Chiral molecules as sensitive probes for direct detection of \( P \)-odd cosmic fields

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Particular advantages of chiral molecules for direct detection of the time-dependence of pseudoscalar CDM particles are highlighted. Such fields are involved in different models for cold dark matter or in the Lorentz-invariance violating standard model extensions and thus are signatures of physics beyond the standard model. The sensitivity of a twenty year old experiment with the molecule CHBrCIF to pseudoscalar cosmic fields as characterized by the parameter \( |b_0| \) is estimated to be \( O(10^{-12}\text{GeV}) \) and allows to predict the sensitivity of future experiments with favorable choices of chiral molecular probes to be \( O(10^{-17}\text{GeV}) \), which will be an improvement of the present best limits by at least two orders of magnitude.

Introduction.—The nature of dark matter (DM), the existence of which is invoked to explain the cosmological motion of visible matter, is considered to be one of the biggest unsolved problems of modern physics (see e.g. Ref. [1]). Among the various DM theories, the cold DM (CDM) variant appears to provide a simple explanation for a wealth of astrophysical observations [2]. Up to now, however, the constituents of CDM are unknown and can range from macroscopic objects such as black holes to new particles like weakly interacting massive particles (WIMPs), axions, sterile neutrinos or dark photons (see e.g. Refs. [3–5]).

The model of CDM has also several shortcomings [6–11]. In order to overcome some of these, so-called fuzzy CDM models, which assume CDM to consist of ultra light particles with masses of \( m_\phi \sim 1 \times 10^{-22}\text{eV}/c^2 \), were proposed [12, 13].

CDM candidates are different types of weakly interacting particles (an overview can be found e.g. in Ref. [14]). Among those, we focus in the following on pseudoscalar and pseudovector particles as they are a source of direct parity (\( P \)) violation.

Pseudoscalar CDM particles behave as axions, which were originally proposed [15–17] as a solution to the so-called strong \( CP \)-problem [18], i.e. the apparently missing \( CP \)-violation in quantum chromodynamics (QCD) although there is a free parameter in QCD that can account for such a violation. The window to search for such particles can be restricted to a defined parameter space, like for the QCD axion (see e.g. [19]) which has to solve the strong \( CP \)-problem, or can be large as for axionic particles that are not bound to solve the strong \( CP \)-problem. The latter are often referred to as axion-like particles (ALPs). Pseudovector fields are important for models such as dark photons [20, 21] and also appear as sources of local Lorentz invariance violation in the Standard Model Extension (SME) [22].

In the last decade many proposals for new experiments and improved bounds on pseudoscalar CDM appeared, some of which employ atomic spectroscopy (see e.g. [23–28]). Among the latter, direct measurement of parity violation with modern atomic precision spectroscopy [26, 29] provided strict limits on static \( P \)-odd fields, where effects of these fields adds to parity violating effects stemming from electroweak electron-nucleus interactions mediated by the \( Z^0 \) boson.

It is well known that such \( P \)-odd effects are strongly enhanced in chiral molecules, as the chiral arrangement of the nuclei leads to helicity in the electron cloud (see e.g. Refs. [30, 31]). This effect can be measured as energy difference between enantiomers of chiral molecules or as resonance frequency differences between the two non-identical mirror-image molecules [32, 33]. As frequency shifts can be measured very accurately, this appears to be a particularly promising tool to search for \( P \)-odd cosmic fields (for recent reviews on molecular parity violation see [30, 31, 34–38]). In the following we show advantages of the use of chiral molecules to search for \( P \)-odd cosmic fields. We estimate the sensitivity on cosmic parity violation of a twenty year old experiment [39] with the chiral methane derivative CHBrCIF [40, 41] and discuss the prospects of modern experiments with chiral molecules.

Theory.—We write the pseudoscalar cosmic field as \( \phi(t) = \phi_0 \cos(\omega_\phi t) \) (see e.g. Ref. [29]), which is supposed to behave non-relativistically \( \hbar \omega_\phi \approx m_\phi c^2 \). The interaction of electrons \( \psi_e \) with such pseudoscalar fields \( \phi(t) \) can be described by the following Lagrangian density (see e.g. [16, 17])

\[
\mathcal{L}_\text{ps}^\phi = g_{\phi\psi_e}(\hbar c \partial_\mu \phi)\bar{\psi}_e \gamma^\mu \gamma^5 \psi_e ,
\]

where \( g_{\phi\psi_e} \) is a coupling constant of dimension \( \text{GeV}^{-1} \). Here the \( 4 \times 4 \) Dirac matrices are defined as

\[
\gamma^0 = \begin{pmatrix} 1_{2\times2} & 0_{2\times2} \\ 0_{2\times2} & -1_{2\times2} \end{pmatrix}, \quad \gamma^k = \begin{pmatrix} 0_{2\times2} & \sigma^k \\ \sigma^k & 0_{2\times2} \end{pmatrix},
\]

where \( \sigma^k \) are the Pauli spin matrices with upper indices \( k = 1, 2, 3 \). The index \( \mu \) runs as \( \mu = 0, 1, 2, 3 \). We define \( \gamma^5 = i\gamma^0 \gamma^1 \gamma^2 \gamma^3 \) with \( i = \sqrt{-1} \) being the imaginary unit. \( \partial_\mu = \frac{\partial}{\partial x^\mu} \) is the first derivative with respect to the four-vector \( x^\mu = (ct, x, y, z) \) and we use Einstein’s sum.
convention here for convenience. The time-derivative of the pseudoscalar field leads to the \( \mathcal{P} \)-odd one-electron Hamiltonian
\[
\hat{h}_\text{ps} = g_{\text{ps}} e \sqrt{2(hc)^3 \rho_{\text{CDM}}} \sin(\omega_c t) \gamma^5, \tag{3}
\]
where \( \rho_{\text{CDM}} \approx \frac{(hc)^3 \rho_{\text{CDM}}}{2(hc)^3} \) is the CDM energy density, for which we assume all ALPs to comprise all of the CDM with a uniform density: \( (hc)^3 \rho_{\text{CDM}} = (hc)^3 0.4 \text{ GeV cm}^{-3} = 7.6 \times 10^{-4} \text{ eV}^4 \) (see Ref. [42]).

Electronic interactions with pseudovector \( \gamma^5 \) cosmic fields can be described by the Lagrangian density
\[
\mathcal{L}^0_{\gamma^5} = -\bf{b}_0^{\mu} \bar{\psi} \gamma^\mu \gamma^5 \psi, \tag{4}
\]
which appears e.g. in the local Lorentz invariance violating Standard Model Extension (SME) (for details see Refs. [22, 43]). The parity non-conserving one-electron interaction Hamiltonian for the temporal component \( \mu = 0 \) is
\[
\hat{h}_{\text{pv}} = b_0^\mu(t) \gamma^5, \tag{5}
\]
where the field can be static \( b_0^\mu(t) = b_0^\mu \) or dynamic \( b_0^\mu(t) = b_0^\mu(t) \sin(\omega_c t) \). Here \( b_0^\mu \) is the interaction strength of the timelike-component of the field with the electrons.

The operators corresponding to electronic interactions with \( \mathcal{P} \)-odd cosmic fields shown above are proportional to \( \gamma^5 \). The electronic expectation value of \( \langle \gamma^5 \rangle \) can be expanded in orders of the fine structure constant \( \alpha \) giving in leading order:
\[
\langle \gamma^5 \rangle \approx \alpha \langle \hat{\sigma} \cdot \hat{p} \rangle, \tag{6}
\]
where \( \hat{p} \) is the electronic linear momentum operator. As \( \hat{\sigma} \cdot \hat{p} \) is an imaginary, electron-spin dependent operator, this expectation value vanishes in the strict electrostatic limit, but it can become non-zero when spin-orbit coupling \( H_{\text{so}} \) is accounted for, similarly to the situation for parity-violation in chiral molecules due to weak neutral currents [44, 45]. Furthermore, it is obvious from eq. (6) that \( \langle \gamma^5 \rangle \) depends on the helicity of the electron cloud. Thus, \( \langle \gamma^5 \rangle \) can be non-zero in a chiral molecule, in which the electrons move in a \( \mathcal{P} \)-odd nuclear potential, whereas in a non-chiral molecule or in an atom \( \langle \gamma^5 \rangle \) vanishes in the absence of additional \( \mathcal{P} \)-odd forces.

It can be shown from perturbation theory that for systems containing two heavy main group elements with nuclear charge numbers \( Z_A \) and \( Z_B \) the following scaling relation holds in lowest order:
\[
\langle \gamma^5 \rangle_{\text{mol}} \sim c_1 \alpha^5 Z_A^2 Z_B^2 + c_2 \alpha^3 Z_A^3 + c_3 \alpha^3 Z_B^3, \tag{7}
\]
Here the factor \( \alpha^2 Z_A^2 \) in the first term emerges from spin-orbit coupling. A detailed derivation together with evidence from numerical studies of several chiral molecules will be provided in a separate publication [46]. From this, and previous studies of the electronic expectation value of \( \gamma^5 \) as a possible total molecular chirality measure [47] (see, however, the critical discussion in Ref. [48] on the utility of pseudoscalar functions as chirality measures), it can be deduced that contributions at the nuclear center dominate the electronic expectation value of \( \gamma^5 \) and let it behave similarly to nuclear-spin independent electroweak electron-nucleon current interactions described by the one-electron Hamiltonian
\[
\hat{h}_{\text{ew}} = \frac{G_F}{2\sqrt{2}} \sum_{A=1}^{N_{\text{nuc}}} Q_W,\alpha A(\vec{r}) \gamma^5 \tag{8}
\]
with \( G_F \) being Fermi’s constant, \( Q_W,\alpha A \) being the weak nuclear charge of nucleus \( A \) with nuclear density distribution \( \rho_A(\vec{r}) \) and the sum running over all \( N_{\text{nuc}} \) nuclei.

Thus, molecular experiments that aim to test parity violation due to weak interactions can also be used for searches of parity violating cosmic fields with a comparable sensitivity.

**Results and Discussion.**—In the following we estimate the expected sensitivity of experiments with chiral molecules to \( \mathcal{P} \)-odd cosmic fields as characterized by the \( b_0^\mu \) parameter from an experiment with CHBrCIF reported by Daussy et. al. [39]. In this experiment the C-F stretching fundamental vibration (\( \nu_4 \)) in enantioenriched samples of CHBrCIF was studied by high-resolution infrared spectroscopy. We are interested in the parity-violating splittings of the vibrational resonance frequency induced by cosmic fields interacting through \( \langle \gamma^5 \rangle \).

Our calculations for CHBrCIF, which are described in more detail in a separate publication [46], were carried out following Ref. [49], which utilized the separable anharmonic adiabatic approximation framework as described in Ref. [50]. Parity-violating molecular properties were computed on the level of two-component ZORA-eGKS with the B3LYP density functional. We reuse electronic densities and Kohn-Sham orbitals as well as vibrational wave functions determined in Ref. [49]. With these, electronic expectation values of \( \gamma^5 = \sum \gamma_i^5 \) and of the nuclear-spin independent electroweak electron-nucleon current interaction term induced by \( \hat{H}_{\text{ew}} = \sum \hat{h}_{\text{ew}}(i) \) were calculated with our ZORA property toolbox approach outlined in Ref. [51]. Vibrational corrections of the properties were computed as described in Ref. [49].

The (negative) outcome of the experimental test for a parity violating frequency shift reported in Ref. [39] is \( |\Delta \nu| = (9.4 \pm 17.9) \text{ Hz} \).

The expectation values of \( \gamma^5 \) for the ground and first excited vibrational states along the C-F stretching mode of (S)-CHBrCIF are computed to be
\[
\langle \gamma^5 | v_4 = 0 \rangle = -8.28 \times 10^{-9} \quad (9)
\]
\[
\langle \gamma^5 | v_4 = 1 \rangle = -7.91 \times 10^{-9} \quad (10)
\]
This leads to an estimate for the splitting between the two enantiomers of CHBrCIF due to the perturbation with \( \gamma^5 \) for the transition between the vibrational ground
and first excited states of $v_4$ of

$$\Delta_{(R,S)} \langle \gamma^5 \rangle = 2 \langle (v_4 = 1 | \gamma^5 | v_4 = 1) \\ - \langle v_4 = 0 | \gamma^5 | v_4 = 0) \rangle \approx 7.4 \times 10^{-10}. \quad (11)$$

As we discuss in more detail in Ref. [46] non-separable anharmonic effects can play a prominent role for the C–F stretching mode in CHBrClF as effects characterized by the first and second derivatives with respect to $q_4$ can be expected to be of the same order as those characterized by first derivatives with respect to $q_R \neq 4$. This can best be seen from a plot of $\langle \gamma^5 \rangle$ on one-dimensional cuts along all modes (see Figure 1). Therein the weak dependence of $\langle \gamma^5 \rangle$ on $q_4$ in comparison to the pronounced dependence on other modes stands out. Therefore, it is not possible to provide a robust theoretical value for $\langle \gamma^5 \rangle$ for the C–F stretching mode, but we give rather the order of magnitude, which is $\Delta_{(R,S)} \langle \gamma^5 \rangle \sim O(10^{-10})$. The sensitivity of this experiment to $b_0^5$ is found to be of the order

$$|b_0^5| \lesssim \frac{12.7 \text{ Hz}}{O(10^{-10})} h \sim O(10^{-12} \text{ GeV}) \quad (12)$$

This sensitivity based on the twenty year old experiment on CHBrClF is about two orders of magnitude inferior to the best limit from modern atomic experiments of $7 \times 10^{-15}$ GeV so far [26]. An improvement in theory, most importantly by consideration of multi-mode effects [46, 52] and additionally by calculations with more sophisticated electronic structure methods, would allow to place a robust limit as we have highlighted in Ref. [46].

The sensitivity of the molecular experiment is supposed to be improvable by two orders of magnitude or better by a different experimental setup as discussed in Refs. [53–55]. The scaling behaviour in (7) suggests that further sensitivity improvements are possible by selecting heavy-elemental chiral molecules. Electroweak $P$-odd effects, which scale like $N_A Z_A^2 Z_\nu^2$ with $N_A$ being the number of neutrons of nucleus $A$, are estimated to give vibrational splittings that can become three orders of magnitude larger in well-chosen heavy-elemental molecules when compared to CHBrClF [49, 54]. Due to the missing $N_A$ scaling, an enhancement by two orders of magnitude can thus be anticipated for $\Delta_{(R,S)} \langle \gamma^5 \rangle$. Furthermore, as indicated in Figure 1 and highlighted in Ref. [46], the sensitivity is improvable by an order of magnitude by choice of a different vibrational transition.

Thus we can estimate that in future $P$-violation experiments with chiral molecules the sensitivity of the 1999 experiment can be improved by at least five orders of magnitude down to $10^{-17}$ GeV, i.e. an improvement of the actual best limit by at least two orders of magnitude. This renders experiments with suitably chosen chiral molecules sensitive probes for physics beyond the Standard Model.

To exploit its full potential, however, a measurement of cosmic parity violation on the background of the larger electroweak frequency splittings would become necessary, which makes additional demands on accuracy of the accompanying computational approaches or calls for experimental schemes to disentangle these two contributions for instance by measuring isotope-dependent electroweak frequency splittings.

The experiment discussed above is sensitive to oscillating $P$-odd interactions of electrons as well. We can exploit the fact that the experiment was performed over a time span of ten days with a well defined set of measurements on each day. In the following we estimate expected sensitivities for this kind of experiments to oscillating pseudoscalar and pseudovector fields. As CHBrClF is not an optimal choice, we do not aim to determine the best possible limit from the actual experiment but rather highlight the applicability of such a type of experiment for the direct detection of oscillating pseudovector fields.

The measured frequency shift due to electronic interactions with ALP fields is proportional to

$$g_{\bar{e}ee} \sqrt{2(hc)^3} \rho_{\text{CDM}} \sim 4 \times 10^{-20} \text{ GeV}^2 g_{\bar{e}ee} \quad (13)$$

For pseudoscalar fields, measurements of the time-derivative of the ALP field as well as the spatial-derivatives are sensitive to the same parameter $g_{\bar{e}ee}$. Thus, it would require static bounds on the order of $10^{-30}$ GeV (i.e. a precision of $\mu$Hz in the CHBrClF experiment) to be competitive with spin precession experiments that set limits of $|g_{\bar{e}ee}| < 10^{-7} \text{ GeV}^{-1}$ (see Refs. [28, 56]). This appears not to be achievable with experiments available today that follow this approach for chiral molecules.

Chiral molecules, however, are directly sensitive to the timelike-component of oscillating pseudovector fields, which is not favorably accessible in spin precession experiments. In the following we discuss briefly the expected sensitivity on $b_0^5$ of oscillating fields that can in principle be obtained from available experiments with chiral molecules.

To obtain a rough estimate for the sensitivity to $b_0^5$ in dependence of $\omega_b$ due to the sinusoidal behavior of $b_0^5(t)$ we assume that the sensitivity is decreasing for larger frequencies with $\omega_b/\nu_{\text{tot}}$. Furthermore we can expect that the experimental uncertainty increases with resulting shorter interrogation times for larger $\omega_b$ as $\sim \sqrt{\omega_b/\nu_{\text{tot}}}$. In the following we estimate not to be sensitive to frequencies with $\omega_b/\nu_{\text{tot}} > n_{\text{tot}}$, where $n_{\text{tot}}$ is the total number of individual measurements. As CDM is supposed to be incoherent for small frequencies $\omega_b < 2\pi/\nu_{\text{tot}}$ we can expect that $b_0^5$ converges to the static limit. The experiment in Ref. [39] was performed on 10 separate days with a total of 580 individual measurements. When assuming a continuous measurement campaign on each day of 58 subsequent measurements we have $\nu_{\text{tot}} \approx 1 \text{ d}$ and $n_{\text{tot}} \approx 58$. In total we arrive at the sensitivities

$$b_0^5 \lesssim \begin{cases} 10^{-12} \text{ GeV}, & \text{if } \frac{\omega_b}{\nu_{\text{tot}}} \leq 1.2 \mu\text{Hz} \\ (\omega_b/\nu_{\text{tot}})^{3/2} 10^{-12} \text{ GeV}, & \text{if } 1.2 \mu\text{Hz} < \frac{\omega_b}{\nu_{\text{tot}}} \leq 0.7 \text{ mHz} \\ \infty, & \text{if } \frac{\omega_b}{\nu_{\text{tot}}} > 0.7 \text{ mHz} \end{cases} \quad (14)$$
The expected sensitivities on $b^0_6$ in CHBrCIF and future experiments in dependence on the pseudovector CDM oscillation frequency $\omega_b$ is shown in Figure 2. It shall be noted that the region of $\omega_b$ to which the experiment is sensitive may be smaller or even extended depending on the actual timing of the measurements. However, robust bounds require an extended theoretical description and a rigorous statistical analysis of the actual data sets as was also discussed in Refs. [57–59].

Conclusion.—We have shown in this letter that $\mathcal{P}$-odd interactions of electrons with cosmic fields are strongly pronounced in chiral molecules. We could demonstrate that chiral molecules are suitable systems to tighten bounds on $\mathcal{P}$-odd electronic interactions of static pseudovector fields that emerge e.g. from the Standard Model Extension. By performing quasi-relativistic calculations of expectation values of $\mathcal{P}$-odd cosmic field interactions in CHBrCIF including vibrational corrections, we demonstrated that the C–F stretching mode is not a good choice to place robust limits on $\mathcal{P}$-odd cosmic fields as the effects are comparatively small and also difficult to prediction due to pronounced multimode contributions. However, we estimated the sensitivity of this mode to the parameter $b^0_6$ to be on the order of $10^{-12}$ GeV in a 20 year old experiment. This sensitivity is inferior by two orders of magnitude to the actual best direct measurements drawn from modern atomic parity violation experiments. We estimate the achievable sensitivity to $\mathcal{P}$-odd cosmic fields with modern high-resolution molecular spectroscopy on suitably chosen chiral molecules to be on the order of $10^{-17}$ GeV for static fields (see Figure 2). This would be an improvement of the current best limit on $b^0_6$ by two orders of magnitude. Furthermore, we discussed possibilities of direct detection of ultra light dark matter by studying oscillating parity violating potentials in chiral molecules. We have shown that without design of a fundamentally new experimental concept limits on electronic interactions of ultra light oscillating pseudovector particles $b^0_6$ with frequencies of around $\omega_b \lesssim 10 \mu$Hz could be pushed to about $10^{-17}$ GeV or better with modern experiments with chiral molecules. This corresponds to a direct detection of CDM masses below $10^{-19} \text{eV}/c^2$ and thus can be interesting for fuzzy dark matter searches.

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Figure 1. Dependence of the expectation value of $\gamma^5$ on the nine different modes in $(S)$–CHBrClF computed at the level of ZORA-cGKS with the B3LYP functional and polynomial fits to $\langle \gamma^5 \rangle$ to fourth order (lines). The C–F stretching mode $\nu_4$ was studied in the experiment in Ref. [39].

Figure 2. Sensitivity on electron couplings with the timelike-component of pseudovector fields $b_0^e$ in dependence of the CDM pseudovector oscillation frequency $\omega_b$ from a twenty year old experiment with CHBrClF[39] compared to the actual best static limit on $b_0^e$ from the Dy experiment (see Ref. [26]). The projected sensitivity is achievable with modern experiments with chiral molecules, assuming an improvement in sensitivity of 5 orders of magnitude compared to the CHBrClF experiment of 1999.