Effects of confinement on permanent electric-dipole moment of Xe atoms in liquid Xe

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Most extensions of the standard model of elementary particles, e.g., supersymmetry, naturally produce permanent electric dipole moments (EDM) of atoms and molecules [1] that are comparable to or larger than present limits (see, e.g., a popular review [2]). For example, the most accurate to date determination of atomic EDM of $^{129}$Xe [M.V. Romalis and M.P. Ledbetter, Phys. Rev. Lett. 87, 067601 (2001)] may significantly improve present limits on the EDMs. To interpret experimental data in terms of CP-violating sources, one must relate measured atomic EDM to various model interactions via electronic-structure calculations. Here we study density dependence of atomic EDMs. The analysis is carried out in the framework of the cell model of the liquid coupled with relativistic electronic-structure calculations. We find that compared to an isolated atom, the EDM of an atom of liquid Xe is suppressed by about 40%.

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magnitude better than the present limits.

Sources of atomic EDM — The conventional atomic Hamiltonian $H_0$ among other symmetries is invariant with respect to space-reflection (P) and time reversal (T). Therefore, on very general grounds, an expectation value of the electric dipole operator $\langle \mathbf{D} \rangle = -\sum_i r_i$ in a non-degenerate atomic state $|\Psi_0\rangle$ vanishes. The tiny CP-violating interactions, here generically denoted as $H_{\text{CP}} = \sum_i h_{\text{CP}}(r_i)$, break the symmetry of the atom and induce a correction to the electronic state $|\Psi\rangle = |\Psi_0\rangle + |\delta\Psi\rangle$. To the lowest order

$$|\delta\Psi\rangle = \sum_k |\Psi_k\rangle \frac{\langle \Psi_k | H_{\text{CP}} | \Psi_0 \rangle}{E_k - E_0},$$

where $E_k$ and $|\Psi_k\rangle$ are eigenvalues and eigenfunctions of $H_0$. Due to selection rules, the $|\delta\Psi\rangle$ admixture has parity opposite to the one of the reference state $|\Psi_0\rangle$. Because of this opposite-parity admixture the atom acquires a permanent EDM

$$d = \langle \bar{\Psi} | \mathbf{D} | \Psi \rangle = 2 \langle \Psi_0 | \mathbf{D} | \delta\Psi \rangle.$$  

Now we specify particular forms of $H_{\text{CP}}$. An analysis [5] shows that for diamagnetic atoms, such as Xe, the EDM predominantly arises due to P,T-odd semileptonic interaction $H_{\text{TN}}$ between electrons and nucleons and also due to interaction $H_{\text{SM}}$ of electrons with the so-called nuclear Schiff moment $\mathbf{D}_N$. Smaller atomic EDM is generated by intrinsic EDM of electrons and

the neighboring atoms. Imposing proper boundary conditions at the cavity radius, first we solve the Dirac-Hartree-Fock (DHF) equations and then employ the relativistic random-phase approximation (RRPA) to account for correlations. To the best of our knowledge, here we report the first ab initio relativistic calculations of properties of a liquid. We find that compared to the EDM of an isolated atom, the resulting EDM of an atom of liquid Xe is suppressed by about 40%. Thus if the experiment with liquid Xe is carried out with the anticipated sensitivity, we expect that the inferred constraints on possible sources of CP-violation would be indeed several orders of magnitude better than the present limits.

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we will not consider this mechanism here. Atomic units 
$|e| = \hbar = m_e = 4\pi\varepsilon_0 \equiv 1$ are used throughout.

Explicitly, the effective P,T-odd semileptonic interaction
Hamiltonian may be represented as

$$h_{\text{TN}}(r_e) = \sqrt{2}G_F \sigma_N \cdot \langle i\gamma_0\gamma_5 \sigma \rangle \rho_N (r_e) \cdot \hat{r}.$$  \hspace{1cm} (3)

Here subscripts $e$ and $N$ distinguish between operators
acting in the space of electronic and nuclear coordinates
respectively. $C_{\text{TN}}$ is a coupling constant to be determined
from an interpretation of EDM measurements and to be
compared with theoretical model-dependent predictions.
Due to averaging over nuclear degrees of freedom, this
interaction depends on nuclear density distribution $\rho_N(r)$.
In the following, we approximate $\rho_N(r)$ as a Fermi
distribution $\rho_N(r) = \rho_0/(1 + \exp[(r - c)/a])$ with $c = 5.6315$
fm and $a = 0.52$ fm. Finally, $G_F \approx 2.22254 \times 10^{-14}$ a.u.
is the Fermi constant.

The interaction of an electron with the nuclear Schiff
moment $S$ has the form

$$h_{\text{SM}}(r_e) = \frac{3}{B_4} \rho_N (r_e) (r_e \cdot \hat{S}),$$  \hspace{1cm} (4)

where $B_4 = \int_0^\infty r^4 \rho_N (r) \, dr$ is the fourth-order moment
of the nuclear distribution. The Schiff moment character-
izes a difference between charge and EDM distributions
inside the nucleus. It depends on a number of important
CP-violating parameters enumerated in the introduction.

Finally, we emphasize that both $h_{\text{TN}}$ and $h_{\text{SM}}$
are contact interactions. They occur when an electron
penetrates the nucleus. The electron speed at the nucleus is
approximately $aZc \approx \frac{1}{2} c (Z = 54)$, i.e., a fully relativis-
tic description of electronic motion is important in this
problem.

Cell model of liquid xenon — Here we employ a simple
cell model (see references therein) to estimate the effects of the environment on permanent EDM of a
given atom. According to the cell model, we confine an
atom to a spherical cavity of radius

$$R_{\text{cav}} = \left( \frac{3}{4\pi} n \right)^{1/3},$$  \hspace{1cm} (5)

$n$ being the number density of the sample. For a densi-
ty of liquid Xe of 500 amagat, $R_{\text{cav}} \approx 4.9$ bohr.
In non-relativistic calculations periodicity requires that
the normal component of the gradient of electronic wave-
function vanishes at the surface of the cell (see, e.g.,

$$\frac{\partial \Psi}{\partial r}(R_{\text{cav}}) = 0.$$  \hspace{1cm} (6)

Before proceeding with a technical question of imple-
menting these boundary conditions in relativistic calcula-
tions, we notice that the cell model implicitly incorporates
an average polarization interaction with the media.
Indeed, the Hamiltonian of an atom placed in the
liquid in addition to the conventional atomic Hamiltoni-
nan $H_0$ includes interaction of electrons with the rest of
the atoms in the media. This interaction is dominated
by polarization potential. An important point is that
the averaged polarization interaction can be expressed as

$$V_p = -1/2(1 - \epsilon^{-1})R_{\text{cav}}^{-1},$$

where $\epsilon$ is the dielectric constant of the media.
This interaction does not depend on electronic coordinate — it is just an additive constant which does not affect calculations of EDM. Thus we may approximate the total Hamiltonian with the traditional atomic Hamiltonian $H_0$.

Further, the spherical symmetry of the cell allows us
to employ traditional methods of atomic structure.
The only modification is due to boundary conditions.
However, in relativistic calculations, a special care should
be taken when implementing this boundary condition.
Indeed, the Dirac bi-spinor may be represented as

$$\varphi_{\kappa\kappa'}(r) = \frac{1}{r} \left( P_{\kappa\kappa'}(r) \frac{\Omega_{\kappa\kappa'}(\hat{r})}{\epsilon} + i Q_{\kappa\kappa'}(r) \frac{\Omega_{\kappa\kappa'}(\hat{r})}{\epsilon} \right),$$  \hspace{1cm} (7)

where $P$ and $Q$ are the large and small radial components
respectively and $\Omega$ is the spherical spinor. The angular
quantum number $\kappa = (l - j) (2j + 1)$. The nonrelativis-
tic boundary condition applied directly to the above
ansatz would lead to two separate constraints on $P$ and $Q$.
This over-specifies boundary conditions and leads to
the Klein paradox.

A possible relativistic generalization of the boundary condition is

$$\frac{d}{dr} P_{\kappa\kappa'}(R_{\text{cav}}) = \frac{d}{dr} Q_{\kappa\kappa'}(R_{\text{cav}}).$$  \hspace{1cm} (8)

Since in the non-relativistic limit the small component
vanishes, this generalization subsumes Eq. (8). Due
to a semi-qualitative nature of our calculations, here we
have chosen to use simpler (MIT bag model) boundary
condition

$$P_{\kappa\kappa'}(R_{\text{cav}}) = Q_{\kappa\kappa'}(R_{\text{cav}}).$$  \hspace{1cm} (9)

Non-relativistically it corresponds to impenetrable cavity
surface. Compared to this condition, the periodic boundary conditions are “softer”, i.e., they modify the free-atom wavefunctions less significantly; we expect that our use of Eq. (8) would somewhat overestimate the effects of confinement in the liquid.

Atom in a cavity: DHF and RRPA solutions — To re-
iterate the discussion so far, within the cell model, the
complex liquid-structure problem is reduced to solving
atomic many-body Dirac equation with boundary condi-
tions. The atomic-structure analysis is simplified by
the fact that Xe is a closed-shell atom. Below we self-
consistently solve the DHF equations inside the cavity.
Then we employ more sophisticated RRPA.

At the DHF level, the atomic wavefunction is repre-
sented by the Slater determinant composed of occupied
(core) orbitals $\varphi_a$. These orbitals are determined from a
set of DHF equations

$$\left( c(\alpha \cdot \hat{p}) + \beta \epsilon + V_{\text{nuc}} + V_{\text{DHF}} \right) \varphi_a = \varepsilon_a \varphi_a,$$  \hspace{1cm} (10)
where \( V_{\text{nuc}} \) is a potential of the Coulomb interaction with a finite-size nucleus of charge density \( \rho_N(r) \) and \( V_{\text{DHF}} \) is non-local self-consistent DHF potential. The DHF potential depends on all the core orbitals. Similar equations may be written for (virtual) excited orbitals \( \phi_m \).

We solved the DHF equations in the cavity using a B-spline basis set technique by Johnson et al. [14]. This technique is based on the Galerkin method: the DHF equations are expressed in terms of an extremum of an action integral \( S_A \). The boundary conditions are incorporated in the \( S_A \) as well. Further, the action integral is expanded in terms of a finite set of basis functions (B-splines). Minimization of such \( S_A \) with respect to expansion coefficients reduces solving integro-differential DHF equations to solving symmetric generalized eigenvalue problem of linear algebra. The resulting set of basis functions is finite and can be considered as numerically complete. In a typical calculation we used a set of basis functions expanded over 100 B-splines.

Given a numerically complete set of DHF eigenfunctions \( \{ \phi_i \} \), the permanent atomic EDM, Eq. (2), may be expressed as

\[
d_{\text{DHF}}^m = 2 \sum_{n,a} \langle \phi_n|\varphi_m\rangle\langle \varphi_m|h_{\text{CP}}|\phi_a\rangle \frac{\varepsilon_m - \varepsilon_a}{\varepsilon_m},
\]

where \( a \) runs over occupied and \( m \) over virtual orbitals. Here \( h_{\text{CP}} \) is either a semileptonic interaction, Eq. (3), or an interaction with the nuclear Schiff moment, Eq. (4). An additional peculiarity related to the Dirac equation is an appearance of negative energy states \( \varepsilon_m < -m_e c^2 \) in the summation over intermediate states in Eq. (11). We have verified that these states introduce a completely negligible correction to the computed EDMS.

To improve upon the DHF approximation, we have also computed EDMS using RRPA method [15]. This approximation describes a dynamic linear response of an atom to a perturbing one-body interaction (e.g., \( H_{\text{CP}} \)). The perturbation modifies core orbitals thus changing the DHF potential. This modification of \( V_{\text{DHF}} \) in turn requires the orbitals to adjust self-consistently. Such a readjustment process defines an infinite series of many-body diagrams, shown, e.g., in Ref. [7]. The RRPA series can be summed to all orders using iterative techniques or solving DHF-like equations. We used an alternative method of solutions based on the use of basis functions [16]. As an input, we used the DHF basis functions generated in the cavity (see discussion above), i.e. the boundary conditions were satisfied automatically. As a result of solving the RRPA equations, we have determined a quasi-complete set of particle-hole excited states and their energies. Then the EDMS are determined using expressions similar to Eq. (11) and (2).

**Discussion and conclusions** — First, we present the results of our calculations for an isolated atom \( (R_{\text{cav}} = \infty) \). For the Schiff-moment-induced EDM, our results,

\[
d_{\text{SM}}^{\text{RRPA}} = 3.78 \left( \frac{S}{e \text{ fm}^3} \right) \times 10^{-18} \text{ e cm},
\]

are in agreement with the recent calculations by Dzuba et al. [8]. For the EDM induced by T,P–odd semileptonic interactions we obtain

\[
d_{\text{DHF}}^{\text{DHF}} = 8.44 \times 10^{-13} \text{ C}_{\text{TN}} \sigma_N \text{ a.u.},
\]

\[
d_{\text{DHF}}^{\text{RRPA}} = 10.7 \times 10^{-13} \text{ C}_{\text{TN}} \sigma_N \text{ a.u.}.
\]

These values are to be compared with the results by Mårtensson-Pendrill [2], \( d_{\text{DHF}}^{\text{DHF}} = 7.764 \) and \( d_{\text{DHF}}^{\text{RRPA}} = 9.808 \) in the same units. The reason for the 10% difference between our results and those from Ref. [7] is not clear.

Before presenting results for finite cavity radii, let us consider individual contributions to EDM from various shells of Xe atom. These contributions for the Schiff-moment-induced EDM of an isolated atom are listed in Table I. A similar table, but for the EDM arising from semileptonic interactions is given in Ref. [7]. From these tables we observe that the dominant contribution to EDMS comes from the outer \( n = 5 \) shell. Thus we anticipate that a noticeable density dependence should occur when \( R_{\text{cav}} \) becomes comparable to the size of external \( n = 5 \) shell. We also notice that the contribution from the outer shell is relatively more important in RRPA calculations than at the DHF level, i.e., the RRPA results should exhibit stronger density dependence.

| n | DHF | RRPA |
|---|---|---|
| 1 | 0.039 | 0.039 |
| 2 | 0.091 | 0.092 |
| 3 | 0.20 | 0.21 |
| 4 | 0.52 | 0.64 |
| 5 | 2.0 | 2.8 |
| Total | 2.88 | 3.78 |

**TABLE I:** Individual contributions from various shells to the EDM of a free \(^{129}\)Xe atom in the DHF and RRPA methods. The EDM is induced by the nuclear Schiff moment and it is given in units of \( S/(e \text{ fm}^3) \times 10^{-18} \text{ e cm} \).
we see that the $R_n$ nucleus. Non-relativistically, as $r \to 0$ the wavefunctions scale as $\varphi_{nlm}(r) \approx N_{nl}(R_{cav}) \times r^l Y_{lm}(\hat{r})$, where $N_{nl}$ are normalization factors. Therefore the dominant contribution to the EDM, Eq.(11) arises from mixing of $s$- and $p$-states. By factorizing the matrix element of $\hat{h}_{\text{CP}}$ as $\langle \varphi_{ns} | \hat{h}_{\text{CP}} | \varphi_{np} \rangle \approx N_{ns}(R_{cav})N_{np}(R_{cav}) \times \langle s | \hat{h}_{\text{CP}} | p \rangle$ we see that the $R_{cav}$-independent factor $\langle s | \hat{h}_{\text{CP}} | p \rangle$ can be pulled out of the summation over atomic orbitals in Eq.(11). Thus, both semileptonic– and Schiff–moment–induced EDMs exhibit approximately the same scaling with the cavity radius. A correction to this “similarity scaling law” may arise, for example, due to different selection rules involved for the two EDM operators.

It is worth emphasizing the semi-qualitative nature of our calculations. The analysis can be improved by employing more realistic models of liquid environment. Even within the cell model we could further refine our analysis. A dense liquid may be considered as a solid with vacancies, i.e., the decrease of the average bond length is negligible, rather the nearest-neighbor occupation numbers are decreased compared to solid. Xe condenses into face-centered cubic structure. The first nearest-neighbor shell contains twelve atoms (partially justifying the spherical symmetry of the elementary cell). The density of the solid Xe is $3.54$ g/cm$^3$, implying the half-radius of this shell of $4.2$ bohr, somewhat smaller than $R_{cav} \approx 4.9$ bohr for liquid Xe. As follows from Fig. 1, this difference leads to more pronounced suppression of the atomic EDM by 40% per cent.

To reiterate, our work was motivated by anticipated significant improvements in sensitivity to atomic EDMs in experiments with liquid $^{129}$Xe [4]. Here we investigated confining effects of the environment on the EDM of Xe atom. We carried out the analysis in the framework of the cell model coupled with relativistic atomic-structure calculations. We found that compared to an isolated atom, the EDM of an atom of liquid Xe is reduced by about 40%. Thus if the experiment with liquid Xe is carried out with the anticipated sensitivity, we expect that the inferred constraints on possible sources of CP-violation would be indeed several orders of magnitude better than the present limits.

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