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Perspectives on electric dipole moments of atoms and molecules

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Abstract. Searches for a permanent electric dipole moment (EDM) of a fundamental particle started with the key idea by Ramsey and Purcell of exploitation of discrete symmetries, such as parity, more than seven decades ago. These searches provide a model-independent test of theoretical frameworks, in particular, the Standard Model of particle physics. Over time, a large number of experimentally suitable systems, also atoms and molecules, provided more stringent limits on possible EDMs. We discuss some perspectives on such experiments in the context of statistics, systematics, and sensitivities to beyond the Standard Model physics.

1 Introduction

Searches for a permanent electric dipole moment (EDM) are executed in composite systems such as atoms and molecules rather than on fundamental particles. The EDM of such a system is a parity (P) and time-reversal (T) violating interaction of its angular momentum $\mathbf{F}$ with the electric field $\mathbf{E}$. The non-relativistic Hamiltonian, including the P,T-conserving interaction with the magnetic field $\mathbf{B}$, is given by:

$$H = -\boldsymbol{\mu} \cdot \mathbf{B} - \mathbf{d} \cdot \mathbf{E}, \tag{1}$$

with $\boldsymbol{\mu} = \mu \mathbf{F}/\hbar$ being the magnetic dipole moment of the system, and $\mathbf{d} = d\mathbf{F}/\hbar$ the EDM.

In 1950 Ramsey and Purcell realized that an EDM breaks P symmetry because of the P-odd nature of the electric field [1]. Searching for an EDM is thus a test of P symmetry, which was until 1957 considered to be a good symmetry of nature. Seven years later, parity violation was demonstrated, not in the context of EDMs, but in weak interactions [2–4]. A non-zero EDM has still not been detected in any system. An EDM is, however, not only P violating, but also T violating. Under the assumption of conservation of the combination of charge conjugation (C), parity and time-reversal symmetry (CPT theorem), T violation is equivalent to CP violation. A major motivation to look for CP violation is the observed matter-antimatter asymmetry in the

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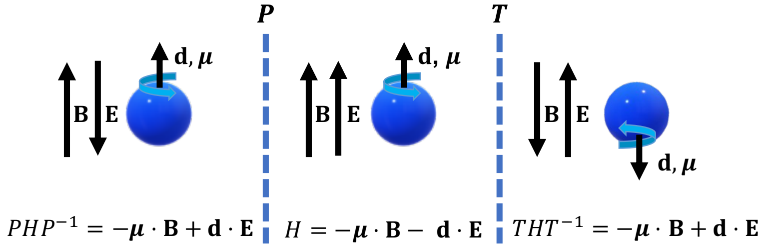

Figure 1. Symmetries of $\boldsymbol{\mu}, \mathbf{d}, \mathbf{E}, \mathbf{B}$ under P and T symmetry. The EDM Hamiltonian changes sign under P and T, which means that the EDM interacting with the electric field is P,T violating. The Zeeman Hamiltonian does not change sign under P,T and is therefore P,T conserving.

universe. Sakharov showed that mechanisms that generate matter and antimatter asymmetrically, need to violate CP symmetry, baryon number symmetry and need to be out of thermal equilibrium [5]. The Standard Model of particle physics (SM) contains CP violation in terms of a complex phase in the Cabibbo–Kobayashi–Maskawa (CKM) matrix [6, 7], describing the mixing of quark flavors. However, the SM CP violation is insufficient to account for the matter-antimatter asymmetry. Another motivation is the apparent absence of CP violation in the strong interaction, parameterized by the QCD theta-term. Furthermore, many of the SM extensions contain CP violation in one form or another [8].

Although the measurement of the EDM (Eq.1) is clear evidence for CP-violating physics, its origin cannot be determined by a single experiment. Fortunately, the experimental approaches are numerous in systems with different sensitivities to EDMs. This ranges from measurements on the neutron, proton, atoms, and molecules. Experiments have been performed employing vapor cells, beams, traps, and condensed matter systems, exploiting a wide range of experimental techniques that exhibit different systematic challenges [9–19]. Searching for CP violation in EDM searches involves (1) high precision, given by the statistics of the measurement, (2) control of systematics, determined by the execution of the experiment, and (3) interpretation of the experimental measurement in terms of CP-violating physics. Progress on these three topics is indispensable for setting limits on new physics theories or eventually claiming a discovery with EDM searches.

2 EDM measurement

The energy scale of a contribution from an EDM is much smaller than all other energy scales in the system of choice. Thus a precision measurement as well as excellent control of many experimental parameters is required. The EDM contribution is unique in its symmetry properties and can therefore be separated in a differential measurement as described in the following. The atomic or molecular system is placed in co-linear electric and magnetic fields. Let us consider two levels with angular momentum F, which differ only by the projection of M_F on the quantization axis given by the direction of $\mathbf{E}$ and $\mathbf{B}$. The energy difference $\hbar\omega$ between the levels depends only on the Zeeman shift due to the magnetic moment μ and the energy shift due to the EDM d

(Eq.1):

$$\omega = (2\mu B \pm 2dE)/\hbar. \tag{2}$$

The sign in front of the EDM contribution refers to parallel (+) and anti-parallel (-) magnetic and electric fields. This energy splitting is most sensitively measured by spin-precession methods, where a superposition of the two spin projections is used [20, 21]. The superposition includes an additional phase ϕ which depends on the electric and magnetic fields. The phase increases with the coherence time T and the energy difference between the two levels:

$$\phi = \omega T. \tag{3}$$

The ultimate precision on the phase ϕ is given by the Quantum Projection Limit (QPL). The projection of the phase difference ϕ in the QPL for N measurements is

$$\delta\phi \geq \frac{1}{\sqrt{N}}. \tag{4}$$

The precision on the frequency is enlarged for large coherence times:

$$\delta\omega \geq \frac{1}{T\sqrt{N}}. \tag{5}$$

The contribution from the P,T-odd EDM can be extracted by executing the measurement for electric and magnetic fields parallel and anti-parallel (Fig. 1):

$$\Delta\omega = \omega(\hat{E} = \hat{B}) - \omega(\hat{E} = -\hat{B}) = 4dE/\hbar. \tag{6}$$

The experimental challenge is achieving an accurate P or T transformation, i.e. realizing the differential measurement as described in Eq.6. Since $\omega(\hat{E} = \hat{B})$ and $\omega(\hat{E} = -\hat{B})$ cannot be measured at the same time and the same location, there can be other differences between the two measurements than only the relative orientation of the magnetic and electric fields. Any change of another parameters between $\omega(\hat{E} = \hat{B})$ and $\omega(\hat{E} = -\hat{B})$ has the potential of being wrongly interpreted as an EDM signal. For example, if the magnetic field for parallel fields is slightly higher than for anti-parallel fields, this results in a larger Zeeman shift for one field orientation. Dealing with these kinds of unavoidable imperfections in the experiment determines the systematic error of the measurement.

EDM measurements look for a P,T-odd change of the Zeeman shift and therefore need a precise magnetic field measurement in combination with an accurate P or T transformation. Large statistics, and stability of and control over magnetic and electric fields are therefore central in EDM searches.

3 Interpretation

The EDM of an atomic or molecular system is defined in terms of a linear Stark shift in the limit of zero electric field [22]:

$$d = \frac{1}{2}\frac{\partial\hbar\omega}{\partial E}\bigg|_{E\to 0}, \tag{7}$$

where $\hbar\omega$ is the energy difference between two levels with opposite angular momentum projection (Eq.2). CP-violating interactions of the system's constituents, either originating from SM physics or beyond the SM (BSM) physics, can induce an EDM of the composite system. The search for BSM physics is systematically organized in an effective field theory (EFT) framework [23, 24], where the CP-violating physics is expressed as:

$$\mathcal{L}_{\mathrm{CPV}} = \mathcal{L}_{\mathrm{SM}}^{\mathrm{CPV}} + \sum_i \frac{c_i^{\mathrm{CPV}}}{\Lambda^{d-4}} \mathcal{O}_i^d. \tag{8}$$

$\mathcal{L}_{\mathrm{SM}}^{\mathrm{CPV}}$ contains the sources of CP violation in the SM, i.e. the CKM and QCD-theta term contributions. $\mathcal{O}_i^d$ are BSM CP-violating operators constructed from the degrees of freedom of the EFT. Λ is the energy scale of new physics and d is the mass-dimension of operator $\mathcal{O}_i$.[1] This framework motivates to express EDMs in terms of the SM contribution and CP-violating BSM EFT coefficients, i.e.

$$d = d^{\mathrm{SM}} + \sum_i R_i c_i^{\mathrm{CPV}}. \tag{9}$$

d^{SM} is the value of the EDM due to SM physics, which has a contribution from the CKM matrix and the QCD-theta term. The CKM contribution is highly suppressed in EDMs [25, 26], and the QCD-theta term is strongly limited by neutron EDM searches [9]. Therefore, $d^{\mathrm{SM}} \approx 0$ at the current experimental sensitivity, i.e. EDMs searches are SM-background free and very sensitive to BSM physics. The CP-violating physics at some higher energy scale beyond the SM is parameterized by the coefficients c_i^{CPV}. R_i are factors incorporating the physics from the energy scale of d to the scale of c_i^{CPV}. Calculating the contributions from the physics at high-energy scales to the low-energy scale of the experiment requires a large theoretical effort. This is done in multiple steps, as we will demonstrate below. We take as an example the EDM of a paramagnetic molecule, which is commonly expressed as a sum of two underlying sources of CP violation, namely the eEDM parameterized by d_e [27, 28]

$$\mathcal{L}^{e\mathrm{EDM}} = -i\frac{d_e}{2}\bar{e}\sigma^{\mu\nu}\gamma_5 e F_{\mu\nu}, \tag{10}$$

and the scalar-pseudoscalar (S-PS) interaction with strength C_S [29]:

$$\mathcal{L}_S = C_S \frac{G_F}{\sqrt{2}}(\bar{e}i\gamma_5 e)(\bar{p}p + \bar{n}n). \tag{11}$$

e, p, n are the electron, proton and neutron field, $\sigma^{\mu\nu} = \frac{1}{2i}[\gamma^\mu, \gamma^\nu]$, $\gamma_5 = \gamma_1\gamma_2\gamma_3\gamma_4$, where γ_i are the Dirac matrices. $F_{\mu\nu}$ is the electromagnetic field strength and G_F is Fermi's constant. These two sources are expected to be the largest contributions to an EDM of a paramagnetic molecule. The corresponding P,T-odd energy shift in an electric field is written as

$$\hbar\omega = (d_e W_d + C_S W_S)\Omega P_n(E), \tag{12}$$

where Ω is the electronic angular momentum quantum number [30] and n labels the spin-rotational state. The molecular EDM is consequently given by

$$d_{\Omega,n} = (d_e W_d + C_S W_S)\Omega \left.\frac{\partial P_n(E)}{\partial E}\right|_{E\to 0}. \tag{13}$$

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[1] We use $\hbar = c = 1$ in the Lagrangian densities, as is customary in particle physics

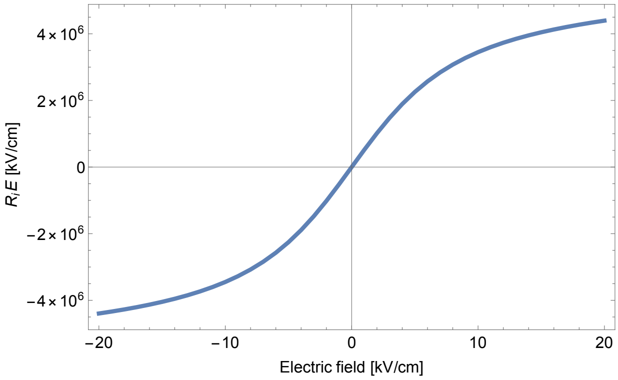

Figure 2. Calculation of the polarization factor $P(E)$ for BaF, showing that CP violation in a paramagnetic molecule leads to a linear Stark shift at $E \to 0$, but saturation at higher electric fields [32].

We identify the two CP-violating EFT coefficients d_e and C_S and their corresponding factors

$$R_i = W_i \Omega \frac{\partial P_n(E)}{\partial E}\bigg|_{E\to 0}, \tag{14}$$

for $i = d, S$, which include the P,T-conserving physics between the scale of $d_{\Omega,n}$, and d_e respectively C_S. W_d and W_S contain the P,T-conserving electromagnetic physics due to the interactions of the electrons in the electronic potential of the nuclei, and can be calculated with electronic structure theory [31]. $P_n(E)$, referred to as the polarization factor, describes the interaction of the molecule with the applied electric field, which results in an alignment of the molecule. This factor is the same for d_e and C_S. It should be noted that a molecular EDM leads to a linear Stark shift only for small electric fields, $E \to 0$. At higher fields, the energy splitting due to CP violation saturates (Fig.2).

The operators in Eq.10 and Eq.11 are defined at different energy scales, both below the energy scale of new physics. The eEDM operator contains solely SM fields below the electroweak symmetry breaking scale $\Lambda_{\text{weak}} \sim 250$ GeV. The S-PS interaction contains nucleons as degrees of freedom, which indicates its energy scale of validity is the nuclear scale $\Lambda \sim 1$ GeV [33]. Therefore, to connect d_e and C_S to CP-violating SM and BSM physics, they have to be in turn expressed in terms of higher-energy contributions according to Eq.9:

$$d_e = d_e^{\text{SM}} + \sum_i R'_i c_i^{\text{CPV}}, \tag{15}$$

and similar for C_S. The Standard Model predicts $d_e^{\text{SM}} = 5.8 \times 10^{-40} e$ cm [26], showing that a non-zero measurement at current experimental sensitivities is clear evidence for new physics. Equations 13 and 15 show how the EDM of the molecule is translated in multiple steps to SM and BSM physics. The different energy scales involved require different expertise. The translation of EDMs into their underlying CP-violating physics parameters at different energy scales allows for the comparison of different experiments. Results of atomic or molecular EDM searches are usually reported in terms of nucleon EDMs, electron-nucleon interactions or the electron

EDM. Translating these in turn to BSM EFT coefficients allows for the comparison of EDM experiments to other experiments, for example collider experiments which also probe the CP-violating coefficients, albeit in completely different experiments and energy scales [34].

Traditionally, some systems are expected to be more sensitive to certain parameters, i.e. paramagnetic molecules provide the stringents limits on d_e while the neutron EDM provides the strongest limit on the QCD theta term. However, all CP-violating interactions can in principle contribute to the EDM of a composite system, but most of them only contribute at high-order in perturbation theory. For example, the limit on the EDM of ThO is usually translated to a limit on d_e, but can also be translated in terms of the QCD theta term. This limit is only two orders of magnitude away from the most stringent limit obtained from the neutron EDM [35]. For every source of CP violation, within the SM or the higher mass-dimension EFT operators, it has to be calculated or estimated how it induces an EDM on the energy scale of the measurement.

By calculating and comparing the factors R_i in different systems, it becomes clear that certain systems are more sensitive to CP violation than others. It is shown that systems containing at least one heavy atom promise large values for R_i [36], which are therefore often referred to as *enhancement* factors. Complex systems such as diatomic and polyatomic molecules promise high values of R_i at small electric fields, because they exhibit opposite parity states which are close in energy [37, 38]. However, the large enhancement factors can only be verified with a non-zero measurement of the EDM, since no alternative measurements are available.

4 Experiments

Various implementations adapt to particular properties of the chosen system, such as particular level structure, state lifetimes and production methods. While the approach to improve phase sensitivity depends on increasing the brightness of the source or the coherence time T, the reach towards new physics requires improvements of electric field strength E and choice of system with favorable enhancement factors R_i. The details of the particular implementations lead in general also to a different set of systematic biases, which motivates strongly the search for EDMs in many systems.

The most stringent limits on EDMs have been determined by three experimental concepts: cell, beam and trap experiments. With cell-type experiments, such as the search for the EDM of the neutron [9], Hg [10] and Xe [39, 40], coherence times T of 100 s or longer are achieved. The sensitivity arises in combination with the much larger number of particles compared to beam or trap experiments. Characteristic systematic challenges originate from collisions with cell walls and other particles in the gas phase [39]. Beam experiments, such as the ones performed with Tl [12], TlF [13], ThO [14], YbF [15] and BaF [16, 41], have the advantage of systems which are not in the ground state or chemically stable, but achievable coherence times T are limited by the particle velocity and length of the interaction zone. Trap measurements, such as the ones performed with Ra [17], Yb [18] and HfF^+ [19, 42], can provide very large coherence times T, which limits the repetition rate of the measurements. However, the number of systems amendable to trapping methods is limited and the number of particles N is small. In Table 1 some characteristic parameters for these experiments are presented. We present the typical sensitivity to the phase ϕ and the corresponding accuracy in the precession frequency $\delta\omega = \delta\phi/T$.

Table 1. Current EDM searches for a number of different systems. Typical coherence times T and the resulting sensitivity in terms of the precession phase $\delta\phi$ and frequency $\delta\omega$ are presented. The sensitivity to the underlying physics R_iE varies over several orders of magnitude for the different systems.

System	$\delta\phi$ [rad]	T [s]	$\delta\omega/2\pi$ [Hz]	R_iE [V/cm]	EDM limit [e cm] (95% c.l.)
Cell					
n [46]	$1 \cdot 10^{-4}$	130	$1.2 \cdot 10^{-7}$	$7 \cdot 10^{3}$	$\lvert d_n \rvert < 3.6 \cdot 10^{-26}$
Hg [10]	$3 \cdot 10^{-8}$	170	$2.9 \cdot 10^{-11}$	$8 \cdot 10^{3}$	$\lvert d_{\mathrm{Hg}} \rvert < 7.4 \cdot 10^{-30}$
Xe [11, 40]	$1 \cdot 10^{-4}$	> 1000	$1.1 \cdot 10^{-8}$	$4 \cdot 10^{3}$	$\lvert d_{\mathrm{Xe}} \rvert < 1.4 \cdot 10^{-27}$
Beam					
Tl [12]	$1 \cdot 10^{-6}$	$2.4 \cdot 10^{-3}$	$6.5 \cdot 10^{-5}$	$1.2 \cdot 10^{5}$	$\lvert d_{\mathrm{Tl}} \rvert < 1.1 \cdot 10^{-24}$
ThO [14]	$3 \cdot 10^{-6}$	$1.0 \cdot 10^{-3}$	$5.0 \cdot 10^{-4}$	$7.8 \cdot 10^{10}$	$\lvert d_e \rvert < 1.31 \cdot 10^{-29}$
YbF [15]	$4 \cdot 10^{-5}$	$0.7 \cdot 10^{-3}$	$8.9 \cdot 10^{-3}$	$1.5 \cdot 10^{10}$	$\lvert d_e \rvert < 1.27 \cdot 10^{-27}$
BaF [16]		$15 \cdot 10^{-3}$		$3.4 \cdot 10^{9}$	proposed
Trap					
Ra [17]	$3 \cdot 10^{-3}$	0.035	$1.6 \cdot 10^{-2}$	$6.5 \cdot 10^{4}$	$\lvert d_{\mathrm{Ra}} \rvert < 5 \cdot 10^{-22}$
Yb [18]	$1 \cdot 10^{-3}$	300	$5.3 \cdot 10^{-7}$	$7.3 \cdot 10^{4}$	$\lvert d_{\mathrm{Yb}} \rvert < 1.5 \cdot 10^{-26}$
HfF^{+} [42]	$1 \cdot 10^{-3}$	3	$5.5 \cdot 10^{-5}$	$2.3 \cdot 10^{10}$	$\lvert d_e \rvert < 5 \cdot 10^{-30}$

Another class of experiments proposed is using atoms and molecules embedded in matrices, such as liquid helium [43] and frozen-gas matrices [44, 45]. These systems promise a large number of particles and long coherence times (T up to seconds) and will come with a new set of systematic challenges due to interactions with the condensed matter.

5 Conclusions

Searches for permanent electric dipole moments rely strongly on the exploitation of symmetries and require a quantification of the ability to perform a mirror version of the experiment, i.e. realize a P or T transformation. In order to search for new CP-violating physics using EDMs, progress has to be made on three frontiers. Firstly, high precision is needed, which motivates improvements in increasing the number of particles and an increase in coherence time T. Secondly, the accuracy of the experiment needs to be improved in parallel with the statistical uncertainty. Here the identification of systematic biases and the development of strategies of accurate determinations is required. Examples are co-magnetometry [9, 12, 40] or the determination of other experimental parameters such as the electric field [41]. Thirdly, the measurement needs an interpretation in terms of fundamental CP-violating physics. Such calculations span vastly different energy scales and require a large theoretical effort ranging from molecular physics to particle physics. In view of this, the perspective for future progress in the active field of CP violation in atomic and molecular systems is very bright. In the near future new exciting experimental ideas are coming to life and they can demonstrate their ability to provide sufficient statistics, good control of systematics and the connection to sources of CP violation in and beyond the Standard Model.

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