Wigner-Eckart Theorem and the False EDM of $^{199}$Hg

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Abstract

In neutron electric dipole moment (EDM) experiments, $^{199}$Hg is used as a comagnetometer. The comagnetometer suffers from a false EDM arising in leading order from a gradient $\partial B_z/\partial z$ in the magnetic field. Our work concerns higher-order multipole corrections to the false EDM of $^{199}$Hg. We show that for spherical traps, all higher-order multipoles are identically zero. We further show that for the usual cylindrical traps used in EDM experiments, selection of quasi-spherical dimensions for the trap can reduce the higher-order contributions. The results are another indication that trap geometry is an important consideration for experiments desiring to control this systematic effect.

Keywords: false electric dipole moment, trapped particles, neutron electric dipole moment

1. False electric dipole moments for particles in traps

In the most precise neutron electric dipole moment (EDM) experiments, ultracold neutrons are stored in a bottle in either parallel or antiparallel electric $E$ and magnetic $B$ fields. Their spin precession frequency $\omega_\pm$ is measured

$$\omega_\pm = (2\mu_n B \pm 2d_n E)/\hbar$$

(1)

for each of the parallel (+) and antiparallel (−) configurations, leading to a determination of the neutron electric dipole moment $d_n$. Here $\mu_n$ is the neutron magnetic moment.

A crucial aspect of the experiment is that the magnetic field be continuously monitored so that any drifts can be corrected. To monitor the field, a second “comagnetometer” atomic species is stored in the cell and its spin
precession frequency is measured optically. Normally $^{199}\text{Hg}$ is used for this purpose \cite{1,2,3}, in part because its true EDM has been constrained to be small \cite{4,5}.

Ideally, the magnetic field $B$ should be uniform so that long free-precession times can be achieved for both the neutrons and Hg atoms. In the previous most precise nEDM experiment \cite{6,7} it was found that a vertical gradient $\partial B_z/\partial z$ in the magnetic field induced false EDM’s for the neutrons and Hg atoms. The frequency shift can be considered as a Ramsey-Bloch-Siegert shift for particles traveling in orbits within the trap \cite{8,9}. The ultracold neutrons traverse the measurement volume slowly enough that their accrued phase can be thought of as a geometric phase which accrues adiabatically \cite{8}.

The false EDM’s can also be calculated by correlation function techniques \cite{10,11}. This led to the realization that false EDM for the Hg atoms could be written in the form

$$df_{f,Hg} = -\frac{h\gamma^2}{2c^2} \langle xB_x + yB_y \rangle$$

using integration by parts \cite{12}. Here, $\gamma = 2\mu/\hbar$ is the gyromagnetic ratio of Hg with $\mu$ being its magnetic moment, and the average is over the storage volume. This form is valid to high precision in the low-frequency (field) limit, even reproducing higher-order effects first studied using Monte Carlo techniques in Ref. \cite{9}.

The false EDM of Hg is also well-understood experimentally, having been characterized using surrounding Cs magnetometers \cite{13} and to higher orders \cite{14}.

The work presented here concerns higher-order corrections beyond the first-order vertical gradient $\partial B_z/\partial z$. We show that for spherical traps, all higher-order terms contributing to Eq. (2) are identically zero. We further show that for the usual cylindrical traps used in EDM experiments, selection of quasi-spherical dimensions for the trap can reduce the higher-order contributions to false EDM of the Hg atoms.

2. Harmonic Decomposition of the Magnetic Field

Within the measurement region of EDM experiments, nonmagnetic components are used so that the field can be measured and controlled precisely. Since bound and free currents are absent from this region, the magnetic field can be written as

$$\vec{B} = -\nabla \Phi_M$$

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where $\Phi_M$ is the magnetic scalar potential. Since $\nabla \cdot \vec{B} = 0$, $\Phi_M$ obeys Laplace’s equation:
\[
\nabla^2 \Phi_M = 0. \tag{4}
\]
The general solution for a boundary-value problem can be written in spherical coordinates $(r, \theta, \phi)$ in terms of spherical harmonics $Y_{\ell m}(\theta, \phi)$ as \[15\]
\[
\Phi_M(r, \theta, \phi) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} \left[ A_{\ell m} r^\ell + B_{\ell m} r^{-(\ell+1)} \right] Y_{\ell m}(\theta, \phi) \tag{5}
\]
where $A_{\ell m}$ and $B_{\ell m}$ are sets of constants determined by the boundary conditions. If we define $r = 0$ to be the center of the trap, the requirement that $\vec{B}$ remain finite enforces $B_{\ell m} = 0$. The spherical harmonics can be written as
\[
Y_{\ell m}(\theta, \phi) = \sqrt{\frac{2\ell + 1}{4\pi} \frac{(\ell - m)!}{(\ell + m)!}} P^m_\ell(\cos \theta) e^{im\phi}, \tag{6}
\]
where $P^m_\ell$ are the associated Legendre polynomials.

The average appearing in Eq. (2) can be recast in terms of the scalar potential as
\[
\langle x B_x + y B_y \rangle = -\left\langle \left( x \frac{\partial}{\partial x} + y \frac{\partial}{\partial y} \right) \Phi_M \right\rangle \tag{7}
\]
which in terms of the spherical harmonics becomes
\[
\langle x B_x + y B_y \rangle = -\sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} A_{\ell m} \left\langle \left( x \frac{\partial}{\partial x} + y \frac{\partial}{\partial y} \right) r^\ell Y_{\ell m} \right\rangle \tag{8}
\]
where again the average is conducted over the measurement cell.

3. Application of the Wigner-Eckart Theorem

We can apply the Wigner-Eckart Theorem to this system by making an analogy to matrix elements in quantum mechanics. The differential operator in Eq. (8) $\left( x \frac{\partial}{\partial x} + y \frac{\partial}{\partial y} \right)$ can be analogized to the operator $(xp_x + yp_y)$ in quantum mechanics.

For a particle in a spherically symmetric potential, the stationary states may be written as eigenstates of the angular momentum operators
\[
L^2 |n\ell m\rangle = \hbar^2 \ell(\ell + 1) |n\ell m\rangle \tag{9}
\]
\[
L_z |n\ell m\rangle = \hbar m |n\ell m\rangle \tag{10}
\]
where $|n\ell m\rangle$ represents the stationary state and $\ell$ and $m$ are quantum numbers. The quantum number $n$ would count energy levels. In the position representation, the states factorize in spherical coordinates as

$$\langle \vec{r} | n\ell m \rangle = R_{n\ell}(r)Y_{\ell m}(\theta, \phi)$$

and thus the states are related to the spherical harmonics.

Eq. (8) therefore has a number of elements which bear a strong similarity with calculations in quantum mechanics. The average in Eq. (8) is over the EDM measurement cell volume. In order to use the Wigner-Eckart theorem, we want to make an analogy to quantum mechanical matrix elements. In order for this analogy be valid, the cell would have to be a spherical cell. In this circumstance, the term under the average is proportional to a quantum mechanical matrix element as

$$\left\langle \left( x \frac{\partial}{\partial x} + y \frac{\partial}{\partial y} \right) r^{\ell m} \right\rangle \sim \langle n'00|(x p_x + y p_y)|n\ell m\rangle$$

where we have inserted a spherically symmetric state with $\ell' = m' = 0$. Being constructed from products of vector operators, we will show that the operator $(x p_x + y p_y)$ can be written as an admixture of spherical tensors of rank 0 and 2.

The Wigner-Eckart theorem relates matrix elements of spherical tensors involving these states to Clebsch-Gordan coefficients [16]. Applied to two states $|n'\ell' m'\rangle$ and $|n\ell m\rangle$, it would read

$$\langle n'\ell' m'|T_{q}^{(k)}|n\ell m\rangle = \langle \ell k; mq|\ell k; \ell' m'\rangle \frac{\langle n'\ell'\ell|T^{(k)}|n\ell\rangle}{\sqrt{2j+1}}$$

where $T_{q}^{(k)}$ is a spherical tensor of rank $k$ with magnetic quantum number $q$. The double-bar matrix element is a reduced matrix element that does not depend on $m$, $m'$, or $q$.

The Clebsch-Gordan coefficient $\langle \ell k; mq|\ell k; \ell' m'\rangle$ can be thought of in the following more familiar way. Imagine two angular momentum operators $\vec{L}$ and $\vec{K}$ with simultaneous eigenstates $|\ell k; mq\rangle$ s.t.

$$L^2|\ell k; mq\rangle = \hbar^2 \ell (\ell + 1)|\ell k; mq\rangle$$
$$L_z|\ell k; mq\rangle = \hbar m|\ell k; mq\rangle$$
$$K^2|\ell k; mq\rangle = \hbar^2 k (k + 1)|\ell k; mq\rangle$$
$$K_z|\ell k; mq\rangle = \hbar q|\ell k; mq\rangle.$$
If we now define a new operator \( \vec{L}' = \vec{L} + \vec{K} \), the Clebsch-Gordan coefficients represent the coefficients transforming to the new basis \(|\ell k; \ell' m'\rangle\) where

\[
L^2|\ell k; mq\rangle = \hbar^2 \ell (\ell + 1)|\ell k; \ell' m'\rangle \tag{18}
\]
\[
K^2|\ell k; mq\rangle = \hbar^2 k (k + 1)|\ell k; \ell' m'\rangle \tag{19}
\]
\[
L'^2|\ell k; mq\rangle = \hbar^2 \ell' (\ell' + 1)|\ell k; \ell' m'\rangle \tag{20}
\]
\[
L'_z|\ell k; mq\rangle = \hbar m'|\ell k; \ell' m'\rangle. \tag{21}
\]

In this way, the Clebsch-Gordan coefficient is related to the addition of angular momentum of the state \(|n\ell m\rangle\) to that of the spherical tensor \(T_q^{(k)}\) and reaching the state \(|n'\ell' m'\rangle\).

The Clebsch-Gordan coefficient is only non-zero if

\[
m + q = m' \tag{22}
\]
and

\[
|\ell - k| \leq \ell' \leq \ell + k \tag{23}
\]

As mentioned earlier the operator \((xp_x + yp_y)\) can be written as an admixture of spherical tensors of rank 0 and 2. The following two spherical tensors may be constructed from the Cartesian components of the vector operators \(\vec{r}\) and \(\vec{p}\):

\[
T_0^{(0)} = -\frac{\vec{r} \cdot \vec{p}}{3} = \frac{-xp_x - yp_y - zp_z}{3} \tag{24}
\]
and

\[
T_0^{(2)} = \frac{3zp_z - \vec{r} \cdot \vec{p}}{\sqrt{6}} = \frac{-xp_x - yp_y + 2zp_z}{\sqrt{6}}. \tag{25}
\]

The operator of interest can then be written as

\[
xp_x + yp_y = -2T_0^{(0)} - \sqrt{\frac{2}{3}} T_0^{(2)} \tag{26}
\]
which are spherical tensors of rank 0 and 2 with magnetic quantum number \(q = 0\).

Finally we note that because these operators are even under the parity transformation, they may only link states of the same parity.

We can now apply Eqs. (22) and (23) to Eq. (8) to find that only terms in the sums with \(m = 0\) and \(\ell = 0, 1, 2\) will contribute. Since the operator in Eq. (8) is a differential operator, the term with \(\ell = 0\) also cannot contribute.
The term with $\ell = 1$ is ruled out by the parity selection rule. We are then left with only one non-zero term arising from the harmonic decomposition of the field:

$$\langle xB_x + yB_y \rangle = -A_{20} \left\langle \left( x \frac{\partial}{\partial x} + y \frac{\partial}{\partial y} \right) r^2 Y_{20} \right\rangle$$

(27)

where the average can be readily carried out since $r^2 Y_{20}$ is a polynomial of degree 2 in Cartesian coordinates.

The suppression of the higher-order terms can be derived in a number of different ways, for example, by the commutation relations of the operators, by integration by parts, and/or by using the known properties of the spherical harmonics/Legendre polynomials. We used this to check the result in a number of ways. This included (a) explicit integration of the $m = 0$ terms for particular higher $\ell$, demonstrating they were all zero, and (b) integration by parts (shifting the derivatives to the left) and then using the orthogonality of the Legendre polynomials to demonstrate that only $\ell = 2$ is permitted.

4. Cylindrical Trap and Suppression of Higher Orders

A requirement of the calculation of the preceding section is that the EDM cell be spherical, which is not an attractive option for EDM experiments. A more typical geometry is a cylindrically symmetric geometry. In this case, it can still readily be demonstrated that only $m = 0$ terms contribute in the second sum in Eq. (27). Switching to cylindrical $(\rho, \phi, z)$ coordinates, the differential operator becomes $x\frac{\partial}{\partial x} + y\frac{\partial}{\partial y} = \rho \frac{\partial}{\partial \rho}$ and since $Y_{\ell m} \sim e^{i m \phi}$ these terms would average to zero unless $m = 0$. Furthermore, terms with $\ell =$ odd are odd in $z$. Thus if the cylindrical cell is centered on $z = 0$, these terms will also average to zero. We are then left with

$$\langle xB_x + yB_y \rangle = - \sum_{\ell=2,4,6,...}^{\infty} A_{\ell 0} \left\langle \left( \rho \frac{\partial}{\partial \rho} \right) r^\ell Y_{\ell 0} \right\rangle$$

(28)

The purpose of this section is to demonstrate that $\ell = 2$ dominates, that for a certain choice of cell dimensions the $\ell = 4$ term can be zeroed, and that for this selection the $\ell = 6$ term can be reduced compared to the typical cell geometry used for the ILL nEDM experiment.
In order to more easily compare to Ref. [12], we introduce a more convenient normalization based on the Legendre polynomials $P_\ell(\cos \theta)$ as

$$\langle xB_x + yB_y \rangle = + \sum_{\ell=2,4,6,...}^\infty \frac{g_{(\ell-1)0}}{\ell} \left\langle \left(\rho \frac{\partial}{\partial \rho} \right) r^\ell P_\ell(\cos \theta) \right\rangle,$$

where the $g_{(\ell-1)0}$ are related by constants (and a sign change) to the $A_{\ell0}$ in Eq. (28). This normalization is preferred because it ensures that the $m = 0$ components of $B_z = -\frac{\partial \Phi_M}{\partial z}$ are polynomials which contain $g_\ell z^\ell$ with $\ell = \text{odd}$. This guarantees, for example, that the leading-order term in the vertical gradient is $\frac{\partial B_z}{\partial z} = g_{10}$, allowing us to easily identify $g_{10}$ itself as the first-order uniform gradient term of Ref. [12].

In general $\frac{1}{r^\ell} P_\ell(\cos \theta)$, when expressed in cylindrical coordinates, is a polynomial in $\rho$ and $z$. For the terms of interest, the polynomials are

$$\frac{1}{2} r^2 P_2(\cos \theta) = \frac{1}{4} (2z^2 - \rho^2)$$

$$\frac{1}{4} r^4 P_4(\cos \theta) = \frac{1}{32} (8z^4 - 24z^2\rho^2 + 3\rho^4), \text{ and (31)}$$

$$\frac{1}{6} r^6 P_6(\cos \theta) = \frac{1}{96} (16z^6 - 120z^4\rho^2 + 90z^2\rho^4 - 5\rho^6).$$

We define the origin of coordinates to lie at the center of a measurement cell with height $H$ and radius $R$. Carrying out the average over the cell, the first three non-zero terms of Eq. (29) become

$$\langle xB_x + yB_y \rangle = -\frac{R^2}{4} \left[ g_{10} + g_{30} \left( \frac{H^2}{4} - \frac{R^2}{2} \right) + g_{50} \left( \frac{H^4}{16} - \frac{5R^2H^2}{12} + \frac{5R^4}{16} \right) \right].$$

The expression for $d_{f,Hk}$ then becomes

$$d_{f,Hk} = \frac{\hbar c^2 R^2}{8e^2} \left[ g_{10} + g_{30} \left( \frac{H^2}{4} - \frac{R^2}{2} \right) + g_{50} \left( \frac{H^4}{16} - \frac{5R^2H^2}{12} + \frac{5R^4}{16} \right) \right].$$

The $g_{10}$ term is in agreement with Ref. [12], and the other two terms have also been derived previously [17]. We now analyze these next two terms.
An immediate observation is that the $g_{30}$ term can be set to zero if the measurement volume is quasi-spherical, i.e., for

$$H = \sqrt{2}R$$  \hspace{1cm} (35)

If this selection is made, then the factor in the $g_{50}$ term becomes

$$\frac{H^4}{16} - \frac{5R^2H^2}{12} + \frac{5R^4}{16} = -\frac{13}{48}R^4$$  \hspace{1cm} (36)

The question now becomes whether this is a reasonable geometry for an nEDM experiment.

5. Discussion and Caveats

5.1. ILL/PSI nEDM experiment geometry

For the ILL/PSI nEDM experiment, the cell dimensions are $R = 23.5$ cm and $H = 12$ cm, and so Eq. (35) is clearly not obeyed. Using the ILL/PSI geometry, the $g_{30}$ term is

$$\frac{H^2}{4} - \frac{R^2}{2} = -240 \text{ cm}^2$$  \hspace{1cm} (37)

and the $g_{50}$ term is

$$\frac{H^4}{16} - \frac{5R^2H^2}{12} + \frac{5R^4}{16} = 6.35 \times 10^4 \text{ cm}^4$$  \hspace{1cm} (38)

The false EDM then becomes

$$d_{f,Hg} = 1.15 \times 10^{-27} \left( g_{10} - g_{30} \cdot 240 + g_{50} \cdot 6.35 \times 10^4 \right) \text{ e} \cdot \text{cm}$$  \hspace{1cm} (39)

where the $g_{\ell 0}$ are expressed in units of pT/cm$^\ell$. The $g_{30}$ term was analyzed and measured experimentally in Ref. [14] and found to be in agreement with expectation.

5.2. Quasi-spherical geometry

In order to compare this result for a realistic experimental geometry to our suggestion of a quasi-spherical cell, we considered several possibilities. One possibility was to keep the radius of the cell the same as for the ILL experiment. This would mean that the size of the pre-factor in Eq. (39) would
be equal. In this case, the height of the cell would necessarily be increased to 
\[ H = \sqrt{2} \cdot 23.5 \text{ cm} \approx 33 \text{ cm} \]. The negative aspect of this suggestion is that a high-voltage nearly a factor of three larger would need to be sustained across the electrodes in order to keep the electric field in the experiment the same. Since the electric field is strongly related to the statistical precision of the experiment, it is not clear that this is a realistic compromise.

Another possibility was to keep the height of the cell the same, which would alleviate any high-voltage issues. In this case the cell radius would need to shrink to 
\[ R = 12 \text{ cm}/\sqrt{2} \approx 8.5 \text{ cm} \]. The negative aspect here is that the volume of the experiment would shrink by a factor of almost eight. In general, the statistical precision of neutron EDM experiments is driven by the neutron density achievable in the cell, and this choice would mean that the number of neutrons loaded into the cell would be reduced by the same factor.

We therefore decided to suggest keeping approximately the same EDM cell volume, setting 
\[ R = 17 \text{ cm} \] and 
\[ H = \sqrt{2} \cdot 17 \text{ cm} \approx 24 \text{ cm} \]. For this choice, the \( \ell = 4 \) term is zero and the expression for the false EDM becomes

\[ d_{f,\text{Hg}} = 6.1 \times 10^{-28} (g_{10} - g_{50} \cdot 2.26 \times 10^4) \text{ e \cdot cm}. \quad (40) \]

For this particular choice, the overall false EDM is smaller, and furthermore the \( g_{50} \) term is reduced relative to the \( g_{10} \) when compared to the usual ILL/PSI geometry.

5.3. Further notes

It is unclear whether this geometry would be realizable experimentally. The main point of the discussion is that the geometry affects the size of the \( g_{30} \) term for the Hg comagnetometer false EDM, and the closer to spherical the less the higher-order terms tend to contribute.

It is also important to consider the overall false-EDM correction scheme used in these experiments. While the false EDM of the mercury comagnetometer tends to dominate the correction, it is by no means the only quantity affected by the higher multipoles of the magnetic field. The neutrons' false EDM, the spin relaxation times of the species, and the electric field independent spin-precession frequencies are also affected by the inhomogeneity.

In particular, the spin-precession frequency ratio (neutrons to Hg) was used in the ILL nEDM experiment to sense the vertical gradient \( g_{10} \) and hence to correct the leading terms in the false EDM's \[ [6, 7] \]. This works because
the ultracold neutrons tend to preferentially sample the bottom of the cell, resulting in an average height difference against gravity. When considered to higher order, other terms contribute to this correction scheme. So while the false EDM of the Hg atoms could be suppressed by a quasi-spherical cell, it is unclear that this is the most important factor in experiment design. Even if the EDM cell could be made spherical, the neutrons still preferentially sample the lower portion of the sphere.

6. Conclusion

We have demonstrated that a spherical EDM measurement cell would reduce the false EDM of Hg to depend on a single multipole of the magnetic field. A cylindrical cell that is quasi-spherical with $H = \sqrt{2}R$ also tends to reduce the higher multipole contributions. Both suggestions are rather difficult to realize experimentally, and do not capture the full correction scheme used in the ILL/PSI nEDM experiment, which must take more into account than simply the false EDM of the Hg comagnetometer. Nonetheless, we think this is an interesting result which points out the dependence of the false EDM contributions on the measurement cell geometry.

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