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Cavity-enhanced parity-nonconserving optical rotation in metastable Xe and Hg

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We propose the measurement of cavity-enhanced parity-nonconserving (PNC) optical rotation in several transitions of metastable Xe and Hg, including Xe ($^2P_{3/2}^o 6s \rightarrow ^2P_{1/2}^o 6s$) $^2[3/2]_2^o \rightarrow ^2[1/2]_1^o$ and Hg ($6s6p \rightarrow 6s6p$) $^3P_2^o \rightarrow ^1P_1^o$, with calculated amplitude ratios of $E_1^{\text{PNC}}/M1 = 11 \times 10^{-8}$ and 10×10^{-8} , respectively. We demonstrate the use of a high-finesse bow-tie cavity with counter-propagating beams and a longitudinal magnetic field, which allows the absolute measurement of chiral optical rotation, with a pathlength enhancement of about 10^4 , necessary for PNC measurement from available column densities of 10^{14} cm^{-2} for metastable Xe or Hg. Rapid PNC-signal reversal, allowing robust background subtraction, is achieved by shifting the cavity resonance to an opposite polarization mode or by inverting the magnetic field. The precise measurement of isotope and nuclear-spin dependent E_1^{PNC} amplitudes provides a sensitive low-energy test of the Standard Model.

Precise measurements of atomic PNC provide a test of the electroweak part of the Standard Model at low-energy scale, in contrast to the bulk of such tests that are carried out at the Z-mass energy scale. Several theoretical and experimental constraints have led to atomic PNC [1] being measured in only a handful of systems [2–8]. The most accurate atomic PNC test performed involves the 6s-7s PNC transition in the only stable isotope of Cs using the Stark-interference technique, for which the E_1^{PNC} amplitude was measured with a precision of 0.35% [2]. The Cs atomic structure calculations have, with efforts spanning over 20 years [9, 10], reached a precision of about 0.25% [11]. Together, experiment and theory constrain new physics beyond the Standard Model, with the most powerful constraint putting a lower bound of $1.4 \text{ TeV}/c^2$ for extra Z' bosons [11]. The optical rotation technique has been used to measure E_1^{PNC} in Tl, Bi, and Pb (see [4–8] and references therein), with Tl being the second-most accurate atomic PNC experiment with a precision of 1% [4]. However, as Tl is essentially a three-valence electron system rather than an alkali, the estimated theoretical accuracy is currently limited to about 3% [12]. Ongoing Stark-interference experiments on Yb have measured the largest E_1^{PNC} amplitude yet observed [3]. However, theoretical precision in Yb is expected to be poor (currently at 13% [13]), therefore future experiments are aimed at measuring Yb PNC precisely for several isotopes, as isotope-ratio measurements can be used as a sensitive test of the Standard Model not relying on precise atomic theory [13, 14]. Proposals have been made for the measurement of atomic PNC in several isotopes of the one-valence electron systems of Ba^+ [15], and Fr and Ra^+ (produced at accelerator facilities such as TRIUMF [16] and KVI [17]) for which E_1^{PNC} is expected to be enhanced compared to Cs [16, 17], and theoretical precision expected to be better than 1%.

Here, we propose the measurement of PNC optical rotation in transitions of metastable Xe and Hg using a novel cavity-enhanced technique, having three main ad-

vantages: theoretical uncertainty for these two-electron excited states is expected to be better than all other systems mentioned except the alkalis; both Xe and Hg have large distributions of stable isotopes ($\Delta N/N = 12/76$ and $8/120$, respectively); and the proposed PNC measurements will be performed in a table-top experiment. These three advantages should motivate all-order atomic-structure calculations for Hg and Xe, to investigate isotope and nuclear-spin-dependent effects, and whether sub-1% theoretical precision is possible.

FIG. 1: Partial energy level diagram of Xe and Hg (not to scale) showing the proposed E_1^{PNC} and $M1$ transitions.

PNC optical rotation φ_{PNC} is caused by the interference of a magnetic dipole ($M1$) transition and a PNC-induced electric dipole (E_1^{PNC}) transition, given by [6]

$$\varphi_{\text{PNC}} = -\frac{4\pi l}{\lambda} (n(\omega) - 1) \mathcal{R}, \quad (1)$$

where $\mathcal{R} \equiv \text{Im}(E_1^{\text{PNC}})/M1$, l is the length of vapor, λ is the optical wavelength, ω is the optical frequency, and

$n(\omega)$ is the refractive index due to the absorption line.

We identify the following favorable PNC transitions in metastable Xe, $(^2P_{3/2}^\circ)6s\ ^2[3/2]_2^\circ \rightarrow (^2P_{1/2}^\circ)6s\ ^2[1/2]_1^\circ$ at 988 nm, and in Hg, $6s6p\ ^3P_J^\circ \rightarrow 6s6p\ ^1P_1^\circ$ at 609 nm ($J = 0$), 682 nm ($J = 1$), and 997 nm ($J = 2$). These first calculations of $\mathcal{R}$, for Xe and Hg, use the particle-hole approach described in Ref. [18]. We use the notation that $|hp\rangle$ describes an excitation of the ground state where an electron in state h forms a hole when it is excited to a state p . When a PNC enhancement factor arises from the presence of an opposite parity state n , nearly degenerate with the final state f in the transition $i \rightarrow f$, the PNC matrix element can be approximated by [13, 14]:

$$E_1^{\text{PNC}} = \frac{1}{\Delta E} \langle f | H_{\text{PNC}} | n \rangle \langle n | D | i \rangle, \quad (2)$$

where ΔE is given in Fig. 1, D is the dipole operator, and appropriate sums over the angular momentum of n are understood. We introduce mixing coefficients n_i , and write $|n\rangle = n_1|5p_{1/2}6p_{3/2}\rangle + n_2|5p_{1/2}6p_{1/2}\rangle + n_3|5p_{3/2}6p_{3/2}\rangle + n_4|5p_{3/2}6p_{1/2}\rangle$. Because H_{PNC} mixes only s and $p_{1/2}$ states, only these components (with coefficients n_2 and n_4) give non-zero matrix elements when $\langle f | H_{\text{PNC}} | n \rangle$ is evaluated. If the final state is written as $|f\rangle = f_1|5p_{3/2}6s\rangle + f_2|5p_{1/2}6s\rangle$, the matrix element of H_{PNC} reduces to the one-body operator $(n_2f_2 + n_4f_1)\langle 6s | H_{\text{PNC}} | 6p_{1/2} \rangle$. Values of the mixing coefficients can be obtained from standard atomic codes, but the level of precision to which they can be calculated is an open question. For the present calculation we use $n_2 = 0.030$, $n_4 = 0.746$, $f_1 = 0.062$, and $f_2 = 0.998$. For these values, $M1 = 0.0042\ e a_0$ and $\mathcal{R} = 11(3) \times 10^{-8}$, where the large uncertainty is caused by the difficulty in evaluating the small mixing terms that occur in H_{PNC} .

For Hg the situation is somewhat simpler, as $|n\rangle = |6s7s\rangle$, though we still need accurate values of f_1 and f_2 in $|f\rangle = f_1|6s6p_{1/2}\rangle + f_2|6s6p_{3/2}\rangle$. In this case we use $f_1 = 0.507$ and $f_2 = 0.862$ and find, for $J=0, 1$, and 2 , $\mathcal{R} = 14(3), 5(1)$, and $10(2) \times 10^{-8}$, and $M1 = 0.0014, 0.0042$, and $0.0057\ e a_0$, respectively. Note that $\mathcal{R}$ for Xe and Hg are similar, as the Xe E_1^{PNC} enhancement from the small ΔE denominator is largely canceled by the small value of n_2 ; also these $\mathcal{R}$ values are comparable to those of Tl, Bi and Pb for which $\mathcal{R}$ is 15, 10, and 10×10^{-8} respectively [4–6]. The estimated error is based on the behavior of low order MBPT calculations. An accurate experimental measurement of PNC in either atom should stimulate theoretical work on a scale comparable to that carried out on Cs.

To obtain measurable PNC optical rotation signals, column densities of $\sim 10^{18}\text{ cm}^{-2}$ thermal atoms are typically required [4–6], so as to achieve about 20 absorption lengths, for which optical rotation is $\sim 5\mathcal{R}$. Such column densities of excited-state atoms are generally not available due to the short excited-state lifetimes. The

FIG. 2: (a) Proposed experimental setup. (b) The laser beam is frequency shifted with an AOM to be on resonance with the nearly degenerate $R_{\text{CW}}-L_{\text{CCW}}$ modes of the cavity. The magnetic field splits the eigenmodes by $2\omega_F = 2\theta_F c/L$, while ω_{PNC} (not to scale) is much smaller than the cavity linewidth. The counterpropagating outputs are recombined into linearly polarized light, and analyzed with linear and circular balanced polarimeters (BP1 and BP2, respectively, using rotating half-wave and quarter-wave plates). (c) Theoretical prediction of PNC optical rotation signal ($2N\varphi_{\text{PNC}}(\mu\text{rad}) \times \text{transmission}$) measured by BP1 and BP2, corresponding to 107 absorption lengths (A.L.) of isotopically pure metastable ^{202}Hg . Alternating between the polarization mode pairs (split by $\sim 0.1\text{ MHz}$), shown in the inset, yields a net polarization difference $2N\varphi_{\text{PNC}}$, occurring at each black point, which are separated by one FSR ($2\pi \times 37.5\text{ MHz}$). (d) similar to (c), but for 20 A.L. of metastable ^{132}Xe (see text).

metastable states Xe 3P_2 and Hg 3P_J have been produced at steady-state densities of about 10^{12} cm^{-3} , using electrical discharge lamps [19, 20] or optical pumping [21], allowing column densities of about 10^{14} cm^{-2} (over a path length of 100 cm). For these column densities and calculated values of $\mathcal{R}$, an enhancement factor of about 10^4 is necessary to obtain signal levels comparable to previous optical rotation experiments [4–6].

Optical cavities have been used to enhance linear birefringence by more than 10^4 , using either polarimetric techniques or frequency metrology, in searches for magnetically-induced birefringence of vacuum [22], characterization of mirror birefringence [23, 24], and measurements of the Cotton-Mouton [25, 26] and Kerr effects in gases [27, 28] with a sensitivity of 3×10^{-13} rad [28]. Linear optical cavities have been used for the enhancement of circular birefringence, such as for the Faraday effect of gases [29], and the optical activity of chiral molecules with the use of intracavity quarter-wave plates [30, 31].

We propose the use of a four-mirror bow-tie cavity with counterpropagating laser beams for the enhancement of PNC measurements, outlined in Fig. 2. For a single pass in the cavity, the light polarization undergoes PNC rotation φ_{PNC} and Faraday rotation θ_F (produced by an applied longitudinal magnetic field). The different symmetry of these rotations under time reversal results in $\varphi_{\text{PNC}} \rightarrow \varphi_{\text{PNC}}$ and $\theta_F \rightarrow -\theta_F$ in the light-propagation frame (and $\varphi_{\text{PNC}} \rightarrow -\varphi_{\text{PNC}}$ and $\theta_F \rightarrow \theta_F$ in the lab frame), causing a difference in rotation for the clockwise (CW) and counterclockwise (CCW) counterpropagating beams: $\alpha_{\text{CW}} = \theta_F + \varphi_{\text{PNC}}$ and $\alpha_{\text{CCW}} = -\theta_F + \varphi_{\text{PNC}}$. This directional symmetry breaking is key for the sensitive measurement of PNC optical rotation, in close analogy to Sagnac interferometry [32].

The Faraday effect splits the cavity spectrum into R and L circular polarization modes by $2\omega_F$, whereas the PNC rotation splits the CW and CCW modes by $2\omega_{\text{PNC}}$, resulting in the cavity modes R_{CW} , L_{CW} , R_{CCW} and L_{CCW} (Fig. 2b). The ratio of these splittings yields $\mathcal{R}$:

$$\mathcal{R} \equiv \frac{\text{Im}(E_1^{\text{PNC}})}{M1} = \frac{\omega_{\text{PNC}}}{\omega_F} D(\omega) B \quad (3)$$

where $D(\omega) = (4\pi h/\mu_B)[(n(\omega) - 1)/(\partial n/\partial \omega)]$ is a line-shape-dependent factor [33], B is the longitudinal magnetic field, $\omega_{\text{PNC}} = \varphi_{\text{PNC}}/L$, and $\omega_F = \theta_F/L$ (where L is the cavity round-trip length).

To measure φ_{PNC} , a linearly polarized laser beam is split into two beams of equal intensity: one beam excites the R_{CW} mode (the reflected part of which is used for locking the laser frequency to the R_{CW} mode using the Pound-Drever-Hall (PDH) scheme [34]) while the other beam excites the nearly degenerate L_{CCW} mode (possible because ω_{PNC} is much smaller than the cavity linewidth ~ 1 kHz). The R_{CW} and L_{CCW} output beams are spatially recombined to produce a linearly polarized beam rotated by $N\varphi_{\text{PNC}}$, where N is the average number of round-trip cavity passes. Our detection system measures the complete set of the Stokes parameters of the output light [35], and therefore directly determines φ_{PNC} and any experimental depolarization effects.

A key point of the experiment is the availability of two rapid experimental reversals: reversing either the magnetic field direction or alternating the frequency of the CW and CCW beams into resonance with the R_{CW} - L_{CCW}

FIG. 3: Numerical analysis of the dependence of the correction factors q and q^2 on δ/α .

and R_{CCW} - L_{CW} mode pairs. These reversals give a signal output of $-N\varphi_{\text{PNC}}$ and a net difference in polarization rotation of $2N\varphi_{\text{PNC}}$. The magnetic field and mode-pair reversals can be performed quickly (up to 100 Hz) to subtract slow experimental drifts. Measurement of the difference in mode-pair frequencies gives $2\omega_F$ (see Fig. 2).

Theoretical calculations for the expected PNC rotation signals are presented in Figs. 2c and 2d. We assume a four mirror bow-tie cavity of round-trip cavity length $L=8$ m (free spectral range, FSR= 37.5 MHz), mirror reflectivity $R=99.99\%$ (enhancement factor $N \sim 10^4$), and a discharge lamp of length $l = 1.3$ m operating with isotopically pure ^{202}Hg (at 400 K) and ^{132}Xe (at 300 K). We note that all even isotopes will have similar spectra, whereas odd isotopes will have hyperfine structure; in addition, all eight Xe and seven Hg stable isotopes are commercially available (each low pressure lamp requires $\sim 1 \mu\text{mol}$ of isotopically pure gas). We plot the PNC optical rotation, $2N\varphi_{\text{PNC}}$, multiplied by the transmission, as a function of the laser frequency about the absorption line center, for metastable ^{202}Hg column densities of $3.8 \times 10^{18} \text{ cm}^{-2}$ (~ 107 absorption lengths)[19], and ^{132}Xe column densities of $1 \times 10^{18} \text{ cm}^{-2}$ (~ 20 absorption lengths)[20]. Collisional line broadenings for Hg and Xe are estimated to be 20 MHz and 10 MHz respectively. The black and red points shown in Figs. 2c and 2d correspond to consecutive longitudinal cavity modes separated by one FSR, so that reasonable spectral resolution can be obtained even for a fixed cavity.

We take advantage of the fact that a large applied circular birefringence strongly suppresses effects of unwanted linear birefringences, which were important sources of systematic errors in past PNC optical rotation experiments, originating from mirror s-p phase shifts [36], window transmission, and from stray transverse magnetic fields [6, 37]. A complete analysis of the polarization properties of the cavity, is performed using a Jones matrix formulation [38]. For a four-mirror cavity with single pass linear birefringence δ and circular birefringence α

the eigensystem is given by:

$$\lambda_{\pm} = \cos \alpha \cos \frac{\delta}{2} \pm \sqrt{\cos^2 \alpha \cos^2 \frac{\delta}{2} - 1} \quad (4)$$

and

$$\nu_{\pm} = \begin{pmatrix} 1 \\ \mp i \csc \alpha \left(\cos \alpha \sin \frac{\delta}{2} + \sqrt{\cos^2 \alpha \cos^2 \frac{\delta}{2} - 1} \right) \end{pmatrix}. \quad (5)$$

Here, for simplicity, we set the reflectivities for the s and p polarizations to be equal, which is a good approximation for near-normal angle-of-incidence bow-tie cavities.

In the ideal case, where $\delta = 0$, the eigenvalues

FIG. 4: Non-resonant Faraday effect of an intracavity SiO₂ window at 800 nm. The cavity ring-down polarization beating frequencies for counterpropagating beams are visibly different (magnetic field of 1000 G). The inset shows the linear and antisymmetric magnetic-field dependence of $\omega_{\text{CW}} - \omega_{\text{CCW}}$.

are equal to $\lambda_{\pm} = e^{\pm i\alpha}$ (with $\alpha_{\text{CW}} = \theta_F + \varphi_{\text{PNC}}$ and $\alpha_{\text{CCW}} = -\theta_F + \varphi_{\text{PNC}}$), which results in the four-mode spectrum described above and in Fig. 2b.

For $\delta \neq 0$, the cavity eigenmodes are elliptical polarization states whose splitting is $\Gamma = \cos^{-1}[\cos \alpha \cos(\delta/2)]$. The non-commutative nature of linear and circular birefringence results in a reduction of the effective amplification of circular birefringence (Faraday and PNC rotation). We express the reduction effect as $\omega'_{\text{PNC}} = q\omega_{\text{PNC}}$, where $q = \alpha/\Gamma$ ranging from 1 to 0 (Fig. 3). Furthermore, with increasing δ the frequency-splitting of the polarization eigenmodes increases as $\omega'_F = (1/q)\omega_F$. Thus, δ reduces the observed ratio $\omega'_{\text{PNC}}/\omega'_F$ by q^2 :

$$\frac{\omega'_{\text{PNC}}}{\omega'_F} = pq^2 \frac{\omega_{\text{PNC}}}{\omega_F}, \quad (6)$$

where p is the degree of linear polarization of the output light which is reduced from 1 for unequal intensities or imperfect overlap of the recombining beams. We plot the correction factors q and q^2 in Fig. 3. Note that for $\delta \gg \alpha$ the circular birefringence (including PNC optical rotation) vanishes, a direct consequence of the transformation

of the eigenpolarization modes into two linear polarization vectors. To ensure $q^2 \simeq 1$ we require $\delta \ll \alpha$. For example, for $q^2 \simeq 0.99$, $\delta = 0.2\alpha$, and for $q^2 \simeq 0.9999$, $\delta = 0.02\alpha$ (inset Fig. 3). For the $M1$ transitions and proposed conditions, the Faraday effect yields $\alpha \simeq 10^{-3}$ rad for a 200 G magnetic field [39], while mirror birefringence $\delta \lesssim 10^{-5}$ rad is achievable. Alternatively, anti-reflection (AR) coated windows with losses of $\lesssim 10^{-4}$ can be used to produce $\alpha \simeq 10^{-2}$ rad for a 1 T magnetic field and window birefringence $\delta \lesssim 10^{-4}$ rad.

To support the proposed experimental setup we demonstrate the mode structure of a bow-tie cavity using polarization-dependent cavity ring-down measurements [31] of the non-resonant Faraday effect of an intracavity AR-coated SiO₂ window of 3 mm thickness. Two linearly-polarized pulsed beams ($\tau_{\text{pulse}} \simeq 35$ fs, $\lambda = 800$ nm, coherent bandwidth ~ 40 nm) are injected into counter-propagating modes of a four-mirror bow-tie cavity ($L = 3.7$ m). After exiting the cavity, the two beams enter separate balanced polarimeters (in contrast to Fig. 2a). By making the cavity non-planar, we introduce a purely geometric chiral rotation (of the same symmetry as PNC rotation) [40]. We adjust the rotation angle to be $\alpha_C = 2.9^\circ$ per round trip, resulting in a large mode splitting of $\omega_C = \alpha_C(c/L) = 4.15$ MHz (for the proposed PNC experiment ω_C must be minimized to be less than a cavity linewidth). The rotation of the polarization appears as a beating in the balanced polarimeters with frequency ω_C [31]. In the presence of Faraday rotation, the polarization beating of the two counter-propagating beams are different, $\omega_{\text{CW}} = \omega_F + \omega_C$ and $\omega_{\text{CCW}} = -\omega_F + \omega_C$, as shown in Fig. 4. We measure ω_{CW} and ω_{CCW} as a function of the magnetic field and show that their difference, $\omega_{\text{CW}} - \omega_{\text{CCW}} = 2\omega_F$, is linear with the magnetic field and changes sign upon magnetic field reversal. The measured Verdet constant of SiO₂ at 800 nm is $(2.41 \pm 0.05) \mu\text{rad G}^{-1} \text{cm}^{-1}$, in agreement with [41].

Our cavity proposal can also be used for PNC optical rotation in other atoms [4–6, 33, 42], searches for permanent electric dipole moments (EDM) [43], and optical rotation or circular dichroism from chiral molecules [31].

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