Electric dipole moment searches using polar molecule clock states

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Electric dipole moments (EDMs) of fundamental particles are indicators of parity and time-reversal violating new physics at energy scales beyond the direct reach of colliders. Almost every EDM experiment to date has searched for a small linear Stark effect in a system with unpaired electron or nuclear spins; consequently, magnetic field fluctuations can cause systematic errors and limit the coherence time of the measurements. We propose an alternative method for EDM measurements, using magnetically-insensitive hyperfine clock states that are available in a wide array of polar molecules. This method may be useful for upcoming electron and nuclear EDM searches using polar molecules.

Polar molecules offer one of the best ways to probe the unknown physics that led to the imbalance between matter and anti-matter in the universe [1,2]. Precise measurements using heavy polar molecules, wherein electron spins experience enormous relativistic electric fields, have set stringent bounds on the parity (P) and time-reversal (T) violating permanent electric dipole moment (EDM) of the electron [3,4]—such experiments constrain the parameter space of new physics models out to energy scales beyond 10 TeV [4,5]. Polar molecules have also been used to constrain new physics by placing bounds on P,T-violation in nuclei [6].

Advances in producing ultracold molecules, such as direct laser-cooling of polar molecules [7,10] and assembly of molecules from trapped ultracold atoms [11,13], have generated interest in applying these techniques to electron and nuclear EDM searches [14,18]. Large ensembles of trapped polar molecules can potentially improve the experimental sensitivity to P,T-violating physics by over two orders of magnitude, due to the long trap lifetimes (>10 s) that can be realized. However, molecules that are amenable to direct laser cooling or ultracold assembly are not considered to be optimal for EDM searches [15]. For example, the majority of diatomic molecules that have been laser-cooled and trapped so far have one non-bonding valence electron in a $^2\Sigma$ electronic ground state, which allows them to scatter many laser photons before ending up in dark states [7,10]. Ultracold assembled molecules that have been proposed for electron EDM searches also typically have $^2\Sigma$ ground states [14,16]. However, EDM searches with molecules in $^2\Sigma$ states have to detect small P,T-violating effects in the presence of the much larger Zeeman interaction of their spins with magnetic fields. Such molecules also lack internal co-magnetometer states, compared to molecules with special parity doublet structure [3,4,19,21]. Therefore, controlling systematic errors due to magnetic fields, and obtaining long coherence times in the presence of magnetic field noise, are challenging in electron EDM searches with $^2\Sigma$ molecules. Similar considerations apply to nuclear EDM searches with molecules in $^1\Sigma$ electronic states, where unpaired nuclear spins can couple to environmental magnetic fields.

One reason for the constrained choice of molecules is that most EDM searches use essentially the same technique—measuring energy shifts between oppositely oriented spin states in molecules—which makes them first-order sensitive to magnetic fields due to the Zeeman effect. As a way to circumvent this constraint, we present an EDM measurement technique that uses magnetically insensitive states in polar molecules: this method enables EDM experiments to have long coherence times commensurate with long trap lifetimes, with reduced sensitivity to systematic errors from magnetic fields. It also enlarges the molecular menu for new EDM search experiments.

Electron EDM searches with molecules.—For background we briefly review electron EDM search methods, using $^{174}$Yb$^{19}$F [22] as an example. In the $^2\Sigma$ electronic ground state of $^{174}$Yb$^{19}$F, the lowest rovibrational manifold (vibrational quantum number $v=0$ and rotational angular momentum $N=0$) contains four hyperfine states obtained by coupling the electron spin $S=1/2$ and the nuclear spin $I=1/2$. The Hamiltonian for the interaction of these states with external electric and magnetic fields is (with $\hbar=1$ everywhere)

$$H'_{\text{int}} = -\mu_E \left( g_S \hat{S} + g_I \hat{I} \right) \cdot \vec{B} - D \hat{n} \cdot \vec{E} + W_{PT} \hat{S} \cdot \hat{n},$$

where $g_S, g_I$ are the electron and nuclear spin g-factors, $D$ is the molecular dipole moment, and $\hat{n}$ is the unit vector along the internuclear axis of the molecule. The term $W_{PT}\hat{S} \cdot \hat{n}$ is an effective low-energy molecular Hamiltonian that parametrizes parity and time reversal violating new physics. The scalar $W_{PT}$ is the sum of two $P,T$-violating terms, $W_{PT} = W_d + W_{SP}$, which are expectation values of the relativistic electron EDM interaction with the electric field inside a fully oriented polar molecule ($W_d$), and the $P,T$-violating scalar-pseudoscalar interaction between the electron and nucleus ($W_{SP}$), in the molecular electronic state (cf. [28]). The quantity $W_d$ is sometimes written as $d_0\mathcal{E}_{\text{ef}}$ in the literature. The goal of electron EDM search experiments is to accurately measure or set limits on the size of $W_{PT}$ [1,2].

In these experiments, a strong electric field $\vec{E} = \mathcal{E}_z \hat{z}$ is applied along with a small bias magnetic field $\vec{B} = B_z \hat{z}$.
we will refer to as a spin interferometer (SI). The SI
ζ
The molecule polarizes in the electric field, leading to a
electric and magnetic fields for a time
τ
|the superposition state
W
ω
⟩
|↑↑⟩
stretched hyperfine states as
states
|m
⟩
|0
|ω
= 0 due to azimuthal symmetry). The interaction Hamiltonian for just the electron and nuclear
spins of freedom is then
H_{\text{int}} = - (g_s S_z + g_l I_z) \mu_B B_z + W_{PT} S_z \zeta. \quad (2)
The molecular orientation \( \zeta = \langle \mathbf{n} \cdot \mathbf{z} \rangle \) depends on the applied electric field \( E_z \), the dipole moment \( D \), and the energy separation between opposite parity states in
the molecule [see Supplementary Material (SM), Sec. A].
The standard electron EDM search method measures the energy difference between the stretched hyperfine states \(|F = 1, m_F = \pm 1\rangle\) (see e.g., [22]). We denote these
stretched hyperfine states as \(|↑↑\rangle, |↓↓\rangle\). The energy difference between these states, \(\omega_B + \omega_{PT} = -(g_s + g_l) \mu_B B_z + W_{PT} \zeta\), is measured using a Ramsey-type technique which we will refer to as a spin interferometer (SI). The SI
method is represented graphically in Fig. 1(a). First, the superposition state \(|X\rangle = \frac{|↑↑⟩+|↓↓⟩}{\sqrt{2}}\) is prepared, starting from \(|F = 0, m_F = 0\rangle\). Under the influence of the electric and magnetic fields for a time \(\tau\), the superposition evolves into the state \(|\Psi\rangle = \frac{|↑↑⟩+e^{i\phi}|↓↓⟩}{\sqrt{2}}\). The phase
\(\phi = (\omega_B + \omega_{PT}) \tau\) is extracted from \(|\Psi\rangle\) by projectively measuring the populations in \(|X\rangle\) and the orthogonal state \(|Y\rangle = \frac{|↑↑⟩-|↓↓⟩}{\sqrt{2}}\). The \(P,T\)-violating phase \((\omega_{PT} \tau)\) is separated from the \(B\)-dependent phase \((\omega_B \tau)\) by measuring \(\phi\) under different combinations of the sign of the magnetic field \(B_z\) and the molecular orientation \(\zeta\). The SI method, with some slight variations, is essentially used in every molecule-based electron EDM search experiment [3 4 18 22 24 25].

Clock transition (CT) method. – Our proposed method uses time-dependent electric and magnetic fields to drive populations between the hyperfine clock states, \(|g\rangle \equiv |F = 0, m_F = 0\rangle = \frac{|↑↑⟩-|↓↓⟩}{\sqrt{2}}\), and \(|e\rangle \equiv |F = 1, m_F = 0\rangle = \frac{|↑↓⟩+|↓↑⟩}{\sqrt{2}}\). We consider time-
dependent magnetic and electric fields applied to the
molecules: \(B_z = B_0 \cos(\omega_E t), E_z = E_0 \cos(\omega_E t + \beta)\), where \(\beta\) is an adjustable phase. The oscillating electric field induces an oscillating molecular orientation with amplitude \(\zeta_0\). The dependence of \(\zeta = \langle \mathbf{n} \cdot \mathbf{z} \rangle\) on \(E_z\) is nonlinear and so \(\zeta(t)\) generally contains odd harmonics of \(\omega_E\) as shown in Fig. 2 [see also Figs. 3, 5 in the SM]. For now, let \(\zeta_0\) refer to the amplitude of oscillations at the fundamental frequency \(\omega_E\). We also assume for now that the electric and magnetic fields are driven at the same frequency, \(\omega_E = \omega_B = \omega\).

We denote the energy separation between the clock states in the presence of \(B_z(t), E_z(t)\) (i.e., including the zero-field hyperfine splitting, and the Zeeman and tensor Stark shifts) as \(\omega_0\), and define the detuning \(\Delta = \omega - \omega_0\). We make the rotating wave approximation for the time-dependent fields (assuming that the Bloch-Siegert shift from the counter-rotating term is also included in \(\omega_0\)). Then the Hamiltonian from Eq. (2), in the two-dimensional subspace spanned by \(|g\rangle\) and \(|e\rangle\), is just
\[H_{\text{int}} = \frac{\Omega_B}{2} \sigma_x + \frac{\Omega_{PT}}{2} (\cos \beta \sigma_x + \sin \beta \sigma_y) + \frac{\Delta}{2} \sigma_z, \quad (3)\]
where \(\sigma_{x,y,z}\) are Pauli matrices, and the Rabi frequencies for the Zeeman and \(P,T\)-violating interactions are \(\Omega_B = -\frac{1}{2} (g_s + g_l) \mu_B B_0\) and \(\Omega_{PT} = \frac{1}{2} W_{PT} \zeta_0\) respectively.

We assume that the molecules are initially prepared in the ground hyperfine clock state \(|g\rangle\). On resonance \((\Delta = 0)\) and under the influence of \(H_{\text{int}}\) for a time \(\tau\), the population in the excited clock state \(|e\rangle\) is \(\rho_{ee}(\tau) = \sin^2 \left( \frac{(\Omega_B + \Omega_{PT} \cos \beta) \tau}{2} \right)\). (We have dropped terms that are quadratic in \(W_{PT}\) ) The \(P,T\)-violating signal is contained in the frequency of the Rabi oscillations of the clock state populations. The measurement of \(W_{PT}\) from the population \(\rho_{ee}\) (of \(\rho_{gg} = 1 - \rho_{ee}\)) is illustrated in Fig. 3. The magnetic field amplitude \(B_0\) and the pulse duration \(\tau\) are set so that \(\Omega_B \tau \approx \pm \pi\) (modulo 2\(\pi\)), and the populations of the clock states are measured for different phase angles \(\beta\). When \(\beta = 0\) (\(\beta = \pi\)) the \(P,T\)-violating term \(\Omega_{PT}\)
FIG. 2: The magnetic field $B_z(t)$ (black, dots) and molecular orientation $\zeta(t)$ (red, solid) in the CT method. The curve for $\zeta(t)$ is the calculated molecular response to an electric field $E_x(t) = E_0 \cos(\omega t + \beta)$ (see SM, Sec. A). The red dashed line shows the first harmonic of $\omega$ in $\zeta(t)$.

FIG. 3: The population in the excited clock state $|e\rangle = \frac{|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle}{\sqrt{2}}$ as a function of time, with the electric and magnetic fields driven at resonance ($\Omega_{PT}$ is greatly exaggerated for illustration). The relative phase $\beta$ between $E_x(t)$ and $B_z(t)$ can be varied to separate the small $P,T$-violating contribution to the Rabi frequency from the larger magnetic-field-induced Rabi frequency.

Hz leads to an electron EDM uncertainty $\delta d_e = 10^{-31}$ e cm.

We have discussed electron EDM searches using molecules with $^2\Sigma$ ground states so far, but the CT method is also applicable to nuclear EDM searches using $^1\Sigma$ diatomic molecules with $m_I = 0$ hyperfine states (e.g., $^{207}\text{Pb}^{17}$O). The hyperfine splitting $\omega_0$ in such molecules arises from dipolar coupling of the two nuclei, and is consequently smaller (typically $\omega_0 \sim 2\pi \times 10$ kHz). For nuclear EDM searches, the quantities $\bar{S}$ and $S_z$ in Equations (1) and (2) are to be replaced by $I_M$ and $I_{M,z}$ respectively, where $I_M$ is the spin of the $P,T$-violation-sensitive nucleus ($^{207}\text{Pb}$ in our example). The quantity $W_{PT}$ is to be understood as the expectation value of the nuclear EDM (arising from $P,T$-violating nuclear interactions and quark EDMs) interacting with the electric field inside a fully oriented molecule. The remainder of our discussion is also applicable to nuclear EDM searches.

Advantages. – Viewed on the Bloch sphere (Fig. 1), it is evident that the CT method is a “rotated” version of the usual SI method. It is therefore unsurprising that the CT and SI methods have comparable statistical sensitivities. However, there are some practical advantages to the CT method:

a) The frequency $\omega_0$ (and thus $\Delta$) is insensitive to magnetic fields to first order, and the Rabi fringe phase is further only quadratically sensitive to $\Delta$ when $\Delta \ll \Omega_B$. Therefore the requirements for shielding low-frequency stray magnetic fields are severely relaxed. Polar molecules with special internal structure (e.g., $\Omega$-doublets or $\ell$-doublets) to track and cancel magnetic field drifts are no longer required. This allows ground-state polar molecules with simple electronic structure (e.g., $^2\Sigma$ or $^1\Sigma$) to be used in competitive electron or nuclear EDM experiments.

b) The coherence times for hyperfine state superpositions, which are insensitive to magnetic noise, are much longer than coherence times for stretched state superpositions. This allows large values of $\tau$ to be practically realized, leading to improved statistical sensitivity.

c) The initial state preparation in this method is simple: it is easy to accurately initialize molecules in the ground hyperfine state, compared to preparing an accurate superposition $|X\rangle$ in the SI method. This feature can improve the duty cycle of experiments and eliminate systematics that arise from imperfect state preparation.

d) Just as stray static magnetic fields lead to spurious phase accumulation in the SI method, stray radio-frequency (rf) magnetic fields in the neighborhood of $\omega_0 \pm 1/\tau$ can shift the Rabi frequency in the
CT method. However, shielding rf magnetic fields is significantly easier than shielding low-frequency magnetic fields.

e) The phase $\beta$ between the electric and magnetic fields can be varied rapidly and smoothly, without correlated switching transients and charging currents that usually accompany reversals of DC electric fields. This allows potential systematics (e.g., from drifts in $\Omega_B$ or non-reversing electric fields) to be cleanly cancelled.

f) This method can be applied to molecular ions, since the hyperfine resonance frequency $\omega_0$ can be chosen to be far away from typical ion trap motional frequencies. Therefore the method enables EDM experiments with a wide class of molecular ions without imposing further constraints, such as requiring $3\Delta_1$ molecular structure, for the sake of magnetic field control. In particular, this opens a feasible approach to EDM searches using nuclei that are short-lived but highly sensitive to $P,T$-violation (e.g., $^{228}\text{Ra}$, $^{229}\text{Pa}$, $^{285}\text{Cu}$ [29]), where there may only be limited options for molecules that can be efficiently created from rare isotope sources.

**Controlling systematics.** – A genuine $W_{PT}$-dependent signal can be identified as the part of $\rho_{ee}$ that changes sign under switches of (i) the initial state ($\langle e \mid \rho \mid g \rangle$), (ii) the phase $\beta$ (0 or $\pi$), and (iii) the pulse area $\Omega_B \tau$ ($\pm \pi/2$ modulo $2\pi$), allowing $B_z$ and $\tau$ to be varied over a large dynamic range to find systematic errors. The $P,T$-violating observable in the CT method is an off-diagonal matrix element or Rabi frequency, rather than an energy matrix element or Rabi frequency, rather than an energy shift as in the SI method. Therefore transition amplitudes that are in phase with the driving electric field can lead to systematic errors.

One strategy for detecting such errors, just as in SI experiments, is to make measurements in states (e.g., $m_P = 0 \rightarrow m_F = 0$ transitions in the $N = 0$ and $N = 1$ rotational manifolds) that have similar responses to magnetic fields but different magnitudes and signs of $\zeta$. Such states can function as adequate internal co-magnetometers even in simple $2\Sigma$ molecules (see e.g., [30]). However a new source of potential systematic errors in the CT method is the displacement current due to the oscillating $E$-field. This induces a $B$-field with amplitude $B_d \sim \frac{\omega_E}{\omega_0} E_0$, where $\ell$ is a length scale on the order of the size of the electrodes. With $\ell \sim 1$ cm, $E_0 \sim 10$ kV/cm and $\omega_E = 2\pi \times 10$ MHz, the displacement $B$-field has amplitude $B_d \sim 50$ mG, which leads to a spurious Rabi frequency $\Omega_d \sim 2\pi \times 100$ kHz that mimics $\Omega_{PT}$. We describe a way to suppress this effect. First, the displacement $B$-field is perpendicular to $E$, which suppresses shifts in $\rho_{ee}$ because $B$-fields in the $xy$-plane only couple $|F = 0, m_F = 0\rangle$ to the $|F' = 1, m_{F'} = \pm 1\rangle$ levels. Further, these transitions are out of resonance with the frequency $\omega_E = \omega_0$ due to the tensor Stark shift in polar molecules [26, 31, 32]. Any residual $B$-field component along $\hat{z}$, due to mis-alignment of the electrodes for example, can be further suppressed using the fact that the induced $B$-field is proportional to the time derivative of $E_z$, and therefore lags the applied $E$-field in phase by $\pi/2$. For example, if $B_d \cdot \hat{z} \neq 0$ and $\beta$ is set to $\pm \pi/2$, there will be a change in $\rho_{ee}$ depending on whether the $E$-field is on or off. Such a shift is only produced when $B_d \cdot \hat{z} \neq 0$, and can therefore be used as a diagnostic for displacement $B$-fields. Despite these, it is conceivable that a combination of phase errors (due to charging currents or cable impedance mismatches) and electrode imperfections could still lead to a residual $B$-field that is both parallel to $\hat{z}$ and in phase with $E_z$: misalignment of the rf electric and magnetic field directions by $\theta = 0.1$ mrad, and a phase error in the electric field drive by $\Delta \beta = 1$ mrad would lead to a residual Rabi frequency $\Omega_d = 0 \Delta \beta$ would lead to a residual Rabi frequency $\Omega_d = 0 \Delta \beta$ that mimics $\Omega_{PT}$.

We point out that there is a general way to further suppress systematic errors generated by the oscillating $E$-field, using the fact that the molecular orientation $\zeta$ is nonlinear in $E_z$. If $E_0$ is large enough to appreciably polarize the molecule, then $\zeta(t)$ also contains higher odd harmonics of $\omega_E$ (see SM, Fig. 4). Therefore, an EDM search experiment can be conducted using, e.g., $\omega_E = \omega_0/3$ and $\omega_B = \omega_0$. Any induced magnetic fields that are linear in $E_z$ (a condition which covers the majority of conceivable systematics) oscillate at $\omega_0/3$, far off resonance from the clock transition, and so their interference with the transition amplitude is greatly suppressed. On the other hand, the Fourier component of $\zeta$ that oscillates at $3\omega_E = \omega_0$ (with amplitude $\zeta_3$) resonantly contributes to the transition probability as $\rho_{ee}(\tau) \sim \sin^2 \left[ \left( \Omega_0 + \Omega_{PT,3} \cos(3\tau) \right) / 2 \right]$, with $\Omega_{PT,3} = \frac{1}{2} W_{PT} \zeta_3$. Therefore, driving the electric field at a sub-harmonic of $\omega_0$ offers a convenient diagnostic for discriminating between systematic errors and real $P,T$-violating signals. Since $\zeta_3 < \zeta_0$ though, it yields lower EDM sensitivity, so we envision that experiments using the CT method will intersperse some measurements with $\omega_E = \omega_0/3$ as systematic checks within larger measurement blocks with $\omega_E = \omega_0$.

We have also considered systematic errors due to effects such as $E_1 - M_1$ mixing: static background electric ($E_{dc}$) and magnetic ($B_{dc}$) fields can admix states in the $N = 0$ and $N = 1$ manifolds. This induces a transition Rabi frequency $\Omega_{E_1 - M_1} \propto E_{dc} B_{dc} \omega_{E_0}$ driven by $E_z(t)$ which mimics $\Omega_{PT}$. Numerical and perturbative calculations for the parameters of YbF (see SM, Sec. A), using realistic estimates of the background fields $E_{dc} = 1$ V/cm and $B_{dc} = 10$ mG, yield $\Omega_{E_1 - M_1} \sim 2\pi \times 10^{-10}$ Hz, which is a negligible systematic error compared to the expected precision of electron EDM experiments with trapped molecules.

In summary, we have devised a measurement technique using hyperfine clock states that offers advantages for
electric dipole moment searches using polar molecules. The use of magnetically insensitive states enables longer coherence times for improved precision, and leads to an enhanced level of systematic error control. Our method opens up a wide selection of polar molecules for use in high-precision EDM search experiments – in particular, this includes simple diatomic molecules that can be directly laser-cooled or assembled out of ultracold atoms.

We thank Wes Campbell, Jonathan Weinstein and David DeMille for helpful comments. M.V. acknowledges support from NSERC and an Ontario Graduate Scholarship. A.M.J. acknowledges support from NSF Grant No. PHY-1912665. A.C.V. acknowledges support from Canada Research Chairs and a Sloan Fellowship.
A. Calculation of the molecular orientation

The field-free Hamiltonian for the $^2\Sigma$ electronic ground state of a molecule such as $^{174}\text{Yb}^{19}\text{F}$ is

$$H_0 = B_{\text{rot}} N (N + 1) + \gamma \vec{S} \cdot \vec{N} + b \vec{S} \cdot \vec{I} + c S_z I_z,$$

(4)

where $\vec{N}, \vec{S}, \vec{I}$ are the molecular rotational angular momentum, electron spin and nuclear spin respectively. $B_{\text{rot}}$ is the rotational constant of the molecule, $\gamma$ is the spin-rotation parameter, and $b, c$ are hyperfine interaction parameters. In an electric field, the interaction Hamiltonian (see Eq. (1)) is $H_{\text{int}} = D \hat{n} \cdot \vec{E}$ where $D$ is the molecular dipole moment.

A characteristic scale for the electric field required to polarize the molecule can be constructed from these constants,

$$E_{\text{pol}} = \frac{2 B_{\text{rot}}}{D}.$$

We used the uncoupled computational basis $|N, m_N; S, m_S; I, m_I\rangle$, including rotational levels up to $N = 20$, and numerically diagonalized $H_0 + H_{\text{int}}$ for different values of $E_z$. The resulting dependence of the molecular orientation $\zeta = \langle \hat{n} \cdot \hat{z} \rangle$ is shown in Fig. 4. For this calculation, we used measured values of $B_{\text{rot}}, \gamma, b, c$ for $^{174}\text{Yb}^{19}\text{F}$\cite{33} – the resulting value of $E_{\text{pol}} = 7.3$ kV/cm. (This value of $E_{\text{pol}}$ lies near the higher end for the list of molecules in Section B.) When the curve of $\zeta$ versus $E_z$ is smoothly interpolated and applied to a sinusoidal electric field $E_z(t) = E_0 \cos(\omega t + \beta)$ (with $E_0 = 3E_{\text{pol}}$), the curve for $\zeta(t)$ in Fig. 2 is obtained.

We also use this numerical model to calculate systematic errors, such as the $E1-M1$ mixing-induced Rabi frequency $\Omega_{E1-M1}$ described in the main text. The numerical calculations confirm the estimate from perturbation theory,

$$\Omega_{E1-M1} \sim D E_0 \left( D E_{dc} \right) \frac{g_S \mu_B B_{dc}}{B_{\text{rot}}} \frac{\gamma^2}{B_{\text{rot}}}.$$

![FIG. 4: Molecular orientation $\zeta = \langle \hat{n} \cdot \hat{z} \rangle$ as a function of the electric field applied to the molecule.](image)

B. Molecules for electron and nuclear EDM searches

The following tables list neutral molecules (Table I) and singly-charged molecular ions (Table II) that can be used for electron EDM and nuclear EDM searches using the CT method. In each table, a combination of species from the two columns forms an EDM-sensitive molecule to which the CT method can be applied. The tables are by no means exhaustive – other analogous molecular systems can potentially be used. The tables include some molecules that have been previously used ($\text{YbF}$\cite{22}) or proposed for use in EDM experiments ($\text{HgF}, \text{HgCl}, \text{HgBr}$\cite{30}, $\text{HgNa}, \text{HgK}, \text{HgRb}$\cite{14}, $\text{RaF}$\cite{34}, $\text{BaF}$\cite{24}, $\text{RaAg}$\cite{16}, $\text{HgCa}$\cite{35}). The CT method can also be applied to other molecules that do not fall into the tables’ categories (e.g., $^{205}\text{Tl}^{19}\text{F}$).
FIG. 5: Molecular orientation $\zeta = \langle \hat{n} \cdot \hat{z} \rangle$ in response to an electric field $\mathcal{E}_z(t) = 3\mathcal{E}_\text{pol} \cos \omega t$. The harmonics contained in $\zeta(t)$ are offset for clarity.

TABLE I: Neutral molecules.

| Electron EDM          | Nuclear EDM  |
|-----------------------|--------------|
| $^{138}\text{Ba}$    | $^{199}\text{Hg}$ |
| $^{174}\text{Yb}$    | $^{17}\text{O}$  |
| $^{202}\text{Hg}$    | $^{207}\text{Pb}$ |
| $^{226}\text{Ra}$    | $^{225}\text{Ra}$ |
| $^{19}\text{F}$      | $^{17}\text{O}$  |
| $^{35}\text{Cl}$     | $^{33}\text{S}$  |
| $^{79}\text{Br}$     | $^{43}\text{Ca}$ |
| $^{107}\text{Ag}$    | $^{33}\text{S}$  |
| $^{16}\text{O}^1\text{H}$ | $^{87}\text{Sr}$ |

TABLE II: Molecular ions.

| Electron EDM          | Nuclear EDM  |
|-----------------------|--------------|
| $^{200}\text{Hg}$    | $^{133}\text{Ba}$ |
| $^{226}\text{Ra}$    | $^{19}\text{F}$  |
| $^{208}\text{Pb}$    | $^{199}\text{Hg}$ |
| $^{232}\text{Th}$    | $^{17}\text{O}$  |
| $^{33}\text{S}$      | $^{35}\text{Cl}$ |
| $^{43}\text{Ca}$     | $^{79}\text{Br}$ |
| $^{16}\text{O}^1\text{H}$ | $^{16}\text{O}^1\text{H}$ |
| $^{87}\text{Sr}$     | $^{229}\text{Pa}$ |
