Invited Article

Cavity-based chiral polarimetry: parity nonconserving optical rotation in Cs, Dy, and HgH

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Abstract

The measurement of single-pass chiral optical rotation and circular dichroism are the most widely used methods for chirality sensing, and are of fundamental importance to many fields in physics, chemistry and biology. The optical rotation method is also one of two successful methods for the measurement of atomic parity nonconservation (PNC), for low-energy tests of the electroweak sector of the standard model. However, PNC optical rotation signals are typically very weak, having been measured only in the most favourable cases of Tl, Bi, and Pb, and their measurement is limited by larger time-dependent backgrounds (such as spurious birefringence) and by imperfect and slow subtraction procedures. Using a novel bowtie cavity with an intracavity Faraday Effect, we have demonstrated three important improvements: (a) the enhancement of the chiral optical rotation angle by the number of the cavity passes (typically \(\approx 1000\)); (b) the suppression of birefringent backgrounds; and (c) the ability to reverse the sign of the chiral optical rotation signal rapidly, allowing the isolation of the PNC signals from backgrounds. We discuss how these cavity-based improvements may allow the extension of the measurement of PNC optical rotation to atomic and molecular systems that would otherwise be inaccessible, for example to Cs and Dy atoms in magneto-optical traps, and to HgH molecules in jet expansions. For these cases, we show that potentially large PNC signals are possible, and we discuss the potential measurement of nuclear-spin-independent, nuclear-spin-dependent, and isotope-dependent PNC effects.

Keywords: atomic parity nonconservation, atomic parity violation, molecular parity nonconservation, molecular parity violation, cavity-enhanced polarimetry

(Some figures may appear in colour only in the online journal)

1. Introduction

Shortly after the discovery of parity nonconservation (PNC) of the weak force \([1, 2]\), Zeldovich proposed that weak neutral currents mix the \(n_{p1/2}\) and \(n_{s1/2}\) states of opposite parity in atomic hydrogen \([3]\), and will allow small electric dipole transition amplitudes \(E_{\text{PNC}}\) between states of the same nominal parity, thus producing parity nonconserving observables such as optical rotation. However, the estimated rotation angle of \(10^{-13}\) rad m\(^{-1}\) was far too small for measurement. In 1974, Bouchiat and Bouchiat showed that for larger atoms \(E_{\text{PNC}}\) scales faster than \(Z^2\) (where \(Z\) is the atomic number), so that large enhancement factors for \(E_{\text{PNC}}\) of about \(10^6\) are possible for atoms with large \(Z\) \([4]\). This large enhancement opened the way for the conception of feasible experiments for the measurement of \(E_{\text{PNC}}\) \([5, 6]\), resulting in

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the first measurements of optical rotation in Bi experiments with measurable signals of order $10^{-6}$ rad m$^{-1}$ [7], as well as Stark-interference experiments in Tl [8, 9].

The weak charge of the nucleus, $Q_w$, can be determined from $E_{\text{IPNC}}$, and precise measurement of $Q_w$ provides a low-energy test of the standard model, complimentary to high-energy measurements [10–12]. We outline the three main goals of atomic PNC: measurement of $Q_{\alpha}$, PNC measurements on isotope chains, and nuclear-spin-dependent PNC measurements (NSD-PNC).

Precise atomic-structure calculations are needed to determine $Q_w$ from $E_{\text{IPNC}}$, and sub-1% precision has only been achieved, so far, for monovalent atoms [13]. Cs has so far proved to be the most successful system for determining $Q_w$ precisely, with $Z$ being large enough to yield measurable signals, while theoretical atomic-structure calculations have achieved, after many years, precision of about 0.5%. The most successful measurement has yielded 0.35% precision for $E_{\text{IPNC}}$ on the 6s–7s transition in Cs [14], and atomic-structure calculations then transforms this to a 0.5% precision determination of $Q_w$ for Cs. This precise determination of $Q_w$ agrees with the standard model, and, until the results in recent years at the Large Hadron Collider, offered the most stringent low-energy tests of the standard model. Experimental efforts to improve the measurement of PNC in monovalent atoms to below 0.35% precision are in progress in Cs, Fr, and Ra$^+$. The new method of the interference of one-photon and two-photon 6s–7s transitions in Cs is being developed and refined in the group of Elliott [15–17]. PNC effects in Fr and Ra$^+$, with larger $Z$, are at least an order of magnitude larger than that in Cs, but experiments with these elements are more difficult as they are radioactive, and most isotopes have short lifetimes; PNC experiments are in progress on Fr in Vancouver [18] and on Ra$^+$ in Groningen [19].

Sidestepping the need for extremely precise atomic-structure calculations, which are currently not possible for most atoms, the dependence of $Q_{\alpha}$ on the number of neutrons in each isotope can be used to test the standard model and its extensions. Yb has been proposed as a favourable system for such isotope measurements, because the $E_{\text{IPNC}}$ amplitude for the 6s$^2$1S_0$\rightarrow$ 5d6s 3D$^1_J$ transition is about 100 times larger than that for the 6s–7s transition in Cs [20]; in addition, Yb has seven stable isotopes, and four of them with zero nuclear-spin. This prediction for a large PNC effect in Yb was confirmed in 2009 [21]. Very recently, PNC measurements of 0.5% precision in isotopes of Yb confirmed the $Q_{\alpha}$ scaling, and offered constraints on possible $Z'$ bosons [22, 23].

The measurement of $E_{\text{IPNC}}$ as a function of resolved hyperfine transitions gives further tests of the standard model, as well as measurement of purely hadronic PNC interactions, which are difficult to measure otherwise [24–26]. To date, nonzero NSD-PNC has only been measured in Cs [14], and this result seems to be in tension with nuclear physics experiments [26].

Successful measurements of atomic PNC have used either the Stark-interference technique, or the optical rotation technique near a magnetic-dipole transition [8]. The main advantage of the Stark-interference technique is the application of several ‘signal reversals’, each of which can invert the sign of the dominant contribution of $E_{\text{IPNC}}$ to the experimental signal (which is typically of order $10^{-6}$–$10^{-4}$ of the total signal). These multiple signal reversals are crucial for isolating the small $E_{\text{IPNC}}$ signal from the dominant non-PNC signal, and have allowed the measurement of $E_{\text{IPNC}}$ with 0.35% precision for Cs [14], and 0.5% precision for several Yb isotopes [22, 23].

The main advantage of the optical rotation technique is the characteristic dispersive lineshape of the PNC optical rotation signal [8], which allows the PNC contribution to be discriminated from Faraday rotation and from spurious optical rotation signals. In addition, a slow background subtraction was performed in this type of experiments, by replacing the gas cell with an empty dummy tube. These procedures were necessary to help isolate the PNC signal from large time-varying backgrounds, and they allowed the measurement of $E_{\text{IPNC}}$ with 1% precision in Tl, Bi, and Pb [27–29]. The two main disadvantages of the traditional optical rotation technique, for PNC measurements, are the lack of any signal reversals, to be able to reduce the sensitivity of the technique further, and the fact that the available atomic systems (Tl, Bi, and Pb) do not satisfy well the three goals of atomic PNC stated above: the atomic-structure calculations for these atoms have been limited to an accuracy of about 3%, so that $Q_w$ cannot be determined precisely; these atoms do not have long chains of stable isotopes; and the 1% experimental precision was not sufficient to measure accurately the NSD-PNC signals.

These experimental shortcomings of the traditional optical rotation technique are the reason why current PNC experiments are using other measurement techniques, with further signal enhancement mechanisms. For example, the Stark-interference experiment on Yb gives $E_{\text{IPNC}}$ of about 100 times larger than that in Cs, due to the fortuitously small energy splitting of 589 cm$^{-1}$ between the excited 5d6s 3D$^1_J$ state and the opposite parity 6s6p 1P$^1_J$ state (which has significant 5d6p character), which enhances the opposite-parity mixing significantly. Enhancing PNC with opposite parity hyperfine states is being taken to the limit in current experiments on BaF molecules, where closely spaced opposite parity hyperfine states are mixed by NSD-PNC in $^{137}$BaF molecules, and this small splitting is tuned to near degeneracy through the Zeeman Effect, giving very large opposite parity-state mixing [30, 31].

In recent years, the authors have proposed [32, 33] and demonstrated [34] a novel ring-cavity-based method for the improvement of the traditional optical rotation method. The cavity offers three distinct improvements: (a) it enhances the effective sample path length by the number of cavity passes $N$ (where $N$ is typically between $10^2$ and $10^4$), thereby enhancing the PNC signals by up to this factor; (b) the birefringence within the sample (and the cavity) is supressed; and (c) at least two rapid signal reversals are introduced. Together, these three improvements promise to improve the sensitivity of the
optical rotation technique significantly, and to allow its application to several new atomic and molecular systems for PNC measurements, that otherwise would not be accessible (in addition to many applications of chirality measurements in chemistry and biology, using essentially the same method).

Previously, we have proposed using cavity-based polarimetry on room-temperature samples of I atoms [33, 35] (from the photodissociation of I₂), and metastable Hg and Xe atoms [32, 33] (in discharge lamps). Of these atomic systems, we showed that I atoms can give large experimental PNC signals, with optical rotation of 1 μrad or larger for about 1000 cavity passes. Although iodine atoms do not have multiple stable isotopes, nor are precise atomic-structure calculations possible, the measurement of NSD-PNC in ¹₂⁷I would test the coupling mechanisms, which have not been applied so far for the measurement of PNC optical rotation.

We consider PNC optical rotation from translationally cold atoms in magneto-optic traps (MOTs), with linewidths a few orders of magnitude narrower than those of room-temperature samples, and a corresponding optical rotation enhancement, as explained in section 2.2; in particular, we examine PNC optical rotation from the favourable cases of Cs and Dy MOTs. For Cs, we consider the ⁶S → ⁵D₁/₂ electric quadrupole transition, using spin-polarized Cs atoms, as described by Cronin [36], while for Dy the ⁵I₈ → ⁵I₇ M₁ transition. In addition, we consider PNC optical rotation from high densities of diatomic radicals, for which the rotational levels provide nearby opposite-parity states separated by a few cm⁻¹, yielding a PNC signal enhancement of about four orders of magnitude; in particular, we examine the PNC optical rotation from ¹⁹⁹HgH radicals, for which Geddes et al have calculated a large NSD-PNC effect [37]. We propose to maximize the optical rotation signal from the rotational and translational cooling achieved through a slit-nozzle expansion. We also note that HgH is being considered for measurements of the electric dipole moment of the electron [38], and of violation of parity (P) and time-reversal (T) invariance [39].

The paper is organized as follows: In section 2.1, we outline how an optical ring-cavity with an intracavity Faraday Effect can be used to enhance the sample path length and PNC signals and introduce at least two signal reversals. In section 2.2 we review the factors, other than path length, that can be used to enhance optical rotation, such as narrow linewidths from samples in cold beams or in MOTs. Finally, in section 3, we describe how cavity-based optical rotation can be applied to transitions in Cs and Dy atoms, and HgH molecules, and we simulate experimental signals in each case, for realistic experimental conditions.

2. Optical-cavity-enhanced polarimetry

2.1. A method description

In recent years, our group has demonstrated [34] how an optical cavity can be used to enhance weak polarimetric signals, by effectively expanding the light-sample interaction pathlength by the number of cavity passes, while employing methods to protect the light polarization state from intracavity birefringence, and also introducing valuable signal reversals. We proposed to extend these ideas to CW lasers and high-finesse cavities for the measurement of extremely weak chiral signals, such as optical rotation from PNC interactions in...
certain atoms and molecules [32, 33]. The general idea is depicted in figure 1: two light beams of common origin are injected into a 4-mirror bowtie cavity, along the two counter-propagating directions (henceforth referred to as clockwise/ CW and counter-clockwise/CCW beams). The chiral sample resides in one arm of the cavity, and the two beams traverse it multiple times, each beam always along the same direction, thus enhancing the weak chiral optical rotation by the number of cavity passes. The resulting polarization rotation for the two beams is of opposite sign, \( \phi_{\text{cw},\text{ccw}} = \pm (2 \mathcal{F} / \pi) \phi_c \), where \( \phi_c \) is the per-pass chiral rotation, and \( \mathcal{F} \) is the cavity finesse. Measuring both rotations and subtracting yields an absolute measure of the chiral rotation, largely relieved from common noise between the two beams. However, there are still two issues with this simple picture: (a) linear birefringence experienced by the beams, due to reflections upon the cavity mirrors or passing through intra-cavity optics, induces an ellipticity to the beam polarization, which is almost invariably significantly larger than the chiral circular birefringence of the sample. The birefringence suppresses the chiral rotation, and for birefringence larger than \( \sim \pi / N \) (where \( N \) is the average number of cavity passes), the rotation angle of the linearly polarized light in the cavity changes sign at least once, so that the total optical rotation cancels, and is near 0. (b) Since chiral signals are very weak, even high levels of common mode noise rejection may not suffice to bring the signal above noise levels, especially at DC.

It is, therefore, desirable to introduce some sort of signal reversal, i.e. some process which can reverse the sign of the chiral rotation signal, and only that, on demand. If we then repeat this process at some steady frequency, the output signal will be modulated at that frequency, with a modulation amplitude directly proportional to the chiral rotation, and we can use phase-sensitive detection on the difference of the CW and CCW signals to isolate the weak chiral OR with extreme precision. Moreover, the measured amplitude will reveal the absolute chirality of the sample; removing the sample to take background measurements, a constant pain of regular polarimeters, will no longer be a necessity.

Both issues can be addressed by using an intra-cavity Faraday effect-induced optical rotation in conjunction with the chiral rotation. By applying a magnetic field on a crystal or on the sample itself, along the beam propagation path, we can induce a Faraday polarization rotation, \( \theta_F \) per-pass, which has the same sign for both the CW and CCW beams, since the sense of rotation in the Faraday effect is dictated by the magnetic field direction. The total rotation for the two beams will then be \( \phi_{\text{cw},\text{ccw}} = (2 \mathcal{F} / \pi) (\theta_F \pm \phi_c) \). If the Faraday-induced rotation is significantly larger than the intra-cavity linear birefringence, \( \alpha \), then it serves to neutralize the effect of the latter, by effectively randomizing the relative angle between the polarization plane of the light and the birefringence axis, something which causes the birefringence to average out to zero, and protects the chiral rotation signal which adds to the larger Faraday rotation, as both are a form of circular birefringence (see [33] for a rigorous treatment).

In addition to the protection offered against birefringence, the intra-cavity Faraday effect also provides a first signal reversal. Indeed, using some type of sign-insensitive polarimetric detection at the output of the cavity (e.g. extinction polarimetry), the detected polarization rotation will be the difference \( \Delta \phi = |\phi_{\text{cw}}| - |\phi_{\text{ccw}}| = (4 \mathcal{F} / \pi) \phi_c \). By reversing the direction of the magnetic field, the Faraday rotation is reversed, but the chiral is not, so \( \phi_{\text{cw}(\text{cw})} = (2 \mathcal{F} / \pi) (\theta_F \pm \phi_c) \). Given that \( \theta_F > \phi_c \), the detected rotation will be \( \Delta \phi = -(4 \mathcal{F} / \pi) \phi_c \). Therefore, reversing the magnetic field reverses the sign of the detected chiral OR, and the swing of the reversal is 4 times the absolute chirality of the sample. We can then use phase-sensitive lock-in detection at the frequency of the magnetic field reversal to measure this absolute chiral rotation, at significantly reduced noise, one of the merits of a phase-locked measurement. Further signal reversals can come about when we study the mode structure of the optical cavity in more detail.

The cavity setup we are proposing is versatile and can adapt to a wide range of sizes, operation wavelengths, and sample configurations, as long as the sample remains weakly absorbing, and the intracavity birefringence is small. We propose to adapt it for the measurement of PNC-induced optical rotation in translationally cooled Cs or Dy atoms (in a MOT) and on rotationally cooled HgH radicals (in a jet). The details of the MOT and the HgH jet are discussed in other parts of this paper, but here we discuss only an implementation detail. In measuring very weak PNC-induced OR signals with optical cavities, a high-finesse, or, equivalently, a high number of passes \( N \) is necessary; we aim for \( N \sim 10^4 \). This requires high-quality, high-reflectivity cavity mirrors with \( R > 0.9999 \). It is also necessary to remove all intracavity optics, for example the Faraday crystal or the windows of the cell containing the sample, since, regardless of how well their surfaces are AR-coated (AR for Anti-Reflection), they still present scattering and absorption losses that do not allow more than about 1000 passes, while also being potential sources of linear birefringence.

The cavity mirrors must be designed to have extremely low birefringence at the wavelength of interest. At the centre of a dielectric mirror’s reflection curve, the linear birefringence changes sign and passes through zero. Therefore, the mirrors must be designed so that this zero point is as close to the desired PNC transition as possible. In practice, this can be achieved to within \( \approx 1 \) nm. Furthermore, the birefringence scales approximately as \( \sin^2 \theta \), where \( \theta \) is the angle of incidence on the mirror. Therefore, making the bowtie cavity angle as small as possible (e.g. \( \theta \approx 5 \)) suppresses the birefringence further. Together, the mirrors can be designed and employed to have less than \( 10^{-4} \) rad birefringence.

Finally, any small remaining birefringence can be suppressed by applying the magnetic field directly across the sample and taking advantage of the Faraday effect of the sample itself. Given the additional fact that both the MOT of Cs or Dy atoms, and the molecular jet of HgH radicals are already under vacuum, the whole cavity setup should also be built under the same vacuum, requiring not cavity windows, as we graphically depict in figure 2.
2.2. Optical rotation signal and broadening mechanisms

The PNC-induced optical rotation caused in a magnetic-dipole-allowed (M1) transition, by a small electric-dipole admixture, \( E_{\text{PNC}} \), is given by:

\[
\varphi = -\frac{4\pi l}{\lambda} (n(\omega) - 1) R,
\]

where \( R = \frac{\text{Im}(E_{\text{PNC}})}{M_1} \) is the ratio of \( E_{\text{PNC}} \) electric dipole to the \( M_1 \) magnetic-dipole amplitude, and \( l \) is the interaction length. The spectral characteristics of the resulting optical rotation signal stem from the frequency dependence of the refractive index, \( n \), which comprises broadening contributions to the transition lineshape from both homogeneous and inhomogeneous processes. Homogeneous processes, e.g. lifetime broadening, pressure broadening etc, result in a characteristic Lorentzian lineshape, while inhomogeneous processes can vary in the lineshapes produced, however the predominant such process is usually the Doppler broadening due to the thermal velocity distribution of atomic or molecular thermal samples, which yields a Gaussian lineshape. When the two types of contributions are comparable, the combined lineshape is a convolution of the two, what is usually called a Voigt profile.

Regardless of the underlying mechanism, lineshape broadening results in a reduction of the optical depth (OD) of the sample, and, by conjugation, of its peak dispersion, which in turn leads to a reduced peak optical rotation signal. This implies that, all things equal, suppressing the lineshape broadening, i.e. reducing the linewidth of the probed transition, should increase its resonant OD, and also the maximum attainable optical rotation signal. This is shown theoretically for a generic gaseous sample in figure 3, where the maximum optical rotation signal, \( 2 \times \varphi(\omega) \times T(\omega) \), is plotted against the ratio of the Doppler to Lorentz linewidth, \( \Gamma_{\text{Dop}} / \Gamma_{\text{Lor}} \), for a constant Lorentz linewidth. In dilute thermal gas samples, transitions in the optical, or even the IR part of the spectrum are usually heavily Doppler-dominated. For example, in the 1315 nm transition of atomic iodine proposed for the measurement of PNC-induced optical rotation in [35], the combined Lorentzian contributions to the linewidth are on the order of 1 MHz, while Doppler broadening is \( \sim 100 \) MHz.

[Figure 2. (a) Schematic of 2D MOT in the bowtie cavity. Parameter values for the Dy MOT have been used; the Cs setup would be completely analogous. (b) Schematic of slit-nozzle expansion intersecting one arm of the bowtie cavity.]

[Figure 3. Plot of calculated maximum PNC optical rotation versus the ratio of Doppler to Lorentz width. For \( \Gamma_{\text{Dop}} / \Gamma_{\text{Lor}} \gg 1 \) the signal decreases approximately as \( 1/\Gamma_{\text{Dop}} \); whereas it becomes largely insensitive to \( \Gamma_{\text{Dop}} \) as the temperature is decreased and \( \Gamma_{\text{Dop}}/\Gamma_{\text{Lor}} \rightarrow 1 \). This particular calculation was performed for \( \Gamma_{\text{Lor}} = 1 \) MHz, \( \lambda = 400 \) nm, and \( R = 10^{-6} \) (in which case the units of the y-axis are \( \mu \text{rad} \)).]
(T \sim 10 \text{ K}), propagating perpendicularly to the laser beam. In both cases, the increase in optical rotation from the decreased Doppler linewidth is significant, especially in the case of the trapped Cs or Dy atoms where the temperature is reduced by some 8 orders of magnitude relative to room-temperature values.

We note here that the gains made by the narrowed line-shape could, in principle, be offset by the lower densities achievable in a MOT or a molecular beam, compared to thermal samples. Lower densities affect the measured optical rotation signal via the linear dependence of the refractive index on the sample density. However, on the one hand this is not true for the HgH (ν = 0; J = 1/2) state, for which the plausible maximal density in a molecular beam (due to rotational cooling) is expected to surpass the highest attainable density in a thermal vapour for the most populated rotational state, while for Cs and Dy, for the currently experimentally feasible MOT configurations, the optical rotation enhancement due to the narrowing of the transition line-shape is still advantageous, as we show in the next Section.

3. PNC optical rotation in CS, Dy, and HgH

3.1. PNC optical rotation in translationally cold atoms

PNC optical rotation has been performed, using the same method and experimental setup, for the cases of Tl, Bi, and Pb [27–29]. An atomic gas density of about \(10^{17} \text{ cm}^{-3}\) is produced in an oven of about 1300 K, along with about 50 mbar of buffer gas. These conditions give Lorentz and Doppler linewidths of \(\sim 250 \text{ MHz}\). With \(R \sim 10^{-7}\) for these systems, PNC optical rotations of \(\sim 1 \mu \text{rad}\) were obtained, for column densities of about \(10^{19} \text{ cm}^{-2}\).

Atomic densities in MOTs are typically of order \(10^{11} \text{ cm}^{-3}\). Even using a 2D MOT of length 10 cm gives a column density of only \(10^{12} \text{ cm}^{-2}\), about 7 orders of magnitude lower than those used in PNC optical rotation experiments, which seems insurmountable. However, if the \(M1\) transition for the atoms in the MOT has a linewidth that is \(\sim 10 \text{ kHz}\) (limited only by the MOT temperature of a few \(\mu \text{K}\)), then the linewidth will be about \(10^4\) narrower than that for the atoms in the oven, and the PNC optical rotation is enhanced by 4 orders of magnitude. In addition, using an optical bowtie cavity can enhance the optical rotation by \(10^3 \sim 10^4\) cavity passes. Therefore, combing a 2D MOT with cavity-enhanced optical rotation can give an overall PNC rotation enhancement of \(7 \sim 8\) orders of magnitude, which can offset the low column densities of the MOT, and give optical rotation signals that are similar to or larger than those from traditional optical rotation experiments.

In addition, the MOT and optical-cavity combination can improve the main problem of the traditional PNC optical rotation experiments: the background subtraction was very slow, involving the replacement of the gas cell with a dummy tube, which took many minutes. This very slow background subtraction was inadequate to account for faster drifts in experimental conditions [40], and was the main limitation to measurement sensitivity. In contrast, the atoms in the MOT can be added or removed on the ms timescale, affording a rapid background subtraction procedure, in addition to the signal reversals offered by the bowtie cavity (see section 2.1). Furthermore, other problems, such as birefringence in the windows of the gas cell or inhomogeneities of the vapour in the oven due to temperature gradients, are not present or greatly reduced in the MOT.

Atomic systems that are well-suited for PNC optical rotation measurements in a MOT must satisfy three criteria: (a) they must have strong trapping transitions, allowing atomic densities in the MOT of about \(10^{11} \text{ cm}^{-3}\), and at temperatures of order \(\mu \text{K}\); (b) they must have a PNC optical rotation transition to an excited state with a long lifetime (preferably of order ms or longer), so that the linewidth is as narrow as possible (preferably limited by the Doppler width, giving linewidths of a few kHz); (c) the PNC optical rotation transition should be sufficiently sensitive to PNC (e.g. the atom has high Z, and the transition has \(R > 10^{-5}\)).

We identify Dy as a good candidate for PNC optical rotation in MOT: It can be trapped in MOT at densities of about \(10^{11} \text{ cm}^{-3}\), and at temperatures of \(\sim 6 \mu \text{K}\); the ground state has an \(M1\) transition at 2419 nm to a long-lived metastable excited state, that can only decay via the same \(M1\) transition (see figure 4(a)); Dy has \(Z = 66\), and the \(M1\) transition is expected to have \(R \sim 10^{-7}\), although this must be verified by atomic theory calculations. Therefore, Dy satisfies all the necessary requirements mentioned above. We also identify Cs as a plausible candidate, as Cs MOT densities can be about an order of magnitude higher than that of Dy. And although Cs does not have a strong \(M1\) transition, it does however have a strong electric quadrupole (E2) transition, the \(6S_{1/2} \rightarrow 5D_{3/2}\) at 690 nm. Cronin [36] showed that optical rotation from \(E1\) – \(E2\) interference is possible, provided the target atoms are polarized. We discuss the peculiarities of the Cs proposal after the discussion on the Dy MOT below.
since the whole setup can be placed in vacuum without any intra-cavity optics.

Since no theoretical atomic-structure calculations have been performed yet for the Dy M1 transition under consideration, we will have to estimate plausible values to the M1 and E1_{PNC} amplitudes. We do so based on the thallium values from [27]: for M1 we use the thallium value, while for E1_{PNC}, we will use a scale-down factor of Z(Dy)³/Z(Tl)³ ≈ 0.5, due to the approximate theoretical Z³ dependence of E1_{PNC}. These values will give the correct order-of-magnitude estimates of the PNC optical rotation signals, however, of course, atomic-structure calculations will be needed to determine precise values.

Finally, the hyperfine constants and g_j factors required for the calculations in the 5^n_J and 5^l_J states of the odd Dy isotopes (¹⁶⁴Dy [42] and ¹⁶⁶Dy) are taken from [42], while the transition linewidth is estimated to be a few kHz, dominated by Doppler broadening, even at 6 μK, since the homogeneous contributions (lifetime and collisions) are expected to be miniscule.

Using these values, and varying the number of cavity passes, we simulate the F = 21/2 → F’ = 19/2 transition of ¹⁶⁴Dy, which is the strongest for this isotope, and plot the calculated optical rotation signal versus frequency detuning from resonance in figure 4. We see that large PNC optical rotation angles are potentially feasible, and therefore this method merits further investigation: the M1 and E1_{PNC} amplitudes should be calculated, and 2D Dy MOTs with the characteristics stated above should be constructed.

3.1.2. Cs. For ¹³³Cs we, again, consider a 2D MOT placed with its long axis along one arm of the cavity, but this time an E2 (rather than an M1) transition is probed, the ⁶²S₁/₂ → ⁵²D₃/₂ at 690 nm. The PNC contribution in this transition comes solely from the S-state, since the PNC interaction only mixes S₁/₂ and P₁/₂ states. The theory for PNC optical rotation E2 – E1_{PNC} interference is presented by Cronin [36], and more details will be given by the authors elsewhere [43]. Due to the symmetry of the transition, the E2 – E1_{PNC} interference does not produce a PNC-induced optical rotation in an isotropically populated vapour of Cs [36], i.e. summing over all excited magnetic substates, and averaging over all equally-populated ground magnetic substates, yields identically zero optical rotation. However, by bringing the population distributions of both the ground and excited states away from the isotropic distribution, e.g. by optical pumping orientation of the ground state and by appropriately dressing the excited state, using the 5D → 7P to create optical transparency, PNC-induced optical rotation signals can be recovered. We illustrate this situation in figure 5: by orienting the populations of the states involved, we can isolate non-zero PNC-induced interference contributions from specific m_f → m’_f transitions, and obtain an optical rotation signal. The simulations in figure 5 have been performed, as in the case of Dy, for 1000, 3000, and 10 000 cavity passes through a 2D MOT of Cs atoms, with a length of 10 cm and a 10¹⁵ cm⁻³ density, and for the F = 4 → F’ = 5 transition, which yields a maximal optical rotation of ~1.5 μrad for these conditions. The simulations have used the Cs hyperfine constants from [44], and the lifetime of the 5d₃/₂ state from [45]. The optical rotation signal depends strongly on the degree and spatial orientation of the Cs polarization. For example, rotating the orientation of the population distributions by π/2 around an axis perpendicular to the beam propagation direction inverts the sign of the optical rotation, but the amplitude changes magnitude, because the contributing interfering terms from smaller m_f are not as strong as those for maximal m_f. Another way to reverse the sign of the optical rotation signal is shown in figure 5, where a different transition is probed, the F = 4 → F’ = 3. In this case, the signal is, again, reversed in sign, but not in amplitude. However, we note that the nuclear-spin-independent (NSI) PNC signal and the nuclear-spin-dependent (NSD) PNC signal behave differently for these two transitions: the larger NSI-PNC signal inverts sign, whereas the NSD-PNC signal (which is about 30 times smaller) does not change very much [46, 47]; this difference
3.2. PNC optical rotation in expansions of diatomic radicals

PNC effects are enhanced in diatomic molecular radicals, due to the closely spaced rotational levels of opposite parity [48, 49], with splittings that are 4–5 orders of magnitude smaller than those typically found in atoms. Such small splitting are being used to create large PNC mixings and large potential PNC signals, using the Stark-interference method applied to a molecular beam of $^{137}$BaF molecules [30, 31], which doesn’t depend crucially on producing large high molecular densities. In contrast, the optical rotation method typically requires large column densities (e.g. $\sim 10^{10}$ cm$^{-2}$ for Tl, Bi, and Pb atoms [27–29]), whereas molecular radicals can only be produced in densities that are several orders of magnitude lower.

Jet-cooled molecular radicals have been shown to be formed via a combination of slit supersonic expansions with pulsed electric discharges. For example, using this method, OH radicals have been produced at densities greater than $10^{14}$ cm$^{-3}$, at rotational temperatures of $T_{rot} \sim 25$ K, and with Doppler linewidths of $\sim 100$ MHz [50]. The low rotational temperature means that most of the population is in the lowest few rotational states of the molecules. HgH ($v = 0$, $J = 1/2^+$) molecules can also be produced at high densities using this method, from a gas mixture of Hg vapour and H$_2$ gas. The discharge (or irradiation with 253.7 nm light) will excite the Hg atoms to Hg($^3P_1$), which will then react with H$_2$ to produce HgH [51]:

$$\text{Hg} (^3P_1) + \text{H}_2 \rightarrow \text{HgH} + \text{H}.$$  

The supersonic expansion then cools the HgH rotationally and translationally to about 10 K, as shown by Mayama et al. for HgH in jet-expansion experiments [51]. We assume that the light inside the optical cavity interacts with a molecular beam of HgH traversing one arm of the cavity. We simulate experimental PNC signals using the parameters mentioned above, and vary the HgH density from $10^{13}$ cm$^{-3}$ up to $2 \times 10^{14}$ cm$^{-3}$.

For the measurement of PNC-induced optical rotation, we propose using the $\frac{1}{2}^2 \Sigma^+ \rightarrow \frac{1}{2}^2 \Pi_{1/2}$ even-to-even parity $M1$ transition, the one closest to the lowest electric-dipole-allowed $Q1$ transition of the $^2\Sigma^+ (v = 0) \rightarrow ^2\Pi_{1/2} (v' = 0)$ vibronic band. Both the $Q1$ and the $M1$ are depicted in figure 6. Given that the $Q_1 (\frac{1}{2}^2 \Sigma^+ \rightarrow \frac{1}{2}^2 \Pi_{3/2})$ is at $24 \, 933.6$ cm$^{-1}$ [51] and the excited state doublet splitting is $\Delta = 3.36$ cm$^{-1}$ [37], the $M1$ transition energy will be $\Delta E_{M1} = 24 \, 930.24$ cm$^{-1}$, corresponding to a wavelength $\lambda = 401.12$ nm. The lifetime of the $^2\Pi_{1/2} (v' = 0)$ is $\tau = 101$ ns [51], yielding a natural linewidth $\Delta f_0 = \frac{1}{2 \pi \tau} = 1.6$ MHz, which is the bulk of the homogeneous contributions to the overall broadening of the lineshape. However, at $T_{rot} = 10$ K, the lineshape is essentially Doppler-dominated, with $\Delta f_D \approx 72$ MHz.

The $M1$ amplitude and $R = \frac{\text{Im}(E_{PNC})}{M1}$ ratio, both necessary for our simulations, were recently calculated for the $^{199}$HgH by Geddes et al. [37], using the relativistic coupled cluster method, yielding $M1 = 1.4 \mu_B$ and $R = 1.5 \times 10^{-6} \kappa$, where $\kappa$ is a...
dimensionless constant determined by the nuclear anapole moment to be extracted from experiment. Since $\kappa$ is unknown, for the calculations we use the trivial value $\kappa = 1$, which means that the calculations we present are of optical rotation per unit $\kappa$.

Finally, two things are worth mentioning. Firstly, the hyperfine structure of the $^{199}$HgH is at a first approximation neglected, since the corresponding energy level splitting is quite smaller than that of the $\Lambda$-doubling. However, we do make a density correction for the calculations as follows: both the ground and excited states of the transition are split into two hyperfine levels with $F = 0, 1$, but only the $F = 1 \rightarrow F' = 1$ transition is allowed [52]. Hence, only this transition is taken into account, and of the total density, only 75% is used for the calculation, which is approximately the ratio of the $F = 1$ population to the total. Another population correction factor comes from the distribution of population within the $v = 0$ vibrational manifold: due to the expansion cooling to $T = 10$ K, the majority of the population is in the desired, lowest vibrational state, $(v = 0, J = 1/2^+)$. Indeed, for the experimental values of the rotational and $\Lambda$-doubling constants of HgH, $B(2^3\Sigma_{1/2}) = 5.3888$ cm$^{-1}$ and $\gamma = 2.14$ cm$^{-1}$, respectively [37, 53], it can be calculated that nearly 60% of the population is in the $(v = 0, J = 1/2^+)$ state. On the other hand, although performing the experiment at room-temperature would allow one to use a longer vapour cell, only about 2.5% of the population is in the $(v = 0, J = 1/2^+)$, which offsets the advantage of a longer interaction path. A simulation for a thermal vapour at the maximal density of HgH molecules is also performed for comparison to the rotationally cooled molecular beam results.

Secondly, the existence of a nearby $E1$-allowed transition (the $Q_1$ at just $\Delta = 3.36$ cm$^{-1}$ away) should barely have any effect on the observation of the $M1$ transition of interest. We provide a brief explanation as to why: away from resonance, dispersion and absorption scale inversely proportionally to the detuning, $1/\delta f$, and the square of the detuning, $1/\delta f^2$, respectively. Additionally, it is the Lorentzian lineshape (with $\Delta f_L = 1.6$ MHz) which dominates for large detunings, since the Doppler exponential decays far more rapidly. So, at $3.36$ cm$^{-1}$, the neighbouring $E1$ transition is already some 60 000 linewidths away, its absorption diminished by the square of that, while the dispersive contribution in the vicinity of the $M1$ transition is not only extremely small, but also its fractional variation across the Doppler profile of the $M1$ transition is on the order of $10^{-7}$.

The projected PNC optical rotation of HgH ($v = 0, J = 1/2^+$) is shown in figure 6, where rotation angles of greater than 10 $\mu$rad are calculated. We note that these are very large rotation angles, more than an order of magnitude larger than those measured in atomic experiments for NSI-PNC. The reason is the combination of the enhancement factors of the HgH molecules and the optical cavity, which more than offset the relatively low sample density. These large PNC signal projections should motivate the construction of an HgH discharge slit expansion, to see whether large HgH ($v = 0, J = 1/2^+$) densities, of order $10^{13}$ cm$^{-3}$ or larger (consistent with literature values for other diatomic radicals), can indeed be produced.

4. Summary and conclusions

We have shown that large optical rotation PNC signals (similar or larger than those of past PNC optical rotation experiments) are potentially achievable, by combining the cavity-enhanced optical rotation method with samples of Cs or Dy atoms trapped in 2D MOTs, or $^{199}$HgH molecules in slit-nozzle expansions. The particularly large optical rotation signals predicted for HgH are due to the exploitation of several enhancement factors (in addition to the usual $\sim 6$ orders of magnitude from the $\sim Z^2$ enhancement for high $Z$): $\sim 4$ orders of magnitude from the small opposite-parity splitting in HgH, $\sim 2$ orders of magnitude from the translational and rotational cooling in the nozzle expansion, and $\sim 3$ orders of magnitude from the number of cavity passes; although we estimate that the column density of HgH ($v = 0, J = 1/2^+$) that can be produced is at least $\sim 4$...
orders of magnitude lower than that for TI, Bi, or Pb in an oven, the remaining enhancement factors allow us to project NSD-PNC optical rotation signals that are a few orders of magnitude larger than those measured in past PNC optical rotation experiments.

The promise of such large PNC signals should be confirmed by a calculation of the transition amplitudes $E_{\text{PNC}}$ and $M1$ for the Dy $5I_6 \rightarrow 5I_7$ transition at 2419 nm, and by the production of $^{199}$HgH molecules in the lowest-rovibrational energy state ($v = 0, J = 1/2^+$) at densities of order $10^{13}$ cm$^{-3}$ or larger, using a slit-nozzle expansion combined with electrical discharge. Such experiments will allow the sensitive measurement of and NSI, NSD, and isotope-dependent PNC effects, in Cs, Dy, and HgH.

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