Search for an Electric Dipole Moment in $^{129}$Xe Atom with Nuclear Spin Oscillator Technique

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Abstract. Development of a nuclear spin oscillator intended for the experimental search for an electric dipole moment in $^{129}$Xe atom is reported. The spin oscillation is brought about by polarizing $^{129}$Xe nuclear spins by spin exchange with optically polarized Rb atoms, detecting the spin precession through the transmission of a circularly polarized laser light, and applying a feedback field according to the detected spins. Until present, a frequency precision, in a measurement time of $T = 45,000$ s, of $\delta \nu = 5.2$ nHz has been achieved.

1. Introduction

Constituents of the matter world and their interactions known from laboratory experiments are surprisingly well described by the standard model (SM) of elementary particles, major part of which was established already in 1980's. Presently, however, most of us cannot avoid feeling dissatisfaction with this model, since there are numbers of questions the model is unable to answer, such as why quarks and leptons exist in three generations, why there are three kinds of interactions, and what is the 95 % of energy content of our universe which seemingly exist but cannot be explained by the SM [1].

The permanent electric dipole moment (EDM) serves as an exclusively suited observable to detect flaws, if any, of the standard model. This prominent role allotted to the EDM stems from the fact that this flavor-diagonal observable violates T and hence CP invariances. Thus, EDMs of particles are predicted to be almost undetectably small in the SM, while most of theories proposed beyond the SM predict their sizes in the regions reachable in not very distant future.

EDMs are being searched for in various sites: in neutron, paramagnetic atoms, diamagnetic atoms, molecules, and even in charged particles such as muon, proton, deuteron and ions. In the present report we describe the present status of our experimental attempt to detect an EDM in the diamagnetic atom $^{129}$Xe.

2. EDM in a diamagnetic atom

Total angular momentum (the atomic spin) of a diamagnetic atom with a singlet cloud of atomic electrons comes solely from its nuclear spin $I$. Since the electron spins are totally paired off, EDMs of electrons give no contribution to the atomic EDM, and thus the EDM of a diamagnetic atom is induced solely from the nucleus. While the Schiff's theorem [2] prohibits direct contribution of a nuclear EDM
to the atomic EDM, there remains a route in which a CP-violating higher-order moment of nucleus, the Schiff moment S as defined shortly below, can give rise to the atomic EDM.

2.1 Atomic EDM and the Schiff moment
Consider a nucleus of point size and charge Ze but with no EDM, in a neutral atom. The nucleus locates itself at the center of the atom, \( r_{\text{nucl}} = 0 \), because the electrostatic potential takes minimum there. Once an EDM \( d_{\text{nucl}} \) is attached to it, the nucleus shifts its position by a displacement \( \Delta r_{\text{nucl}} = -d_{\text{nucl}}/Ze \) so that its effective position of charge

\[
\rho_{\text{ch}}(r) \mathbf{e} = \frac{1}{Ze} \int \rho_{\text{ch}}(r - r_{\text{nucl}}) \mathbf{r} \, d^3 r
\]

\[
= \frac{1}{Ze} \int \left[ Ze \delta(r - r_{\text{nucl}}) + q \delta(r - r_{\text{nucl}} - \frac{1}{2} a) - q \delta(r - r_{\text{nucl}} + \frac{1}{2} a) \right] \mathbf{r} \, d^3 r
\]

\[
= r_{\text{nucl}} + \frac{1}{Ze} d_{\text{nucl}}
\]

becomes zero (the center of the atom). Here \( r_{\text{nucl}} \), \( d_{\text{nucl}} = qa \), and \( \rho_{\text{ch}}(r) \) denote the position, EDM, and charge distribution of the nucleus, respectively. Since the effective position of charge remains at the center anyhow, the atom does not "know" that the nucleus possesses an EDM \( d_{\text{nucl}} \) (Schiff's theorem). Now we take a finite nuclear size into account. The effect of having \( d_{\text{nucl}} \) is again simply a position shift \( \Delta r_{\text{nucl}} = -d_{\text{nucl}}/Ze \), but in this case atomic electrons "feel" that the electrostatic interaction with the nucleus \( \Phi(R) \) has changed [3]:

\[
\Delta \Phi(R) \equiv \int \rho_{\text{ch}}(r) \mathbf{r} \, d^3 r - \int \rho_{\text{ch}}(r - \Delta r_{\text{nucl}}) \mathbf{r} \, d^3 r = \frac{1}{Ze} \left( d_{\text{nucl}} \cdot \nabla_R \right) \int \rho_{\text{ch}}(r) \, d^3 r
\]

where \( R \) is the position of electron. Taking the lowest order term (setting an octupole term aside), Eq. (2) is equivalent to an electric field

\[
E_{\text{Schiff}} \approx \frac{15}{R_{\text{nucl}}} S
\]

acting inside the nuclear volume to the electrons [3], where \( R_{\text{nucl}} \) is the nuclear radius, and the Schiff moment \( S \) is defined as

\[
S = \frac{1}{10} \int_{\text{nucleus}} \rho_{\text{ch}}(r) r \left( r^2 - \frac{5}{3} \left\langle r^2 \right\rangle_{\text{ch}} \right) \, d^3 r
\]

and \( \left\langle r^2 \right\rangle_{\text{ch}} \) is the mean square charge radius of the nucleus. Electric field (2) polarizes the cloud of atomic electrons, leading to a finite EDM of atom

\[
d_{\text{atom}} = \alpha S,
\]

where the constant of proportionality \( \alpha \) is obtained from atomic physics calculations [4,5] for individual atoms.

2.2 Schiff moment from fundamental processes
The Schiff moment is induced by CP violating processes in the nucleus. In a nuclear physics terminology, the relevant processes would be the mixing of small CP violating amplitude into the nuclear ground state wave function, and CP violating nucleon-nucleon interactions that produce the mixings. Thus, nuclear structure calculations give the Schiff moment \( S \) as a function of isoscaler, isovector, and isotensor components of CP violating \( \pi NN \) coupling constants \( \bar{g}^{(0)}_{\pi NN}, \bar{g}^{(1)}_{\pi NN}, \) and \( \bar{g}^{(2)}_{\pi NN} \) through a perturbation expression

\[
S \approx \sum_i \frac{\langle \text{g.s.}|i\rangle M_{\text{CPV}} \langle i|\text{g.s.} \rangle}{E_0 - E_i}
\]

and CP violating one-pion exchange \( NN \) interactions

\[
V_{\text{CPV}} = \frac{g_{\pi NN}}{8\pi m_N} \sum_i \left[ -\frac{1}{2} \bar{g}^{(0)}_{\pi NN} (r_i \cdot r_j) + \bar{g}^{(1)}_{\pi NN} (3r_i \cdot r_j - r_i \cdot \tau_j) \right] \left( \sigma_i - \sigma_j \right) + \bar{g}^{(2)}_{\pi NN} (r_i - r_j) \left( \sigma_i + \sigma_j \right) \cdot \nabla_i \frac{\exp(-m_N|\mathbf{r}_i - \mathbf{r}_j|)}{|\mathbf{r}_i - \mathbf{r}_j|},
\]

while particle physics analyses [6,7] provide the latter constants in terms of quark chromoelectric dipole moment \( \bar{d}_u, \bar{d}_d, \) and \( \bar{d}_s \) of up, down, and strange quarks. Analyses in Ref. [6] give (assuming the Peccei-Quinn symmetry [8] and inserting numerical values therein) \( \bar{g}^{(0)}_{\pi NN} \approx -1.02 (\bar{d}_u + \bar{d}_d) \), \( \bar{g}^{(1)}_{\pi NN} \approx -9.2 (\bar{d}_u - \bar{d}_d) \) and \( \bar{g}^{(2)}_{\pi NN} \approx 0 \), where \( \bar{d}_q \) are in GeV.

2.3 Recent status

EDMs of diamagnetic atoms have been studied for \(^{129}\text{Xe}\) and \(^{199}\text{Hg}\). Also, there are experimental plans for radioactive \(^{209}\text{Rn}\) and \(^{225}\text{Ra}\) isotopes [9,10], in which enhancements of EDM due to octupole deformation are actively discussed [11]. The most studied theoretically is the case of \(^{199}\text{Hg}\), on which the recent experiment [12] has placed very stringent limit, \( d(\text{Hg}) < 3.1 \times 10^{-29} \text{ cm} \). Atomic physics calculation [13] gives \( d(\text{Hg}) = -2.8 \times 10^{-17} \text{ S in e fm}^3 \) cm. As for the Schiff moment \( S(\text{Hg}) \), the earlier calculation [5] provided the relation

\[
S(\text{Hg}) = g_{\pi NN} \left( -0.0010 \bar{g}^{(0)}_{\pi NN} + 0.074 \bar{g}^{(1)}_{\pi NN} + 0.018 \bar{g}^{(2)}_{\pi NN} \right) \text{ efm}^3.
\]

The above experimental limit thus has been supposed to give an upper limit on a combination of the quark chromoelectric dipole moment, \( \bar{d}_u - \bar{d}_d < 6 \times 10^{-27} \text{ cm} \). Very recently, however, fully self-consistent calculations [14] of the Schiff moment has given a result which turns out quite different from the earlier one:

\[
S^{\text{New}}(\text{Hg}) = g_{\pi NN} \left( 0.016 \bar{g}^{(0)}_{\pi NN} - 0.006 \bar{g}^{(1)}_{\pi NN} + 0.019 \bar{g}^{(2)}_{\pi NN} \right) \text{ efm}^3.
\]

Note that the previously perceived dominance of the isovector component \( \bar{g}^{(1)}_{\pi NN} \) has now disappeared, weakening the above obtained constraint on \( \bar{d}_u - \bar{d}_d \) by an order of magnitude, and even its sign has changed.

In the case of \(^{129}\text{Xe}\) atom, atomic physics calculations [13,15] give \( d(\text{Xe}) = 0.38 \times 10^{-17} \text{ S in e fm}^3 \) cm, whereas the relation of \( S(\text{Xe}) \) to the CP violating \( \pi NN \) coupling constants \( \bar{g}^{(0)}_{\pi NN}, \bar{g}^{(1)}_{\pi NN}, \) and \( \bar{g}^{(2)}_{\pi NN} \)
and $B_{x^2NN}$ was calculated more than two decades ago with a rather simple account of CP violating $NN$ interactions and core polarization effects. Thus, new calculation of $S(129\text{Xe})$ is strongly awaited.

3. Experiment

We aim to measure the atomic EDM of $^{129}\text{Xe}$. $^{129}\text{Xe}$ is a stable nucleus so that a macroscopic number of particles can be subjected to the measurement. It has spin $I = \frac{1}{2}$ and exists in a form of a rare gas, and there is a well established method to obtain a high degree of spin polarization by using the spin-exchange with optically pumped alkali atoms. Furthermore, the $^{129}\text{Xe}$ spins show quite long transverse relaxation times (spin coherent times) [16]. These advantageous features of $^{129}\text{Xe}$ have made us choose it as a physical system where an EDM is looked for, even with supposed disadvantages that it is not as heavy as the other candidates $^{199}\text{Hg}$, $^{209}\text{Rn}$, and $^{225}\text{Ra}$ and there is no enhancement due to octupole collectivity expected.

3.1 EDM measurement with a nuclear spin oscillator

The detection of an EDM is, in most cases, nothing other than a detection of extremely small change in frequency of the spin precession upon a reversal of the applied electric field. The key issue for a sensitive EDM detection is therefore a high precision determination of frequency with well stabilized (or monitored) strength of the applied magnetic field. Thus, the EDM $d$ is deduced from the observed frequency change $\Delta \nu = \nu_+ - \nu_-$, as

$$d = \frac{h\Delta \nu}{4E}. \quad (10)$$

Usually the observation time for a single shot of continued spin precession is limited by the transverse spin relaxation time $T_2$. The measurement is made by repeating $n$ times the shot of precession for a

![Figure 1. Setup for the spin oscillator experiment.](image-url)
time period $T$, having the total measurement time to be $T_{\text{total}} = nT$, with $T \sim T_2$. The frequency precision for such a measurement is given by

$$\delta \nu = \delta \nu_{\text{single}} / \sqrt{n} \propto (T_{\text{total}})^{-1/2}. \quad (11)$$

If, on the other hand, the spin coherence time is elongated up to the full measurement time $T_{\text{total}}$, the precision $\delta \nu$ should go as

$$\delta \nu \propto (T_{\text{total}})^{-3/2}, \quad (12)$$

enabling a much faster improvement in $\delta \nu$. In order to realize such an elongation of the spin precession time, we employ the technique of a nuclear spin oscillator with feedback by optical spin detection [17].

Figure 1 shows the experimental setup for the spin oscillator experiment. A $^{129}$Xe gas contained in a 18 mm diameter spherical glass cell together with a small amount of Rb vapor is illuminated by a circularly polarized laser light of 794.7 nm wavelength. By spin exchange with the optically pumped Rb atoms, the $^{129}$Xe spins are polarized in the direction of a static magnetic field $B_0 = 30.6$ mG. Once the $^{129}$Xe spins are tilted, they start precession about $B_0$, which is observed through transmission of a probe laser light traversing the cell in the direction perpendicular to $B_0$. Thus, the precession signal obtained at a Si photodiode is lock-in amplified, digitalized, processed, and used for generating a feedback field $B_{FB}$ applied on the $^{129}$Xe cell. The $B_{FB}$ field is so arranged that it is kept 90 degree in advance from the transverse component of the precessing $^{129}$Xe spins, thus acting to increase transverse component of $^{129}$Xe spins to maintain the precession. Detailed explanation of the operation of the spin oscillator is given in Ref. [17].

3.2 Frequency characteristics

After succeeding in the realization of steady state oscillation, detailed studies of operation

![Graph 1: Frequency precision attained by operating the spin maser for time periods T. Blue squares and red circles represent, respectively, data before and after the current source was replaced by a new one.

Fig. 2: Frequency precision attained by operating the spin maser for time periods $T$. Blue squares and red circles represent, respectively, data before and after the current source was replaced by a new one.

![Graph 2: Time variations of the maser frequency (a), and the environmental field (b). A clear correlation between (a) and (b) is seen.

Fig. 3: Time variations of the maser frequency (a), and the environmental field (b). A clear correlation between (a) and (b) is seen.
characteristics such as frequency and amplitude stabilities, their dependences on the cell temperature, pumping laser power, environmental field, and strength of the $B_0$ and $B_{FB}$ fields were conducted. In particular, the frequency drifts and fluctuations were found to occur from several different sources. Some part of drifts appears to be increased regularly in the midday when the ambient temperature raises, while some part shows a clear dependence on drifts in the current of the solenoid coil for applying $B_0$. Figure 2 shows the frequency precision, $\delta \nu$, attained in long-time operations of the spin oscillator. The blue squares represent the data in the measurements before the current source for the $B_0$ solenoid was replaced by a new one. Although the expected improvement of frequency precision, $\delta \nu \propto (T_{\text{total}})^{-3/2}$ was observed in the region up to 1000 s, it became flattened for $T = 1,000 - 5,000$ s, and then started to be improved faster. In about 30,000 s, $\delta \nu$ reached the best value of $\delta \nu = 9.3$ nHz which corresponded to an EDM precision of $9 \times 10^{-28}$ cm provided that an electric field of $E = \pm 10$ kV/cm was applied. Detailed inspection revealed that in the region of time $T = 1,000 - 5,000$ s frequency drifts were largest. Since there seemed several sources of drifts and fluctuations as stated earlier, we made several measurements using different setups: i) the current source was replaced with a new one which was equipped with specially designed feedback stabilizer, ii) a lower cell temperature of $50^\circ$C was chosen at which Rb density in the cell was calculated to be low, and iii) the frequency was corrected for the drift in the environmental field measured with a flux gate magnetometer. The result is shown in Fig. 2 by the red filled circles. Now $\delta \nu$ remains almost in the $(T_{\text{total}})^{-3/2}$ dependence until 10,000 s, and reached an improved limit of $\delta \nu = 5.2$ nHz corresponding to a $5 \times 10^{-28}$ cm EDM precision. In Fig. 3, the observed frequency is plotted as a function of time, in comparison with the simultaneously monitored environmental field $B_{\text{env}}$. A close resemblance between the behaviors of $\delta \nu$ and $B_{\text{env}}$ is clearly seen. We are now preparing for an active compensation of the environmental field.

4. Outlook

After spending a rather long term of "groping in the dark" to find out sources of frequency drifts and fluctuations, we have reached a point to establish the final setup for the EDM search with the best tuned nuclear spin oscillator. To this end, we are preparing for cells with electrodes for the $E$ field application, a reinforced pumping laser, a fluid circulation system for the cell temperature stabilization, and high precision magnetometry using nonlinear magneto-optical rotation [18].

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