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A new avenue in the search for CP violation: Mössbauer spectroscopy of $^{227}\mathrm{Ac}$

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Abstract This work proposes a new avenue in the search for CP-violating odd-electric and even-magnetic nuclear moments. A promising candidate to find such moments in the ground state is the quadrupole-deformed and octupolecorrelated nucleus 227-actinium. In this nucleus, the 27.4keV E1 transition that connects the $3/2^+$ parity-doublet partner and the 3/2⁻ ground state is perfectly suited to apply the sensitive technique of recoil-free selfabsorption, commonly known as Mössbauer spectroscopy. In this experimental approach, the lifetime of the $3/2^+$ upper parity-doublet partner allows an estimate of the lower limit of $\Delta E = 2 \cdot \Gamma_{\nu} =$ $23.7(1) \times 10^{-9}$ eV for the achievable energy resolution to be made. This resolution must be exceeded by the interaction of a *CP*-violating moment and the corresponding multipole moment of the field distribution in the lattice. This work presents the first ideas for patterns caused by CP-violating moments on the expected quadrupole splitting and nuclear Zeeman effect.

1 Introduction

The observation of enhanced $B(E3, 0^+ \rightarrow 3_1^-)$ excitation strength in several lanthanide [1,2] and actinide [3–6] isotopes indicates octupole correlations in the quadrupole-deformed ground state of at least some of these nuclei.

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The resulting quadrupole-octupole deformed pear shape is predicted to enhance the possible CP-violating laboratory Schiff moment [7–13].

Interestingly, a long-standing theme investigated in (γ, γ') photon-scattering experiments is the E1 strength of the $[2^+ \otimes 3^-]_{1^-}$ quadrupole-octupole coupled (QOC) 1_1^- levels [14]. In particular, Kneissl, Pitz, and coworkers established in stable nuclei comprehensive systematics of these, in general, lowest-lying 1_1^- levels [15,16]. The collective nature of these 1 levels is evidenced by their energy systematics (Fig. 1); these systematics display a smooth behaviour, which can be summarised as follows. At/near closed shells, the energy E_{1-} of the QOC 1⁻ level corresponds nearly to the sum $\Sigma = E_{2_1^+} + E_{3_1^-}$ of the excitation energies of the first 2_1^+ and 3_1^- levels, but decreases relative to Σ with the onset of quadrupole correlations. Transitional nuclei with ground-state quadrupole correlations, but no well developed quadrupole deformation, exhibit a near degeneracy of $\mathbf{1}_1^-$ and 3⁻ levels. Once static quadrupole deformation is present, levels 1_1^- and 1_2^- become the band-heads of the K=0 and K=1 octupole bands. Furthermore, the $B(E1, 0^+ \rightarrow 1_1^-)$ strength remains in the same order of magnitude over a wide range of nuclei with varying underlying quadrupole deformation (Fig. 2). In addition, in spherical nuclei it has been shown that the strength of the two-phonon creating/annihilating transition connecting this 1- state with the ground state scales with the $3_1^- \rightarrow 2_1^+$ two-phonon exchanging transition [17], whereas in well-deformed nuclei the branching behaviour predicted by the Alaga rules is observed [18]. Interestingly, an enhanced E1 strength of up to $20 \times 10^{-3} \text{e}^2 \text{fm}^2$ is noticeable for semi-magic nuclei and prolate-deformed



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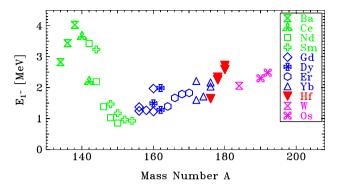


Fig. 1 Systematics of the excitation energy E_{1-} of the first excited 1_{1}^{-} level in the lanthanide/rare-earth region. Figure is taken from Ref. [19]

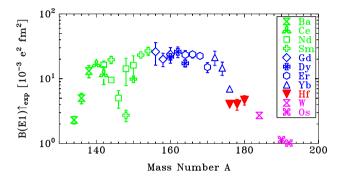
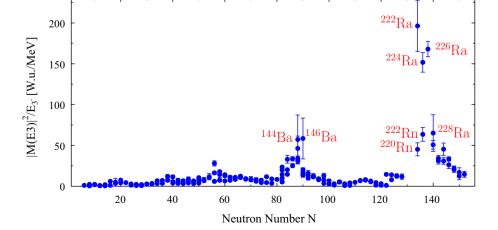


Fig. 2 Systematics of $B(E1, 0_{gs}^+ \to 1_1^-)$ strength in the lanthanide /rare-earth region. Figure is taken from Ref. [19]

nuclei, which is almost an order of magnitude reduced in the transitional region.

In spherical nuclei, for which the 2_1^+ and 3_1^- levels are interpreted as phonons, the QOC 1^- level is the low-spin member of a quintuplet of negative-parity levels with spins ranging from $J^{\pi}=1^-$ to 5^- . However, candidate levels for the full multiplet are proposed for only a few nuclei, see, e.g., Refs. [20,21]. While quadrupole deformation in the ground state of the nuclear many-body quantum system is well established, only a few candidates with enhanced octupole correlations and possibly even deformation were proposed follow-

Fig. 3 Inverse energy-weighted $B(E3, 0^+ \rightarrow 3_1^-)$ strength as a function of the mass number A. The systematics include nuclei for which the B(E3) strength is known [27]. The recent data points for 144,146 Ba [1,2], 146 Ba, 220,222 Rn [3,5], and 222,224,226,228 Ra [4-6] are highlighted



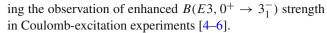


Figure 3 shows the inverse energy-weighted B(E3) strengths. This quantity combines the two most relevant characteristics of octupole correlations/deformations and emphasises the special nature of the radium nuclei 222,224,226 Ra. Furthermore, for 228 Th, indirect experimental evidence [22] suggests enhanced octupole correlations. The interplay of quadrupole deformation and octupole correlations, for the above mentioned nuclei in the ground state, results in the nucleus adopting a pear shape, in which the odd-electric (E1, E3, and E5) and even-magnetic (M2 and M4) moments are present in the intrinsic reference frame. Indeed, quadrupole-octupole coupling is predicted to enhance a possible nuclear Schiff moment [7–12] and magnetic quadrupole (M2) moment [13] caused by CP-violating physics [23–26].

The projection of the pear shape [28] from the intrinsic to the laboratory reference frame results in good-parity wave functions, that are linear combinations of the pear shape pointing to the left Ψ_l and to the right Ψ_r . Assuming that Ψ_l and Ψ_r are orthogonal, the linear combinations are

$$\Psi^{+} = \frac{1}{\sqrt{2}} (\Psi_l + \Psi_r), \qquad \Psi^{-} = \frac{1}{\sqrt{2}} (\Psi_l - \Psi_r).$$
(1)

The linear combination Ψ^+ is invariant under the space-inversion P operation. However, the Ψ^- linear combination inverts its sign and, therefore, has negative parity $\pi=-$. In an odd-mass nucleus, the coupling of the unpaired particle to these two states leads to the presence of parity doublets, which are two levels with identical angular momentum but opposite parity. Indeed, as shown in Table 1, several odd-mass nuclei in the region near Z=88/90 and N=134/136 exhibit parity-doublet candidates. Clearly, the data in Table 1 suffer from uncertainties concerning spin and, especially, parity assignment, and unobserved upper partner levels. Nevertheless, at present 227 Ac is the nucleus with the lowest established parity-doublet energy difference ΔE_{PD} of 27.4 keV.



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Table 1 Data for selected odd-mass nuclei in the $A\approx 224$ mass region exhibiting parity-doublet candidates. The table presents the isotope, its halflife $T_{1/2,gs}$, spin and parity of the ground state J_0^π , energy difference to the lowest-lying possible parity-doublet partner ΔE_{PD} , and the lifetime $T_{1/2,ul}$ of the upper level. Data are taken from the NNDC data base [29] and Ref. [30]

Nucleus	$T_{1/2,gs}$	J^{π}	ΔE_{PD} [keV]	$T_{1/2,ul}$ [ns]
²²³ Fr	22.00(7) m	3/2 ⁽⁻⁾	134.48(4)	
²²⁵ Fr	3.95(14) m	3/2-	142.59(3)	
²²⁷ Fr	2.47(3) m	1/2+	59.10(5)	
²²¹ Ra	28(2) s	5/2+	103.61(11)	
²²³ Ra	11.43(5) d	3/2+	50.128(9)	0.63(7)
²²⁵ Ra	14.9(2) d	1/2+	55.16(6)	
²²⁷ Ra	42.2(5) m	3/2+	90.034(2)	0.254(9)
²²³ Ac	2.10(5) m	$(5/2^{-})$	64.62(4)	≤ 0.250
²²⁵ Ac	9.920(3) d	$(3/2^{-})$	40.10(4)	0.72(3)
²²⁷ Ac	21.772(3) y	3/2-	27.369(11)	38.52(19) ^a
²²⁹ Ac	62.7(5) m	$(3/2^+)$	104.3(4)	
²²⁹ Th	7880(120) y	5/2+	146.357(2)	
²³¹ Th	25.52(1) h	5/2+	185.718(2)	1.07(8)
²²⁹ Pa	1.50(5) d	$(5/2^+)$	99.3(4)	
²³¹ Pa	32760(11) y	3/2-	102.269(2)	≤ 0.7

^aThis value is taken from the most recent evaluation [30]

In phenomenological estimates (see Refs. [23,24] and references therein), the enhancement of the intrinsic nuclear Schiff moment, $\langle \hat{S} \rangle$, is predicted to scale with the quadrupole β_2 and the square of the octupole β_3 deformations, as well as inversely with the energy difference ΔE_{PD} between the parity doublet partners

$$\langle \hat{S} \rangle \propto \frac{\beta_2(\beta_3)^2}{\Delta E_{PD}}.$$
 (2)

The quadrupole-octupole coupling contributes with the factor $\beta_2 \cdot \beta_3$ and the expected CP-violating interaction with an additional β_3 factor. However, at present it is not possible to disentangle the static and dynamic contributions to the β_3 deformation parameter in a model-independent way. Nevertheless, self-consistent calculations in well quadrupole-octupole deformed nuclei allow a determination of $\langle \hat{S} \rangle$ values directly, without passing through the estimates of deformations β_2 and β_3 .

The above mentioned enhanced B(E3) probabilities may indicate the presence of a non-zero intrinsic E3 moment in the ground state of Ra isotopes and, consequently, a possibility of the additional non-zero intrinsic E1 and E5 as well as E3 and E4 moments. We can thus expect that a E4 corresponding interaction may induce enhanced values of the corresponding symmetry-violating laboratory moments.

2 Experimental motivation for studying ²²⁷Ac

Given the high charge number of Z=89 and low energy of the transition connecting the parity-partner levels in 227 Ac, and considering that the internal conversion coefficients (ICCs) exhibit a strong multipolarity dependence, a measurement of the ICCs appear to be the obvious way to search for CP-violating physics. If parity was no longer a good quantum number, the E1 transition connecting the parity-doublet partners would contain a M1 component. However, it can be assumed that the P-/T-odd effect scales as a 10^{-7} contribution of the weak interaction to the nuclear force. Hence, any signal of such physics will be well below the associated uncertainty in the calculation of CCs. For the 27.4-keV E1 transition in 227 Ac, the value of the ICC is $\alpha_C=3.54(5)$ [31], which still allows for ≈ 22 % of all decays to proceed via γ -ray emission.

The low γ -ray energy, and hence the low momentum transfer in the emission and absorption process, enables the nuclear photonics technique of recoil-free resonant absorption (known as Mössbauer spectroscopy [32–34]) to be used as a method to investigate the E1 transition in question and, subsequently, to pin down the properties of the two parity-doublet partners. Mössbauer spectroscopy exploits the fact that, for a nucleus embedded in a crystal lattice, there is a probability that the recoil momentum transfer in the emission and absorption of a γ ray is absorbed by the entire crystal. If both processes are recoil-free, the theoretical achievable energy-resolution $\Delta E \approx 2 \cdot \Gamma_{\gamma}$ is limited only by the natural line width Γ , which for 227 Ac is $\Gamma_{\gamma} = \frac{\hbar \ln(2)}{T_{1/2,ul}} = 11.8(1) \times 10^{-9}$ eV.

Other advantages of studying 227 Ac are its presence in the decay chain of 235 U and a comparably long half-life ($T_{1/2} \approx 21.8$ years [29]). These may allow a chemical separation of a sufficient amount of the target material to manufacture an absorber target, whose lattice would be tailored to the specific requirements of Mössbauer spectroscopy.

Mössbauer spectroscopy can be performed in fluorescence or absorption. The latter, for which the detector is situated in the extension of the emitter-absorber line, has the advantage that the detector can be retracted from the absorber/emitter samples. This prevents pile-up events due to a too high detector count rate. Given the relatively small expected size of the radioactive samples, the resulting reduction of the solid angle would not adversely impact the count rate of good events.

A further benefit of 227 Ac is that the upper parity-doublet level is strongly populated following either the α decay of 231 Pa ($T_{1/2}=32760(110)$ years) [35] or the 227 Ra β decay ($T_{1/2}=42.2(5)$ min) [36]. While practical considerations involving the lifetime of 231 Pa favour the population via α decay, the recoil experienced by the 227 Ac daughter might be devastating in terms of a well-defined position of the emitting



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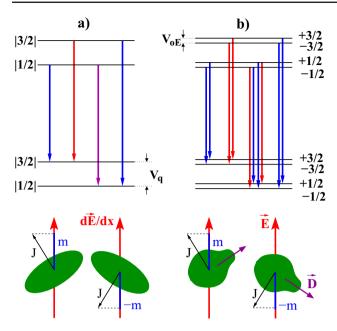


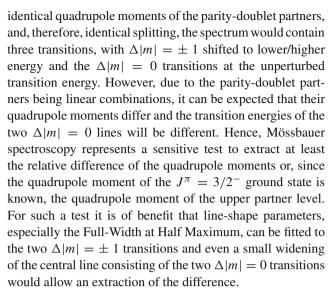
Fig. 4 Part **a** shows the level scheme of a nucleus with a $J^{\pi} = 3/2^{\pm}$ parity doublet under the influence of quadrupole splitting. Part **b** includes the influence of the interaction V_{oE} of an EL odd-electric moment with the corresponding L^{th} derivative of the electric potential of the crystal lattice at the nuclear coordinates. This CP-violating physics lifts the |m| degeneracy of the time-reversal orbits with magnetic quantum numbers m and -m. For a detailed discussion see text

²²⁷Ac nucleus within the lattice structure and, therefore, local field distribution.

3 *CP*-violating moments and Mössbauer spectroscopy

Traditionally, three effects can be observed in Mössbauer spectroscopy, namely, isomer shift, quadrupole splitting, and nuclear Zeeman effect. Each of them slightly shifts the energies of the involved levels and, consequently, alters the energy of the emitted and/or absorbed γ rays. The resulting energy difference can be compensated by mounting the emitter or absorber source on a drive inducing a velocity-dependent Doppler shift. For example, for 227 Ac an energy shift of 11.74×10^{-9} eV corresponds to a sample velocity of 1 cm/s.

The isomer shift corresponds to a slight shift of the Coulomb energy of a nuclear level due to the electron density and, consequently, electrostatic potential at the nuclear coordinates; it is observed, if emitter and absorber nuclei are embedded in different crystal lattices. The second effect of quadrupole splitting is observed if, due to the crystal composition at the coordinates of the absorbing nucleus, an electric-field gradient is present. This situation is shown in Fig. 4a for a nucleus with a $J^{\pi}=3/2^{\pm}$ parity doublet. The reflection symmetry of the quadrupole shape results in a |m| degeneracy of the time-reversal m and -m orbits. For



Far more intriguing is that, if the investigated nucleus is truly reflection asymmetric, any residual interaction of an odd-electric moment with a higher moment of the multipole expansion of the lattice charge distribution at the nuclear coordinates (e.g., E1 moment with the electric field or the E3 moment with the curvature of the electric field) would lift the |m| degeneracy. Again, given that EL moments of the two physical states represented by the linear combinations differ, a different interaction V_{oE} of EL odd-electric moment and Lth multipole order of the lattice charge distribution can be anticipated for each doublet partner level. Consequently, a split in the transitions will be observed. With the exception of the $\Delta m = \pm 1$ transitions of the $\Delta |m| = 0$, namely the $m_{\rm upper} = \pm 1/2 \rightarrow m_{\rm lower} = \mp 1/2$ transitions, for all transitions the splitting results in the same energy shift. Only these $\Delta m = \pm 1$ transitions experience a stronger shift and the $\Delta |m| = 0$ transition will, in addition to the quadrupole splitting exhibit a further splitting into a quadruplet. In the quadruplet the two satellite lines are expected to show half the intensity of the lesser shifted $\Delta |m| = 0$, $\Delta m = 0$ transitions. It is helpful that the two $\Delta |m| = \pm 1$ lines are not affected in the same way and allow a fit of the peak shape. Consequently, even if V_{oE} is less than ΔE , an alteration of the central line's peak shape yields evidence for CP-violating physics possibly even an order of magnitude below the experimental resolution ΔE . Concerning the interaction of a nuclear E1 moment and the electric field at the nuclear coordinates, it must be mentioned that, in a given lattice structure, the nucleus will position itself at coordinates for which the net electric field vanishes. Therefore, the E3 moment should be the lowest odd-electric moment contributing. Eventually, the use of a piezzo-electric lattice will allow access to the E1 moment.

Finally, the combination of the sensitivity of Mössbauer spectroscopy and the magnetic field at the nuclear coordinates in a lattice allow the observation of the nuclear Zeeman



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effect. If the magnetic moments of the parity-doublet partners are identical, the three-line pattern of the normal Zeeman effect will be observed. However, due to the two parity partners being linear combinations, their magnetic properties differ and the pattern of the anomalous Zeeman effect is expected. Here, for a J=3/2 parity doublet, ten transitions can be observed. However, these lines will exhibit a centroid symmetry. Given it is present, the interaction of a CP-forbidden magnetic quadrupole M2 moment [13] with the magnetic-field gradient perturbs the expected pattern. Since the additional interaction is direction-dependent, it can be expected that the otherwise m-independent Zeeman splitting between two levels with m and $m \pm 1$ receives an additional m-dependent term. For such a m-dependent splitting, the centroid-symmetric pattern will be disturbed.

To extract a quantitative result for a possible *CP*-violating interaction, knowledge of the electric and magnetic field distribution, specifically the field values and higher derivatives at the position of the nucleus, is required. These values can be calculated for specific materials using modern density functional theory [37], and this has already been successfully used to obtain electric field gradient and hyperfine fields for a variety of materials (see, e.g., [38–40]).

4 Summary and outlook

To summarise, in this contribution, we proposed Mössbauer spectroscopy of the octupole-correlated $^{227}\mathrm{Ac}$ nucleus as a new avenue to search for CP-violating physics. This work provides first thoughts in relation to the way in which a residual interaction associated with an odd-electric or even-magnetic moment alters the effects of quadrupole splitting or the nuclear Zeeman effect. In a long-term future outlook, this project would benefit from a sufficiently mono-energetic source with $\Delta E_{\gamma} \ll \Gamma$ of a fully-polarised γ -ray beam. This would allow the elimination of effects associated with recoils in the radioactive decays and in addition the selective population of m substates can be used to test the involved m substates.

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Data availability statement This manuscript has no associated data or the data will not be deposited. [Authors' comment: The paper outlines a novel experimental approach in the search for CP violation. So far, no experimental effort has been made and no data has been recorded.]

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