Searching for the electron EDM in a storage ring

D Kawall
Department of Physics, University of Massachusetts, Amherst, MA 01003, USA
E-mail: kawall@physics.umass.edu

Abstract. Searches for permanent electric dipole moments (EDM) of fundamental particles have been underway for more than 50 years with null results. Still, such searches are of great interest because EDMs arise from radiative corrections involving processes that violate parity and time-reversal symmetries, and through the CPT theorem, are sensitive to CP-violation. New models of physics beyond the standard model predict new sources of CP-violation leading to dramatically enhanced EDMs possibly within the reach of a new generation of experiments. We describe a new approach to electron EDM searches using molecular ions stored in a tabletop electrostatic storage ring. Molecular ions with long-lived paramagnetic states such as tungsten nitride WN⁺ can be injected and stored in larger numbers and with longer coherence times than competing experiments, leading to high sensitivity to an electron EDM. Systematic effects mimicking an EDM such as those due to motional magnetic fields and geometric phases are found not to limit the approach in the short term, and sensitivities of \( \delta |d_e| \approx 10^{-30} \text{e-cm/day} \) appear possible under conservative conditions.

1. Introduction and motivation
An electric dipole moment (EDM) of a fundamental particle would lie along its spin, \( d = d\sigma \), and lead to a linear Stark shift in an electric field, \( H_{\text{EDM}} = -d \cdot E \), analogous to a Zeeman shift, \( H_Z = -\mu \cdot B \). Unlike the Zeeman interaction, the EDM interaction is odd under parity \( P \) and time reversal \( T \) symmetries, and will be zero unless \( P \) and \( T \) symmetries are broken [1].

While \( P \) violation is inherent in the weak interaction, \( T \) violation has only been observed in decays of \( K \) and \( B \) mesons and is accommodated in the Standard Model (SM) with a single complex phase in the Cabibbo-Kobayashi-Maskawa quark mixing matrix [2, 3]. Thus non-zero EDMs are expected in the Standard Model (SM) from \( P \) and \( T \)-violating radiative corrections.

The SM prediction for an electron EDM is at the level of \( < 10^{-38} \text{e-cm} \) [4], more than 11 orders of magnitude smaller than the current limit. This extreme suppression occurs because at least three quantum loops are required to link the electron to the sole \( T \)-violating phase in the SM. However, theorists now predict dramatically enhanced EDMs, within a few orders of magnitude of current experimental limits [5, 6, 7]. These enhancements are consequences of new physics required to solve several hierarchy problems in the standard model. For instance supersymmetric (SUSY) theories solve some problems in the SM at the expense of introducing new particles and new \( T \)-violating phases, yielding much larger expectations for EDMs. Dimensional analysis suggests the scaling :

\[
d_e \approx \left( \frac{300 \text{ GeV}}{M} \right)^2 \sin \phi \times 10^{-25} \text{e-cm},
\]

Published under licence by IOP Publishing Ltd
where $M$ is the mass scale of new physics, and $\phi$ is a new $CP$-violating phase [6]. If an electron EDM were not found at the level of $10^{-30}$ e-cm, the $CP$ phase would have to be $\approx 10^{-5}$, which is unnaturally small. Alternatively, such a limit would rule out new physics at the scale of 100 TeV, beyond the reach of the Large Hadron Collider (LHC).

In addition to the expectations of new physics required to solve the hierarchy problems of the SM, $CP$-violation in the SM is more than six order of magnitude too small to explain the observed dominance of matter over anti-matter [8, 9, 10]. Thus new physics is expected, and almost certain to come with significant new sources of $CP$-violation beyond that in the SM, leading to EDMs of the electron naturally expected within a few orders of magnitude of the current limit [6, 7].

This prospect motivates a search for an electron EDM $d_e$ using a new approach: molecular ions stored in an electrostatic storage ring. The new approach promises to reduce the limits on the electron EDM by three to four orders of magnitude to $10^{-30} - 10^{-31}$ e-cm. This represents an increase in sensitivity of three to four orders of magnitude over the current limit of $|d_e| \lesssim 1.6 \times 10^{-27}$ e-cm [11], and would be complementary to direct searches for new physics at the LHC. If an electron EDM were not found at this level of sensitivity, many theories of physics beyond the SM would be overturned.

2. EDM measurement approach in paramagnetic molecules

The proposed experiment is similar to others using beams of neutral molecules, [12, 13, 14], and with one proposing to use trapped molecular ions [15], but with differences resulting in gains in statistical sensitivity.

Electron EDM experiments search for an energy shift from the electron EDM $d_e$ interacting with the large internal electric field $E_{\text{int}}$ in a heavy polar molecule. The field $E_{\text{int}} = E_{\text{int}} \hat{n}$ lies along the internuclear axis $\hat{n}$. If $d_e \neq 0$, unpaired electrons which are spin polarized along $\hat{n}$ will exhibit a linear Stark shift, described by the non-relativistic Hamiltonian $H_{\text{EDM}} = -d_e \cdot E_{\text{int}} = -d_e \sigma \cdot E_{\text{int}}$.

In a nutshell, the experiment proposes to search for an electron EDM by injecting $WN^+$ ions moving at $\approx 4 \times 10^4$ m/s into an electrostatic storage ring of roughly 1 m radius. The electrodes, separated by 4 cm, are biased to roughly $\pm 60$ V and curved to confine the ions radially and vertically. The radial electric field of the storage ring polarizes the large internal electric field of the molecule along the radial direction. Lasers are then used to polarize the unpaired electron spin perpendicular to the internal electric field, where the spins precess if $d_e \neq 0$. After a storage time of 100 ms (limited by collisions with residual gas), the precession angle is measured and used to set a limit on $d_e$.

This search will use states with total angular momentum $J=1$ in the $^3\Delta_1$ ground state of the molecular ion $WN^+$. The states are labeled by kets $|J, M, \Omega\rangle$, where $M$ is the projection of $J$ on the space-fixed (laboratory) $\hat{Z}$ axis, and $\Omega$ is the projection of electronic angular momentum $J_e$ on $\hat{n}$, $\Omega \equiv J_e \cdot \hat{n} = \pm 1$. Even and odd combinations of states with the same $J$ and $M$ but opposite $\Omega$ form degenerate eigenstates of parity, $P$. The degeneracy is lifted by Coriolis-like couplings between spin and rotational angular momentum. This splits the $J=1$ level by $\Delta E_\Omega \equiv \Delta \Omega$ into closely spaced states of opposite parity, labeled $e$ for $P = (-1)^J$, $f$ for $P = (-1)^{J+1}$ [16].

For the EDM measurement, an external field $E$ along $\hat{z}$ mixes the $\Omega$-doublet states to form eigenstates of definite $\Omega$, polarizing the electronic angular momentum along $\hat{n}$ (and hence along $E_{\text{int}}$), $P_E = \langle J_e \cdot \hat{n} \rangle \approx \pm 1$. This leads to complete mixing of the $|J, M, P = +1\rangle$ and $|J, M, P = -1\rangle$ states, and ordinary Stark shifts from $H_{\text{St}} = -\mu_e \cdot E$. Here $\mu_e = \mu_e \hat{n}$ is the molecule-fixed dipole moment of the $^3\Delta_1$ state, estimated as $3.27$ D or $1.64$ MHz/(V/cm). (Since $WN^+$ is isoelectronic to tungsten carbide (WC), many of its electronic properties will be similar and we expect $\Delta \Omega(WN^+) \approx$ few kHz, and $E_{\text{int}} \approx 50$ GV/cm [13, 16]). While the $M=0$ states do not mix, for $\mu_e \cdot E \gg \Delta \Omega$, the $|J| > 0$ $\Omega$-doublet states are mixed fully,
Figure 1. The $J=1$ levels of the $X^3\Delta_1$ ground state of WN$^+$ and their shifts in external fields $E = E\hat{z}$ and $B = B\hat{z}$ are shown. The $\Omega$-doublet splitting is indicated by $\Delta E_\Omega$. Dashed lines (black) indicate the levels when $E = 0$ and $B = 0$. Applying $E$ results in Stark shifts $\Delta E_{\text{St}}$ to the dotted lines (black). Applying $B$ results in Zeeman shifts $\Delta E_{\text{Z}}$ to the dash-dot lines (red). If $d_e \neq 0$, the levels shift further to the solid lines (blue). The $M=0$ levels are unchanged in all cases, and the Zeeman, $\Omega$-doubling, and EDM shifts are highly exaggerated.

electrically polarizing the molecule along the external field $E$ and leading to a well defined polarization direction $N = \text{sgn}(\mu_a \cdot E)$. It is convenient to describe the states in a new basis $|J,M,N\rangle \equiv \frac{1}{\sqrt{2}} [ |J,M,P=+1\rangle - (1)^J |J,M,P=-1\rangle ]$, where $\Omega = N \cdot \text{sgn}(M)$ [17]. Then the upper (lower) $|M| > 0$ levels are characterized by $N=1$ ($N=-1$). This quantity is important because states with the same $N$ have $E_{\text{int}}$ oriented in the same direction.

Applying a magnetic field $B = B\hat{z}$ leads to Zeeman shifts $\Delta E_Z = g_u,l \mu_B B M$. If $d_e \neq 0$, the $|M| > 0$ levels are shifted further by $\Delta E_{\text{EDM}} = d_e E_{\text{int}} M N$. The levels are shown in figure 1.

The EDM shifts can be measured by preparing a coherent superposition of $|J,M,N\rangle$ for $N=+1$ or $N=-1$. In this superposition, the spin $\langle \mathbf{S} \rangle \perp \hat{z}$, and the magnetic moment and electric dipole moment experience torques from the $B$ and $E_{\text{int}}$ fields. For the superposition of levels with the opposite sign of $N$, the torque from $E_{\text{int}}$ reverses, leading to a difference in precession frequency which depends on the magnitude $d_e E_{\text{int}}$. Equivalently, the two states involved in either of the $N=\pm 1$ superpositions differ in energy by $\Delta E(N) = 2g_N \mu_B B - 2d_e E_{\text{int}} N$. Assuming $g_{N=+1} = g_{N=-1}$, then the EDM shift can be isolated from the Zeeman shifts by measuring the splittings $\Delta E(N)$ for $N=\pm 1$ and taking the difference: $\Delta E(N = +1) - \Delta E(N = -1) = 4d_e E_{\text{int}}$. This comparison between the oppositely polarized members of the $\Omega$-doublet is extremely powerful for isolating the tiny EDM shift from other effects. It was proposed by V. Flambaum [18], and first incorporated in experiments by D. DeMille [19].

To detect the EDM, we measure the precession frequencies $\Delta E(N)/\hbar$ for $N = \pm 1$. The technique is identical to that proposed by the ACME collaboration experiment using ThO [12]. We prepare the superposition in the $J=1$ levels:

$$|\psi_N(t=0)\rangle = \frac{1}{\sqrt{2}} [ |M=1,N\rangle + |M=-1,N\rangle ] .$$
This evolves after time $\tau$ to:

$$|\psi_N(t = \tau)\rangle = \frac{1}{\sqrt{2}} \left[ |M = 1, N\rangle + e^{i\phi_N} |M = -1, N\rangle \right]$$

$$= \frac{1}{2} \left( 1 + e^{i\phi_N} \right) |X\rangle + \frac{1}{2} \left( 1 - e^{i\phi_N} \right) |Y\rangle,$$

where

$$|X\rangle \equiv \frac{1}{\sqrt{2}} \left[ |M = 1, N\rangle + |M = -1, N\rangle \right] \quad \text{and} \quad |Y\rangle \equiv \frac{1}{\sqrt{2}} \left[ |M = 1, N\rangle - |M = -1, N\rangle \right]$$

and the phase shift $\phi_N$ is given by $\phi_N = \Delta E(N)\tau / \hbar$. To measure this precession angle, we effectively measure the components of $|\psi_N(t = \tau)\rangle$ along $\hat{x}$ and $\hat{y}$.

This can be done by driving a transition from the superposition state to a higher lying electronic excited state, $D$, with $J'=1$, and detecting the fluorescence from its decay. By exciting with a resonant laser with wavevector $k$ along $\hat{y}$ and polarized along $\epsilon = \hat{x}$, we find:

$$\langle D, J' = 1, M' = 0 | \hat{x} | X\rangle|^2 \propto n_0 (1 + \cos \phi)/2,$$

where $n_0$ is the number of the molecules in the superposition state. Similarly, using $\epsilon = \hat{y}$ and wavevector $\hat{x}$ we find:

$$\langle D, J' = 1, M' = 0 | \hat{y} | Y\rangle|^2 \propto n_0 (1 - \cos \phi)/2.$$  

Finally we note $\langle D, J' = 1, M' = 0 | \hat{x} | X\rangle|^2 = 0$, and $\langle D, J' = 1, M' = 0 | \hat{y} | Y\rangle|^2 = 0$. The asymmetry $A$ formed from the difference divided by the sum of the fluorescence is then given by $A_N = \cos \phi_N / n_0$.

Maximum sensitivity to $\phi_N$ occurs for $\phi_N = \pi/2$, which can be achieved by adjusting the magnetic field strength $B$ along $\hat{z}$. Defining the number of photons detected with $\hat{i}$ polarized light as $N_i$, then:

$$A_N = \frac{N_X^N - N_Y^N}{N_X^N + N_Y^N} = \cos \phi_N = \cos (\Delta E(N)\tau / \hbar) \quad (1)$$

$$= \cos (2g_N \mu_B B \tau / \hbar) \cos (2d_e \epsilon_{\text{int}} N \tau / \hbar) + \sin (2g_N \mu_B B \tau / \hbar) \sin (2d_e \epsilon_{\text{int}} N \tau / \hbar) \quad (2)$$

$$\approx 2d_e \epsilon_{\text{int}} \tau \cdot \text{sgn}(B) \cdot \text{sgn}(\epsilon) \cdot \text{sgn}(N) \tau / \hbar. \quad (3)$$

The shot-noise limit on $d_e$ per measurement cycle is given by:

$$\delta d_e \approx \frac{\hbar}{2\epsilon_{\text{int}} \tau \sqrt{N_i}}, \quad (4)$$

where $N_i = N_X^N + N_Y^N$ is the total number of photons detected. Sensitivity is maximized by large $\epsilon_{\text{int}}$, long coherence times $\tau$, and measuring the final state of as many molecules as possible. Note that the asymmetry $A$ is largely independent of fluctuations in the beam source.

3. **EDM measurement with WN$^+$ in an electrostatic storage ring**

The electronic structure of WN$^+$ was studied using the density functional method B3LYP [20, 21] with the SDD basis set [22] and Gaussian03 program [23]. The studies predict a $^3\Delta_1$ ground-state with occupation $5\sigma^2 2\pi^4 1\delta^4 6\sigma^4$, in agreement with the only published calculation [24], and consistent with the experimentally confirmed $^3\Delta_1$ ground state of the isoelectronic WC [25]. (The structure calculation also predicts a $^3\Sigma^-$ state, nearly degenerate within calculational uncertainties with the $^3\Delta_1$ state with occupation $5\sigma^2 2\pi^4 1\delta^2$, which cannot be excluded as the ground state. Note that the $^3\Sigma^+$ state could also be used for the EDM search, with reduced sensitivity due to its larger magnetic moment.)

1 With Ricardo Metz, Department of Chemistry, University of Massachusetts Amherst, Amherst MA 01003, USA
The EDM measurement will occur in the $X^3\Delta_1(v = 1, J = 1)$ ground state of WN$^+$. This will be populated via optical-optical double resonance by excitation from $X^3\Delta_1(v'' = 0, J'' = 1, M'' = 0)$ to the electronic excited state (predicted from the calculations) $D^3\Pi(v = 0, J = 2, M = 0)$ using $\hat{z}$ polarized light, and simultaneous stimulated emission into a coherent superposition of $X^3\Delta_1(v' = 1, J' = 1, M' = \pm 1)$ (i.e. one of the $|\psi_{N=\pm 1}\rangle$ states) using counter-propagating $\hat{x}$ polarized light. The excitation will occur on WN$^+$ ions stored in the electrostatic ring using chopped CW lasers. The lasers counter-propagate along $\hat{y}$, perpendicular to both the storage ring radial electric field along $\hat{z}$ and the ion velocity vectors along $\hat{x}$.

The excitation efficiency, defined as the ratio of ions in the superposition state to the total number initially in the $X(v' = 0, J' = 1)$ state, should be at least 5%.

Detection of the $|X\rangle$ state will occur by driving $X(v' = 1) \rightarrow D(v = 0, 1, \text{ or } 2)$ with $\hat{x}$-polarized light, and detecting the fluorescence back to $X(v'' = 0)$ (or $X(v'' = 1)$). The $|Y\rangle$ state will be detected in the same manner but with $\hat{y}$-polarized light separated slightly in time. To minimize Doppler shifts, the excitation lasers propagate along the radial direction ($\hat{z}$), transverse to the ion motion. See figure 2 for the excitation and detection schemes.

The WN ions for the experiment will be produced by laser ablation of a solid WN or WN$_2$ precursor, which thermalize in a buffer gas of He at 4.2 K. This technique has been well developed by J. Doyle’s group at Harvard [26, 27]. The number of WN neutral molecules thermalized in a buffer gas, from pulsed laser ablation of a solid WN precursor with a doubled Nd:YAG, 25 mJ/pulse, $\approx 10^{10}$ W/cm$^2$, should be similar to yields of other diatomics. In the case of PbO these yields are roughly $10^{12}$/pulse [27]. The fraction of WN$^+$ ions can be taken as at least 10% [28], or $10^{11}$ per pulse. The ablation occurs in a cell with He buffer gas held at 4.2 K and number density $n > 10^{16} - 10^{17}$ cm$^{-3}$. The ions thermalize with the buffer gas in 100-200 collisions, to 4.2 K in $\approx 100$ $\mu$s. The fraction of thermalized WN$^+$ ions in the $X^3\Delta_1(v'' = 0, J'' = 1)$ state is roughly $B_e/k_B T \approx 0.15$, where $B_e$ is the rotational constant, 14.4 GHz. This suggests $\approx 1.5 \times 10^{10}$ WN$^+$ produced in the lowest quantum state per ablation pulse.

A small electric field in the cell is used to drift the ions out of small hole of a few mm in diameter. This will cause some reheating, and the number in the $v' = 0, J' = 1$ state is reduced to $\approx 0.5 \times 10^{10}$ at the cell exit.

At the exit, electrostatic lenses collimate and direct the ions towards a quadrupole mass filter.

![Figure 2](image1.png)

Figure 2. The state preparation scheme is shown on the left. By tuning the laser which stimulates emission from the $D$ state to the $X(v'' = 1)$ by the Stark splittings, both the upper and the lower superposition states ($N = \pm 1$) can be populated. The detection scheme is shown on the right. The $|X\rangle$ and $|Y\rangle$ states can be detected in steps by exciting into the $D$ state and detecting the fluorescence to $X(v'' = 0)$, which is to the blue of the excitation.
Figure 3. A schematic top view of the storage ring and injection scheme are shown on the left. On the right a cross-section of the electrodes is shown. The radii of curvature of roughly 100-104 cm combine radial and vertical focusing.

(QMF). The QMF is used in a wide-band mode to pass the WN isotopes $^{182}$W$^{14}$N, $^{184}$W$^{14}$N, and $^{186}$W$^{14}$N, with natural abundances of 26.5%, 30.6%, and 28.4% respectively. The ions are accelerated after the QMF to their final velocity of $\approx 4 \times 10^4$ m/s (requiring $\approx 1600$ V) and directed to the storage ring. Turbo pumps bring the pressure in the storage ring vacuum chamber to 10$^{-8}$ Torr or less. To store the ions, 1/8 of the ring is turned off, and the ions are injected through a small hole in the outer ring electrode in this grounded section. The ions eventually encounter the radial field and move in a circular orbit. The grounded section is turned on before the ions complete 7/8 of a revolution, and the hole is plugged (see figure 3). The ions are put in the superposition state, left for 100 ms, then the EDM precession angle is measured.

4. Estimated statistical sensitivity of the electron EDM experiment

The number of ions in the lowest quantum state from cell is $n_{\text{cold}} \approx 0.5 \times 10^{10}$. A conservative estimate is that 2% of them will be stored ($\epsilon_{\text{store}} = 0.02$). Thus each ablation pulse should yield about $10^8$ stored ions. The fraction put into the excited state is estimated as $\epsilon_{\text{exc}} \approx 5\%$. This state is detected with an efficiency $\epsilon_{\text{det}} = BF \times QE \times SA = 0.1 \times 0.2 \times 0.1 \approx 2 \times 10^{-3}$ where BF is the branching fraction of the decay fluorescence to the blue of the excitation, QE is the quantum efficiency and SA is the solid angle. Per ablation pulse the number of photons detected is $n_{\text{cold}} \times \epsilon_{\text{store}} \times \epsilon_{\text{exc}} \times \epsilon_{\text{det}} = 1 \times 10^4$. At a repetition rate of 10 Hz, this becomes $n_\gamma = 10^5$ s$^{-1}$. This yields a shot noise limit from Eqn. 4 of $\delta d_e = 2 \times 10^{-28} e \cdot cm/\sqrt{Hz}$, and $\delta \phi_e = 7 \times 10^{-31} e \cdot cm/\sqrt{day}$, assuming 100% efficiency, and where we have assumed $E_{\text{int}} = 50$ GV/cm [13, 29] Even assuming 50% efficiency, and only one isotope detected, $\delta d_e = 1.7 \times 10^{-30} e \cdot cm/\sqrt{day}$. This is 3 orders of magnitude below the current limit of $1.6 \times 10^{-27} e \cdot cm$ [11]. This enormous gain in sensitivity is due primarily to the extraordinarily long coherence times. The high statistical sensitivity makes possible the investigation of systematic effects at same level.

5. Systematics leading to a false EDM

Before discussing some systematic uncertainties, note that for $E_{\text{int}}$ of 50 GV/cm, a limit at $10^{-31} e \cdot cm$ corresponds to a precession frequency of 1.2 $\mu$Hz. In a coherence time of 100 ms, this corresponds to an EDM-induced precession angle of $7.5 \times 10^{-7}$ radians.

Effects such as those due to electric field gradients, the molecular electric quadrupole of WN$^+$, motional magnetic fields, and leakage currents can all be shown to lead to effects smaller than $10^{-31} e \cdot cm$. The most serious effects are due to geometric phase effects, and the fact that unlike other EDM experiments, the electric field can not be reversed.

The measurement protocol instead involves measuring the phase in the upper and lower doublets, and taking the difference, $A_N - A_{-N} \approx 4d_e E_{\text{int}} \tau / \hbar$. However, the $g$ factors of the
doublets are slightly different, $\Delta g(\mathcal{E})/g \approx 3\mu_e\mathcal{E}/(20B_0) \approx 2.5 \times 10^{-4}$ [17] (we have omitted the $\mathcal{E} = 0$ difference which is of order $\Delta\Omega/B_0$ [30] which disappears at as $\Delta\Omega/(\mu_e\mathcal{E})$ [17]). For a $B_0 \approx 10\mu$G, this leads to a frequency shift of $\Delta \Omega = \Delta g(\mathcal{E})\mu_B B_0 \approx 350 \mu$Hz that does not cancel in the comparison between doublets. Reversing the magnetic field to $B'_r \approx -B_r$, measuring in both doublets, and then comparing with the original result yields:

$$(A^B_N - A^B_{-N}) - (A^B_N - A^B_{-N}) = 4d\mathcal{E}_{\text{int}} + 2(g_{N=1} - g_{N=-1})\mu_B(B_r - B'_r).$$

If the field can be reversed to a few parts in $10^{-4}$, then a limit at $10^{-31} \text{e}\cdot\text{cm}$ is possible. Of course the reversal of the current in the field coils can easily be done this well, the problem will be with imperfect shielding leading to a net ambient radial field $B_{\text{amb}}$ of 10 nG that does not reverse with the current in the coils. Magnetic shielding at the level of $10^3$ has been achieved over large volumes [31] which would almost be sufficient. We propose to combine shielding with an array of atomic magnetometers to measure the radial field. Relatively simple systems are capable of much better than 1 nG/$\sqrt{\text{Hz}}$ [32], which would easily meet our requirements, and enable a systematic limit from imperfect $B$ field reversal coupled with a difference of doublet $g$ factors, to be below $10^{-31} \text{e}\cdot\text{cm}$.

**Geometric Phases**: The electric and magnetic fields of the storage ring are static, but in the rest frame of the ions, appear to undergo several cyclic motions. The external field $\mathcal{E}$ appears to rotate in the $\hat{x} - \hat{z}$ plane at the cyclotron angular frequency $\omega_{\text{cyc}}$. Further, there are betatron oscillations in the vertical plane from vertical focusing. For a ring of 1 m radius, and ion velocity of $4 \times 10^4$ m/s, the cyclotron frequency is $\omega_{\text{cyc}} = 2\pi \times 6.4 \text{ kHz}$. This is much less than the Stark shift, $\omega_{\text{Stark}} \approx 2\pi \times 25 \text{ MHz}$, so the system responds adiabatically to the field rotation ($\omega_{\text{cyc}}/\omega_{\text{Stark}} \approx 2.5 \times 10^{-4}$). This holds true for the betatron motion too, as $\omega_{\beta} \lesssim \omega_{\text{cyc}}$.

It is known that in such cases, in addition to the dynamic phase from the electric and magnetic fields, the quantum system acquires a geometric phase associated with the variation of the Hamiltonian [33, 34].

In the case where the variation is adiabatic, the additional phase factor is $\exp[-iMd\Omega]$ where $d\Omega$ is the solid angle subtended by the electric field vector. Corrections due to internal structure (nearby rotational levels) are small since $\omega_{\text{cyc}} \ll 2\pi B_e = 2\pi \times 14.4 \text{ GHz}$ [35]. The correction to Berry’s result to first order in the adiabaticity parameter after $N_t$ turns in the ring is $\approx 2\pi N_t M\omega_{\text{cyc}}/(2\omega_{\text{Stark}}) \approx 0.5$ radians [35].

The phase factors are troublesome as they are different for the $M = \pm 1$ levels of the superposition state, and appear as an energy shift [36]. However, since these geometric phases just depend on $M$, they cancel in the comparison between the upper and lower doublets, and do not lead to a false EDM.

A second potential problem occurs because the ions in the ring follow slightly different orbits, so the geometric phases are different, which can lead to dephasing and loss of sensitivity. In the case of the adiabaticity correction, after 600 turns the accumulated phase is 0.5 radians. However, since $\omega_{\text{cyc}}$ varies by $< 0.5\%$, the width of this corrections is $< 2.5$ mrad, so the dephasing is negligible.

Betatron oscillations do not cause dephasing. The solid angle subtended after $N_t$ turns is:

$$d\Omega \approx 2\pi N_t - \Delta \frac{\omega_{\text{cyc}}}{\omega_{\beta}} \left[ \sin \left( \frac{\omega_{\beta}}{\omega_{\text{cyc}}} 2\pi N_t + \phi_0 \right) - \sin \phi_0 \right].$$

The term $\Delta \leq |\mathcal{E}|/\mathcal{E}_r < 0.0025$ is the ratio of the maximum vertical electric field to the radial field for stored ions, and $\phi_0$ is a phase. The betatron angular frequency is $\omega_{\beta} \approx \sqrt{(q\mathcal{E}_z/dz)/m} \approx 3.5 \times 10^4 \text{ rad/s}$, where $d\mathcal{E}_z/dz \approx -0.25 \text{ V/cm}^2$, and $m$ is the WN$^+$ ion mass. For each ion, $\Delta$ will differ, but is bounded by 0.003. The phase also differs for each ion. This leads to a maximum range of phases of $4\Delta\omega_{\text{cyc}}/\omega_{\beta} \lesssim 0.012$ radians, with an average value of zero. Thus betatron oscillations do not lead to dephasing.
Distortions in the ring can lead to geometric phases, but are too small to lead to dephasing as each ion sees the same perturbation. Note that the first order geometric phase from betatron oscillations, and phases due to distortions in the construction of the ring can be measured by injecting the ions in the reverse direction. Changing the electric field in the ring and the ion velocity, allows a measurement of the correction depending on $\omega_{\text{cyc}}/\omega_{\text{Stark}}$. It should be repeated that these geometric phases should be very stable, do not lead to dephasing, can be measured, and cancel in the difference between doublets.

6. Conclusions

A systematic limit at the level of $10^{-31} \text{e}\cdot\text{cm}$ appears feasible. Note also that the EDM can be measured in the three isotopes simultaneously, and that a 3 to 4 order of magnitude improvement appears possible in $d_e$ using $WN^+$ ions in an electrostatic storage ring.

7. References

[1] Khriplovich I B and Lamoreaux S K 1997 CP Violation Without Strangeness Electric Dipoles Moments of Particles, Atom and Molecules (Berlin: Springer-Verlag)
[2] Kobayashi M and Maskawa T 1973 Prog. Theor. Phys. 49 652
[3] Nakamura K and Group P D 2010 J. Phys. G: Nucl. Part. Phys. 37 075021
[4] Pospelov M E and Khriplovich I B 1991 Sov. J. Nucl. Phys. 53 638
[5] Fortson N, Sandars P and Barr S 2003 Physic Today 56 33–39
[6] Abel S A and Lebedev O 2006 JHEP 01 011
[7] Pospelov M and Ritz A 2005 Ann. Phys. (N.Y.) 318 119–169
[8] Sakharov A 1967 Sov. Phys. JETP. Lett. 5 24
[9] Peskin M 2002 Nature 419 24–26
[10] Huet P and Sather E 1995 Phys. Rev. D 51 379–394
[11] Regan B, Commings E, Schmidt C and DeMille D 2002 Phys. Rev. Lett. 88 071805
[12] Vutha A C, Campbell W C, Gurevich Y V, Hutzler N R, Parsons M, Patterson D, Petrik E, Spaun B, Doyle J M, Gabrielse G and DeMille D 2010 J. Phys. B 43 074007
[13] Lee J, Meyer E R, Faudel R, Bohn J L and Leanhardt A E 2009 J. Mod. Opt. 56 2005
[14] Hudson J J, Sauer B E, Tarbutt M R and Hinds E A 2002 Phys. Rev. Lett. 89 023003
[15] Stutz R and Cornell E 2004 Bull. Am. Soc. Phys. 89 76
[16] Brown J M and Carrington A 2003 Rotational Spectra of Diatomic Molecules (Cambridge: Cambridge University Press)
[17] Bickman S, Hamilton P, Jiang Y and DeMille D 2009 Phys. Rev. A 80 023418
[18] Flambaum V 1987 Ph.D. thesis Institute of Nuclear Physics Novosibirsk
[19] DeMille D, Bay F, Bickman S, Kallaw D, Krause D, Maxwell S E and Hunter L R 2000 Phys. Rev. A 61 052507
[20] Becke A D 1993 J. Chem. Phys. 98 5648
[21] Lee C, Yang W and Parr R G 1988 Phys. Rev. B 37 785
[22] Schwerdtfeger P, Dolg M, Schwarz W H E, Bowmaker G A and Boyd P D W 1989 J. Chem. Phys. 91 1762
[23] Frisch M J et al. Gaussian 03, Revision C.02 Gaussian, Inc., Wallingford, CT, 2004
[24] Horng B, Cheng L, Wang M Y and Wu Z J 2010 Mol. Phys. 108 25
[25] Sickafoose S M, Smith A W and Morse M D 2001 J. Chem. Phys. 116 993
[26] deCarvalho R, Doyle J M, Friedrich B, Guillet T, Kim J, Patterson D and Weinstein J D 1999 Eur. Phys. J. D 7 289
[27] Egorov D, Weinstein J D, Patterson D, Friedrich B and Doyle J M 2001 Phys. Rev. A. 63 030501(R)
[28] Thestrup B, Toftmann B, Schou J, Doggett B and Lunney J G 2002 Appl. Surf. Sci. 197-198 175–180
[29] Meyer E R and Bohn J L 2008 Phys. Rev. A 78 010502(R)
[30] TNelis, Beaton S P, Evenson K M and Brown J M 1991 J. Mol. Spect. 148 462–478
[31] Xu S, Rochester S M, Yaschuk V V, Donaldon M H and Budker D 2006 Rev. Sci. Instrum. 77 083106
[32] Budker D and Romanis M 2007 Nature Phys. 3 227–234
[33] Berry M V 1984 Proc. R. Soc. London Ser. A 392 45–57
[34] Holstein B R 1989 Am. J. Phys. 57 1079–1084
[35] Meyer E R, Leanhardt A E, Cornell E A and Bohn J L 2009 Phys. Rev. A 80 062110
[36] Vutha A and DeMille D 2009 (Preprint arXiv:0907:5116)