Nuclear-Spin Dependent Parity Violation in Optically Trapped Polyatomic Molecules

E. B. Norrgard, D. S. Barker, S. Eckel, J. A. Fedchak, N. N. Klimov, J. Scherschligt
Sensor Sciences Division, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

We investigate using optically trapped linear polyatomic molecules as probes of nuclear spin-dependent parity violation. The presence of closely spaced, opposite-parity \( \ell \)-doublets is a general feature of such molecules, allowing parity-violation-sensitive pairs of levels to be brought to degeneracy in magnetic fields typically 100 times smaller than in diatomics. Assuming laser cooling and trapping of polyatomics at the current state-of-the-art for diatomics, we expect to measure nuclear spin-dependent parity-violating matrix elements \( iW \) with 70 times better sensitivity than the current best measurements. Our scheme should allow for 10\% measurements of \( iW \) in nuclei as light as Be or as heavy as Yb, with averaging times on order the of 10 days and 1 second, respectively.

Measurements of nuclear spin-independent (NSI) and nuclear spin-dependent (NSD) parity violation (PV) are a means to probe Standard Model (SM) electroweak interactions on a tabletop scale [1]. NSI-PV has been measured in protons and a number of heavy atoms [2,7] and found to be in good agreement with SM PV predictions due to the weak charge \( Q_W \). However, the only non-zero measurement (14\% relative uncertainty) of NSD-PV in an atomic system comes from Cs [8], and this result implies constraints on SM meson-nucleon couplings which are in disagreement with other atomic PV measurements [9,10]. NSD-PV arises primarily from two interactions: vector electron-axial nucleon electroweak current coupling \( (V_A) \), and the nuclear anapole moment. The \( V_A \) effect is described by two parameters \( C_{2u} \) and \( C_{2d} \) relating to spin-dependent \( Z_0 \) boson exchange between an electron and an up or down quark, respectively. These parameters are among the most poorly measured in the SM, with relative uncertainties 300\% and 70\%, respectively [11]. PV measurements may also probe beyond standard model physics [12,13]. Searches for oscillating PV signals have been proposed as a means to detect axion-like particles, a leading dark matter candidate [14].

A beam of cold diatomic molecules has been demonstrated [15,19] to be a highly sensitive system for measuring NSD-PV effects. Mixing of opposite-parity quantum states from PV effects is amplified when the states have nearly the same energy [1]. The lowest two rotational states of diatomic molecules have opposite parity and may be brought to near degeneracy using a large magnetic field \( B \). While this method is quite general, current systematic uncertainties are roughly 100 times too large to measure NSD-PV in the lightest nuclei where nuclear structure calculations are tractable [21].

Recent advances in laser cooling have lead to optically trapped diatomic molecules with sub-Doppler temperature and single-molecule detection efficiency [21], while similar strides with polyatomic molecules have followed closely behind [22,23]. Moreover, polyatomic molecules have been proposed as exquisite systems for precision measurements of fundamental symmetries [26,28] and time variation of fundamental constants [24,25].

Here, we show that linear asymmetric polyatomic molecules in an optical trap are well-suited for measurement of NSD-PV. Polyatomic molecules possess opposite-parity states 10 to 1000 times closer in energy than diatomics, requiring similarly smaller \( B \)-fields. Systematic uncertainties are reduced compared to beam experiments due to the lower magnetic field and a smaller interaction volume. Furthermore, these smaller fields may be produced without superconducting magnets, allowing trivial \( B \)-field reversal for detection and mitigation of systematic effects. We show that these molecules may be optically trapped using “magic” conditions where differential light shifts are small enough for a precise PV measurement. The obvious advantage of performing a precision measurement on trapped species compared to a beam is the increased interaction time \( \tau \). The sensitivity to any PV matrix element \( iW \) is \( \delta W/W = 1/\tau \sqrt{N_m} \), where \( N_m \) is the total number of measurements \( (iW) \) is purely imaginary due to conservation of time-reversal symmetry. Assuming optical trapping of polyatomic molecules at the current state-of-the-art for diatomics [21,31,32], we expect at least a factor of 70 increase in PV sensitivity over the state-of-the-art BaF measurement [18,33]. Our method is applicable to all laser-coolable polyatomic molecules with \( ^2\Sigma \) ground states, and possibly others.

Consider the properties of linear asymmetric molecules with a \( ^2\Sigma^+ \) electronic ground state. If a bending vibrational mode (with vibrational constant \( \omega_b \) and quantum number \( v_b \)) is excited and all other vibrational modes are in their ground state, the molecule’s rotational angular momentum \( N \) has a projection along the molecular axis \( \ell = \pm v_b, \pm (v_b - 2), \ldots, \pm 1 \) or 0. Within this vibrational manifold, the effective Hamiltonian is

\[
H = B(N^2 - \ell^2) \pm (1) N \nu_b N^2 + \gamma N \cdot S + b I \cdot S + c I_z S_z - c T^2(\nabla E) \cdot T^2(Q) + (\mu_{BS} \mathbf{B} \cdot \mathbf{S}) + (\mu_{B} \mathbf{g}_{I} \mathbf{B} \cdot \mathbf{I})
\]

(1)

where \( B \) is the rotational constant, \( \gamma \) is the spin-rotation (SR) constant, \( b \) and \( c \) are hyperfine (HF) constants, \( e \) is the electron charge, \( T^2(\nabla E) \cdot T^2(Q) \) is a scalar product of rank-2 spherical tensors describing the elec-

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* Electronic address: eric.norrgard@nist.gov
The NSD-PV Hamiltonian is proportional to the strength of the nuclear anapole moment and is proportional to \( \kappa_b a \). Here, \( \kappa_b \) is the nuclear anapole moment, \( a \) is the unit vector along the molecular axis, and \( | \nabla \times \mathbf{A} | \) is the nuclear anapole moment. In a given nucleus, various \( \kappa_b \) are proportional to the strength of the \( \mathbf{V}_C \mathbf{A}_n \) coupling, and is independent of nuclear mass \( A \) (for a typical nucleus, \( | \kappa_b | \approx 0.05 \)). \( \kappa_a \) is proportional to the nuclear anapole moment and is proportional to \( A^{2/3} \). \( \kappa_Q \) is due to the combined effects of nuclear weak charge and normal hyperfine structure, and is negligible compared to \( \kappa_2 \) and \( \kappa_a \). Measurements in several nuclei are required to distinguish among the different NSD-PV effects.

Ultimately, the ability to precisely determine \( \kappa \) and its underlying contributions will be limited to the accuracy of theoretical values of \( W_p \). Calculations of \( W_p \) have been performed on several diatomic molecules via Dirac-Hartree-Fock and relativistic density-functional \( \kappa \), quasi-relativistic zero-order regular approximation \( \kappa_2 \), and (with an estimated 1.5% accuracy) relativistic coupled-cluster \( \kappa_a \) methods. While such calculations are beyond the scope of this proposal, a semiempirical method may be used to calculate \( W_p \) for any species to approximately 10%, assuming the SR/HF constants in Eq.1 are known \( \kappa_2 \). The SR/HF constants relating to a typical metal atom M differ by a approximately 10% between laser-coolable monofluorides MF and MX molecules, where X is a suitable ligand with charge state -1 (e.g., OH, NC, or CCH). Thus, we expect similar 10% accuracy when estimating \( W_p \) for MX, using either MX SR/HF constants for a semiempirical calculation or a more detailed method described above for the corresponding MF. In Table I, we give a list of laser-coolable polyatomic molecules MX, with \( W_p \) from calculations on MX where available or MF otherwise.

### Table I. Parameters for PV measurement in polyatomic molecules.

| Species | \( I \) | \( W_p/2\pi \) (Hz) | \( W''/2\pi \) (Hz) |
|---------|------|----------------|----------------|
| \( ^{171}\text{Yb} \) | 3/2 | 4.1 | 3.9 |
| \( ^{87}\text{Sr} \) | 9/2 | 4.0 | 3.9 |
| \( ^{43}\text{Ca} \) | 7/2 | 4.1 | 4.0 |
| \( ^{42}\text{Mg} \) | 5/2 | 4.0 | 3.9 |
| \( ^{34}\text{Si} \) | 3/2 | 3.9 | 3.9 |
| \( ^{133}\text{Ba} \) | 3/2 | 3.9 | 3.9 |

In general, Eq.2 should be summed over all nuclei \( i \) with spin \( I_i \geq 1/2 \) in a molecule. The PV signal is easiest to interpret when the unpaired electron is centered on one atom in the molecule, i.e. \( W_p(\ell) \approx 0 \) for all but one atom. A single-atom-centered unpaired electron is also a defining characteristic of laser-coolable molecules: this electron interacts negligibly with the nuclear vibration of the molecule, leading to electronic transitions which are highly diagonal in vibrational quantum number (and thus requires a small number of vibrational repump lasers for cooling) \( \text{[41]} \). Laser cooling schemes have been proposed for molecules with an electron centered on a wide range of mass (as light as \( ^{11}\text{Be} \) and \( ^{12}\text{B} \), and as heavy as \( ^{171}\text{Yb} \) and \( ^{209}\text{Tl} \)), and extension to polyatomic species, while technically more complicated, is straightforward \( \text{[24]} \).

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(where appropriate) the constants \( B, \omega b=2, \gamma, b(H), \) and \( c(H) \) from the \(^{174}\)YbOH isotopologue \((I^{(174)\text{Yb}}=0) \)[17]; by assuming constants \( b(\text{Yb}) \) and \( c(\text{Yb}) \) to be the same as in chemically similar \(^{171}\)YbF; and by taking \( q_b = -2B/\omega_b \). With these parameters, the \( \ell \)-doublet in \( |v_\ell^t = 1, N = 1 \rangle \) is smaller than the SR and Yb HF interactions. For all possible PV pairs, the value of \( \langle (s \times i(\text{Yb})) \cdot \mathbf{n} \rangle \) is only nonzero due to the small state mixing from the HF interaction with the H nucleus. However, in \( |v_\ell^t = 1, N = 2, 3 \rangle \), the \( \ell \)-doublet is larger than the Yb HF splitting and multiple PV pairs with \( \langle (s \times i(\text{Yb})) \cdot \mathbf{n} \rangle \sim 0.1 \) exist.

We now propose a procedure to trap polyatomic molecules and measure matrix elements \( iW \) of \( H_{a}^{\text{eff}} \). A cryogenic buffer gas beam source creates a slow, cold beam of the desired molecule species \([48, 49]\). A cyrogenic buffer gas beam source creates a slow, cold beam of the desired molecule species \([48, 49]\). We apply a static magnetic field \( B = B Z \) to shift a particular PV pair to near degeneracy. We denote the time-dependent probability amplitudes of these states \( c_{\pm}(t) \), and assume an initial state \( c_{\pm}(0) = 1 \), \( c_{\pm}(0) = 0 \). An oscillating electric field \( \mathcal{E} = \mathcal{E}_0 \cos(\omega_\mathcal{E} t) \) is applied to drive the transition between the near degenerate levels. The effective Hamiltonian \( H_{a}^{\text{eff}} \) for the two level system can be written \([55]\)

\[
H_{a}^{\text{eff}} = \left( \frac{\Delta}{d \mathcal{E}_0 \cos(\omega_\mathcal{E} t)} - iW - \alpha' \mathcal{E}_0^2 \cos^2(\omega_\mathcal{E} t)/2 \right) .
\]

Here \( \Delta \) is the small detuning from degeneracy, \( d \) is the transition dipole moment, and \( \alpha' \) is the differential polarizability of the two states. In the limit where \( W \ll d \mathcal{E}_0, \Delta \ll \omega_\mathcal{E} \), and assuming for now that \( \alpha' = 0 \), the PV signal \( S = |c_{-}(t)|^2 \) is

\[
S \approx 4 \left[ \frac{2Wd \mathcal{E}_0}{\Delta \omega_\mathcal{E}} + \left( \frac{d \mathcal{E}_0}{\omega_\mathcal{E}} \right)^2 \right] \sin^2 \left( \frac{\Delta t}{2} \right) .
\]

From Eq.\(4\) we see that the PV transition amplitude (first term in square brackets) interferes with the E1 transition amplitude (second term in square brackets). The interference term changes sign under a reversal of either \( \mathcal{E}, \) \( B, \) or \( \Delta \). The PV matrix element \( W \) may be extracted through an asymmetry measurement \([17]\)

\[
A = \frac{S(+ \mathcal{E}_0) - S(- \mathcal{E}_0)}{S(+ \mathcal{E}_0) + S(- \mathcal{E}_0)} = 2 \frac{W \mathcal{E}_0}{\Delta d \mathcal{E}_0} + \ldots .
\]

where the ellipse denotes higher order terms in \( W/\Delta \). Detection using optical cycling should provide shot noise-limited readout.

Sensitivity to \( W \) is expected to be limited by homogeneous broadening of \( \Delta \). For optically trapped molecules, the detuning uncertainty \( \delta \Delta \) is dominated by the differential stark shift due to the optical trap. Employing certain “magic” polarization conditions in an ODT \([55, 56]\) will set \( \delta \Delta = \delta U = \frac{W}{dE \mathcal{E}_0} \delta \theta \), where \( \delta \Delta \) is the trap depth and \( \theta \) is an angle related to the polarization. We investigated 3 such magic conditions for PV pairs in \( \text{MgNC} \) and \( \text{YbOH} \) and find \( \delta \Delta \approx 7 \mu \text{J} \) for the largest \( \delta U / U \approx 10^{-4} \) is possible using high-quality Glan-type polarizers. We therefore expect \( \delta U / U \approx 10^{-4} \) using a magic angle trap \([55, 56]\), where the ODT linear polarization is rotated by \( \theta_{\text{magic}} = \cos^{-1}(1/\sqrt{3}) \approx 54.7^\circ \) with respect to the quantization axis. For \( U = 2 \times 1 \text{ MHz} \), \( \delta U = 2 \pi \times 100 \text{ Hz} \).

An inhomogeneous \( B \)-field will also produce broadening: typically \( \delta \Delta \approx \mu_B \delta B \) \([10]\). We require \( \delta B \ll 1/5U \mu_B \lesssim 1 \text{nT} \) for \( B \)-field inhomogeneity to not limit sensitivity. This implies \( \delta B / B = 10^{-7} \) for the largest fields we may require, \( B = 10 \text{nT} \). For comparison, the recent BaF NSD-PV measurement \( \delta B / B = 10^{-8} \), even with the much more experimentally challenging \( B \approx 400 \text{ mT} \) \([18, 19]\). We expect that the ability to easily reverse a smaller field will further aid in det-
tecting and eliminating B-field inhomogeneities. Moreover, the field must only be homogenous over the interaction volume, which is approximately 100 times smaller in an ODT compared a beam. Systematic uncertainties involving field gradients should similarly be reduced by a factor of 100 from the smaller interaction volume.

From Eq. (2) we see that Stark and NSD-PV amplitudes are \( \pi/2 \) out of phase, and there is no interference in a static \( E \)-field. However, the presence of a non-reversing \( E \)-field \( \hat{E}_n \) still poses an issue. In the molecule frame, a static \( \hat{E}_n \) has significant frequency components at axial and radial ODT frequencies \( \omega_z, \omega_r \), and multiples, sums, and differences thereof. Assuming uncorrelated trap oscillations, this effect will lead to a homogeneous broadening much smaller than that of the differential ac Stark shift. Accurate measurement of \( \hat{E}_n \) is possible by Stark interference with a reversible pulsed dc field \( \hat{E}_r \), or by microwave depletion spectroscopy \( \text{[60]} \). Finally, investigating multiple PV pairs in the same molecule provides strong systematic error rejection by varying the ratio \( \left\langle (\hat{S} \times \hat{I}) \cdot \hat{n} \right\rangle /d \) by a calculable, possibly large, amount \( \text{[54]} \). For example, in PV pairs with different signs of \( \left\langle (\hat{S} \times \hat{I}) \cdot \hat{n} \right\rangle /d \), contributions to \( A \) from actual NSD-PV will switch sign, but contributions from \( \hat{E}_n \) will not.

Other relaxation mechanisms are expected to lead to negligible broadening compared to differential ac Stark shifts. For example, in a beam experiment, \( \Delta \sigma \) is typically limited by interaction time \( \tau \), with \( \tau \approx 100 \mu s \) \( \text{[13]} \). In an ODT, \( \tau \) may easily exceed 1 s. Trapped molecule lifetimes \( \tau_{\text{trap}} \approx 0.5 \text{ s to 25 s} \) have been reported in a variety of traps \( \text{[21, 52, 61, 62]} \). With near ideal vacuum conditions, we expect trap lifetimes \( \tau_{\text{trap}} \approx 10 \text{ s} \), limited by vibrational decay. The loss rate due to off-resonant scattering from the trapping laser can typically be made \( R_{\text{sc}} \leq 1 \text{ s}^{-1} \) by using standard mid-infrared wavelength fiber lasers. Typical inelastic collision cross sections are expected to be \( \sigma_{\text{in}} \leq 10^{-3} \text{ cm}^2/\text{s} \). Comparing with the trapping conditions of Ref. \( \text{[32]} \) with \( N = 1300 \) molecules at density \( n = 6 \times 10^{8} \text{ cm}^{-3} \), we estimate an inelastic molecule-molecule collision rate of \( R_{\text{in}} \approx 0.5 \text{ s}^{-1} \). Therefore, collisions will become important when trapped molecule number \( N \gtrsim 10^3 \), or with additional cooling.

We now estimate the sensitivity of our method to NSD-PV matrix elements, \( \delta W = 1/\tau \sqrt{\mathcal{R}NT} \), where \( \mathcal{R} \) is the repetition rate, \( N \) the number of trapped molecules per measurement, and \( T \) the total measurement time. We assume molecules are trapped in a \( U = 2\pi \times 1 \text{ MHz} \) deep magic angle trap, with \( \delta \theta/\theta = 10^{-4} \). The combined effects of all relaxation times considered should allow for interaction times of \( \tau \gtrsim 1 \text{ s} \), but peak sensitivity is achieved with \( \tau = 1/\delta \Delta \approx 1/(U \delta \theta) \approx 1.6 \text{ ms} \). Allowing \( t_{\text{MOT}} = 50 \text{ ms} \) to load the MOT and \( t_{\text{trans}} = 40 \text{ ms} \) for state transfer, repetition rates \( \mathcal{R} = 10 \text{ s}^{-1} \) should be possible. We expect that with molecules produced from an isotopically enriched source, \( N \approx 1000 \) for all species; this would be equivalent to the best to-date sample of directly cooled molecules in an ODT \( \text{[52]} \). Under these conditions, our expected experimental sensitivity is \( \delta W \approx 2\pi \times 1 \text{ Hz}/\sqrt{\text{Hz}} \). This represents a factor of 70 improvement over the best to-date NSD-PV measurement in BaF \( \text{[18, 12, 33]} \). More ambitiously, one could plausibly expect \( N = 10^5 \) to \( 10^6 \) could be achieved with improved loading and cooling efficiency \( \text{[28, 63]} \).

With the proposed sensitivity, it should be possible to separate contributions to \( \kappa \) from nuclear anapole \( (\kappa_a \propto A^{2/3}) \) and \( V_c A_n \) \( (\kappa_2 A\text{-independent}) \) effects by measuring NSD-PV in a variety of nuclei. With the only non-zero NSD-PV measurement to-date in the heavy \( ^{133}\text{Cs} \), a precise measurement of PV in a light system would be especially illuminating. In light nuclei, \( \kappa = \kappa_2 \). As stated, \( \kappa_2 \) depends upon \( C_{2u,d} \) which are among the most poorly known SM parameters and are suppressed at tree level. Thus, a precise measurement of NSD-PV in light systems could potentially be sensitive to beyond SM physics above the 1 TeV scale \( \text{[12]} \). In molecules such as BeNC and MgNC the nuclear and molecular calculations are highly tractable. Furthermore, \( W_p(N) \sim W_p(C) \sim 2 \times 10 \text{ mHz} \) in these systems; a single species could provide a 10\% measurement of \( \kappa \) for three nuclei \( (^{13}\text{C}, ^{14}\text{N}), \) and either \(^{8}\text{Be}, ^{10}\text{Be}, \) or \(^{25}\text{Mg}) \) with \( T \lesssim 100 \text{ hours} \) per nucleus. The nuclear structure of \(^{14}\text{N}\) is of special interest and well studied due to the anomalously long \(^{14}\text{C}\rightarrow^{14}\text{N}\) half-life and their role in radiocarbon dating \( \text{[20]} \).

Because \( iW \) is enhanced by \( \approx Z^2A^{2/3} \) in heavy species \( \text{[38]} \), RaOH appears especially promising \( \text{[28]} \). However, the high mass is a hindrance to effective laser-slowing by the standard methods for molecules \( \text{[43, 50]} \). Moreover, the longest-lived Ra isotope possessing nuclear spin \( ^{225}\text{Ra} \) has a half-life of only 15 days. Nevertheless, MOTs of atomic \(^{225}\text{Ra} \) with typical \( N = 1000 \) have been produced for atomic electric dipole measurements \( \text{[64]} \); with our expected sensitivity, only \( N = 300 \) total molecules would need to be detected for \( \delta W/W = 0.1 \).

We have shown that optically trapped polyatomic molecules offer a dramatically enhanced sensitivity to parity-violating effects and additional checks of systematic errors. Restriction to laser-coolable species still allows for measurement of NSD-PV in nuclei with a wide range of masses, necessary for determination of key SM parameters and tests of beyond SM physics. The improved sensitivity should enable measurements of NSD-PV even in light nuclei where calculations are highly accurate.

Here, we have only considered linear asymmetric molecules. Symmetric top molecules possess \( k \)-doublets of opposite parity (similar to \( \ell \)-doublets) even in their vibrational ground state. For most \( ^2A_1 \) states (analogue of \( ^2\Sigma \)), the \( k \)-doublet splitting \( (\sim 10 \text{ kHz}) \) is smaller than SR/HF and thus not suitable for Zeeman tuning PV pairs to degeneracy for a NSD-PV measurement. However, the close spacing and miniscule differential Zeeman and ac Stark shifts of \( k \)-doublets may make symmetric top molecules in a magnetic or optical trap ideal for measuring NSI-PV.
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