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Scheck, Marcus, Chapman, Robert, Dobaczewski, Jacek Jan orcid.org/0000-0002-4158-3770 et al. (5 more authors) (2023) A new avenue in the search for CP violation : Mössbauer spectroscopy of 227Ac. *European Physical Journal A : Hadrons and Nuclei*. 116. ISSN 1434-601X

<https://doi.org/10.1140/epja/s10050-023-01000-z>

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

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Received: date / Revised version: date

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 nuclear moments in the ground state is the quadrupole-deformed and octupole-correlated nucleus ^{227}Ac . In this nucleus, the 27.4-keV $E1$ 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 to estimate a lower limit of $\Delta E = 2 \cdot \Gamma_\gamma = 23.7(1) \times 10^{-9}$ eV energy resolution. This work presents the first ideas for patterns caused by CP-violating moments on the expected quadrupole splitting and nuclear Zeeman effect.

PACS. XX.XX.XX No PACS code given

1 Introduction

The recent observation of enhanced $B(E3, 0^+ \rightarrow 3_1^-)$ excitation strength in several lanthanide [1, 2] and actinide [3, 4, 5, 6] isotopes indicate octupole correlations in the quadrupole-deformed ground state of at least some of these nuclei. The resulting quadrupole-octupole deformed pear shape is predicted to enhance the possible CP-violating laboratory Schiff moment [7, 8, 9, 10, 11, 12, 13].

Interestingly, a long-standing theme investigated in the (γ, γ') photon-scattering experiments is the $E1$ strength of $[2^+ \otimes 3^-]_{1^-}$ quadrupole-octupole coupled (QOC) 1_1^- levels [14]. Especially, Kneissl, Pitz, and coworkers established in stable nuclei a comprehensive systematic of these, in general, lowest-lying 1_1^- levels [15, 16]. The collective nature of these 1^- levels is evidenced by their energy-systematic (Fig. 1). This systematic displays 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 1_1^- and 3_1^- 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 nuclei.

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, only for a few nuclei candidate levels for the full multiplet are proposed, 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 following the observation of enhanced

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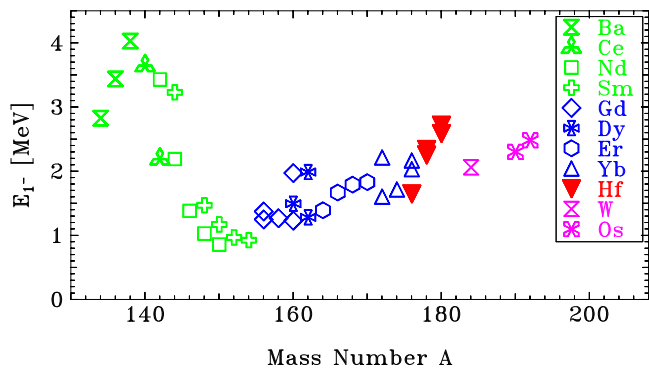


Figure 1. Systematic of the excitation energy E_{1-} of the first excited 1_{1-} level in the lanthanide/rare-earth region. Figure is taken from Ref. [19].

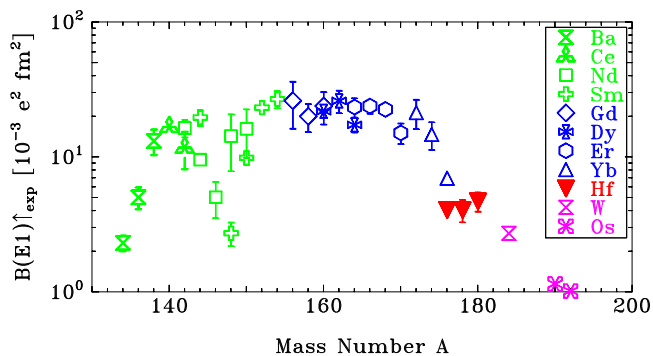


Figure 2. Systematic of $B(E1, 0_{g_s}^+ \rightarrow 1_{1-})$ strength in the lanthanide/rare-earth region. Figure is taken from Ref. [19].

$B(E3, 0^+ \rightarrow 3_{1-})$ strength in Coulomb-excitation experiments [4, 5, 6].

Figure 3 depicts the inverse energy-weighted $B(E3)$ strengths. This quantity combines the two most relevant ground-state characteristics of octupole correlations/deformations and emphasises the special nature of the radium nuclei $^{222,224,226}\text{Ra}$. Furthermore, for ^{228}Th , an 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 above mentioned 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, 8, 9, 10, 11, 12] and magnetic quadrupole ($M2$) moment [13] caused by CP-violating physics [23, 24].

The projection of the pear shape [26] from the intrinsic to laboratory reference frame results in good-parity wave functions that are linear combinations of the pear pointing to the left Ψ_l and to the right Ψ_r . Assuming that Ψ_l and Ψ_r are orthogonal, we have

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

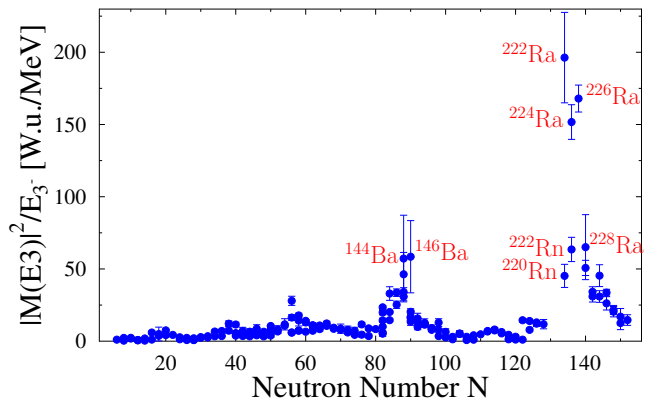


Figure 3. Inverse energy-weighted $B(E3, 0^+ \rightarrow 3_{1-})$ strength as function of the mass number A . The systematic includes the nuclei for which the $B(E3)$ strength is known [25]. The recently obtained data points for ^{144}Ba [1], ^{146}Ba [2], $^{220,222}\text{Rn}$ [5, 3], and $^{222,224,226,228}\text{Ra}$ [4, 5, 6] are highlighted.

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 about 27 keV.

In phenomenological estimates [27], 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 inversely with the energy difference ΔE_{PD} between the parity doublet partners

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

Hereby 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 [28]. Nevertheless, self-consistent calculations in well quadrupole-octupole deformed nuclei allow for determining values of $\langle \hat{S} \rangle$ 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 $M2$ and $M4$ moments. We can thus expect that CP-violating interaction may induce enhanced val-

Table 1. Data for selected odd-mass nuclei in the $A \approx 224$ mass region exhibiting parity-doublet candidates. Given is the isotope, its half-life $T_{1/2,gs}$, 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].

ues of the corresponding symmetry-violating laboratory moments.

2 Experimental motivation for ^{227}Ac

Given the high charge number of $Z \approx 89$ and low energy of the transition connecting the parity-partner levels in ^{227}Ac , and considering that the conversion coefficients (CCs) exhibit a strong multipolarity dependence, the CCs appear to be the obvious choice 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 an $M1$ component. However, it can be assumed [31] 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 percent-level uncertainty for the calculation of CCs. For the 27.4 keV $E1$ transition in ^{227}Ac , the value of the CC amounts to $\alpha_C = 3.54(5)$ [32], which still allows for $\approx 22\%$ 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, qualify the nuclear photonics technique of recoil-free resonant absorption, which is known as Mössbauer spectroscopy [33, 34, 35], as a method of choice 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 or absorption of a γ ray is absorbed by the entire crystal. Consequently, both processes are recoil-free and the theoretical achievable energy-resolution $\Delta E \approx 2 \cdot \Gamma_\gamma$ is limited only by the natural line width, which for ^{227}Ac is $\Gamma_\gamma = \frac{\hbar \ln(2)}{T_{1/2,ul}} = 11.8(1) \times 10^{-9}$ eV.

Another advantage of studying ^{227}Ac is its presence in the decay chain of ^{235}U and a long half-life ($T_{1/2} \approx 21.8$ years [29]). These may allow for a chemical separation of a sufficient amount of the target material to manufacture an absorber target, whose lattice would be tailored to the 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 count rate. Given a 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) [36] or the ^{227}Ra β decay [$T_{1/2} = 42.2(5)$ min] [37]. 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 ^{227}Ac 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, the 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 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 levels 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. 4 a) for a nucleus with $J^\pi = 3/2^\pm$ parity doublet. The reflection symmetry of the quadrupole shape results in a m degeneracy of the time-reversal m and $-m$ orbits. For identical quadrupole moments of the parity-doublet partners, and, therefore, identical splitting, the

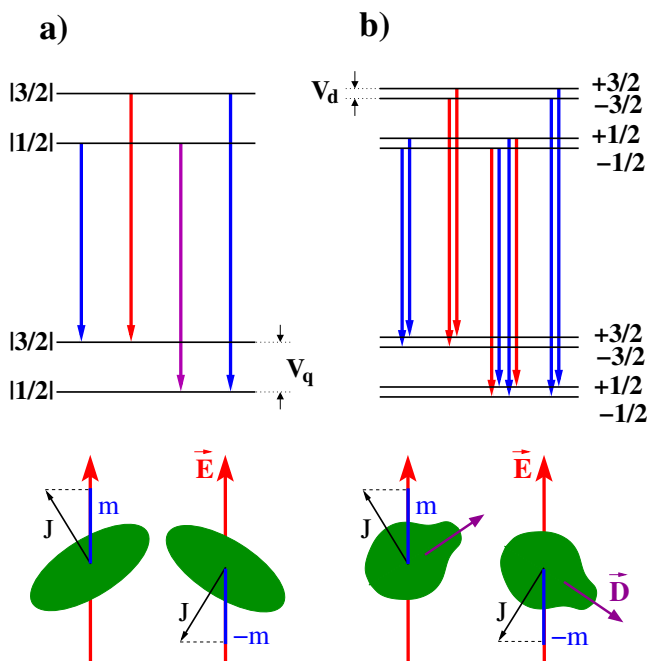


Figure 4. Part a) shows the level scheme of a nucleus with a $J^\pi = 3/2^\pm$ parity doublet under the influence of a for Mössbauer spectroscopy typical quadrupole splitting. Part b) includes the influence of the interaction of an EL odd-electric moment with the corresponding L^{th} derivative of the 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 $-m$.

spectrum would contain three transitions, with $\Delta|m| = \pm 1$ shifted to lower/higher energy and the $\Delta|m| = 0$ transitions at the unperturbed transition energy. However, even for parity-doublet partners, it can be expected that their quadrupole moments differ and the transition energies of the two $\Delta|m| = 0$ lines will be different. Mössbauer spectroscopy represents a sensitive test to extract at least the relative difference. Beneficial for such a test is that the line-shape parameters, such as the Full-Width at Half Maximum, can be fitted to the two $\Delta|m| = \pm 1$ transitions and even a small widening of the central line would allow for extracting a difference of the quadrupole moments.

Far more intriguing, 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. The different EL moments would result in a different interaction for each partner level and, consequently, a split in the transitions will be observed. However, for all transitions but the $\Delta|m| = \pm 1$ transitions of the $\Delta|m| = 0$ transitions, that are, the $m_{\text{upper}} = \pm 1/2 \rightarrow m_{\text{lower}} = \mp 1/2$ transitions experience a stronger shift. In consequence the central doublet line will split in a quadruplet with the two satellite lines having half the intensity of the lesser shifted $\Delta|m| = 0$, $\Delta m = 0$ transitions. Hence, the $m_{\text{upper}} = \pm 1/2 \rightarrow m_{\text{lower}} = \mp 1/2$

transitions experience an enhanced shift proportional to the interaction energy V_{oE} of EL odd-electric moment and L^{th} multipole order of the lattice charge distribution. Beneficial is that the two $\Delta|m| = \pm 1$ lines are not affected in the same way and allow to fit 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 to access the $E1$ moment.

Finally, the combination of the sensitivity of Mössbauer spectroscopy and the magnetic field at the nuclear coordinates in a lattice allow for the observation of the **nuclear Zeeman effect**. If the magnetic moments of the parity-doublet partners are identical the three-line pattern of the normal Zeeman effect would be observed. However, due to the two parity partners being linear combinations, their magnetic properties do differ and the pattern the anomalous Zeeman effect be expected. Here up to 10 lines can be observed. However, these lines would exhibit a centroid symmetry. Given it is present in the nuclear force, the interaction of CP-forbidden magnetic quadrupole $M2$ moment [13] with the gradient of the magnetic field perturbs the expected pattern. Since this additional interaction is direction-dependent, it can be expected that the otherwise m -independent Zeeman splitting between two levels with $m = 0$ 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 [38], which has already been successfully used to obtain electric field gradient and hyperfine fields for a variety of materials (see, e.g., [39, 40, 41]).

4 Summary and Outlook

To summarise, in this contribution, we proposed Mössbauer spectroscopy of the octupole-correlated ^{227}Ac as a new avenue to search for CP-violating physics. This work provides first thoughts in which way a residual interaction associated with an odd-electric or even-magnetic moment alters the effects of quadrupole splitting or the nuclear Zeeman effect. Obviously, the sample preparation is a great obstacle in the practical realisation, but given the interesting physics, it is an avenue worthwhile to pursue. 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 to eliminate effects associated with recoils in the radioactive decays and the selective population of m substates can be used to test the involved m substates.

5 Acknowledgements

We acknowledge financial support by the UK-STFC Grant Nos. ST/P005101/1, ST/P003885/1, and ST/V001035/1, by the Polish National Science Centre under Contract No. 2018/31/B/ST2/02220, and by a Leverhulme Trust Research Project Grant. We acknowledge the CSC-IT Center for Science Ltd., Finland, for the allocation of computational resources. This project was partly undertaken on the Viking Cluster, which is a high performance compute facility provided by the University of York. We are grateful for computational support from the University of York High Performance Computing service, Viking and the Research Computing team.

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