DEUTSCHES ELEKTRONEN-SYNCHROTRON
Ein Forschungszentrum der Helmholtz-Gemeinschaft

DESY 20-183
arXiv:2010.13760
October 2020

Hunting for the CP Violating ALP

L. Di Luzio

Deutsches Elektronen-Synchrotron DESY, Hamburg

R. Gröber, P. Paradisi

Dipartimento di Fisica e Astronomia 'G. Galilei',
Università di Padova, Italy
and
Istituto Nazionale di Fisica Nucleare, Sezione di Padova, Italy

ISSN 0418-9833

NOTKESTRASSE 85 - 22607 HAMBURG

Hunting for the CP violating ALP

Luca Di Luzio,[1] Ramona Gröber,[2,3] and Paride Paradisi[2,3]

[1]*DESY, Notkestraße 85, D-22607 Hamburg, Germany*
[2]*Dipartimento di Fisica e Astronomia 'G. Galilei', Università di Padova, Italy*
[3]*Istituto Nazionale Fisica Nucleare, Sezione di Padova, I–35131 Padova, Italy*

The impact of axion-like particles (ALPs) on the search of permanent electric dipole moments (EDMs) of molecules, atoms, nuclei and nucleons is systematically investigated. We classify first the full set of CP-violating Jarlskog invariants emerging in the ALP effective field theory (EFT) containing operators up to dimension-5. Then, we evaluate the leading short-distance effects to the EDMs up to two-loop order. The high sensitivity of EDMs to CP-violating ALP interactions is emphasised exploiting both the current and projected experimental sensitivities.

I. Introduction. The lack for heavy new physics (NP) at the LHC, mostly motivated by the weak-scale hierarchy problem, has triggered a shift of paradigm towards alternative scenarios, with new light mediators, that are receiving increasing attention both theoretically and experimentally. NP scenarios with light pseudoscalar bosons, referred to as axion-like-particles (ALPs) [1] are prominent examples. The lightness of ALPs can be naturally explained if they are identified with the pseudo-Nambu-Goldstone bosons of an approximate global symmetry. Interestingly, ALPs can be invoked to address a number of fundamental open questions in particle physics such as the strong CP problem [2], the origin of dark matter [3], as well the flavor [4] and hierarchy [5] problems. Furthermore, various anomalies can be solved by the ALPs such as the longstanding discrepancy of the anomalous magnetic moment of the muon [6, 7], the excess in excited Beryllium decays $^8\mathrm{Be}^* \to {}^8\mathrm{Be} + e^+e^-$ [8–10] and that of electronic recoil events with an energy of $\mathcal{O}(\mathrm{keV})$ observed by the XENON1T collaboration [11, 12].

Since the relation between ALP mass and couplings depends on the specific ultraviolet (UV) completion, it is customary to take a model-independent approach where ALPs are treated as a generalization of the QCD axion, with mass and couplings being free parameters to be probed experimentally. In this framework, ALP interactions with Standard Model (SM) fermions and gauge bosons are described via an effective Lagrangian built with operators up to dimension-5 [13]. This approach still captures general features of a broad class of models.

For ALP masses below the MeV scale, a vast experimental program, intertwined with cosmology and astrophysics, is currently ongoing [1]. That ranges from "wave-like" approaches to ALP searches in the sub-eV region (such as haloscopes, helioscopes and optical/EM setups), to beam-dump experiments stretching up to the GeV scale [14]. Collider experiments have also probed ALP masses ranging from the GeV scale up to the electroweak scale, through searches of ALPs associated production with photons, jets and electroweak gauge bosons [15]. Searches for the exotic, on-shell Higgs and Z decays into ALPs were also shown to probe regions of the parameter space previously unconstrained [16]. Other low-energy observables which are extremely sensitive to ALPs are flavor-changing neutral-currents (FCNC) processes both in the quark [17] and lepton sectors [18]. Indeed, since there is no fundamental reason for the ALP interactions to respect the SM flavor group, ALPs can induce FCNC already at tree level.

Rather surprisingly, CP-violating (CPV) signatures of ALPs received so far much less attention [7, 19]. The CP symmetry is violated as long as ALP couplings to photons entail both $\phi F\tilde{F}$ and ϕFF interactions (where ϕ is the ALP, F the QED field strength tensor and $\tilde{F}$ its dual) and/or if ALP couplings to fermions (f) include both $\phi\bar{f}\gamma_5 f$ and $\phi\bar{f}f$ interactions. As we will see, these conditions require that the global shift symmetry (responsible for the lightness of the ALP) as well as CP are broken in the UV sector and such a symmetry breaking is eventually communicated to the infrared dynamics by some mediators. The required UV dynamics can arise quite naturally in strongly-coupled theories, and in fact an explicit realization is provided by the SM itself. Consider for definiteness the effective interactions of the neutral pion field π^0 below the GeV scale (see e.g. [22]). The role of the ALP is played by π^0, while quark masses are responsible for the breaking of the shift symmetry and the source of CP violation is the QCD θ term. The mediators from the strong sector to the π^0 are instead the electromagnetic (EM) interactions. CP-even pion interactions contain the terms $A_1\frac{\pi^0}{f_\pi}F\tilde{F} + A_2\frac{\partial_\mu\pi^0}{f_\pi}\bar{e}\gamma^\mu\gamma_5 e$ where $A_1 = \frac{\alpha}{4\pi}$ is the Wess-Zumino-Witten term and A_2 is generated radiatively from A_1 via EM interactions so that $A_2 \sim (\frac{\alpha}{4\pi})^2$. CP-odd pion interactions $C_1\frac{\pi^0}{f_\pi}FF + C_2 m_e\frac{\pi^0}{f_\pi}\bar{e}e$ are instead sourced by the QCD θ term $C_1 \sim \theta$ while C_2 is generated radiatively from C_1 via EM and therefore $C_2 \sim \theta\frac{\alpha}{4\pi}$. A strongly coupled dynamics at the scale Λ that resembles the pion dynamics of the SM can hence be conceived in analogy.

Other scenarios providing a strong motivation for the study of a CPV ALP are relaxion models [5] which propose a new solution to the weak-scale hierarchy problem by introducing an ALP field, the relaxion, which scans the Higgs boson mass in the early universe from an initial value comparable to the cutoff scale $M \gg 1$ TeV to the final value of $\mathcal{O}(v)$. The simultaneous presence of the relaxion-Higgs mixing and the relaxion-photon/gluon couplings violates CP.

From the experimental side, there is an extraordinary ongoing program aiming at improving the current limits of permanent electric dipole moments (EDMs) of molecules, atoms, nuclei and nucleons by many orders of magnitude [23]. In the light of the above considerations, we believe that a still-missing comprehensive exploration of the phenomenological implications of a CPV ALP is mandatory. The main motivation of the present work is to fill this gap.

After introducing the ALP effective field theory (EFT) containing operators up to dimension-5, we systematically classify the physical CPV phases of the theory. Then, we evaluate the leading short-distance effects to the EDMs up to two-loop order. This will enable us to highlight the high sensitivity of EDMs to CPV ALP interactions once the current and projected experimental sensitivities on EDMs are exploited.

II. ALPs with CP violating couplings. The most general $SU(3)_c \times U(1)_{\rm em}$ invariant dimension-5 effective Lagrangian accounting for CPV ALP interactions with photons, gluons and SM fermions, reads

$$\mathcal{L}_\phi = e^2 \frac{C_\gamma}{\Lambda} \phi F F + e^2 \frac{\tilde{C}_\gamma}{\Lambda} \phi F \tilde{F} + g_s^2 \frac{C_g}{\Lambda} \phi G G + g_s^2 \frac{\tilde{C}_g}{\Lambda} \phi G \tilde{G} + \frac{v}{\Lambda} y_S^{ij} \phi \bar{f}_i f_j + i \frac{v}{\Lambda} y_P^{ij} \phi \bar{f}_i \gamma_5 f_j \,, \quad (1)$$

where Λ is the EFT cutoff, $f \in (e, u, d)$ denotes SM fermions in the mass basis, i, j are flavor indices and, by construction, the matrices y_S and y_P are hermitian. F and G stand for the QED and QCD field strenght tensors, respectively, while $\tilde{F}$ and $\tilde{G}$ are their duals. Note that the last two terms in eq. (1) can be written in a shift-symmetric way through the dimension-5 operators $\frac{\partial_\mu \phi}{\Lambda} \bar{f} \gamma^\mu f$ (only for flavor off-diagonal components) and $\frac{\partial_\mu \phi}{\Lambda} \bar{f} \gamma^\mu \gamma_5 f$ upon integrating by parts and applying the equations of motion. This justifies the normalization factor $\frac{v}{\Lambda}$ (with $v = 246$ GeV) which traces back their effective origin from dimension-5 operators. Instead all the other terms in eq. (1) break the ϕ shift symmetry. Hereafter, we assume that $m_\phi \gtrsim 1$ GeV so that QCD can be treated perturbatively. Moreover, although we take $\Lambda \gtrsim 1$ TeV, we focus only on EM and strong interactions as weak interactions play a subleading role in our analysis. Finally, in eq. (1) we factor out the gauge couplings e^2 and g_s^2 in order to make the coefficients $C_{\gamma,g}$ and $\tilde{C}_{\gamma,g}$ scale invariant at one-loop order.

Since the operators XX and $X\tilde{X}$ ($X = F, G$), as well as scalar and pseudoscalar operators, have opposite CP transformation properties, it is clear that $\mathcal{L}_\phi$ violates CP irrespectively of the CP nature of the spin-0 field ϕ. In particular, the full set of CPV Jarlskog invariants of our ALP EFT emerging at dimension-6 level reads

$$C_a \tilde{C}_b, \quad y_S^i \tilde{C}_a, \quad y_P^i C_a, \quad y_S^i y_P^j, \quad y_S^{ik} y_{\rm SM}^k y_P^{ki} \,, \quad (2)$$

where $a, b = \gamma, g$ and the double superscript "ik" is specified only for flavour off-diagonal Yukawa couplings. Notice that only the last invariant of eq. (2) is sensitive to flavor-violating effects. Moreover, as we will see, at the two-loop level all the above invariants will be generated.

III. Effective Lagrangian for EDMs. The leading low-energy CPV Lagrangian relevant for EDMs of molecules, atoms, nuclei and nucleons reads [24]

$$\mathcal{L}_{\rm CPV} = \sum_{i,j=u,d,e} C_{ij} (\bar{f}_i f_i)(\bar{f}_j i\gamma_5 f_j) + \alpha_s C_{Ge}\, GG\, \bar{e} i\gamma_5 e + \alpha_s C_{\tilde{G}e}\, G\tilde{G}\, \bar{e}e - \frac{i}{2} \sum_{i=u,d,e} d_i \bar{f}_i (F\cdot\sigma)\gamma_5 f_i - \frac{i}{2} \sum_{i=u,d} g_s d_i^C \bar{f}_i (G\cdot\sigma)\gamma_5 f_i + \frac{d_G}{3} f^{abc} G^a \tilde{G}^b G^c \,, \quad (3)$$

where we omitted color-octet 4-quark operators (as they are induced only at one-loop level in the ALP framework) and the dim-4 $G\tilde{G}$ operator. The latter is assumed to be absent thanks to a UV mechanism solving the strong CP problem. Within our EFT, C_{ij}, C_{Ge} and $C_{\tilde{G}e}$ are generated by the Feynman diagrams of fig. 1 and read

$$C_{ij} \simeq \frac{v^2}{\Lambda^2} \frac{y_S^{ii} y_P^{jj}}{m_\phi^2} \,, \qquad C_{Ge} = \frac{4\pi}{m_\phi^2} \frac{v}{\Lambda^2} C_g y_P^e \,, \quad (4)$$

while $C_{\tilde{G}e} \leftrightarrow C_{Ge}$ via the replacement $C_g y_P^e \leftrightarrow \tilde{C}_g y_S^e$. The leptonic (pseudo)scalar couplings, including one-loop EM corrections, are given by

$$y_S^\ell \simeq y_S^\ell(\Lambda) + \frac{6\alpha}{\pi} \frac{m_\ell}{v} e^2 C_\gamma \log \frac{\Lambda}{m_\phi} \,, \quad (5)$$

$$y_P^\ell \simeq y_P^\ell(\Lambda) - \frac{6\alpha}{\pi} \frac{m_\ell}{v} e^2 \tilde{C}_\gamma \log \frac{\Lambda}{m_\phi} \,. \quad (6)$$

In the quark sector, also QCD interactions are at work and we obtain the following leading order result

$$y_S^q \simeq y_S^q(\Lambda) + \frac{m_q}{v} \left(\frac{6\alpha}{\pi} Q_q^2 e^2 C_\gamma + \frac{8\alpha_s}{\pi} g_s^2 C_g \right) \log \frac{\Lambda}{m_\phi} \,, \quad (7)$$

$$y_P^q \simeq y_P^q(\Lambda) - \frac{m_q}{v} \left(\frac{6\alpha}{\pi} Q_q^2 e^2 \tilde{C}_\gamma + \frac{8\alpha_s}{\pi} g_s^2 \tilde{C}_g \right) \log \frac{\Lambda}{m_\phi} \,, \quad (8)$$

for $q = u, d, s$. Note that QED (QCD) loop-corrections in eqs. (5)-(8) are significantly larger than the expectations of naive dimensional analysis. Top and bottom contributions are taken into account by means of the QCD trace anomaly in the gluon-gluon-ALP vertex after they have been integrated out (see fig. 1). The resulting effect is

$$g_s^2 C_g \simeq g_s^2 C_g(\Lambda) + \frac{\alpha_s}{12\pi} \sum_{q=t,b} \frac{v y_S^q}{m_q} \,, \quad (9)$$

$$g_s^2 \tilde{C}_g \simeq g_s^2 \tilde{C}_g(\Lambda) - \frac{\alpha_s}{8\pi} \sum_{q=t,b} \frac{v y_P^q}{m_q} \,, \quad (10)$$

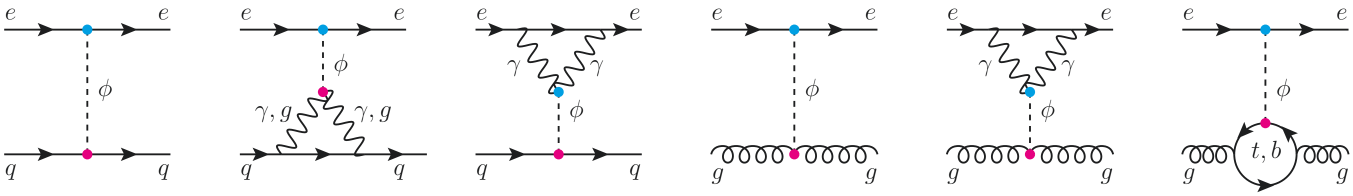

FIG. 1. Leading contributions to the semi-leptonic nucleon-electron operators. The combination of light-blue and purple blobs refer to CPV effective interaction vertices.

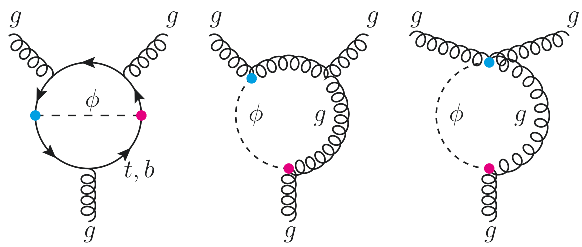

FIG. 2. Leading contributions to the Weinberg operator.

in agreement with the Higgs low-energy theorem [25].

The last term of eq. (3) refers to the Weinberg operator which is generated by the representative diagrams shown in fig. 2. The related Wilson coefficient d_G reads

$$d_G \simeq \frac{g_s \alpha_s}{(4\pi)^3} \sum_{i=t,b} \frac{v^2}{\Lambda^2} \frac{y_S^i y_P^i}{4 m_i^2} + \frac{3 g_s}{\pi^2} \frac{g_s^4 C_g \tilde{C}_g}{\Lambda^2} \log \frac{\Lambda}{m_\phi} \,, \quad (11)$$

where in the first term, which refers to the two-loop diagram, it is assumed that $m_\phi \ll m_{t,b}$. Instead, the second term of eq. (11) arises from the one-loop diagrams of fig. 2 and enjoy a very large enhancement factor with respect to the naive dimensional analysis expectation. As a result, we anticipate that d_G will provide the by far dominant effects to EDMs as induced by $C_g \tilde{C}_g$.

Finally, we analyse the fermionic (C)EDMs induced by ALP interactions. The leading contributions stem from the Feynman diagrams reported in fig. 3 and read

$$\begin{aligned}\frac{d_i}{e} \simeq &- \sum_{k>i} \frac{Q_k}{16\pi^2} \frac{m_k}{m_\phi^2} \frac{v^2}{\Lambda^2} \mathrm{Re}(y_S^{ik} y_P^{ki}) \frac{(3 - 4x_k + x_k^2 + 2\log x_k)}{(1-x_k)^3} \\ &- \sum_k \frac{N_c^k \alpha Q_i Q_k^2}{8\pi^3 m_k} \frac{v^2}{\Lambda^2} \left(y_P^i y_S^k f(x_k) + y_S^i y_P^k g(x_k) \right) \\ &- \frac{Q_i}{2\pi^2} \frac{v}{\Lambda^2} e^2 (y_S^i \tilde{C}_\gamma - C_\gamma y_P^i) \log \frac{\Lambda}{m_\phi} \\ &- \frac{3\alpha Q_i^3}{\pi^3} \frac{m_i}{\Lambda^2} e^4 C_\gamma \tilde{C}_\gamma \log^2 \frac{\Lambda}{m_\phi} \\ &- \delta_{qi} \frac{2\alpha_s Q_i}{\pi^3} \frac{m_i}{\Lambda^2} e^2 g_s^2 (C_\gamma \tilde{C}_g + C_g \tilde{C}_\gamma) \log^2 \frac{\Lambda}{m_\phi} \,, \quad (12)\end{aligned}$$

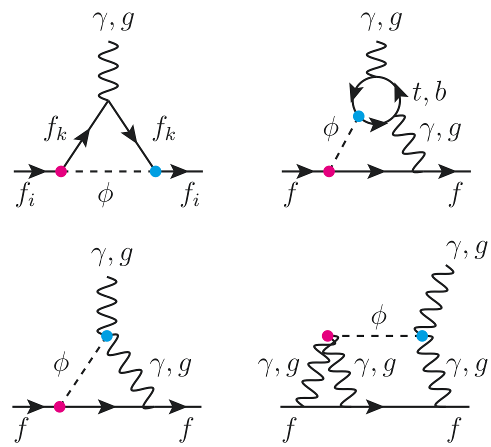

FIG. 3. Leading contributions to the fermionic (C)EDMs.

in the EDMs case (where $i = e, u, d$ and $q = u, d$) and

$$\begin{aligned}d_i^C \simeq &- \sum_{k>i} \frac{1}{16\pi^2} \frac{m_k}{m_\phi^2} \frac{v^2}{\Lambda^2} \mathrm{Re}(y_S^{ik} y_P^{ki}) \frac{(3 - 4x_k + x_k^2 + 2\log x_k)}{(1-x_k)^3} \\ &- \sum_k \frac{\alpha_s}{16\pi^3 m_k} \frac{v^2}{\Lambda^2} \left(y_P^i y_S^k f(x_k) + y_S^i y_P^k g(x_k) \right) \\ &- \frac{1}{2\pi^2} \frac{v}{\Lambda^2} g_s^2 (y_S^i \tilde{C}_g - C_g y_P^i) \log \frac{\Lambda}{m_\phi} \\ &- \frac{4\alpha_s}{\pi^3} \frac{m_i}{\Lambda^2} g_s^4 C_g \tilde{C}_g \log^2 \frac{\Lambda}{m_\phi} \\ &- \frac{3\alpha Q_i^2}{2\pi^3} \frac{m_i}{\Lambda^2} e^2 g_s^2 (C_\gamma \tilde{C}_g + C_g \tilde{C}_\gamma) \log^2 \frac{\Lambda}{m_\phi} \,, \quad (13)\end{aligned}$$

for the CEDMs (where $i = u, d$). The loop functions are $f(x) \approx (6\log x + 13)/18$ and $g(x) \approx (\log x + 2)/2$, in the asymptotic limit $x \gg 1$ where $x_k = m_k^2/m_\phi^2$. While the contributions to the electron EDM stemming from the third and fourth diagrams of fig. 3 were already considered in [7], the expressions of quark (C)EDMs are new. Moreover, we also consider here flavor-violating effects for the first diagram and Barr-Zee two-loop contributions (second diagram). The well-known flavour-diagonal effects for the first diagram can be found instead in [26].

Although eqs. (5)-(13) capture only the leading-order short-distance effects of our ALP model, the bounds of Table I have been obtained taking into account also one-loop QCD running effects (improved with a two-loop running of α_s and quark masses) from $\Lambda = 1$ TeV down to m_ϕ, which is assumed to coincide with the hadronic scale $\mu_{\rm had} = 1$ GeV. For $m_\phi > 1$ GeV, the running of $\mathcal{L}_{\rm CPV}$ down to $\mu_{\rm had}$ should be also included.

Constraints on d_e, as well as on the coefficients C_{ij} and C_{Ge} in eq. (3), are set by using the polar molecule ThO. The electron spin-precession frequency receives contributions from both d_e and CP-odd electron–nucleon (N) interactions $\mathcal{L} \supset -\frac{G_F}{\sqrt{2}} C_S \bar{N} N \bar{e} i\gamma_5 e$ [27]

$$\omega_{\rm ThO} = 1.2\,{\rm mrad/s}\left(\frac{d_e}{10^{-29}\,e\,{\rm cm}}\right) + 1.8\,{\rm mrad/s}\left(\frac{C_S}{10^{-9}}\right)$$

with a theoretical error of few percent and the experimental limit $\omega_{\rm ThO} < 1.3\,{\rm mrad/s}$ (90% C.L.) [28]. The coefficient C_S is related to C_{ij} and C_{Ge} as $C_S/v^2 \simeq -17(C_{ue} + C_{de}) + 4.7\,{\rm GeV}\,C_{Ge}$. The neutron EDM is induced by quark (C)EDMs, the Weinberg operator and 4-quark operators [29–31]

$$\begin{aligned} d_n &= 0.784(28)\,d_u - 0.204(11)\,d_d - 0.55(28)\,e\,d_u^C \\ &\quad - 1.10(55)\,e\,d_d^C + 50(40)\,{\rm MeV}\,e\,d_G \\ &\quad + 30(30)\,{\rm MeV}\,e\,(C_{ud} - C_{du})\,, \end{aligned} \qquad (14)$$

while the experimental bound is $d_n < 1.8 \cdot 10^{-26}\,e$ cm (90% C.L.) [32]. The EDM of the diamagnetic atom ^{199}Hg gets contributions from both nuclear and leptonic CP-odd interactions [27, 30]

$$d_{\rm Hg} \simeq 4.0 \cdot 10^{-4} d_n - [2.8\,C_S - 2.1\,C_P]\,10^{-22}\,e\,{\rm cm} \qquad (15)$$

with $C_P \simeq C_P^{(0)} - C_P^{(1)}$ defined in terms of the Lagrangian $\mathcal{L} \supset -\frac{G_F}{\sqrt{2}} \bar{N}(C_P^{(0)} + \tau_3 C_P^{(1)}) i\gamma_5 N \bar{e}e$. To set bounds we employ $C_P/v^2 = 350(C_{eu} + C_{ed}) + 1.1\,{\rm GeV}\,C_{\tilde{G}e}$ and the experimental limit $d_{\rm Hg} < 6 \cdot 10^{-30} e$ cm (90% C.L.) [33].

IV. Probing ALPs with EDMs. The sensitivity of physical EDMs to the CPV invariants classified in eq. (2) are reported in Table I, where we employed central values for theoretical predictions. While the bounds on $|C_\gamma \tilde{C}_\gamma|$ and $|y_S^e \tilde{C}_\gamma - y_P^e C_\gamma|$ were already studied in [7], all the other bounds are new. As shown in Table I, 4-fermion operators provide the most stringent bounds on several CP invariants. Similarly, the Weinberg operator sets tight limits on ALP couplings to the top and bottom quarks as well as to gluons which were previously unconstrained. Finally, we remark that also flavor-violating contributions to the EDMs are quite effective. Indeed, despite the suppression arising from flavor mixing angles –which are otherwise constrained by FCNC processes– (C)EDMs enjoy a chiral enhancement m_k/m_i, for $k > i$, which is absent in the case of flavor-conserving interactions.

CPV invariant	Bound	Observable
$\lvert C_\gamma \tilde{C}_\gamma\rvert$	6.2×10^{-3}	$\omega_{\rm ThO}(d_e)$
$\lvert C_g \tilde{C}_g\rvert$	8.5×10^{-7}	$d_n, d_{\rm Hg}(d_G)$
$\lvert C_\gamma \tilde{C}_g\rvert$	4.6×10^{-2}	$d_{\rm Hg}(C_P)$
$\lvert C_g \tilde{C}_\gamma\rvert$	1.8×10^{-4}	$\omega_{\rm ThO}(C_S)$
$\lvert y_S^e \tilde{C}_\gamma - y_P^e C_\gamma\rvert$	7.1×10^{-11}	$\omega_{\rm ThO}(d_e)$
$\lvert y_S^u \tilde{C}_g - y_P^u C_g\rvert$	9.0×10^{-9}	$d_n, d_{\rm Hg}(d_u^C)$
$\lvert y_S^d \tilde{C}_g - y_P^d C_g\rvert$	4.6×10^{-9}	$d_n, d_{\rm Hg}(d_d^C)$
$\lvert y_P^e C_g\rvert$	2.3×10^{-12}	$\omega_{\rm ThO}(C_S)$
$\lvert y_S^e \tilde{C}_g\rvert$	1.5×10^{-10}	$d_{\rm Hg}(C_P)$
$\lvert y_S^u y_P^d - y_S^d y_P^u\rvert$	5.0×10^{-10}	$d_n, d_{\rm Hg}(C_{ud} - C_{du})$
$\lvert y_S^e y_P^u\rvert$, $\lvert y_S^e y_P^d\rvert$	2.3×10^{-14}	$d_{\rm Hg}(C_P)$
$\lvert y_S^u y_P^e\rvert$, $\lvert y_S^d y_P^e\rvert$	1.1×10^{-14}	$\omega_{\rm ThO}(C_S)$
$\lvert y_S^t y_P^e\rvert$	2.7×10^{-10}	$\omega_{\rm ThO}(C_S)$
$\lvert y_S^b y_P^e\rvert$	6.6×10^{-12}	$\omega_{\rm ThO}(C_S)$
$\lvert y_S^e y_P^t\rvert$	6.6×10^{-9}	$\omega_{\rm ThO}(d_e)$
$\lvert y_S^e y_P^b\rvert$	8.1×10^{-10}	$d_{\rm Hg}(C_P)$
$\lvert y_S^t y_P^t\rvert$	0.22	$d_n, d_{\rm Hg}(d_G)$
$\lvert y_S^b y_P^b\rvert$	1.2×10^{-4}	$d_n, d_{\rm Hg}(d_G)$
$\lvert y_S^e\, y_P^e\rvert$	1.0×10^{-10}	$\omega_{\rm ThO}(d_e)$
$\lvert y_S^{e\mu}\, y_P^{\mu e}\rvert$	2.2×10^{-12}	$\omega_{\rm ThO}(d_e)$
$\lvert y_S^{e\tau}\, y_P^{\tau e}\rvert$	3.1×10^{-12}	$\omega_{\rm ThO}(d_e)$
$\lvert y_S^u\, y_P^u\rvert$	1.1×10^{-7}	$d_n, d_{\rm Hg}(d_u^C)$
$\lvert y_S^{uc}\, y_P^{cu}\rvert$	2.2×10^{-8}	$d_n, d_{\rm Hg}(d_u^C)$
$\lvert y_S^{ut}\, y_P^{tu}\rvert$	2.2×10^{-6}	$d_n, d_{\rm Hg}(d_u^C)$
$\lvert y_S^d\, y_P^d\rvert$	2.3×10^{-8}	$d_n, d_{\rm Hg}(d_d^C)$
$\lvert y_S^{ds}\, y_P^{sd}\rvert$	3.4×10^{-9}	$d_n, d_{\rm Hg}(d_d^C)$
$\lvert y_S^{db}\, y_P^{bd}\rvert$	9.4×10^{-9}	$d_n, d_{\rm Hg}(d_d^C)$

TABLE I. Bounds on CPV invariants for $\Lambda = 1$ TeV and $m_\phi = 1$ GeV. In the 3rd column we specify the observable and (in brackets) the leading operator setting the bound.

Although the relative importance of the above contributions to the physical EDMs will depend on the relative strength of the microscopic parameters $C_{\gamma(g)}$, $\tilde{C}_{\gamma(g)}$, y_S and y_P and therefore on the specific ALP model, let us consider as an example the case where $y_{S,P}^{ii} \propto \frac{m_i}{v}$, $C_{\gamma(g)}$ and $\tilde{C}_{\gamma(g)} \propto \frac{1}{16\pi^2}$. In such a setup it turns out that 4-fermion operators are the by far best probes of CP violation followed by the electron EDM and the Weinberg operator which have comparable sensitivities, as it can be easily checked from Table I.

The expected sensitivities of future EDM experiments will greatly improve the current resolutions. The neutron EDM measurement should be improved by more than two orders of magnitude, $d_n \lesssim 10^{-28}e$ cm [23]. There are also plans to measure the EDMs of charged nuclei such as the proton and deuteron in EM storage rings with expected resolutions of $d_{p,D} \lesssim 10^{-29}e$ cm [23]. Moreover, we expect also one order of magnitude improvement on the current measurement of molecular systems, such as the polar molecule ThO, which give rise to the most stringent constraints on the electron EDM and electron-nucleon couplings. If this is the case, the bounds on d_G and quark (C)EDMs will improve by roughly three orders of magnitude while the bounds on the electron EDM and 4-fermion contributions will become one order of magnitude more stringent.

V. Conclusions. In this Letter, we have studied for the first time the full set of contributions of ALPs to permanent EDMs of molecules, atoms, nuclei and nucleons. After classifying the CPV Jarlskog invariants emerging in the ALP EFT, we have evaluated the leading short-distance effects to EDMs up to two-loop order. Our main result is that 4-fermion and Weinberg operators, so far neglected in the literature, provide by far the largest contributions to the EDMs.

Our work can be generalised in several directions. For instance, it would be interesting to extend our analysis to ALP masses in the sub-GeV region where QCD cannot be treated perturbatively. Moreover, it could be worth to investigate possible UV completions of our EFT that resemble the strong dynamics of the pion in the SM. Furthermore, since relaxion models addressing the hierarchy problem share many similarities with our EFT, it would be interesting to check whether these scenarios can be probed by means of the new EDM observables studied here. Finally, another ambitious project would be to investigate whether a successful baryogenesis can be driven by CPV ALP interactions.

In summary, a CPV ALP can be related to many fundamental open questions in particle physics. It is very exciting that the outstanding experimental progress, which is expected in the next years, on the searches for permanent EDMs will likely shed light on some of them.

Acknowledgments. We thank Stefano Bertolini, Martin Jung and Fabrizio Nesti for useful discussions. The work of LDL is supported by the Marie Skłodowska-Curie Individual Fellowship grant AXIONRUSH (GA 840791) and the Deutsche Forschungsgemeinschaft under Germany's Excellence Strategy - EXC 2121 Quantum Universe - 390833306.

[1] J. Jaeckel and A. Ringwald, Ann. Rev. Nucl. Part. Sci. **60**, 405 (2010); D. J. E. Marsh, Phys. Rept. **643**, 1-79 (2016); I. G. Irastorza and J. Redondo, Prog. Part. Nucl. Phys. **102** (2018) 89; L. Di Luzio, M. Giannotti, E. Nardi and L. Visinelli, Phys. Rept. **870**, 1-117 (2020).

[2] R. D. Peccei and H. R. Quinn, Phys. Rev. Lett. **38**, 1440 (1977); Phys. Rev. D **16**, 1791-1797 (1977); S. Weinberg, Phys. Rev. Lett. **40**, 223 (1978); F. Wilczek, Phys. Rev. Lett. **40**, 279 (1978).

[3] J. Preskill, M. B. Wise and F. Wilczek, Phys. Lett. **120B**, 127 (1983); L. F. Abbott and P. Sikivie, Phys. Lett. **120B**, 133 (1983); M. Dine and W. Fischler, Phys. Lett. **120B**, 137 (1983); R. L. Davis, Phys. Lett. B **180**, 225 (1986).

[4] A. Davidson and K. C. Wali, Phys. Rev. Lett. **48** (1982) 11; F. Wilczek, Phys. Rev. Lett. **49**, 1549 (1982); Y. Ema, K. Hamaguchi, T. Moroi and K. Nakayama, JHEP **1701** (2017) 096; L. Calibbi, F. Goertz, D. Redigolo, R. Ziegler and J. Zupan, Phys. Rev. D **95**, no. 9, 095009 (2017).

[5] P. W. Graham, D. E. Kaplan and S. Rajendran, Phys. Rev. Lett. **115**, no. 22, 221801 (2015).

[6] D. Chang, W. F. Chang, C. H. Chou and W. Y. Keung, Phys. Rev. D **63** (2001) 091301.

[7] W. J. Marciano, A. Masiero, P. Paradisi and M. Passera, Phys. Rev. D **94** (2016) no.11, 115033.

[8] A. J. Krasznahorkay *et al.*, Phys. Rev. Lett. **116** (2016) no.4, 042501.

[9] J. L. Feng, B. Fornal, I. Galon, S. Gardner, J. Smolinsky, T. M. P. Tait and P. Tanedo, Phys. Rev. D **95** (2017) no.3, 035017.

[10] U. Ellwanger and S. Moretti, JHEP **1611** (2016) 039.

[11] E. Aprile *et al.* [XENON Collaboration], Phys. Rev. D **102**, no.7, 072004, (2020).

[12] F. Takahashi, M. Yamada and W. Yin, Phys. Rev. Lett. **125**, no.16, 161801 (2020); I. M. Bloch, A. Caputo, R. Essig, D. Redigolo, M. Sholapurkar and T. Volansky, arXiv:2006.14521 [hep-ph].

[13] H. Georgi, D. B. Kaplan and L. Randall, Phys. Lett. **169B**, 73 (1986).

[14] B. Döbrich, J. Jaeckel and T. Spadaro, JHEP **05**, 213 (2019) [erratum: JHEP **10**, 046 (2020)].

[15] J. Jaeckel and M. Spannowsky, Phys. Lett. B **753**, 482 (2016); S. Knapen, T. Lin, H. K. Lou and T. Melia, Phys. Rev. Lett. **118**, no. 17, 171801 (2017); I. Brivio, *et al.*, Eur. Phys. J. C **77**, no. 8, 572 (2017); A. Mariotti, D. Redigolo, F. Sala and K. Tobioka, Phys. Lett. B **783** (2018) 13; X. Cid Vidal, A. Mariotti, D. Redigolo, F. Sala and K. Tobioka, JHEP **1901** (2019) 113 Erratum: [JHEP **2006** (2020) 141]; D. Aloni, Y. Soreq and M. Williams, Phys. Rev. Lett. **123** (2019) no.3, 031803; D. Aloni, C. Fanelli, Y. Soreq and M. Williams, Phys. Rev. Lett. **123**, no. 7, 071801 (2019).

[16] M. Bauer, M. Neubert and A. Thamm, JHEP **1712**, 044 (2017); Phys. Rev. Lett. **119** (2017) no.3, 031802.

[17] B. Batell, M. Pospelov and A. Ritz, Phys. Rev. D **83** (2011) 054005; M. B. Gavela, R. Houtz, P. Quilez, R. Del Rey and O. Sumensari, Eur. Phys. J. C **79** (2019) no.5, 369; J. Martin Camalich, M. Pospelov, P. N. H. Vuong, R. Ziegler and J. Zupan, Phys. Rev. D **102**, no.1, 015023 (2020).

[18] M. Bauer, M. Neubert, S. Renner, M. Schnubel and A. Thamm, Phys. Rev. Lett. **124** (2020) no.21, 211803; C. Cornella, P. Paradisi and O. Sumensari, JHEP **2001** (2020) 158; L. Calibbi, D. Redigolo, R. Ziegler and J. Zupan, arXiv:2006.04795 [hep-ph].

[19] CPV interactions have been investigated in the context of long-range forces for the QCD axion [20] and for scalar-pseudoscalar ALP couplings to nucleons and electrons [21].

[20] J. E. Moody and F. Wilczek, Phys. Rev. D **30**, 130 (1984); G. Raffelt, Phys. Rev. D **86**, 015001 (2012); S. Bertolini, L. Di Luzio and F. Nesti, [arXiv:2006.12508 [hep-ph]]; C. A. J. O'Hare and E. Vitagliano, [arXiv:2010.03889 [hep-ph]].
[21] Y. V. Stadnik, V. A. Dzuba and V. V. Flambaum, Phys. Rev. Lett. **120**, no.1, 013202 (2018); V. A. Dzuba, V. V. Flambaum, I. B. Samsonov and Y. V. Stadnik, Phys. Rev. D **98**, no.3, 035048 (2018).
[22] K. Choi and J. y. Hong, Phys. Lett. B **259** (1991) 340.
[23] T. Chupp, P. Fierlinger, M. Ramsey-Musolf and J. Singh, Rev. Mod. Phys. **91**, no.1, 015001 (2019).
[24] M. Pospelov and A. Ritz, Annals Phys. **318** (2005) 119; M. Pospelov and A. Ritz, Phys. Rev. D **63**, 073015 (2001); O. Lebedev, K. A. Olive, M. Pospelov and A. Ritz, Phys. Rev. D **70**, 016003 (2004); J. R. Ellis, J. S. Lee and A. Pilaftsis, JHEP **10**, 049 (2008).
[25] M. A. Shifman, A. I. Vainshtein and V. I. Zakharov, Phys. Lett. B **78**, 443-446 (1978).
[26] C. Y. Chen, H. Davoudiasl, W. J. Marciano and C. Zhang, Phys. Rev. D **93** (2016) no.3, 035006.
[27] W. Dekens, J. de Vries, M. Jung and K. K. Vos, JHEP **1901** (2019) 069.
[28] V. Andreev *et al.* [ACME Collaboration], Nature **562** (2018) no.7727, 355; J. Baron *et al.* [ACME Collaboration], Science **343** (2014) 269.
[29] J. Hisano, K. Tsumura and M. J. S. Yang, Phys. Lett. B **713**, 473-480 (2012).
[30] V. Cirigliano, A. Crivellin, W. Dekens, J. de Vries, M. Hoferichter and E. Mereghetti, Phys. Rev. Lett. **123** (2019) no.5, 051801.
[31] S. Bertolini, A. Maiezza and F. Nesti, Phys. Rev. D **101**, no.3, 035036 (2020).
[32] C. Abel *et al.* [nEDM], Phys. Rev. Lett. **124**, no.8, 081803 (2020); J. M. Pendlebury *et al.*, Phys. Rev. D **92** (2015) no.9, 092003.
[33] B. Graner, Y. Chen, E. G. Lindahl and B. R. Heckel, Phys. Rev. Lett. **116** (2016) no.16, 161601 Erratum: [Phys. Rev. Lett. **119** (2017) no.11, 119901]; W. C. Griffith, M. D. Swallows, T. H. Loftus, M. V. Romalis, B. R. Heckel and E. N. Fortson, Phys. Rev. Lett. **102** (2009) 101601.