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Engineering field-insensitive molecular clock transitions for symmetry violation searches

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Molecules are a powerful platform to probe fundamental symmetry violations beyond the Standard Model, as they offer both large amplification factors and robustness against systematic errors. As experimental sensitivities improve, it is important to develop new methods to suppress sensitivity to external electromagnetic fields, as limits on the ability to control these fields are a major experimental concern. Here we show that sensitivity to both external magnetic and electric fields can be simultaneously suppressed using engineered radio frequency, microwave, or two-photon transitions that maintain large amplification of CP-violating effects. By performing a clock measurement on these transitions, CP-violating observables including the electron electric dipole moment, nuclear Schiff moment, and magnetic quadrupole moment can be measured with suppression of external field sensitivity of $\gtrsim 100$ generically, and even more in many cases. Furthermore, the method is compatible with traditional Ramsey measurements, offers internal co-magnetometry, and is useful for systems with large angular momentum commonly present in molecular searches for nuclear CP-violation.

Precision measurements of heavy atomic and molecular systems have proven to be a powerful probe of physics beyond the Standard Model (BSM) [1]. For example, searches for the electron’s electric dipole moment (eEDM) in ThO and HfF⁺ probe charge-parity (CP) violating new physics at TeV energy scales [2–4]. Their high sensitivities rely on the enhancement of CP-violating observables in the internal molecular electromagnetic environment, combined with the advantageous experimental features arising from their unique molecular structures. The sensitivity of these molecular CP-violation (CPV) searches will continue to improve as new methods such as laser cooling are developed to increase count rates and coherence times [1, 5, 6].

The two most sensitive eEDM experiments [2–4] rely on a particular molecular structure (³Δ₁ state) which offers two critical features: a small magnetic moment, and “internal co-magnetometers.” The small magnetic moment of these states makes them largely insensitive to uncontrolled magnetic fields, which are a major challenge for EDM experiments with atoms [7] and neutrons [8]. Internal co-magnetometers give the ability to reverse the desired CPV energy shifts without changing lab fields, resulting in robustness against systematic effects arising from the applied electric and magnetic fields.

However, molecules which are sensitive to CPV and have this molecular structure are not laser coolable – a feature which would be advantageous for improving sensitivity through advanced quantum control. Polyatomic molecules offer internal co-magnetometers generically, including molecules which can be laser-cooled [5, 9]. Laser-coolable, eEDM sensitive molecules have a single, metal-centered *s*-type valence electron [10, 11], which means that they have magnetic moments on the order of the Bohr magneton μ_B ; this is $\gtrsim 100$ times larger than ³Δ₁ states, and correspondingly more sensitive to mag-

netic fields. A recent demonstration [12] showed that it is possible to tune the magnetic sensitivity in polyatomic molecules to very low values while still maintaining eEDM sensitivity. Another proposal to reduce field sensitivity is to use magnetically-insensitive “clock” transitions, which occur in many molecules [13].

In this manuscript, we discuss a generic method to engineer “clock” transitions which have reduced sensitivity to both magnetic *and* electric fields while maintaining large sensitivity to CPV in molecules. The basic idea behind these CPV-sensitive field-insensitive transitions (CP-FITs) is similar to magic conditions in atomic clocks [14, 15] and precision spectroscopy in molecules [16]. We find CP-FITs in a wide range of polyatomic and diatomic molecules with applications to search for the eEDM, nuclear Schiff moments (NSM), and nuclear magnetic quadrupole moments (MQM). This technique is particularly useful for molecules with large nuclear spins and complicated hyperfine structure for nuclear CPV searches. Furthermore, because CP-FITs do not involve traditional $M = 0 \rightarrow 0$ clock transitions, where M is the angular momentum projection quantum number, driving the same transitions with the opposite signs of M provides a co-magnetometry method to reject systematic errors. Finally, since the states involved have static energy shifts sensitive to CPV, they can be probed with traditional Ramsey spectroscopy using RF, microwaves, or lasers.

The Hamiltonian governing the CPV energy shifts [17] in a molecule interacting with external fields is

$$H = -D\mathbf{n} \cdot \boldsymbol{\mathcal{E}} - g\mu_B\mathbf{S} \cdot \boldsymbol{\mathcal{B}} + W_d d_e \mathbf{S} \cdot \mathbf{n} + W_Q \frac{Q}{I} \mathbf{I} \cdot \mathbf{n} - W_M \frac{M}{2I(2I-1)} \mathbf{S} \hat{T} \mathbf{n}. \quad (1)$$

The first line is the external field interaction; D is the

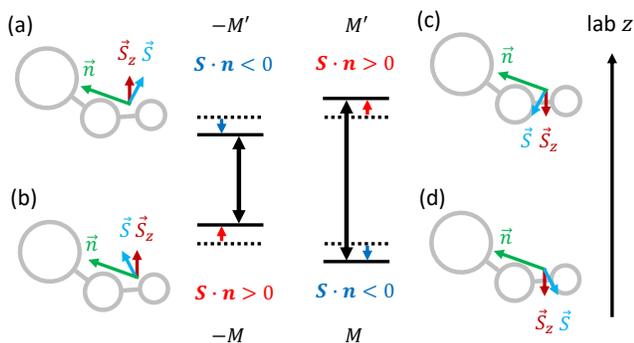


FIG. 1. The CP-FITs with eEDM sensitivity in partially polarized molecules where the molecule axis \mathbf{n} and the lab z axis are not fully aligned. States (a) and (b) have the same orientation of \mathbf{n} with respect to z as well as the same projection S_z of \mathbf{S} on z , and thus have the same response to electric and magnetic fields. On the other hand, they have different orientations of \mathbf{S} on \mathbf{n} , and thus different eEDM energy shifts (short arrows). This results in transitions (long arrows) sensitive to eEDM interactions but highly insensitive to electric and magnetic fields. The transition connecting states (c) and (d) is similar though it has the opposite eEDM shift, serving as an internal co-magnetometer. The same mechanism works for CP-FITs with NSM and MQM sensitivity.

molecular dipole moment, \mathbf{n} is a unit vector along the molecular axis, g is the magnetic g -factor, and \mathbf{S} is the effective electron spin. **Here we define the molecular axis as the symmetry axis, along which the dipole moment lies.** The second line is the CPV effects; d_e is the eEDM, Q is the NSM, \mathbf{I} is the nuclear spin, M is the MQM, and \mathbf{T} is the rank 2 tensor operator $T_{ij} = I_i I_j + I_j I_i - \frac{2}{3} \delta_{ij} I(I+1)$. W_d, W_Q , and W_M are sensitivities to eEDM, NSM, and MQM, which are determined by electronic structure, and are the same within the same electronic state to good approximation. On the other hand, $P_d \equiv \langle \mathbf{S} \cdot \mathbf{n} \rangle$, $P_Q \equiv \frac{1}{I} \langle \mathbf{I} \cdot \mathbf{n} \rangle$, and $P_M \equiv \frac{1}{2I(2I-1)} \langle \mathbf{S} \mathbf{T} \mathbf{n} \rangle$ are measures of the eEDM, NSM, and MQM sensitivities which depend on each particular quantum state, and in general have different orientations of \mathbf{S} , \mathbf{I} , and \mathbf{n} . In this manuscript, we refer to these state-dependent sensitivities P_d, P_Q , and P_M as “CPV sensitivities” as we shall restrict our discussion to transitions within a given electronic state.

The existence of CP-FITs – pairs of states having similar electric and magnetic field sensitivities but different CPV sensitivities – can be intuitively understood as arising from the ability to access different orientations of the molecular \mathbf{n} and lab z axes. Figure 1 shows a time-reversed pair of two CP-FITs in a partially polarized

As a specific example, we examine CP-FITs in the $^{173}\text{YbOH}$ metastable bending mode of the electronic ground state, $\tilde{X}^2\Sigma_{1/2}^+(010)$. The combination of parity doublets from the mechanical bending vibration and the heavy, quadrupole-deformed ^{173}Yb ($I = 5/2$) nucleus offers good sensitivity to the eEDM, MQM, and NSM [9, 23, 24]. We shall find CP-FITs in this molecule,

molecule where \mathbf{n} and the lab z axis are not fully aligned. The field sensitivities arise from the projections of \mathbf{n} and \mathbf{S} on the *lab* axis whereas the CPV sensitivities arise from the projections of \mathbf{S} and \mathbf{I} on the *molecular* axis. The interactions between \mathbf{S} , \mathbf{I} , and the angular momentum associated with the molecular axis (e.g. spin-rotation interaction), all of which generically exist in species with non-zero electron and nuclear spin, offer different orientations of \mathbf{S} and \mathbf{I} with respect to \mathbf{n} . Thus, when molecules are partially polarized, states with the same projections of $\mathbf{n} \cdot z$ and $\mathbf{S} \cdot z$ (field sensitivities) can have different projections of $\mathbf{S} \cdot \mathbf{n}$, $\mathbf{I} \cdot \mathbf{n}$, and $\mathbf{S} \mathbf{T} \mathbf{n}$ (CPV sensitivities).

As the molecular Stark shifts begin to approach the magnitude of other splittings in the molecule, the coupling of the spins and dipole moment can transition from being molecule-quantized to lab-quantized, resulting in changes to the behavior of Stark and Zeeman shifts, including the appearance of pairs of states with the same absolute or differential (“magic”) field sensitivity. This has been observed in molecules [16, 18], and is, for example, the intuitive reason behind magnetically-insensitive states and transitions [19] in alkali atoms. In this manuscript, we show that these exist rather generically, and that we can find transitions that have large CPV-sensitivity.

Although this mechanism is similar to what gives rise to eEDM-sensitive, magnetically-insensitive states in polyatomic molecules [9, 12], here we are exploring *transitions* between states which may have large electromagnetic field sensitivity, but whose shifts are similar, making transition frequencies largely insensitive to electromagnetic fields while still sensitive to CPV. Similar types of field-insensitive transitions in atoms and molecules have been explored in other contexts, including for clocks [20], quantum computing [21], searches for ultralight dark matter [22], and precision spectroscopy of polyatomic molecules [16], and in the latter case have proven their experimental power.

A time-reversed pair of CP-FITs (that is, transitions between $M \leftrightarrow M'$ and $-M \leftrightarrow -M'$, see Figs. 1 and 2a) are degenerate in the absence of a magnetic field (and CPV), and have exactly the same projection $\mathbf{n} \cdot z$, but opposite projections of \mathbf{S} and \mathbf{I} on \mathbf{n} – that is, opposite CPV sensitivity. By comparing these two transition frequencies without changing lab fields, shifts due to the electric field are canceled while CPV energy shifts are not. Therefore, this scheme provides an opportunity for internal co-magnetometry, even for species and states without parity doublet structures, as well as a number of others.

We diagonalize the molecular effective Hamiltonian [18, 25] including Stark and Zeeman terms to compute quantum state energy levels and magnetic, electric, eEDM, NSM, and MQM sensitivities at each electric field, as described in detail in the Supplemental Material [26]. The nuclear spin and rotational Zeeman terms

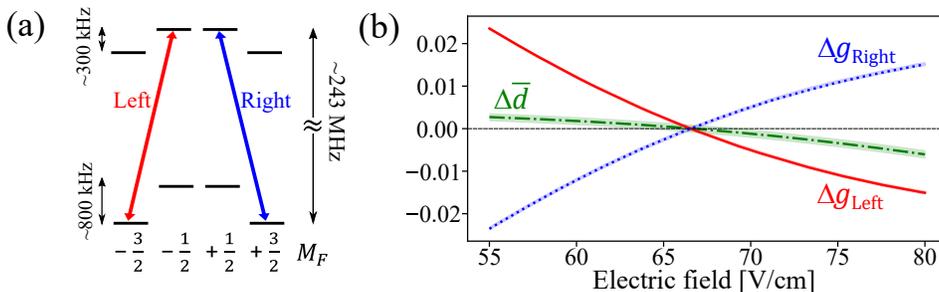


FIG. 2. An example CP-FIT where both $\Delta\bar{d}$ and Δg cross zero near 66.7 V/cm. (a): Level diagram of states and transitions involved. (b): $\Delta\bar{d}$ and Δg as a function of the electric field. The shaded regions indicate the uncertainties from the measurement uncertainties of the spectroscopic parameters.

are neglected since their contributions are expected to be smaller by around three orders of magnitude than the electron spin term [31, 32]. To represent the magnetic and electric sensitivity, the g -factor and dipole moment are computed by taking the first derivative of energies as a function of magnetic and electric fields, respectively.

We are able to find many values of the electric field supporting CP-FITs – pairs of states which have large CPV sensitivity, small g -factor and dipole moment, and which differ by $\Delta M \leq 2$, which makes them easier to drive in the laboratory. Table I lists several examples in $^{173}\text{YbOH}$, and Figure 2 shows one in detail. We characterize these transitions in this and other molecules by computing the differential g -factor between the two states, $\Delta g = g_1 - g_2$, the differential dipole moment $\Delta\bar{d} = (d_1 - d_2)/D$, normalized to the molecular dipole moment, and the differential CPV sensitivities. For ease of comparison, we typically display normalized CPV sensitivities, for example \bar{P}_d , defined to be P_d divided by the value to which P_d saturates in the limit of full mixing of the parity doublets. Note that these values can be larger than 1 since there can be local maxima in sensitivity, as has been observed elsewhere [12, 33], and because the transitions are between two states, which can have additive sensitivity greater than that of a single state. There are more than 250 other CP-FITs in $^{173}\text{YbOH}$ where both $|\Delta\bar{g}|$ and $|\Delta\bar{d}|$ are < 0.01 and $|\Delta\bar{P}_M| > 30\%$. More CP-FITs can be found when one or two of the three requirements for $|\Delta g|$, $|\Delta\bar{d}|$, and CPV sensitivity are relaxed. There are also transitions with large field sensitivity and small CPV sensitivity, providing further opportunities for robust systematic error detection and rejection. Furthermore, these transitions have different relative sensitivities to the eEDM, NSM, and MQM, which is important for disentangling their observable effects.

To quantify the reduction of electromagnetic sensitivity, we calculate decoherence times τ_{EM} due to inhomogeneous field fluctuations $\delta\mathcal{E} \sim 1$ mV/cm and $\delta\mathcal{B} \sim 1$ μG across the molecular sample, which are typical experimental values [34]. Note the experimental coherence time may be limited to smaller values by several factors, including radiative decay of the bending state, which is estimated to be around 800 ms [35]. An example of a CP-FIT shown in Fig. 2 has $\tau_{\text{EM}} \sim 2900$ s at ~ 66.7 V/cm. Both Δg and $\Delta\bar{d}$ are suppressed to below 0.001, and therefore have $\tau_{\text{EM}} \gtrsim 4$ s over a 1.2 V/cm

TABLE I. Example CP-FITs in $^{173}\text{YbOH}$. The top eight transitions have good CPV sensitivity and low field sensitivity (CP-FITs). The bottom two transitions have either large electric or magnetic field sensitivity, but small CPV sensitivity. Note that our uncertainty on Δg is $\sim 10^{-3}$

\mathcal{E} V/cm	f MHz	$ \Delta g $	$ \Delta\bar{d} $	$ \Delta\bar{P}_d $ %	$ \Delta\bar{P}_Q $ %	$ \Delta\bar{P}_M $ %
194.98	58	1×10^{-3}	0	24	39	40
194.48	58	0	3×10^{-4}	24	39	40
66.71	243	2×10^{-3}	0	31	42	42
66.55	243	0	5×10^{-5}	31	42	42
143.1	5,132	3×10^{-3}	0	112	117	141
238.1	23,754	3×10^{-4}	0	93	48	95
533.91	29,520	2×10^{-4}	0	56	86	84
533.97	29,520	0	2×10^{-4}	56	86	84
23.3	5,597	1.1	0	1.0	0.4	0.1
650.6	28,550	0	0.7	0.1	0.2	0.2

range of applied field. Typical, non-field-insensitive transitions have $|\Delta g|, |\Delta\bar{d}| \sim \mathcal{O}(1)$, which would result in $\tau_{\text{EM}} \lesssim 1$ ms. At the same time, the magnitude of the normalized CPV sensitivity maintains reasonably large values of $|\Delta\bar{P}_M| \approx 42\%$, $|\Delta\bar{P}_d| \approx 31\%$, and $|\Delta\bar{P}_Q| \approx 42\%$.

Since there is field dependence of Δg and $\Delta\bar{d}$, there will be residual electromagnetic sensitivity in the presence of any field imperfections or noise, though it is also suppressed. We find $(d\Delta g/d\mathcal{E}) \sim 2 \times 10^{-4}/(\text{V/cm})$ and $(d\Delta\bar{d}/d\mathcal{E}) \sim 1 \times 10^{-3}/(\text{V/cm})$, indicating that high suppression of Δg and $\Delta\bar{d}$ is maintained over more than a few V/cm electric field range and therefore the CP-FIT is robust against the fields gradients and fluctuations. The dependencies of Δg and $\Delta\bar{d}$ on the electric (magnetic) field are linear over a few V/cm (Gauss) around the zero crossing point, so the differential energy shifts are mostly quadratic. Note that this dependence is factored in when calculating τ_{EM} .

An applied magnetic field **parallel (or anti-parallel) to the electric field** can be also utilized to further tune Δg and $\Delta\bar{d}$. For instance, with a CP-FIT where $\Delta\bar{d}$ crosses zero but Δg does not, Δg can further be suppressed or even tuned to zero. An example of this exists at electric field of ~ 143 V/cm and magnetic field of ~ 106 mG in

TABLE II. Measured spectroscopic parameters for the $\tilde{X}^2\Sigma^+(01^1_0)$ state of YbOH.

Parameter		$^{171}\text{YbOH}$	$^{173}\text{YbOH}$
T_0/cm^{-1}	Origin energy	319.90861(6)	319.90932(6)
B/MHz	Rotation	7340.9(3)	7334.1(4)
γ/MHz	Spin-rotation	-90(2)	-87(3)
γ_G/MHz	Axial spin-rotation	12(6)	14(4)
q_G/MHz	ℓ -doubling	-12.6(3)	-12.5(5)
p_G/MHz	P-odd ℓ -doubling	-11(3)	-13(5)
b_F/MHz	Fermi contact	6795(3)	-1881.0(8)
c/MHz	Spin dipolar	281(21)	-92(10)
e^2Qq_0/MHz	Quadrupole	-	-3322(27)

$^{173}\text{YbOH}$, where both the electric and magnetic sensitivities vanish. Note that by applying a magnetic field, the exact cancellation of $\Delta\bar{d}$ and the frequency between time-reversed transitions is lost; however, $\Delta\bar{d}$ only changes by $\sim 1 \times 10^{-5}$. **Although here we only consider magnetic fields parallel (or anti-parallel) to the electric field, applying a magnetic field at a different angle relative to the electric field, or even to the polarization of an optical dipole trap [36, 37], may provide further tuning of field sensitivities.**

While CP-FITs exist quite generically, their location and properties depend strongly on the molecular structure. To understand these transitions in $^{173}\text{YbOH}$, we performed high-resolution spectroscopy on the $\tilde{X}^2\Sigma^+(01^1_0)$ bending mode in $^{173}\text{YbOH}$ and $^{171}\text{YbOH}$ to accurately determine the molecular constants. We studied the $\tilde{A}^2\Pi_{1/2}(000) - \tilde{X}^2\Sigma^+(01^1_0)$ band with the same experimental technique and apparatus employed in $^{174}\text{YbOH}$ recently [18]. Briefly, a cryogenic buffer gas beam is used to perform laser-induced fluorescence spectroscopy under electric and magnetic fields. The chemical enhancement enables identification of each isotopologue's spectral features [25]. We fit the features to an effective Hamiltonian including Stark, Zeeman, rotation, spin-rotation, ℓ -doubling, and hyperfine terms, which are shown in table II. Further details can be found in the Supplemental Material [26].

We varied the spectroscopic parameters in the effective Hamiltonian and confirmed that the CP-FITs still exist with different values of parameters within the experimental uncertainties, with only small changes in their location and properties, as shown in Fig. 2. This indicates that the CP-FITs of a particular species can be understood with only moderate uncertainty on spectroscopic parameters, and exist quite generally without reliance on finely-tuned properties.

We then calculated CP-FITs in several species which are candidates for CPV searches and whose spectroscopic parameters have been measured or calcu-

TABLE III. Examples of CP-FITs in various species. See Supplemental Material for details [26].

Species	I	$\mathcal{E}/(\text{V}/\text{cm})$	$ \Delta\bar{P}_{CPV} /\%$	$ \Delta g $	$ \Delta\bar{d} $
$^2\Sigma^+(01^1_0)$ states					
$^{137}\text{BaOH}$	$\frac{3}{2}$	45.76	173 (MQM)	9×10^{-4}	0
		45.74	173 (MQM)	0	9×10^{-4}
$^{171}\text{YbOH}$	$\frac{1}{2}$	226.3	81 (NSM)	2×10^{-3}	0
$^{88}\text{SrOH}$	0	223.10	94 (eEDM)	1×10^{-2}	0
		223.12	94 (eEDM)	0	4×10^{-3}
$^{174}\text{YbOH}$	0	57.9	136 (eEDM)	5×10^{-2}	0
$^{175}\text{LuOH}^+$	$\frac{7}{2}$	519	98 (MQM)	3×10^{-3}	0
$^2\Sigma^+(v=0)$ states					
^{173}YbF	$\frac{5}{2}$	29,976	33 (MQM)	4×10^{-2}	0
^{171}YbF	$\frac{1}{2}$	28,170	27 (NSM)	0	5×10^{-6}
^{225}RaF	$\frac{1}{2}$	10,215	44 (NSM)	1×10^{-1}	0
^{225}RaF	$\frac{1}{2}$	24,568	43 (NSM)	5×10^{-3}	0
		24,576	43 (NSM)	0	6×10^{-3}
$^3\Delta_1(v=0)$ states					
$^{232}\text{ThF}^+$	0	18.6	56 (eEDM)	1×10^{-4}	0

lated, such as $^{137}\text{BaOH}$ [23, 38, 39], ^{173}YbF [40–42], ^{171}YbF [42, 43], $^{171}\text{YbOH}$ (measured here), $^{174}\text{YbOH}$ [18], $^{88}\text{SrOH}$ [38, 44], $^{175}\text{LuOH}^+$ [45, 46], ^{225}RaF [47, 48], and $^{232}\text{ThF}^+$ [49–51], as shown in table III. We find CP-FITs in all of these species, indicating that they appear to exist quite generally. However, for species without parity doublets, the electric fields required are orders of magnitude higher than those with parity doublets; this is expected as appreciable mixing of the rotational levels must occur for appreciable CPV sensitivity.

Implementing a CPV measurement utilizing these CP-FITs has the advantage of being immediately compatible with Ramsey methods used, for example, by ACME [2] and demonstrated in CaOH [12], which use either coherent population trapping or microwave pulses, respectively, to create and read out the necessary superpositions. There are many CP-FITs having $\Delta M \leq 2$ (which is a restriction we impose on the CP-FITs considered here), and the transition dipole moment of the two CP-FITs shown in Fig. 2, for instance, is ~ 0.2 Debye, meaning that it can be easily driven with the aforementioned methods. Reversing the signs of the applied fields, polarizations, etc. can be used to isolate the CPV and non-CPV signals. The same measurement could be performed on the time-reversed pair to make use of the internal co-magnetometer feature; simultaneous measurement of both transitions could provide very strong robustness.

While the ability to suppress the effects of electromagnetic fields is valuable, any CPV measurement requires a careful and thorough consideration of systematic effects,

which will be unique to each system. Here we mention two example relevant effects.

First, the residual, quadratic field sensitivity mentioned earlier will result in false EDMs due to non-reversing (nr) background fields [34], which can be estimated to be $\{\mu_B \frac{d\Delta g}{d\mathcal{E}} + D \frac{d\Delta \vec{d}}{d\mathcal{B}}\} \mathcal{E}_{nr} \mathcal{B}_{nr} \sim 0.3 \mu\text{Hz}$, or $\lesssim 10^{-31}$ e cm, using $\mathcal{E}_{nr} = 1$ mV/cm and $\mathcal{B}_{nr} = 1 \mu\text{G}$, and can be suppressed by using the internal co-magnetometers. Furthermore, there are different CP-FITs where $\frac{d\Delta g}{d\mathcal{E}} / \Delta P_{CPV}$ differs by a factor of ~ 100 . This means that the false vs. true CPV signals will be different for these transitions. Note that molecules in optical traps will have tensor AC Stark residual moments as well, as was observed in CaOH [12], which could be addressed using the same methods discussed therein.

Second, transverse magnetic fields will introduce coupling between different M_F states and cause unwanted precession and dephasing. These states are typically split by $\delta \gtrsim 100$ kHz due to fine and hyperfine structure, so a transverse field of $\sim \mu\text{G}$ will give rise to a transverse coupling of $\Omega \sim 1$ Hz and second order shift on the order of $\frac{\Omega^2}{4\delta} \sim \frac{(1\text{Hz})^2}{4(100\text{kHz})} \sim \mu\text{Hz}$. Transitions involving $|M_F| = 1/2$ states will have a degenerate partner state directly coupled by a transverse field; such a degeneracy could be lifted by applying a small bias field, with ~ 1 mG giving rise to a second order shift of $\lesssim \text{mHz}$.

In summary, we find transitions in molecules with electric and magnetic field sensitivity suppressed by over a hundred-fold while maintaining good sensitivity to CP-violation. These transitions offer internal co-magnetometry, and can be driven with RF, microwave, or two-photon transitions, and are therefore compatible with Ramsey measurements in both beams and traps. It is also suitable for systems with large angular momentum commonly found in nuclear CPV searches where hyperfine structure increases the internal complexity and congestion. No particular molecular structure is required, though in species with parity doublets the transitions exist at moderate electric fields $\lesssim 1$ kV/cm. It is likely that similar transitions exist in a wide range of species, including atoms [52], and both symmetric and asymmetric tops. They may also be compatible with spin-squeezing methods to decrease the noise limit. Finally, the ability to polarize molecules while suppressing decoherence from electromagnetic field noise with these transitions can be of great interest in other fields including quantum simulation, where a long-range dipole-dipole interaction with a long interaction time is extremely advantageous [53–55].

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