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Scheck, Marcus; Chapman, Robert; Dobaczewski, Jacek; Ederer, Claude; Ivanov, Peter; Lorusso, Guiseppe; O'Donnell, David; Schröder, Christian

*Published in:*  
European Physical Journal A

*DOI:*  
[10.1140/epja/s10050-023-01000-z](https://doi.org/10.1140/epja/s10050-023-01000-z)

Published: 26/05/2023

*Document Version*  
Publisher's PDF, also known as Version of record

[Link to publication on the UWS Academic Portal](#)

#### *Citation for published version (APA):*

Scheck, M., Chapman, R., Dobaczewski, J., Ederer, C., Ivanov, P., Lorusso, G., O'Donnell, D., & Schröder, C. (2023). A new avenue in the search for *CP* violation: Mössbauer spectroscopy of <sup>227</sup>Ac. *European Physical Journal A*, 59(5), [116]. <https://doi.org/10.1140/epja/s10050-023-01000-z>

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

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Received: 18 November 2022 / Accepted: 5 April 2023

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Communicated by Calin Alexandru Ur

**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 octupole-correlated nucleus  $^{227}\text{actinium}$ . 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 an estimate of the lower limit of  $\Delta E = 2 \cdot \Gamma_{\gamma} = 23.7(1) \times 10^{-9}$  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

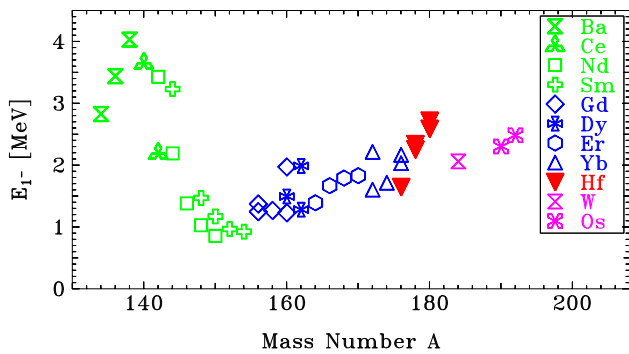
The observation of enhanced  $B(E3, 0^+ \rightarrow 3_1^-)$  excitation strength in several lanthanide [1,2] and actinide [3–6] isotopes indicates octupole correlations in the quadrupole-deformed ground state of at least some of these nuclei.

Topical Issue: Nuclear Photonics: Overview and Perspectives Maria Jose Garcia Borge, Kazuo A. Tanaka, Calin Ur and Andreas Zilges (Guest editors).

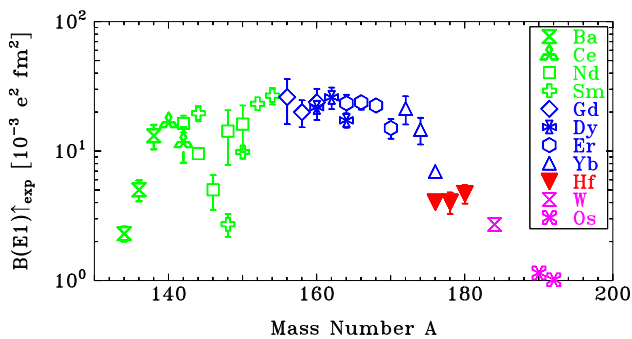
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The resulting quadrupole-octupole deformed pear shape is predicted to enhance the possible  $CP$ -violating laboratory Schiff moment [7–13].

Interestingly, a long-standing theme investigated in  $(\gamma, \gamma')$  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  $1_1^-$  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  $E_{1_1^-}$  of the QOC  $1_1^-$  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  $\Sigma$  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  $1_1^-$  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



**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]

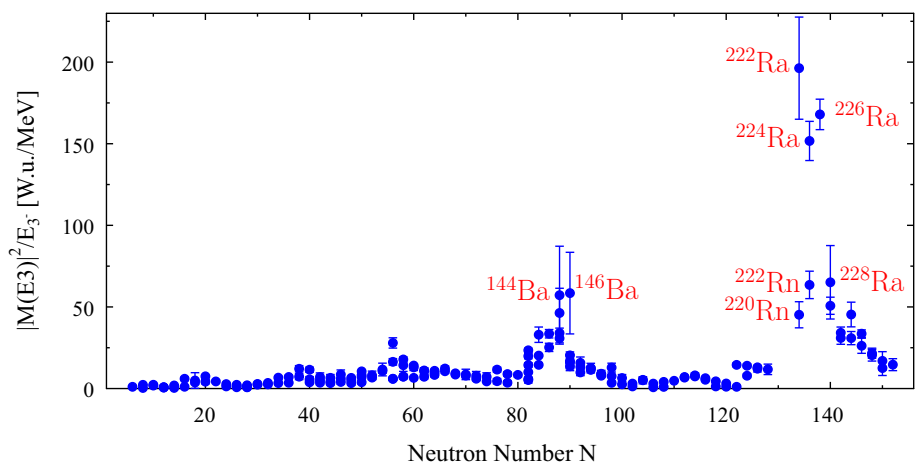


**Fig. 2** Systematics of  $B(E1, 0_{gs}^+ \rightarrow 1_{1-}^-)$  strength in the lanthanide/rare-earth region. Figure is taken from Ref. [19]

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  $^{144,146}\text{Ba}$  [1,2],  $^{146}\text{Ba}$ ,  $^{220,222}\text{Rn}$  [3,5], and  $^{222,224,226,228}\text{Ra}$  [4–6] are highlighted



ing the observation of enhanced  $B(E3, 0^+ \rightarrow 3_{1-}^-)$  strength in Coulomb-excitation experiments [4–6].

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  $^{222,224,226}\text{Ra}$ . Furthermore, for  $^{228}\text{Th}$ , 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  $\Psi_l$  and to the right  $\Psi_r$ . Assuming that  $\Psi_l$  and  $\Psi_r$  are orthogonal, the linear combinations are

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

The linear combination  $\Psi^+$  is invariant under the space-inversion  $P$  operation. However, the  $\Psi^-$  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  $^{227}\text{Ac}$  is the nucleus with the lowest established parity-doublet energy difference  $\Delta E_{PD}$  of 27.4 keV.

**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 half-life  $T_{1/2,gs}$ , spin and parity of the ground state  $J_0^\pi$ , energy difference to the lowest-lying possible parity-doublet partner  $\Delta E_{PD}$ , and the lifetime  $T_{1/2,ul}$  of the upper level. Data are taken from the NNDC data base [29] and Ref. [30]

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

<sup>a</sup>This 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  $\beta_2$  and the square of the octupole  $\beta_3$  deformations, as well as inversely with the energy difference  $\Delta E_{PD}$  between the parity doublet partners

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

The quadrupole-octupole coupling contributes with the factor  $\beta_2 \cdot \beta_3$  and the expected  $CP$ -violating interaction with an additional  $\beta_3$  factor. However, at present it is not possible to disentangle the static and dynamic contributions to the  $\beta_3$  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  $\beta_2$  and  $\beta_3$ .

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 a  $CP$ -violating interaction may induce enhanced values of the corresponding symmetry-violating laboratory moments.

## 2 Experimental motivation for studying $^{227}\text{Ac}$

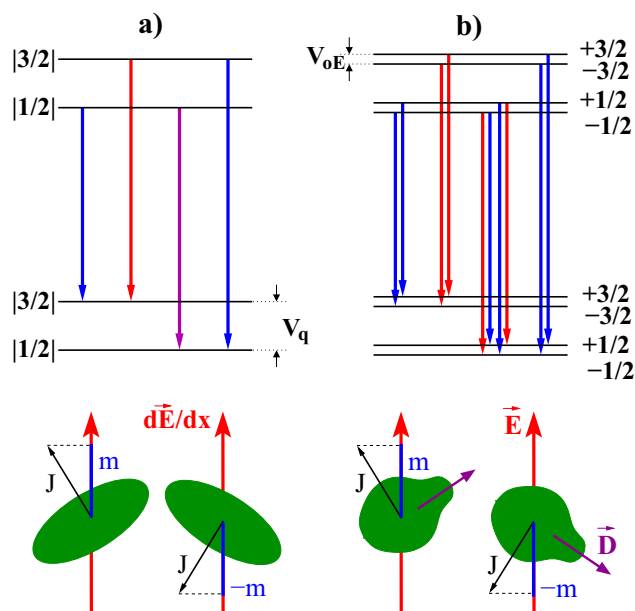
Given the high charge number of  $Z = 89$  and low energy of the transition connecting the parity-partner levels in  $^{227}\text{Ac}$ , and considering that the internal conversion coefficients (ICCs) exhibit a strong multipolarity dependence, a measurement of the ICCs appear to be the obvious way to 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 ICCs. For the 27.4-keV  $E1$  transition in  $^{227}\text{Ac}$ , the value of the ICC is  $\alpha_C = 3.54(5)$  [31], which still allows for  $\approx 22\%$  of all decays to proceed via  $\gamma$ -ray emission.

The low  $\gamma$ -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  $\gamma$  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  $\Gamma$ , which for  $^{227}\text{Ac}$  is  $\Gamma_\gamma = \frac{\hbar \ln(2)}{T_{1/2,ul}} = 11.8(1) \times 10^{-9}$  eV.

Other advantages of studying  $^{227}\text{Ac}$  are its presence in the decay chain of  $^{235}\text{U}$  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  $^{227}\text{Ac}$  is that the upper parity-doublet level is strongly populated following either the  $\alpha$  decay of  $^{231}\text{Pa}$  ( $T_{1/2} = 32760(110)$  years) [35] or the  $^{227}\text{Ra}$   $\beta$  decay ( $T_{1/2} = 42.2(5)$  min) [36]. While practical considerations involving the lifetime of  $^{231}\text{Pa}$  favour the population via  $\alpha$  decay, the recoil experienced by the  $^{227}\text{Ac}$  daughter might be devastating in terms of a well-defined position of the emitting



**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

$^{227}\text{Ac}$  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, 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  $\gamma$  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  $^{227}\text{Ac}$  an energy shift of  $11.74 \times 10^{-9}$  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

identical quadrupole moments of the parity-doublet partners, and, therefore, identical splitting, the 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, due to the parity-doublet partners being linear combinations, it can be expected that their quadrupole moments differ and the transition energies of the two  $\Delta|m| = 0$  lines will be different. Hence, Mössbauer spectroscopy represents a sensitive test to extract at least the relative difference of the quadrupole moments or, since the quadrupole moment of the  $J^\pi = 3/2^-$  ground state is known, the quadrupole moment of the upper partner level. For such a test it is of benefit that line-shape parameters, especially 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 consisting of the two  $\Delta|m| = 0$  transitions would allow an extraction of the difference.

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  $L^{th}$  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  $m_{\text{upper}} = \pm 1/2 \rightarrow m_{\text{lower}} = \mp 1/2$  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  $\Delta E$ , 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  $\Delta E$ . 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

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  $^{227}\text{Ac}$  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  $\gamma$ -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.

**Acknowledgements** The authors would like to dedicate this article to the memory of the late Professor Dr. Ulrich Kneissl. As a teacher, he introduced two of the authors (MS and CE) to the secrets of Mössbauer spectroscopy; as a distinguished scientist and academic father of the corresponding author, he provided the foundations for the experimental ideas outlined in this contribution. 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 computing 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.

**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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