in the decoupled basis for the radium spin, which is diagonal in $|N, K, S, J, m_J\rangle |\Gamma, I_H, m_{IH}\rangle$:

$$\begin{aligned}
 & \langle I'_M m'_{IM} | \hat{H}_{z,IM} | I_M m_{IM} \rangle = \\
 & -g_N \mu_N \sum_p (-1)^p \langle I'_M, m'_{IM} | T_p^1(\mathbf{I}_M) | I_M, m_{IM} \rangle \langle T_{-p}^1(\mathbf{B}) \rangle, \tag{S28}
 \end{aligned}$$

where the nuclear spin angular momentum matrix element for a single nucleus is, generically,

$$\begin{aligned}
 \langle I', m'_I | T_p^1(\mathbf{I}) | I, m_I \rangle &= (-1)^{I' - m'_I} \begin{pmatrix} I' & 1 & I \\ -m'_I & p & m_I \end{pmatrix} \\
 & \quad \times \delta_{I', I} \sqrt{I(I + 1)(2I + 1)}. \tag{S29}
 \end{aligned}$$

For the coupling to hydrogen spins, we evaluate the parameterized $T^1(\mathbf{I}_0)$ matrix elements derived in the previous section, which results in a near-identical expression

to eq. (S29),

$$\begin{aligned} & \langle I_M m_{IM} | \hat{H}_{z, I_M} | I_M m_{IM} \rangle = \\ & - g_N \mu_N \sum_p [(-1)^p \langle \Gamma', I'_H, m'_{IH} | T_p^1(\mathbf{I}_0) | \Gamma, I_H, m_{IH} \rangle \\ & \times \langle T_{-p}^1(\mathbf{B}) \rangle]. \end{aligned} \quad (\text{S30})$$

The rotational Zeeman term [36] is:

$$\begin{aligned} \hat{H}_{z, R} &= g_R \mu_N \mathbf{N} \cdot \mathbf{B} \quad (\text{S31}) \\ &= \mu_N B_0 \sum_k (-1)^k \sqrt{\frac{2k+1}{3}} T_0^1(g_r^k, \mathbf{N}) \\ &= \mu_N B_0 \sum_k (-1)^{2k+1} \sqrt{2k+1} \\ & \times \sum_p \frac{1}{2} [T_p^k(g_r) T_{-p}^1(\mathbf{N}) + (-1)^k T_p^1(\mathbf{N}) T_{-p}^k(g_r)] \\ & \times \begin{pmatrix} k & 1 & 1 \\ p & -p & 0 \end{pmatrix}. \end{aligned} \quad (\text{S32})$$

Relativistic *ab initio* values for the rotational g-tensor g_r^k have been computed using the four-component linear response within elimination of small component (LRESC) approach of Aucar et al. [37] as implemented in the DIRAC19 code [38]. Electron correlation is treated at the level of DFT with a B3LYP functional [16] and a dyall.v4z basis is used [39]. See table I in main text for specific values.

Nuclear Spin-Rotation Coupling

The nuclear spin-rotation coupling is expressed similarly to the electron spin-rotation elements:

$$H_{\text{nsr}} = \frac{1}{2} \sum_{\alpha, \beta} C_{\alpha, \beta} (N_\alpha I_\beta + I_\beta N_\alpha) \quad (\text{S33})$$

$$= \frac{1}{2} \sum_{k=0}^2 [T^k(C) \cdot T^k(\mathbf{N}, \mathbf{I}) + T^k(\mathbf{N}, \mathbf{I}) \cdot T^k(C)] \quad (\text{S34})$$

$$= \frac{1}{2} \sum_{k=0}^2 \sum_p^k [T_p^k(C) T_{-p}^k(\mathbf{N}, \mathbf{I}) + T_p^k(\mathbf{N}, \mathbf{I}) T_{-p}^k(C)], \quad (\text{S35})$$

where the product is decomposed as

$$T_p^k(\mathbf{N}, \mathbf{I}) = (-1)^p \sqrt{2k+1} \sum_{p_1, p_2} T_{p_1}^1(\mathbf{N}) T_{p_2}^1(\mathbf{I}) \begin{pmatrix} 1 & 1 & k \\ p_1 & p_2 & -p \end{pmatrix}. \quad (\text{S36})$$

In $^{225}\text{RaOCH}_3^+$, the nuclear spin rotation interaction is divided into two components: the interaction between rotation and the spin-1/2 ^{225}Ra nucleus, and the interaction between rotation and the hydrogen spins in the methyl group.

Let us consider the spin-rotation coupling for the single radium spin \mathbf{I}_M . We write out eq. (S35), in the decoupled basis $|N, K, S, J, m_J\rangle |I_M, m_{IM}\rangle | \Gamma, I_H, m_{IH} \rangle$, taking care to sum over a complete set of states between $T(C)$ and $T(\mathbf{N})$,

$$\begin{aligned} & \langle N' K' S' J' m'_J; I'_M m'_{IM}; \Gamma I_H m_{IH} | H_{\text{nsr-M}} | N K S J m_J; I_M m_{IM}; \Gamma I_H m_{IH} \rangle \\ &= \frac{1}{2} \sum_{k, p} (-1)^p \sqrt{2k+1} \sum_{p_1, p_2} \left[\sum_{\eta''} \delta_{J', J} \frac{(-1)^{J'-J''}}{2J'+1} \langle N' K' S' J' m'_J | T_p^k(C) | N'' K'' S'' J'' m''_J \rangle \right. \\ & \times \langle N'' K'' S'' J'' m''_J | T_{p_1}^1(\mathbf{N}) | N K S J m_J \rangle \langle I'_M m'_{IM} | T_{p_2}^1(\mathbf{I}_M) | I_M m_{IM} \rangle \begin{pmatrix} 1 & 1 & k \\ p_1 & p_2 & p \end{pmatrix} \\ & + \sum_{\eta''} \delta_{J', J} \frac{(-1)^{J'-J''}}{2J'+1} \langle N' K' S' J' m'_J | T_{p_1}^1(\mathbf{N}) | N'' K'' S'' J'' m''_J \rangle \\ & \left. \times \langle \Gamma' I'_M m'_{IM} | T_{p_2}^1(\mathbf{I}_M) | \Gamma I_M m_{IM} \rangle \langle N'' K'' S'' J'' m''_J | T_{-p}^k(C) | N K S J m_J \rangle \begin{pmatrix} 1 & 1 & k \\ p_1 & p_2 & -p \end{pmatrix} \right]. \end{aligned} \quad (\text{S37})$$

Note that eq. (S37) is diagonal with respect to the $| \Gamma, I_H, m_{IH} \rangle$ spin states.

Evaluating the nuclear spin-rotation and rotational angular momentum matrix elements yields

$$\begin{aligned} & \langle N' K' S' J' m'_J | T_p^1(\mathbf{N}) | N K S J m_J \rangle = \\ & \times (-1)^{J'-m'_J} \begin{pmatrix} J' & 1 & J \\ -m'_J & p & m_J \end{pmatrix} \\ & \times \delta_{S', S} (-1)^{J+N'+1+S'} \sqrt{(2J'+1)(2J+1)} \\ & \times \begin{Bmatrix} N & J & S \\ J' & N' & 1 \end{Bmatrix} \delta_{N', N} \sqrt{(N(N+1)(2N+1))}, \end{aligned} \quad (\text{S38})$$

$$\begin{aligned}
& \langle N'K'S'J'm'_J | T_p^k(C) | NK SJ m_J \rangle = \\
& (-1)^{J'-m'_J} \begin{pmatrix} J' & k & J \\ -m'_J & p & m_J \end{pmatrix} \\
& \times \delta_{S',S} (-1)^{J+N'+k+S'} \sqrt{(2J'+1)(2J+1)} \begin{Bmatrix} N & J & S \\ J' & N' & k \end{Bmatrix} \\
& \times \sum_q [(-1)^{N'-K'} \sqrt{(2N'+1)(2N+1)} \\
& \times \begin{pmatrix} N' & k & N \\ -K' & q & K \end{pmatrix} T_q^k(C)], \tag{S39}
\end{aligned}$$

where, for an on-axis spin, the non-zero spherical tensor elements for the nuclear spin-rotation coupling are:

$$T_0^0(C) = -(C_{xx} + C_{yy} + C_{zz})/\sqrt{3}, \tag{S40}$$

$$T_0^2(C) = (2C_{zz} - C_{xx} - C_{yy})/\sqrt{6}, \tag{S41}$$

$$T_{\pm 2}^2(C) = (C_{xx} - C_{yy})/2. \tag{S42}$$

The lab-frame matrix element $T_p^1(\mathbf{I}_M)$ can be evaluated as eq. (S29) for a single spin.

Now, we consider the coupling to rotation for the composite hydrogen spins (\mathbf{I}_H). We can utilize the parameterized nuclear spin operators and coupling tensors derived earlier. This adds an additional summation over the parameter α ,

$$\begin{aligned}
H_{\text{nsr-H}} &= \frac{1}{2} \sum_{\alpha} \sum_{k=0}^2 \sum_p^k [T_p^k(C_{\alpha}) T_{-p}^k(\mathbf{N}, \mathbf{I}_{-\alpha}) \\
&+ T_p^k(\mathbf{N}, \mathbf{I}_{-\alpha}) T_{-p}^k(C_{\alpha})]. \tag{S43}
\end{aligned}$$

We can thus generalize eq. (S37) to the case of multiple spins,

$$\begin{aligned}
& \langle N'K'S'J'm'_J; \Gamma' I' m'_I | H_{\text{nsr-H}} | NK SJ m_J; \Gamma I m_I \rangle = \sum_{\alpha} \frac{1}{2} \sum_{k,p} (-1)^p \sqrt{2k+1} \\
& \times \sum_{p_1, p_2} \left[\sum_{\eta''} \delta_{J',J} \frac{(-1)^{J'-J''}}{2J'+1} \langle N'K'S'J'm'_J | T_p^k(C_{\alpha}) | N''K''S''J''m''_J \rangle \right. \\
& \times \langle N''K''S''J''m''_J | T_{p_1}^1(\mathbf{N}) | NK SJ m_J \rangle \langle \Gamma' I' m'_I | T_{p_2}^1(\mathbf{I}_{-\alpha}) | \Gamma, I, m_I \rangle \begin{pmatrix} 1 & 1 & k \\ p_1 & p_2 & p \end{pmatrix} \\
& + \sum_{\eta''} \delta_{J',J} \frac{(-1)^{J'-J''}}{2J'+1} \langle N'K'S'J'm'_J | T_{p_1}^1(\mathbf{N}) | N''K''S''J''m''_J \rangle \\
& \left. \times \langle \Gamma' I' m'_I | T_{p_2}^1(\mathbf{I}_{-\alpha}) | \Gamma I m_I \rangle \langle N''K''S''J''m''_J | T_{-p}^k(C_{\alpha}) | NK SJ m_J \rangle \begin{pmatrix} 1 & 1 & k \\ p_1 & p_2 & -p \end{pmatrix} \right]. \tag{S44}
\end{aligned}$$

where $\langle \Gamma' I' m'_I | T_p^1(\mathbf{I}_{\alpha}) | \Gamma I m_I \rangle$ is evaluated using Wigner-Eckart and the reduced matrix elements listed in eqs. (S17) – (S19). The $T_p^k(C_{\alpha})$ tensors can be obtained with the appropriate parameterization listed in eqs. (S17) – (S18).

Relativistic *ab initio* values for the $T(C)$ nuclear spin-rotation tensors are also obtained with a four-component LRESC approach [40, 41] in the DIRAC19 code [38] using a dyall.v4z basis set [39]. Correlation effects are treated at the level of DFT with the local density approximation and popular hybrid functions (B3LYP [16], PBE0 [42]). The full results are listed in Table S3. While validating these methods is challenging, due to the absence of experimental data (particularly for atoms heavier than group 4), benchmarks with diatomic species in refs. [37, 41] suggests that these predictions are accurate within 10 percent.

The contributions from nuclear spin rotation couplings to the K -doubling can be extracted from the anisotropy in the $T(C)$ tensors. As the radium spin-rotation coupling is diagonal in $|\Gamma, I_H, m_{IH}\rangle$, the $H_{\text{nsr-M}}$ Hamiltonian does not couple off-diagonally between $|E_+\rangle$ and $|E_-\rangle$ hydrogen spin states, and therefore does not contribute to the breaking of K -degeneracy. By contrast, the hydrogen spin-rotation coupling does couple $|E_+\rangle$ and $|E_-\rangle$ hydrogen spin states. The computed anisotropic contribution to K -doubling is $|T_{xx} - T_{yy}| \sim 0.3$ kHz, as noted in Table I of the main text.

Nuclear Spin Dipolar Coupling

For coupling two generic spins I_1 and I_2 , the matrix element is

$$H_{II} = \frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi} \left[\frac{\mathbf{I}_1 \cdot \mathbf{I}_2}{r^3} - \frac{3(\mathbf{I}_1 \cdot \mathbf{r})(\mathbf{I}_2 \cdot \mathbf{r})}{r^5} \right] \quad (\text{S45})$$

$$= -\sqrt{6} \frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi} T^2(C) \cdot T^2(\mathbf{I}_1, \mathbf{I}_2) \quad (\text{S46})$$

$$= -\sqrt{6} \frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi} \sum_p T_p^2(C) T_{-p}^2(\mathbf{I}_1, \mathbf{I}_2), \quad (\text{S47})$$

where μ_0 is the vacuum permeability and $\gamma_{1,2}$ are the nuclear gyromagnetic ratios. $T_q^2(C)$ is the spherical harmonic tensor $\langle C_q^2(\theta, \phi) r^{-3} \rangle$, where the polar angles

(θ, ϕ) parameterize the vector from I_1 to I_2 . Meanwhile, $T^2(\mathbf{I}_1, \mathbf{I}_2)$ can be decomposed as

$$T_p^2(\mathbf{I}_1, \mathbf{I}_2) = (-1)^p \sqrt{5} \sum_{p_1, p_2} T_{p_1}^1(\mathbf{I}_1) T_{p_2}^1(\mathbf{I}_2) \begin{pmatrix} 2 & 1 & 1 \\ -p & p_1 & p_2 \end{pmatrix}. \quad (\text{S48})$$

Let us consider the spin-spin interaction between the ^{225}Ra nucleus and the hydrogen nuclei. We write the matrix elements in the decoupled basis $|N, K, S, J, m_J\rangle |I_M, m_{IM}\rangle |\Gamma, I_H, m_{IH}\rangle$ and then evaluate the spherical harmonic and nuclear spin-spin tensors

$$\begin{aligned} & \langle N' K' S' J' m'_J; I'_M m'_{IM}; \Gamma', I'_H m'_{IH} | H_{\text{ss}} | N K S J m_J; I_M m_{IM}; \Gamma I_H m_{IH} \rangle \\ &= -\sqrt{6} \frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi} \sum_p \langle N' K' S' J' m'_J | T_p^2(C) | N K S J m_J \rangle \\ & \times \langle I'_M m'_{IM}; \Gamma' I'_H m'_{IH} | T_{-p}^2(\mathbf{I}_M, \mathbf{I}_H) | I_M m_{IM}; \Gamma I_H m_{IH} \rangle \\ &= -\sqrt{6} \frac{\mu_0 \gamma_1 \gamma_2 \hbar^2}{4\pi} \sum_{\alpha} \sum_p \langle N' K' S' J' m'_J | T_p^2(\mathbf{C}_{\alpha}) | N K S J m_J \rangle \\ & \times \langle I'_M m'_{IM}; \Gamma' I'_H m'_{IH} | T_{-p}^2(\mathbf{I}_M, \mathbf{I}_{-\alpha}) | I_M m_{IM}; \Gamma I_H m_{IH} \rangle, \end{aligned} \quad (\text{S49})$$

$$\begin{aligned} & \langle N' K' S' J' m'_J | T_p^2(\mathbf{C}_{\alpha}) | N K S J m_J \rangle \\ &= (-1)^{J'-m'_J} \begin{pmatrix} J' & 2 & J \\ -m'_J & p & m_J \end{pmatrix} \delta_{S', S} (-1)^{J+N'+S'+2} \sqrt{(2J'+1)(2J+1)} \begin{Bmatrix} N & J & S \\ J' & N' & 2 \end{Bmatrix} \\ & \times \langle N' K' | \sum_q \mathcal{D}_{pq}^2(\omega) \cdot T_q^2(\mathbf{C}_{\alpha}) | N K \rangle \end{aligned} \quad (\text{S50})$$

$$\begin{aligned} &= (-1)^{J'-m'_J} \begin{pmatrix} J' & 2 & J \\ -m'_J & p & m_J \end{pmatrix} \delta_{S', S} (-1)^{J+N'+S'+2} \sqrt{(2J'+1)(2J+1)} \begin{Bmatrix} N & J & S \\ J' & N' & 2 \end{Bmatrix} \\ & \times \sum_q (-1)^{N'-K'} \sqrt{(2N'+1)(2N+1)} \begin{pmatrix} N' & 2 & N \\ -K' & q & K \end{pmatrix} T_q^2(\mathbf{C}_{\alpha}) \end{aligned} \quad (\text{S51})$$

$$\begin{aligned} & \langle I'_M m'_{IM}; \Gamma' I'_H m'_{IH} | T_p^2(\mathbf{I}_M, \mathbf{I}_{\alpha}) | I_M m_{IM}; \Gamma I_H m_{IH} \rangle \\ &= (-1)^p \sqrt{5} \sum_{p_1, p_2} \langle I'_M m'_{IM} | T_{p_1}^1(\mathbf{I}_M) | I_M m_{IM} \rangle \langle \Gamma' I'_H m'_{IH} | T_{p_2}^1(\mathbf{I}_{\alpha}) | \Gamma I_H m_{IH} \rangle \begin{pmatrix} 2 & 1 & 1 \\ -p & p_1 & p_2 \end{pmatrix}. \end{aligned} \quad (\text{S52})$$

The molecule-frame $T_q^2(\mathbf{C}_{\alpha})$ reduced matrix element can be evaluated using the appropriate sums of spherical harmonics that parameterize the vector between the i th hydrogen and the radium atom,

$$T_q^2(\mathbf{C}_0) = \sum_{i=1}^3 \sqrt{\frac{4\pi}{5}} Y_q^2(\theta_i, \phi_i) r^{-3}, \quad (\text{S53})$$

$$\begin{aligned} T_q^2(\mathbf{C}_{\pm}) &= \sqrt{\frac{4\pi}{5}} [Y_q^2(\theta_1, \phi_1) + e^{\pm 2\pi i/3} Y_q^2(\theta_2, \phi_2) \\ & + e^{\mp 2\pi i/3} Y_q^2(\theta_3, \phi_3)] r^{-3}. \end{aligned} \quad (\text{S54})$$

By convention [33], the Cartesian form of the single-hydrogen dipolar tensor T_1 (of the form eq. (S11)) is taken to be traceless $\text{tr}[T] = 0$ and symmetric, which

yields the following simplifications from eqs. (S22)–(S23), which are invoked in table I in the main text:

$$\alpha_{dip} \cdot T_0^2(\mathbf{C}_0) = 3T_{zz}/\sqrt{6}, \quad (\text{S55})$$

$$\alpha_{dip} \cdot T_{\pm}^2(\mathbf{C}_{\mp}) = \mp T_{xz}, \quad (\text{S56})$$

$$\alpha_{dip} \cdot T_{\pm 2}^2(\mathbf{C}_{\pm}) = (T_{xx} - T_{yy})/2. \quad (\text{S57})$$

where we define a scaling parameter $\alpha_{dip} = -\sqrt{6}\mu_0\gamma_H\gamma_{Ra}\hbar^2/4\pi$.

The radium-hydrogen spin-spin interaction couples the opposite symmetry states $|E_+\rangle$ and $|E_-\rangle$. This leads to $|T_{xx} - T_{yy}\rangle \sim 0.4$ kHz K -doubling terms which couple the $|N, K, S, J, m_J\rangle|I_M, m_{IM}\rangle|I_H, m_{IH}\rangle$ and $|N, -K, S, J, m_J\rangle|I_M, m_{IM}\rangle|I_H, m_{IH}\rangle$ states. The

spin-spin interaction between the hydrogen spins themselves are only non-zero between the ‘‘ortho’’ stretched spin states of A character. This term can therefore be ignored in the $|N, |K| = 1\rangle$ manifold.

SCHIFF MOMENT

Effective molecular sensitivity

The effective Schiff moment sensitivity is proportional to $\langle \mathbf{I}_M \cdot \hat{\mathbf{n}} \rangle$, where I_M is the metal spin and \hat{n} is the internuclear axis [43]. In the decoupled basis, this is written as:

$$\begin{aligned} & \langle N', K', S', J', m'_J; I'_M, m'_{IM}; I'_H, m'_{IH} | \mathbf{I}_M \cdot \hat{\mathbf{n}} | N, K, S, J, m_J; I_M, m_{IM}; I_H, m_{IH} \rangle \\ &= \delta_{I'_H, I_H} \delta_{m'_{IH}, m_{IH}} \langle I'_M, m'_{IM} | \mathbf{I}_M | I_M, m_{IM} \rangle \langle N', K', S', J', m'_J | \hat{n} | N, K, S, J, m_J \rangle \\ &= \delta_{I'_H, I_H} \delta_{m'_{IH}, m_{IH}} \langle I'_M, m'_{IM} | \mathbf{I}_M | I_M, m_{IM} \rangle (-1)^{J'-m'_J} \begin{pmatrix} J' & 1 & J \\ -m_J & 0 & m_J \end{pmatrix} \\ &\quad \times \delta_{S', S} (-1)^{J+N'+S'+1} \sqrt{(2J'+1)(2J+1)} \left\{ \begin{matrix} N & J & S' \\ J' & N' & 1 \end{matrix} \right\} \langle N' | \hat{\mathbf{n}} | N \rangle \\ &= \delta_{I'_H, I_H} \delta_{m'_{IH}, m_{IH}} (-1)^{I'_M - m'_{IM}} \begin{pmatrix} I'_M & 1 & I_M \\ -m'_{IM} & 0 & m_{IM} \end{pmatrix} \delta_{I'_M, I_M} \sqrt{I_M(I_M+1)(2I_M+1)} \\ &\quad \times (-1)^{J'-m'_J} \begin{pmatrix} J' & 1 & J \\ -m_J & 0 & m_J \end{pmatrix} \delta_{S', S} (-1)^{J+N'+S'+1} \sqrt{(2J'+1)(2J+1)} \left\{ \begin{matrix} N & J & S' \\ J' & N' & 1 \end{matrix} \right\} \\ &\quad \times (-1)^{N'-K'} \sqrt{(2N'+1)(2N+1)} \begin{pmatrix} N' & 1 & N \\ -K' & 0 & K \end{pmatrix} \end{aligned} \quad (\text{S58})$$

In zero field, there is no defined orientation of the molecule, and the Schiff moment sensitivity is zero. In the decoupled limit ($\gtrsim 1$ V/cm), the value of $\langle \mathbf{I}_M \cdot \hat{\mathbf{n}} \rangle$ is $K \cdot m_N \cdot m_{IM} \cdot 1/2$ for the $N = 1$ and $|K| = 1$ manifold. The $1/2$ factor arises from the $K/N(N+1)$ pre-factor to the Stark energy. The states with ‘‘stretched’’ sensitivity therefore take on the values $\langle \mathbf{I}_M \cdot \hat{\mathbf{n}} \rangle = \pm 1/4$.

BSM Sensitivity

The frequency sensitivity of a spin-precession measurement on a molecule or atom with coherence time τ and repetition n is expressed as $\delta\omega = [\tau\sqrt{n}]^{-1}$. In the main text, we assume a single trapped $^{225}\text{RaOCH}_3^+$ ion with 5 sec coherence time, which provides a frequency sensitivity of $\delta\omega = 7.5$ mrad $\text{s}^{-1}/\sqrt{\text{hour}}$.

Dobaczewski and Engel calculated $|\mathbf{S}^{(225}\text{Ra})|$ in the framework of T, P -violating interactions between nucleons mediated by a pion using a variety of Skyrme energy functionals [44]. The average of their results,

which was expressed in terms of πNN vertices, was reparameterized by ref. [43] in terms of QCD $\bar{\theta}$ and the quark-chromo EDMs \tilde{d}_u and \tilde{d}_d :

$$|\mathbf{S}^{(225}\text{Ra})| = 1.0\bar{\theta} e \text{ fm}^3 \quad (\text{S59})$$

$$= 10^4(0.50\tilde{d}_u - 0.54\tilde{d}_d) e \text{ fm}^2 \quad (\text{S60})$$

The energy shift resulting from a Schiff moment \mathbf{S} and a coupling constant W_s is $H_{\text{sm}} = W_s (\mathbf{I}_M \cdot \hat{\mathbf{n}}) |\mathbf{S}|/|\mathbf{I}|$. For a differential measurement performed between states of opposite effective sensitivity ($\langle \mathbf{I}_M \cdot \hat{\mathbf{n}} \rangle = \pm 1/4$), we arrive at

$$\delta\bar{\theta} = \frac{2\hbar\delta\omega}{W_s(S/\bar{\theta})} \quad (\text{S61})$$

where $S/\bar{\theta} = 1.0 e \text{ fm}^3$ and the factor of two arises from $\Delta[\langle \mathbf{I}_M \cdot \hat{\mathbf{n}} \rangle] = 1/2$. Given the estimate of $W_s(\text{RaOCH}_3^+) \approx 30,000$ a.u. [43] (where a.u. $\equiv e/4\pi\epsilon_0 a_0^4$), we have $\delta\bar{\theta}/\delta\omega = 24 \times 10^{-8}$ s. Two weeks of data taking would thus result in the model-dependent sensitivity of $\delta\bar{\theta} < \times 10^{-10}$.

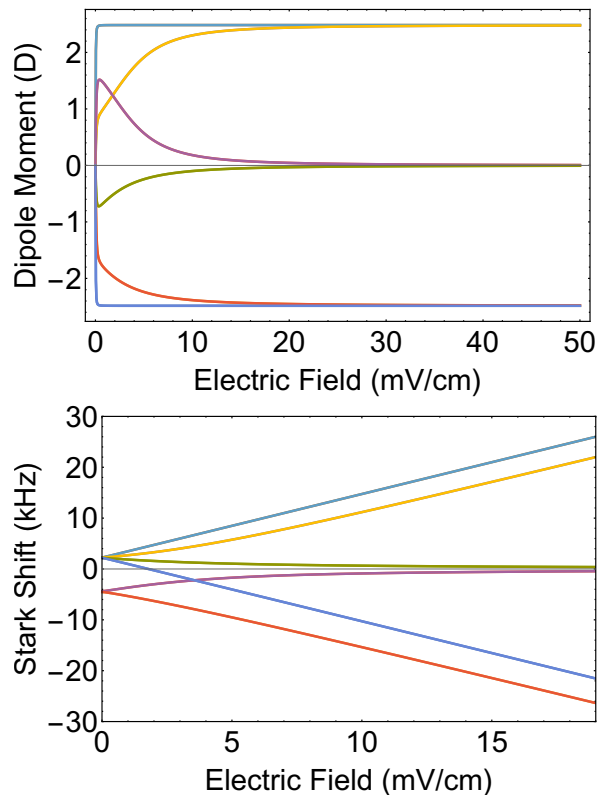


Figure S2. Lab frame dipole moment (top) and Stark shifts (bottom) of hyperfine states of the $|N = 1, |K| = 1\rangle$ manifold in the fully polarized limit for $^{226}\text{RaOCH}_3^+$. High, low, and no field seekers correspond to states with negative, positive, and zero dipole moment ($K \times m_N = +1, 0,$ and -1). The jumps indicate avoided crossings between the Stark states.

This discussion only considers in detail the limit on $\bar{\theta}$ as an example, though the other quantities to which Schiff moments are sensitive are also of interest. Due to the many possible hadronic sources, measurements in different systems are needed to obtain robust bounds [45]

SPIN ZERO ISOTOPOLOGUES

We briefly consider the hyperfine structure for $^{226}\text{RaOCH}_3^+$ in the $|N = 1, |K| = 1\rangle$ manifold, which yields 12 unique levels. This is the isotopologue recently produced in an ion trap [46], and could serve as a valuable platform for spectroscopy and development. As the ^{226}Ra nucleus is spin-0, the only hyperfine term to consider is the spin-rotation interaction between the ortho-hydrogen state and the molecular rotation, and the hyperfine basis reduces to $|N, K, S, J, m_J\rangle|\Gamma, I_H, m_{IH}\rangle$ with the radium spin terms omitted. Fig. S2 shows the Stark shifts and dipole moments for the molecule up to the fully polarized limit, and fig. S3 shows the level structure grouped by the projection of total angular momentum ($m_F = m_N + m_{IH}$) and their Stark manifold ($K \times m_N$).

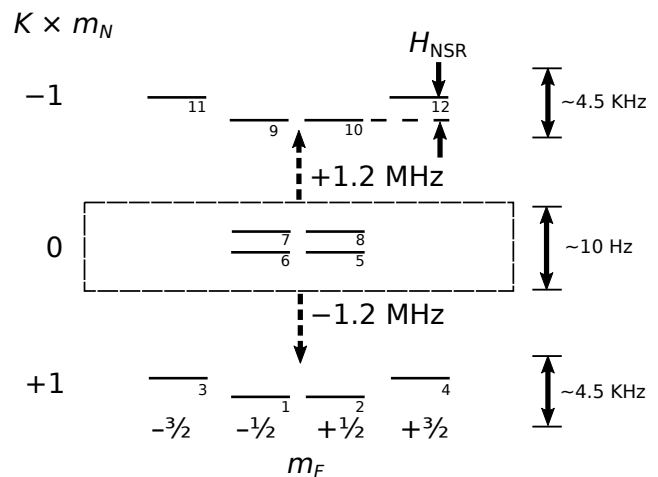


Figure S3. Level structure for the 12 hyperfine states of $^{226}\text{RaOCH}_3^+$ in the $|N = 1, |K| = 1\rangle$ manifold in the decoupled regime (1 V/cm), grouped by the projection of total angular momentum $m_F = m_N + m_{IH}$ and their Stark manifold ($K \times m_N$). The states are numbered in order of energy.

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Method:	LRESC(B3LYP)	LRESC(PBE0)	LRESC(LDA)	LRESC(DHF)
Basis:	dyall.v4z	dyall.v4z	dyall.v4z	dyall.v4z
NSR (H)				
$T(C_{\text{nsr}})_{zz}$	15.27503794	15.23988653	15.05936414	15.54351836
$T(C_{\text{nsr}})_{yy}$	0.29172044	0.29285025	0.27717575	0.31026183
$T(C_{\text{nsr}})_{xx}$	-0.00937583	-0.00725048	-0.02242052	0.01481825
$T(C_{\text{nsr}})_{zy}$	-0.00000000	-0.00000000	-0.00000000	-0.00000000
$T(C_{\text{nsr}})_{yz}$	-0.00000012	-0.00000012	-0.00000010	-0.00000014
$T(C_{\text{nsr}})_{yx}$	-0.00000005	-0.00000005	-0.00000005	-0.00000005
$T(C_{\text{nsr}})_{xy}$	0.00000005	0.00000005	0.00000005	0.00000005
$T(C_{\text{nsr}})_{xz}$	253.81827070	253.90232417	254.10917668	253.71558497
$T(C_{\text{nsr}})_{zx}$	-3.38051883	-3.37994662	-3.37277269	-3.38404825
NSR (Ra)				
$T(C_{\text{nsr}})_{xx}$	3.67255479	3.52587436	4.86603582	1.71006450
$T(C_{\text{nsr}})_{yy}$	2.41881565	2.49385127	3.24531289	1.35943566
$T(C_{\text{nsr}})_{zz}$	2.41881768	2.49385648	3.24531347	1.35945538
$T(C_{\text{nsr}})_{zy}$	-0.00000003	-0.00000004	-0.00000005	-0.00000002
$T(C_{\text{nsr}})_{yz}$	-0.00000275	-0.00000284	-0.00000369	-0.00000155
$T(C_{\text{nsr}})_{yx}$	0.00000000	-0.00000000	-0.00000000	0.00000000
$T(C_{\text{nsr}})_{xy}$	0.00000000	-0.00000000	0.00000000	-0.00000000
$T(C_{\text{nsr}})_{xz}$	-0.00000158	-0.00002529	0.00000641	-0.00013274
$T(C_{\text{nsr}})_{zx}$	0.00000009	-0.00000060	-0.00000024	0.00000726

Table S3. Nuclear spin-rotation constants computed with optimized geometries from table I. Units in kHz.

State	Energy	Mixing	Schiff Sensitivity
1	-1.2545 MHz	$[-1, -1\rangle + 1/2\rangle E_+, +1/2\rangle - + 1, +1\rangle - 1/2\rangle E_-, -1/2\rangle]/\sqrt{2}$	0
2	-1.2545 MHz	$[-1, -1\rangle + 1/2\rangle E_+, +1/2\rangle + + 1, +1\rangle - 1/2\rangle E_-, -1/2\rangle]/\sqrt{2}$	0
3	-1.2515 MHz	$[-1, -1\rangle - 1/2\rangle E_+, +1/2\rangle$	-1/4
4	-1.2515 MHz	$+ + 1, +1\rangle + 1/2\rangle E_-, -1/2\rangle$	+1/4
5	-1.24926 MHz	$[-1, -1\rangle + 1/2\rangle E_+, -1/2\rangle$	+1/4
6	-1.24926 MHz	$+ + 1, +1\rangle - 1/2\rangle E_+, -1/2\rangle$	-1/4
7	-1.24781 MHz	$[+ 1, +1\rangle + 1/2\rangle E_-, +1/2\rangle$	+1/4
8	-1.24781 MHz	$- - 1, -1\rangle - 1/2\rangle E_+, -1/2\rangle$	-1/4
9	-932.31 Hz	$[-1, 0\rangle + 1/2\rangle E_+, -1/2\rangle + - 1, 0\rangle - 1/2\rangle E_+, +1/2\rangle - + 1, 0\rangle + 1/2\rangle E_-, -1/2\rangle - + 1, 0\rangle - 1/2\rangle E_-, +1/2\rangle]/2$	0
10	-932.287 Hz	$[-1, 0\rangle + 1/2\rangle E_+, -1/2\rangle - - 1, 0\rangle - 1/2\rangle E_+, +1/2\rangle - + 1, 0\rangle + 1/2\rangle E_-, -1/2\rangle + + 1, 0\rangle - 1/2\rangle E_-, +1/2\rangle]/2$	0
11	-619.612 Hz	$[-1, 0\rangle + 1/2\rangle E_+, -1/2\rangle + - 1, 0\rangle - 1/2\rangle E_+, +1/2\rangle + + 1, 0\rangle + 1/2\rangle E_-, -1/2\rangle + + 1, 0\rangle - 1/2\rangle E_-, +1/2\rangle]/2$	0
12	-619.584 Hz	$[-1, 0\rangle + 1/2\rangle E_+, -1/2\rangle - - 1, 0\rangle - 1/2\rangle E_+, +1/2\rangle + + 1, 0\rangle + 1/2\rangle E_-, -1/2\rangle - + 1, 0\rangle - 1/2\rangle E_-, +1/2\rangle]/2$	0
13	619.367 Hz	$[- 1, 0\rangle + 1/2\rangle E_+, +1/2\rangle + + 1, 0\rangle + 1/2\rangle E_-, +1/2\rangle]/\sqrt{2}$	0
14	619.367 Hz	$[- 1, 0\rangle - 1/2\rangle E_+, -1/2\rangle + + 1, 0\rangle - 1/2\rangle E_-, -1/2\rangle]/\sqrt{2}$	0
15	932.466 Hz	$[- 1, 0\rangle + 1/2\rangle E_+, +1/2\rangle - + 1, 0\rangle + 1/2\rangle E_-, +1/2\rangle]/\sqrt{2}$	0
16	932.466 Hz	$[- 1, 0\rangle - 1/2\rangle E_+, -1/2\rangle - + 1, 0\rangle - 1/2\rangle E_-, -1/2\rangle]/\sqrt{2}$	0
17	1.24705 MHz	$[- 1, +1\rangle - 1/2\rangle E_+, -1/2\rangle + + 1, -1\rangle + 1/2\rangle E_-, +1/2\rangle]/\sqrt{2}$	0
18	1.24705 MHz	$[- 1, +1\rangle - 1/2\rangle E_+, -1/2\rangle - + 1, -1\rangle + 1/2\rangle E_-, +1/2\rangle]/\sqrt{2}$	0
19	1.25004 MHz	$[+ 1, -1\rangle - 1/2\rangle E_-, +1/2\rangle$	+1/4
20	1.25004 MHz	$- - 1, +1\rangle + 1/2\rangle E_+, -1/2\rangle$	-1/4
21	1.25227 MHz	$[- 1, -1\rangle - 1/2\rangle E_+, +1/2\rangle$	-1/4
22	1.25227 MHz	$+ + 1, -1\rangle + 1/2\rangle E_-, -1/2\rangle$	+1/4
23	1.25371 MHz	$[+ 1, -1\rangle - 1/2\rangle E_-, -1/2\rangle$	+1/4
24	1.25371 MHz	$- - 1, +1\rangle + 1/2\rangle E_+, +1/2\rangle$	-1/4

Table S4. Energy, mixings, and Schiff sensitivity of hyperfine states of $^{225}\text{RaOCH}_3^+$ in the $|N=1, |K|=1\rangle$ manifold in the high-field regime (1 V/cm). The mixings are written in the decoupled basis $|K, m_N\rangle|m_{IM}\rangle|\Gamma, m_{IH}\rangle$. The quantum numbers $N=1$, $I_M=1/2$, and $I_H=1/2$ are omitted for brevity.