

Nucleon Electric Dipole Moments in Paramagnetic Molecules through Effective Field Theory

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Electric dipole moment (EDM) measurements using paramagnetic molecules have significantly advanced over the last decade. Traditionally, these experiments have been analyzed in terms of the electron EDM. However, paramagnetic molecules are also sensitive to hadronic sources of charge-parity (CP) violation, highlighting the need for a new framework to interpret the experimental results. In this Letter, we introduce an effective field theory framework to relate molecular EDMs to the EDMs of neutrons and protons. We identify the dominant contributions through power counting and pinpoint the necessary nuclear matrix elements. As a practical application, we employ the nuclear shell model to calculate these nuclear matrix elements for the polar molecule BaF. Finally, we estimate the limits on the nucleon EDMs set by current molecular EDM experiments.

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Introduction—Electric dipole moment (EDM) experiments are extremely sensitive probes of new sources of charge-parity (CP) violation and indirectly probe beyond-the-Standard-Model (BSM) physics at very high scales of up to ~ 100 TeV [1,2]. Recent years have seen impressive experimental progress using polar molecules, which benefit from large internal electric fields that amplify the CP -violating signal [3–7]. Among them, EDMs of paramagnetic systems, which have one unpaired electron, are mainly interpreted in terms of the electron EDM. Current measurements lead to a strong bound on the electron EDM, $|d_e| < 4.1 \times 10^{-30} e \text{ cm}$ [7], and future experiments aim to improve this by one to two orders of magnitude [6–13]. This constraint is four orders of magnitude more stringent than the neutron EDM limit [14].

Traditionally, paramagnetic systems have not been used to constrain hadronic sources of CP violation, such as the quantum chromodynamics (QCD) $\bar{\theta}$ term within the SM or

higher-dimensional quark-gluon operators that arise from heavy BSM physics. This is because of the assumption that far stricter limits can be obtained through the EDMs of the neutron or diamagnetic atoms. That being said, paramagnetic systems are sensitive to hadronic sources of CP violation through the CP -odd electron-nuclear force they induce [15–17]. While this force is typically strongly suppressed, the rapid progress in paramagnetic EDM experiments might make it the best way to search for hadronic sources of CP violation in the future. However, the current theoretical description of the CP -odd electron-nuclear force is still at a very rudimentary stage.

In this Letter, we systematically derive this force as induced by the EDMs of neutrons and protons, making it possible to constrain these EDMs with paramagnetic molecular EDM experiments. As the problem involves a multitude of well-separated energy scales, it can be systematically described using effective-field-theory (EFT) techniques. We show that this connection requires the calculation of a set of nuclear matrix elements (NMEs) that are different from the ones involved in the Schiff moments of diamagnetic systems [18–20]. As an explicit example, we compute the NMEs for the polar molecule BaF, which is being targeted by the NL-eEDM collaboration [8].

Effective field theory—The calculation of molecular EDMs in terms of fundamental sources of CP violation involves widely separated energy scales. These range from

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the BSM and electroweak scales (Λ and M_W) to low-energy scales such as the electron mass or electron binding energy $\mathcal{O}(\alpha_{\text{em}}^2 m_e)$. The atomic nucleus gives rise to additional scales associated with the chiral-symmetry-breaking scale $\Lambda_\chi \sim m_N \sim 1$ GeV (comparable to the nucleon mass), the pion mass $m_\pi \sim \gamma \sim 100$ MeV (comparable to the nuclear binding momentum), and the scale of nuclear excitations $m_\pi^2/m_N \sim \mathcal{O}(\text{MeV})$.

Within the SM, the most relevant source of CP violation is the QCD $\bar{\theta}$ term, as (paramagnetic) EDMs induced by the phase in the Cabibbo-Kobayashi-Maskawa (CKM) matrix are orders of magnitude too small to be detected by current and envisioned experiments [21]. BSM sources are described by dimension-six operators [22–25], matched at Λ_χ to a χ EFT Lagrangian for light mesons, nucleons, photons, electrons [1,22], with the most relevant hadronic operators being quark (chromo-)EDMs, the Weinberg operator [26], and CP -odd four-quark interactions [22]. For our purposes, the most relevant interactions are given by

$$\mathcal{L}_\chi = \bar{g}_0 \bar{N} \tau^a N \pi^a + \bar{g}_1 \bar{N} N \pi^0 + \bar{g}_{0\eta} \bar{N} N \eta + 2\bar{N}(d_0 + d_1 \tau^3) v^\mu S^\nu N F_{\mu\nu}, \quad (1)$$

where the first line describes three CP -odd meson-nucleon interactions, and the second line the isoscalar and isovector nucleon EDM. We use the nonrelativistic nucleon doublet $N = (p, n)^T$ with spin $S^\mu = (0, \boldsymbol{\sigma}/2)$ and velocity $v^\mu = (1, \mathbf{0})$, as well as the pion triplet π^a and the eta meson η .

The paramagnetic EDMs induced by the meson-nucleon interactions in Eq. (1) arise mainly through intermediate CP -odd electron-nucleon interactions, which take on the form

$$\mathcal{L} = \frac{G_F}{\sqrt{2}} \bar{e} i \gamma_5 e \bar{N} (C_{\text{SP}}^0 + C_{\text{SP}}^1 \tau^3) N. \quad (2)$$

The nucleon EDMs in Eq. (1) give rise to contributions at longer distance scales through the diagrams in Fig. 1. They induce effective interactions between the nucleus and the electrons, i.e. the nuclear equivalent of $C_{\text{SP}}^{0,1}$, which we denote by $\bar{C}_{\text{SP}}$, see Eq. (7). To systematically compute the various contributions, it is useful to consider different photon modes depending on the scaling of their momentum $q_\gamma^\mu = (q_\gamma^0, \mathbf{q}_\gamma)$.

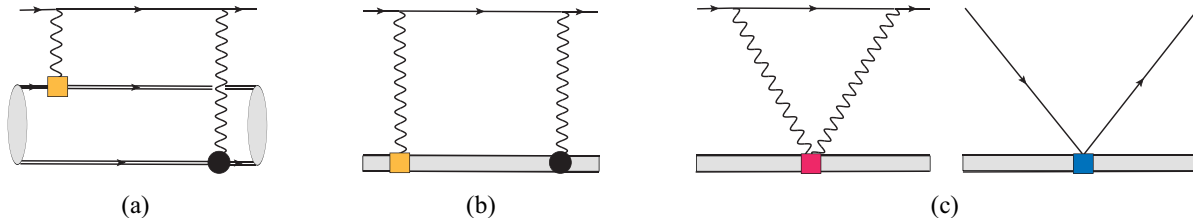


FIG. 1. Contributions to $\bar{C}_{\text{SP}}$ arising from the nucleon EDMs. We denote electrons by single and nucleons by double straight lines, nuclei by gray ovals [in (a)] or bars [in (b),(c)], and photons by wavy lines. The black circle stands for the nucleon MDM, while the yellow, magenta, and blue squares indicate the CP -violating vertices: nucleon EDM, β_v and $\bar{C}_{\text{SP}}$ effective vertices [see Eq. (7)], respectively. (a) Shows the two-nucleon potential-region contribution, and (b) the ultrasoft one. (c) Shows the two diagrams relevant to the matching and running of diagram (b) in an EFT with the nuclear ground state as the remaining degree of freedom.

We identify three regions that give relevant contributions, (1) soft photons: $q_\gamma^0 \sim |\mathbf{q}_\gamma| \sim m_\pi$, (2) ultrasoft photons: $q_\gamma^0 \sim |\mathbf{q}_\gamma| \sim m_\pi^2/m_N$, (3) potential photons: $q_\gamma^0 \sim |\mathbf{q}_\gamma|^2/m_N$, $|\mathbf{q}_\gamma| \sim m_\pi$, and we define $Q \sim m_\pi \sim \gamma$ and $q \sim Q^2/m_N$.

The CP -odd meson-nucleon interactions in Eq. (1) contribute to $\bar{C}_{\text{SP}}^{0,1}$ through diagrams involving a meson exchange or a pion loop in combinations with the exchange of two photons in the ultrasoft or soft region. These diagrams were first considered in Ref. [15] and later computed with heavy-baryon chiral perturbation theory in Ref. [17]. In addition, integrating out the mesons leads to renormalization of nucleon EDMs [27–29], effectively shifting $d_{0,1} \rightarrow \bar{d}_{0,1}$, where the bar denotes the renormalized low energy constants (LECs). In what follows, we use $\bar{d}_{0,1}$ as the physical nucleon EDMs.

In this Letter, we focus on additional contributions to $\bar{C}_{\text{SP}}$ from the nucleon EDMs, which arise through the topologies shown in Figs. 1(a) and 1(b). These diagrams are captured by an effective action of the form

$$\langle h_f(p_f) e(p_e') | i S_{\text{eff}} | h_i(p_i) e(p_e) \rangle = \frac{e^3}{2} \int_{x_i} \langle h_f e | T [\bar{e} \mathcal{A} e(x_1) \bar{e} \mathcal{A} e(x_2) \mathcal{L}_\chi^{(\bar{d}_{0,1})}(x_3) (A_\mu J_{\text{em}}^\mu)(x_4)] | e h_i \rangle, \quad (3)$$

where we integrate over all $x_{1,2,3,4}$, $h_{i,f}$ denote the initial and final nuclear states (for EDMs we have the nuclear ground state $|h_i\rangle = |h_f\rangle = |0^+\rangle$) and J_{em}^μ denotes the nuclear electromagnetic current. Diagrams involving nucleon EDMs and photons with soft momenta are sub-leading as they require the inclusion of additional pions. Power counting gives the expected size of the potential and ultrasoft contributions

$$\left\{ C_{\text{SP}}^{(\text{pot})}, C_{\text{SP}}^{(\text{usoft})} \right\} = \frac{m_e \alpha^2 \mu_i \bar{d}_i}{e G_F m_N} \left\{ \frac{4\pi}{Q}, \frac{1}{q} \right\}, \quad (4)$$

where μ_i are the nucleon magnetic dipole moments (MDMs) in units of the nuclear magneton. Numerically $4\pi q \sim Q$ and these estimates are rather close, but, as we will see, they do not capture possible coherent enhancements.

Potential region—To evaluate the potential contributions, we can use the so-called method of regions to expand the amplitude in small ratios of scales, such as $q_\gamma^0/|\mathbf{q}_\gamma|$. After doing so, there are no contributions from diagrams where the nucleon EDM and J_{em} attach to the same nucleon (the potential region arises from picking up the poles of nucleon propagators, which can always be avoided in these one-body diagrams). There are, however, two-nucleon effects through the diagram in Fig. 1(a). Because of spin and parity constraints, the first contributions arise from the nucleon magnetic moments, which appear in J_{em}^μ at next-to-leading order $\mathcal{O}(Q/m_N)$. This results in the following contribution to the amplitude [30]

$$\mathcal{A}_{\text{pot}} = -\langle h_f | V | h_i \rangle \bar{u}(p'_e) \left(1 - \frac{v \cdot (p'_e - p_e)}{2m_e} \not{v} \right) i\gamma_5 u(p_e),$$

where we take the limit $p'_e - p_e \ll m_e$ in what follows, while V denotes the potential between the two interacting nucleons. In momentum space [31]

$$V = \frac{4e^4 m_e}{9m_N} \sum_{i \neq j} \frac{\mu^{(i)} D^{(j)}}{|\mathbf{q}|^4} \left[\boldsymbol{\sigma}^{(i)} \cdot \boldsymbol{\sigma}^{(j)} - \frac{1}{4} S^{(ij)} \right], \quad (5)$$

where i, j label the nucleons, $\mathbf{q}_i = \mathbf{p}'_i - \mathbf{p}_i$ is the exchanged momentum and we define the combination $\mathbf{q} = (\mathbf{q}_i - \mathbf{q}_j)/2$. In addition, $D = (\vec{d}_0 + \vec{d}_1 \tau_3)/e$, $\mu \equiv [(1 + \kappa_0)/2] + [(1 + \kappa_1)/2]\tau_3$ describe the EDM and MDM operators (with $\kappa_0 = -0.12$ and $\kappa_1 = 3.7$), while $S^{(ij)} = \boldsymbol{\sigma}^{(i)} \cdot \boldsymbol{\sigma}^{(j)} - 3\mathbf{q} \cdot \boldsymbol{\sigma}^{(i)} \mathbf{q} \cdot \boldsymbol{\sigma}^{(j)} / \mathbf{q}^2$. These contributions are thus determined by the NME of V , which scales as $1/|\mathbf{q}|^4$. The form of this two-body potential is similar to the NMEs appearing in neutrinoless double- β decay [33] or radiative corrections to superallowed β decays [32,34] although with different isospin and/or $\mathbf{q}$ dependence. The many-body techniques needed to compute such NMEs, including *ab initio* approaches, have developed significantly in the last decade and can be directly applied to these EDM calculations [33,35].

Ultraviolet region—In this region, we expand in small ratios of scales, such as $q_\gamma^0/m_\pi \sim |\mathbf{q}_\gamma|/m_\pi$ and m_e/m_π . Photons with this (small) momentum scaling can be thought of as coupling to the nucleus as a whole, instead of the individual nucleons. After inserting a complete set of (nuclear) states between the hadronic operators and working out the time-ordered product in Eq. (3), we again find that the leading contributions involve the magnetic moments,

$$\begin{aligned} \mathcal{A}_{\text{usoft}} = & \frac{-2ie^4}{m_N} \int \frac{d^4 k}{(2\pi)^4} \frac{\bar{u}(p'_e) \gamma_\lambda (k + m_e) \gamma_\rho u(p_e) \epsilon_{\sigma\alpha\beta\eta} v^\beta}{(k^2 - m_e^2)(k - p_e)^2(k - p'_e)^2} \sum_n \left[(p_e - k)^\sigma (p'_e - k)^\mu g^{\nu\lambda} g^{\alpha\rho} \right. \\ & \times \left(\frac{\langle h_f | D_{\mu\nu} | n \rangle \langle n | M^\rho | h_i \rangle}{v \cdot l_+ - E_n + i\epsilon} + \frac{\langle h_f | M^\rho | n \rangle \langle n | D_{\mu\nu} | h_i \rangle}{v \cdot l_- - E_n + i\epsilon} \right) + \{ p_e \leftrightarrow p'_e, l_+ \leftrightarrow l_-, \alpha \leftrightarrow \nu \} \Big], \end{aligned} \quad (6)$$

where $|n\rangle$ denote intermediate nuclear $|1^+\rangle$ states with energies E_n , while $l_+ = p_i + p_e - k$, $l_- = p_i - p'_e + k$. Furthermore, $M^\rho \equiv \bar{N} \mu S^\rho N(0)$ and $D_{\mu\nu} \equiv \bar{N} D(v_\mu S_\nu - v_\nu S_\mu) N(0)$ denote the MDM and EDM operators.

Although the appearing integrals can be evaluated using known techniques [36], the expressions are rather unwieldy. They greatly simplify if there is a hierarchy between the nuclear excited states and the electron momenta, $p_e \sim m_e \ll \Delta_n = |E_n - E_i|$, which is a good approximation for ^{138}Ba as discussed below. Likewise, the relevant excited states in magnetic-dipole transitions—also driven by the spin operator—in isotopes of Yb, Hf, and Th with an even number of neutrons also enter at about 2 MeV or higher energies [37–39]. In this case, Eq. (6) can be captured by a low-energy nuclear EFT in which the excited nuclear states have been integrated out, but which still contains electrons, ultrasoft photons, and the ground state of the nucleus. The relevant interactions in this theory can be written as

$$\mathcal{L}_\Psi = \Psi_i^\dagger \left[\frac{G_F}{\sqrt{2}} \bar{C}_{\text{SP}} \bar{e} i\gamma_5 e + \beta_v v^\alpha F_{\alpha\beta} v_\lambda \epsilon^{\beta\lambda\mu\nu} F_{\mu\nu} \right] \Psi_i, \quad (7)$$

where Ψ_i denotes the spin-0 field describing the nucleus [40], $\bar{C}_{\text{SP}}$ describes the nuclear version of $C_{\text{SP}}^{0,1}$, while β_v has a similar form as the nuclear polarizability but violates CP . At the scale $\lambda = \Delta_n$, β_v obtains a contribution from integrating out the excited states at tree level, while $\bar{C}_{\text{SP}}$ arises from Eq. (6). After expanding in $m_e/|E_n - E_i|$, this expression simplifies and the remaining integrals can be calculated using techniques from Ref. [41]. All in all, matching the nucleon-level theory to the EFT without excited states then gives at a scale $\lambda \simeq \Delta_n$,

$$\begin{aligned} \frac{G_F}{\sqrt{2}} \bar{C}_{\text{SP}} = & -\frac{e^4 m_e}{4\pi^2 m_N} \sum_n \frac{A_n}{\Delta_n} \left(4 - 3 \log \frac{4\Delta_n^2}{\lambda^2} \right), \\ \beta_v = & \frac{e^2}{m_N} \sum_n \frac{A_n}{\Delta_n}, \quad A_n = -\frac{\langle h_i | D \boldsymbol{\sigma} | n \rangle \cdot \langle n | \mu \boldsymbol{\sigma} | h_i \rangle}{12}. \end{aligned} \quad (8)$$

These interactions can be evolved from $\lambda \sim \Delta_n$ to lower energies, $\lambda_e \sim m_e$, using the renormalization group equation, which arises through the loop diagram of Fig. 1(c). The amplitude at low scales, $\lambda_e \sim m_e$, can finally be expressed as the sum of $\bar{C}_{\text{SP}}(m_e)$ and a loop involving β_v .

We capture the total combination of the ultrasoft and potential contributions by an effective contact interaction, such that $\mathcal{A}_{\text{total}} = (G_F/\sqrt{2})\bar{C}_{\text{SP}}^{\text{eff}}\bar{u}(p'_e)i\gamma_5 u(p_e)$ with

$$\bar{C}_{\text{SP}}^{\text{eff}} = -\frac{\sqrt{2}}{G_F} \left[\frac{4\alpha^2 m_e}{m_N} \sum_n \frac{A_n}{\Delta_n} \left(3 \ln \frac{m_e^2}{4\Delta_n^2} - 1 \right) + \langle h_i | V | h_i \rangle \right], \quad (9)$$

which is independent of the renormalization scale λ . Additional details of the derivation can be found in Supplemental Material [42]. We stress that this is the effective interaction between electrons and the nucleus as a whole and differs from Eq. (2), which is the coupling to individual nucleons. Evaluating the ultrasoft region thus requires the excited state energies, Δ_n , and the set of nuclear matrix elements of the one-body operator $\sim \langle h_i | \sigma | n \rangle$ contained in A_n . These have a form similar to the leading two-neutrino double β , and double magnetic-dipole NMEs [46–48] and of subleading NMEs of the neutrinoless double- β decay [49,50]. Therefore, similar many-body methods used in these studies can be applied here. Equation (9) is the main result of this Letter and makes it possible to connect nucleon EDMs to measurements of paramagnetic molecules.

Nuclear matrix elements—We now focus on the polar molecule BaF, which is targeted by the NL-eEDM collaboration [8]. The heaviest atom in the molecule, ^{138}Ba , has a magic neutron number, and it is just two neutrons away from ^{136}Ba , the well-studied [51] final state of the double- β decay of ^{136}Xe . We calculate excitation energies and NMEs with the nuclear shell model [52], using the codes [53,54], a configuration space of $1d_{5/2}, 0g_{7/2}, 2s_{1/2}, 1d_{3/2}, 0h_{11/2}$ orbitals with a ^{100}Sn core, and three effective interactions: GCN5082 [55], QX [56], and Sn100pn [57]; details are in Supplemental Material [42]. The value of the ultrasoft NME is

$$\bar{C}_{\text{SP}}^{\text{usoft}} = (67 \pm 28) d_p \text{ (e fm)}^{-1}, \quad (10)$$

where $d_p = \bar{d}_0 + \bar{d}_1$. We only find sensitivity to the proton EDM because, in our calculation, the 82 neutrons form a closed shell, as ^{138}Ba is magic in neutrons. This is also why the first intermediate 1^+ excited state appears around 2.5 MeV. The largest contribution to the ultrasoft NME arises from states around $E_n = 4.5 \text{ MeV} \gg m_e$, justifying our approximation, and higher-energy states only contribute mildly. We show the cumulative contribution from the excited-state spectrum in Supplemental Material [42].

The potential contribution evaluates to

$$\bar{C}_{\text{SP}}^{\text{pot}} = [(-433 \pm 5)d_p + (387 \pm 0.4)d_n] \text{ (e fm)}^{-1}, \quad (11)$$

where $d_n = \bar{d}_0 - \bar{d}_1$. Compared to the ultrasoft regime, the potential contribution is dominant. This is because of the

coherent nature of the potential NME, which leads to a linear scaling with the total number of protons, Z (d_p term), or neutrons, N (d_n term), in the nucleus. The coherence appears as most NME contributions stem from proton-proton and neutron-neutron spin-zero pairs, prevalent in nuclei due to the attractive pairing interaction, and also dominant for the long-range potential (proportional to $|\mathbf{r}_i - \mathbf{r}_j|$ in coordinate space). This scaling is in rough agreement with the estimate of Ref. [15], as well as an evaluation of the potential of Eq. (5) in a Fermi gas state [58]. Our many-body calculations, which also cover nuclei lighter than ^{138}Ba , suggest that nucleus-dependent effects can correct this estimate by up to 20%. The coherent character makes potential NMEs less dependent on the details of the nuclear structure, reducing their relative uncertainty with respect to ultrasoft NMEs (the very small error in the d_n potential NME is because in our calculation the 82 neutrons form a closed shell). We provide more details in Supplemental Material [42]. While we do not expect any breakdown of the scaling behavior discussed above for the potential contribution, in nuclei with particularly strong low-lying M1 excitations, the ultrasoft contributions could be enhanced [59]. Explicit calculations of ultrasoft and potential NMEs in heavier systems such as Th or Hf would be very interesting.

The uncertainties in Eqs. (10) and (11) were estimated by considering the differences between several nuclear Hamiltonians, see Supplemental Material [42] for details. In addition, a fully quantified theory error should take into account the errors due to the many-body method and missing orders in chiral EFT. The latter are expected to give $\mathcal{O}(m_\pi/\Lambda_\chi)$ corrections, while more solid estimates require explicit calculations, e.g., by computing the corrections to the potential in Eq. (5). We leave a more complete analysis of theory uncertainties to future work.

The expected sensitivity of the BaF experiment is an electron EDM equivalent of $d_e \leq 10^{-30} \text{ e cm}$ [8]. Using the NME calculations of this Letter, this would correspond to a sensitivity to the nucleon EDMs $|d_p|_{\text{BaF}} < 8.4 \times 10^{-24} \text{ e cm}$ and $|d_n|_{\text{BaF}} < 8 \times 10^{-24} \text{ e cm}$. While we do not have shell-model calculations for the most precise experiment based on HfF^+ , we can use the linear Z and N dependence of the potential NME, where nucleus-dependent corrections are expected at the $\sim 20\%$ level, with additional higher-order chiral corrections of $\mathcal{O}(m_\pi/\Lambda_\chi)$, to estimate

$$|d_p|_{\text{HfF}^+} \lesssim 1.6 \times 10^{-23} \text{ e cm}, \quad |d_n|_{\text{HfF}^+} \lesssim 1.6 \times 10^{-23} \text{ e cm}, \quad (12)$$

roughly two orders of magnitude weaker than the proton EDM limit set by ^{199}Hg [60] and three orders than the direct neutron EDM limit [14]. Both of these limits have been stable within an order of magnitude in the last decade. Based on the prospects for direct neutron EDM experiments [11],

an order of magnitude improvement can be expected. Experiments with radioactive molecules could provide very stringent limits, but are still under development [12]. Considering the past and anticipated progress in paramagnetic polar molecular EDM experiments, with projected improvements of two to three orders of magnitude within a decade [11], the precision gaps in the limits on $|d_p|$ and $|d_n|$ are not insurmountable.

Discussion—A nonzero EDM measurement raises the question of the underlying CP -violating mechanism. Ratios of paramagnetic systems disentangle electron EDM from CP -odd electron-nucleon contributions [61,62], and nuclear-to-nucleon EDM ratios separate quark-gluon sources [63]. We devise a complementary strategy to identify the underlying source of CP violation. Besides the diagrams calculated in this Letter, there appear contributions from meson-exchange diagrams [15,17]. Depending on the underlying CP -violating source, the ratio of meson to nucleon EDM contributions varies. We can be most concrete for the QCD $\bar{\theta}$ term, where the sizes of the low-energy constants in Eq. (1) are known relatively well [64–66]. For BaF, the meson diagrams give a contribution $\bar{C}_{\text{SP}}^{\text{meson}}(\text{BaF}) = (220 \pm 62) \times 10^{-2} \bar{\theta}$ [17]. Inserting lattice-QCD nucleon EDMs [67,68] gives

$$\bar{C}_{\text{SP}}^{\text{pot+usoft}} = -(196 \pm 54) \times 10^{-2} \bar{\theta}, \quad (13)$$

comparable in magnitude to the meson-exchange diagrams but with *opposite* sign—an accidental cancellation specific to the $\bar{\theta}$ term. We combine all contributions to compute the *equivalent* electron EDM $d_e^{\text{equiv}} \equiv r_{\text{mol}} \bar{C}_{\text{SP}} / A$ [69], which is convenient as paramagnetic EDM searches are usually interpreted as limits on d_e . For BaF, $r_{\text{BaF}} = 4.46[18] \times 10^{-21} \text{ e cm}$ [70], and we obtain $d_e^{\text{equiv}}(\bar{\theta}) = (7.5 \pm 27) \times 10^{-24} \bar{\theta} \text{ e cm}$.

For other hadronic sources of CP violation, power counting arguments give insight into the ratio of meson-to-nucleon-EDM contributions. For example, the quark chromo-EDM breaks CP and isospin symmetry. The meson-nucleon interactions in Eq. (1) are the leading CP -violating hadronic interactions [22,71] and their contributions dominate the paramagnetic EDMs. On the other hand, for quark EDMs and the Weinberg operator the mesonic interactions are suppressed by, respectively, α/π (electromagnetic suppression) and m_π^2/Λ_χ^2 (chiral suppression) [22]. As such, the ratio of $\bar{C}_{\text{SP}}^{\text{eff}}$ (and thus d_e^{equiv}) to the neutron EDM is different. We illustrate this in Fig. 2, where we plot the d_e^{equiv} , including both mesonic and nucleon EDM contributions, against d_n . The bands correspond to scenarios where d_e^{equiv} and d_n are sourced, respectively, by the $\bar{\theta}$ term, the up quark EDM d_u , and the up quark chromo-EDM $\tilde{d}_u$. Remarkably, the ratio of neutron to *paramagnetic* EDMs can identify the underlying hadronic source of CP violation. We emphasize that this identification is

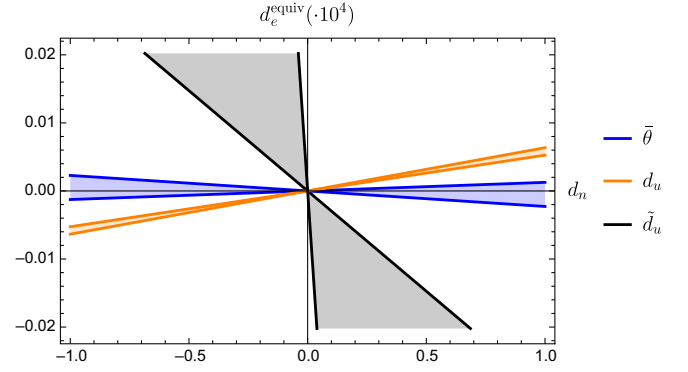


FIG. 2. Ratio d_e^{equiv}/d_n as a function of d_n (in arbitrary units), for three underlying CP -violating sources: the $\bar{\theta}$ term (blue band), the up quark EDM d_u (orange) or chromo-EDM $\tilde{d}_u$ (gray). Each band shows the predicted ratio assuming a given source; the width reflects hadronic uncertainties. The QCD matrix elements of Refs. [71–73] were used to connect the CP -violating sources to the hadronic couplings, $d_{n,p}$ and $\bar{g}_{0,1,0\eta}$.

independent of the numerical value of the CP -odd source, be it $\bar{\theta}$, d_q , or $\tilde{d}_q$, as these drop out in the ratio. As an example, consider a scenario where d_n is measured at 10^{-27} e cm in a future experiment. Figure 2 shows that if $\tilde{d}_u$ is the underlying CP -odd source, we would expect d_e^{equiv} to be negative and of order 10^{-32} – 10^{-33} e cm , while d_u should lead to a positive d_e^{equiv} of order 10^{-34} e cm , and $\bar{\theta}$ to even smaller values.

Conclusions—The EFT approach presented in this Letter allows one to derive the contribution from nucleon EDMs to paramagnetic EDMs in a systematic way. We have identified the novel NMEs needed to interpret paramagnetic EDMs in terms of hadronic CP violation. While power counting arguments indicate that similar contributions would arise from potential and ultrasoft virtual photons, explicit shell-model calculations show that potential NMEs dominate because of the coherent contribution of most protons and neutrons in the nucleus. Experimental improvements of two to three orders of magnitude in paramagnetic molecular systems are needed to set competitive limits on nucleon EDMs. Finally, we have shown that ratios of paramagnetic-to-neutron EDMs point toward the underlying mechanism of CP violation.

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Data availability—The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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