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Measurement of Quadrupole Deformation using E2 and M1+E2 Transitions in Heavy Isotopes in the Mass Range of $150 < A < 250$

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The measurement of a permanent electric dipole moment (EDM) in atoms is crucial for understanding the origins of CP-violation. Quadrupole and octupole deformed nuclei significantly enhance atomic EDM. However, accurate interpretation of the EDM in such systems requires the characterization of their deformation. While nuclear deformation is indicated in various structure models, there is substantial mutual disagreement between the theoretical models or between theoretical models and experimental values. Experimental confirmation of the same, particularly in heavy isotopes essential for EDM measurements, is lacking.

Nuclear E2 transitions allow access to quantify quadrupole deformation, but these transitions are often mixed with M1 transitions. Both E2 and M1 transitions are well characterized by Weisskopf estimates, which rely on a single-particle approximation, but are affected by collective nuclear deformation. Previously, Weisskopf estimates were only available for the mass range $A < 150$, and in this work we have extended the Weisskopf estimates of both E2 and M1 transition lifetimes to the mass range of $150 \leq A \leq 250$.

This allowed us to comprehensively study 91 candidate isotopes, by comparing their E2 and M1+E2 transition lifetimes to their nearest even-even counterparts, whose E2 transition strengths are very well understood. Estimates of collective nuclear quadrupole deformation in 67 of these isotopes were obtained, either from E2 or M1+E2 transition lifetimes, and in 32 cases they were obtained from both types of transitions independently. We find that the quadrupole deformation extracted from the two different types of transitions are mutually consistent, as well as that they follow the trends established in theory. We thereby identify the isotopes ^{223,225}Fr, ^{221,223}Ra, ^{223,225,227}Ac and ^{229}Pa, where EDM measurements are foreseen and information on nuclear deformation is needed, for which no measurement of nuclear quadrupole deformation has been made.

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1. Introduction

The amount of charge-parity (CP) violation in the Standard Model, arising from the weak sector, is insufficient [1] to explain the observed baryon asymmetry of the universe [2, 3]. Measurement of a statistically significant CP violating electric dipole moment (EDM) in multiple systems, not only provides a means to quantify the amount of CP violation from various sources [4], but also directly probes the CP violation arising from the strong sector [5].

Even though measurement of the EDM for the neutron was the first to be pioneered [6, 7], atoms and molecules have emerged as lucrative systems in which to measure CP violating parameters of the atomic EDM [8, 9] and the nuclear magnetic quadrupole moment (MQM) [10, 11]. The EDM of atoms with quadrupole and octupole deformed nuclei are dramatically enhanced [11–16]. Nuclear quadrupole deformation is a prerequisite for this enhancement of the EDM, both via the nuclear MQM [17, 18], as well as the nuclear Schiff moment [13, 14]. The particulars of the enhancement have been described in further detail in *Refs.* [19, 20]. In this work, we are focused on nuclear quadrupole deformation, accessed via nuclear γ-ray spectroscopy measurements, that are already available.

The nuclear quadrupole moment is characterized by β_2, which is the coefficient associated with the quadrupole term of the multipole expansion defining the surface of a nucleus given by [21]

$$R = c_V R_A \left(1 + \sum_{\lambda=2} \beta_\lambda Y_\lambda^{m=0}\right), \tag{1}$$

where $c_V = 1 - \{(1/\sqrt{4\pi}) \sum_{\lambda=2} \beta_\lambda^2\}$ is the volume normalization such that $R_A = R_0 A^{1/3} = 1.2\ \text{fm} \cdot A^{1/3}$, A is the total number of nucleons in the nucleus, β_λ are the 2^λ-pole structure deformation coefficients, and Y_λ^m are spherical harmonic functions. The quadrupole deformation of even-even nuclei has been well vetted [22, 23], and can be accessed in the NNDC database [24].

Nuclear quadrupole deformation and shape collectivity have been well measured [25, 26] for a large range of nuclei; however, the same are only scantly known for isotopes with $A > 200$ [27]. Especially since the octupole deformed region around $A \sim 223$ is of great interest to the EDM community [20], it is absolutely necessary to characterize the deformation of these isotopes. Nuclear theory models which predict the deformation are mature [28–30]. Nonetheless, there are inconsistencies between the various theoretical models themselves, *e.g.* for ^{224}Ra, $\beta_2 = 0.177$ in *Ref.* [28], $\beta_2 = 0.18$ in *Ref.* [29], and $\beta_2 = 0.143$ in *Ref.* [30]. There are also inconsistencies between measurements and theoretical models, *e.g.* the measurements indicate that the quadrupole deformation of ^{224}Ra is $\beta_2 = 0.2022(25)$ [22]. Precisely knowing the deformation is critical to interpreting the EDM in terms of more fundamental sources of CP violation arising from individual nucleons [10, 17] and their mutual interactions [4, 8]. This provides additional motivation for explicitly measuring the deformation, as opposed to relying solely on the theoretical models.

2. Electromagnetic Transition Lifetimes

In order to better understand the nuclear transition energy spectra, we began by studying the Weisskopf estimates [31, 32] for single particle electromagnetic transitions in heavy isotopes, $150 < A < 250$. The Weisskopf estimates give ballpark estimates of the decay width for nuclear transitions under the assumption that a single nucleon is participating in the process [33]. The Weisskopf estimates further assume that the initial and final states of the single particle participating in the transition has an angular momentum of $l = 1/2$. This assumption is relaxed by incorporating multiplicity into the Weisskopf estimates, as done by the Moszkowski estimates [34].

Nuclear transitions can be electric or magnetic in type, where the single nucleon participating in the transition interacts with the electric or magnetic fields (arising from the core of the nucleus), respectively. These transitions are characterized by the inverse of the decay lifetime (τ), or the width (Γ). In order to remove the dependence on the energy of the photon emitted ($\mathcal{E}_\gamma$), it is convenient to use strengths of the transition, $B(i\lambda)$, defined by [35]

$$\frac{\mathrm{Ln}(2)}{\tau_{1/2}(i\lambda)}\frac{1}{\mathcal{E}_\gamma} = \frac{\Gamma(i\lambda)}{\mathcal{E}_\gamma} = \frac{8\pi(\lambda+1)}{\hbar\lambda\,((2\lambda+1)!!)^2}\left(\frac{\mathcal{E}_\gamma}{\hbar c}\right)^{2\lambda+1}\cdot\left(B_{w:i\lambda}+B_\beta(i\lambda)+\mathcal{O}\right), \tag{2}$$

where $i\in\{E,M\}$, B_w are single particle Weisskopf estimates, B_β are the strengths due to collective deformation, $\tau_{1/2}$ is the half-life of the transition, which is closely linked to the lifetime, τ, as $\tau_{1/2}=\mathrm{Ln}(2)\tau$ as well as the transition width, $\Gamma(i\lambda)$, as $\Gamma(i\lambda)=1/\tau$, and $\mathcal{O}$ encompasses any N-body contributions [36, 37].

Quadruple deformation is ubiquitous across the nuclide chart [20]. The Weisskopf estimates only give a ballpark measure of the transition lifetime, mostly because it is dominated by contributions from nuclear deformation, indicated by B_β in Eq. 2. As one might expect, the transition strength of the electric Eλ transition is directly linked to the 2^λ-pole nuclear deformation [38, 39]

$$B_\beta(E\lambda:j_i\to j_f) = \frac{2\lambda+1}{16\pi}\left\|\langle j_i,0;\lambda,0|j_f,0\rangle\right\|^2 Q^2_{\lambda,0}, \tag{3}$$

where the inner product is the Clebsch-Gordan coefficient, and $Q_{\lambda,0}$ is the electric 2^λ-pole moment of the nucleus given by [40]

$$Q_{\lambda\geq2,0} = \frac{3}{\sqrt{(2\lambda+1)\pi}}Z\mathrm{e}R_A^\lambda\bar{\beta}_\lambda. \tag{4}$$

To the first order $\bar{\beta}_\lambda\approx\beta_\lambda$, where β_λ are defined in Eq. 1, and Z is the number of protons in the nucleus. However, higher order corrections to $\bar{\beta}_\lambda$ are available in *Ref.* [40].

3. Extending Weisskopf Estimates in the Range $150<A<250$

The Weisskopf estimates for M1 and E2 transitions have been extensively studied for lighter isotopes. A global analysis of these estimates maybe found in *Refs.* [41–43] for $A\in[4,150]$. In this section, we will attempt to extend these single particle estimates over a range $150<A<250$, which will help us experimentally confirm the quadrupole deformation in isotopes relevant for EDM measurements, from existing E2 and M1+E2 transition lifetimes [24].

For obtaining Weisskopf estimates, we choose to use nuclei with spin-0 ground state, given that such systems provide for the simplest case. In the case of spin-0 nuclei, higher order electromagnetic multi-polar corrections are absent [44, 45]. We further exclude nuclei with non-band transitions or inter-band transitions, and consider candidate isotopes with pure M1 and E2 transitions directly to the ground state, so that the transition energy is adequately represented by the energy of the emitted photon in Eq. 2. In addition, since E2 transition strength is strongly dependent on nuclear quadrupole deformation, we also selected the isotopes which are known to be minimally quadrupole deformed ($\beta_2<0.1$) in finite range droplet macroscopic model (FRDM) calculations [30], such that the contribution from quadrupole deformation to the E2 strength is small (Eqs. 2,3). However, such a filter involving the theoretical quadrupole deformation was not applied to the list of isotopes with M1 transitions since M1 transition strength is not directly affected by nuclear quadrupole deformation. These pre-conditions severely constrain the available measurements in the mass range of $150<A<250$.

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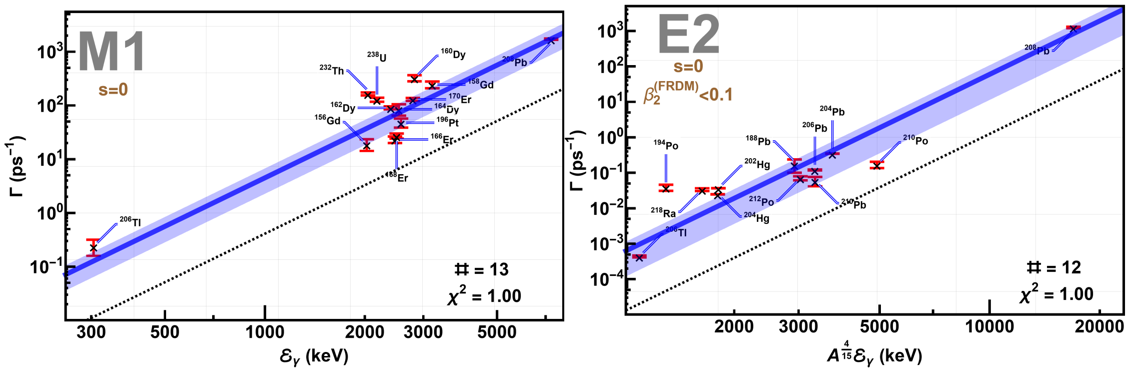

Figure 1: Plot showing the pure M1 (E2) transition decay widths as a function of transition energy, $\mathcal{E}_\gamma$ ($A^{4/15}\mathcal{E}_\gamma$), in the Left (Right) panel for 13 (12) candidate isotopes with spin a 0 ground state. In the case of E2 transition decay widths, in the Bottom panel, the candidate isotopes were minimally quadrupole deformed, $\beta_2^{\text{(FRDM)}} < 0.1$, according to *Ref.* [30].

In order to obtain the Weisskopf estimates, we used 13 candidate nuclei for M1 transitions and 12 candidate nuclei for E2 transitions, all of which satisfy the above requirements. The energy difference between the nuclear levels involved in the γ-ray transition ($\mathcal{E}_\gamma$) has been plotted as a function of their transition widths in Figure 1, for candidate isotopes with both M1 and E2 transitions. We fitted the transition width to, $\mathcal{E}_\gamma^3$ for M1 transitions, and $\mathcal{E}_\gamma^5$ for E2 transitions. We thus obtained weighted fits for M1 and E2 transitions, respectively, of

$$\Gamma_{w:\text{M1}}(\mathcal{E}_\gamma) = \underbrace{2.3(9) \times 10^{-9}}_{m_{w:\text{M1}}} \left(\frac{\mathcal{E}_\gamma}{\text{keV}}\right)^3 \text{ps}^{-1}, (5) \quad \Gamma_{w:\text{E2}}(\mathcal{E}_\gamma) \lesssim \underbrace{6.0(4) \times 10^{-19}}_{m_{w:\text{E2}}} A^{\frac{4}{3}} \left(\frac{\mathcal{E}_\gamma}{\text{keV}}\right)^5 \text{ps}^{-1}, \quad (6)$$

where the uncertainty of the slope represents the 68.3% C.I. of the standard error on the mean, such that the reduced-$\chi^2 \approx 1$, and m_i represents the respective slopes. Typically the relative uncertainty of the measured transition lifetime dominated, compared to that of the photon energy. Since the candidate list is limited in Figure 1, we also tested the resilience of the slope values above by checking if removing a single data point changes the values significantly. We note that even though ^{206}Tl and ^{208}Pb fall on the extreme sides of both the plots in Figure 1, the removal of these two data points does not impact the slope values in Eqs. 5 and 6 significantly. Even though the Weisskopf estimate for E2 transitions, obtained in Eq. 6, used only theoretically mildly quadrupole deformed isotopes, it is still an overestimate due to the pervasive quadrupole deformation present in the heavy isotopes in the range of $150 < A < 250$. A careful analysis, described in detail in arXiv: [2410.09272], considers the effect of the quadrupole deformation and updated the slope to

$$m_{w:\text{E2}} = 1.13(5) \times 10^{-19}. \quad (7)$$

4. Measurement of Quadrupole Deformation

The empirically measured E2 transition lifetime corrected for single particle estimates, can be converted directly to the strength of the transition due to quadrupole deformation, via Eq. 2, which gives us access to the quadrupole deformation parameter of β_2, via Eqs. 3 and 4. However, this typically leads to overestimation of the strength, which in turn leads to an overestimation of the quadrupole deformation. The contributions from N-body interactions can in fact be quite large [36, 37]. In order to minimize such spurious contributions, we use the ratio of the E2 transition lifetime of the candidate isotope with that of the nearest even-even nuclei,

$$\frac{\tau_{1/2}^{\text{(even-even)}}}{\tau_{1/2}^*} = \frac{\Gamma^*(\text{E2}:j_i \to j_f)}{\Gamma^{\text{(even-even)}}(\text{E2}:0^+ \to 2^+)} \approx \frac{B_{w:\text{E2}}^*(\mathcal{E}_\gamma^*) + B_\beta^*(\text{E2}:j_i \to j_f)}{B_{w:\text{E2}}^{\text{(even-even)}}(\mathcal{E}_\gamma^{\text{(even-even)}}) + B_{\text{adopted}-\beta}^{\text{(even-even)}}(\text{E2}:0^+ \to 2^+)}, \quad (8)$$

where the parameters marked with an asterisk are the candidate isotopes we have considered, and those marked (even-even) are from the nearest even-even nuclei. When using the ratio of E2 transition lifetimes in Eq. 8, we also correct the E2 transition strength of the nearest even-even nuclei with its corresponding contribution from the Weisskopf estimate using the appropriate energy of the photon linked to the $2^+ \to 0^+$ transition ($\mathcal{E}_\gamma^{\text{(even-even)}}$). The strength associated with the single particle estimates for the E2 transition lifetime obtained in Eq. 7 was calculated using Eq. 2. The E2 transition strength associated with quadrupole deformation of the even-even nuclei was obtained using the corresponding adopted $B(\text{E2})$ (or adopted values of β_2), from a combination of measurements and theory, as reported in *Refs.* [22, 23]. The extraction of quadrupole deformation from M1+E2 transition lifetime in candidate nuclei was analogous to pure E2, but included the contribution of $B^*_\beta(\text{M1} : j_i \to j_f)$ in addition.

The majority of isotopes relevant for EDM searches are odd-even nuclei which have a ground state spin of at least $1/2$ [20]. Therefore, when going from the so extracted E2 transition strength to the coefficient characterizing the nuclear quadrupole deformation, β_2, via Eqs. 3 and 4, we ensured that the right transition matrix elements (in Eq. 3), corresponding to the initial and final spin states involved in the E2 transition to the ground state, were used. The transition matrix element used not only the appropriate Clebsch-Gordan coefficients, but also the correction factors arising from relativistic effects and those arising from the interaction of the angular momentum with the spin (see Tables A.1 to A.3 in *ref.* [46]). These values of β_2 are listed in arXiv:[2410.09272], along with the recommended values for the corresponding E2 transition strengths, from both pure E2 and M1+E2 transitions.

5. Conclusion

Interpreting atomic EDM measurements in terms of the EDM of the constituent particles [10, 17] or the mutual CP violating interactions [4, 8] between them requires knowledge of nuclear deformation. While the theoretical models that predict the nuclear deformations are mature [28–30], there are however mutual inconsistencies between the various theoretical models, as well as between the models and measurements. This demands the explicit measurement of the nuclear deformations relevant for EDM measurements [19, 20]. Fortunately, E2 nuclear transitions to the ground state of various isotopes has already been measured [24]. In this work we have used the existing data on E2 transitions to infer the quadrupole deformation, before setting out on a campaign to measure those where such measurements are not available a-priori.

The values for the quadrupole deformation we obtained through the study of these two different kinds of transitions, are remarkably consistent, demonstrating the versatility of the analysis. However, it is important to note that the uncertainty in our estimates of the quadrupole deformation is mainly dictated by the uncertainty associated with the lifetime of the transition, and to a negligible degree the uncertainty associated with the transition energy difference. The final uncertainties in the quadrupole deformation also have significant contribution from our Weisskopf estimates of the transition lifetime. Due to this, the uncertainty of the deformation estimates for mildly to moderately deformed species like ^{189,191,193}Ir, ^{195}Pt, and ^{223,225}Ra are heavily subject to the Weisskopf estimates, and therefore underestimated. The final estimates have no contributions from the N-body interactions, and therefore are underestimated. We have also chosen to neglect the effects of nuclear surface diffuseness [48], that further modify the transition matrix elements in Eq. 3. Furthermore, especially in the octupole deformation region, the quadrupole deformation of the isotopes of ^{229}Th, ^{231}Pa, and ^{239}Pu, could not be precisely characterized due to interactions of the higher order de-

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formations. In these species, the uncertainty associated with the quadrupole deformation has been under-estimated. In the isotope ^{195}Pt, whose lifetimes are relatively well measured, the other effects we have neglected here contribute significantly, therefore the uncertainty in the extracted quadrupole deformation is underestimated.

The quadrupole deformation of certain isotopes could not be characterized in this method even though the data regarding their transition lifetimes was available. Isotopes of ^{151}Eu, ^{167}Er, ^{183}W, ^{187,197}Pt, ^{185,196,197,198}Au, ^{195,197,199,201}Hg, ^{200}Tl, ^{219}Rn, and ^{229}Ra were left out since they all involved non-band energy levels in the transition measurements. In isotopes such as ^{152}Eu, ^{160}Tb, ^{171}Hf, ^{177}W, ^{186}Re, ^{177}Os and ^{185}Ir, the transition lifetimes were unusually large, larger than that allowed by the Weisskopf estimates, indicating other effects in play, *e.g.* unavailability of low-lying spin states from which to E2 transition down to the ground state.

Typically, the isotopes relevant for EDM searches [7] have a spin non-zero ground state. Using the E2 or M1+E2 transition lifetime to their ground state, and comparing them against the E2 transition strength from the very well vetted measurements in the nearest even-even nuclei [22, 23], $B(E2 : 2^+ \to 0^+)$, we were able to infer the quadrupole deformation of 67 isotopes in the mass range of $150 \leq A \leq 250$. Such a comparison was made possible since the spin of most of our candidate nuclei, the vast majority of which had a ground state spins greater than 0, was carried by a single valence nucleon, leaving a core of 0 spin [49]. Yet, we showed that in such cases special care must be taken with the application of appropriate correction factors (Tables A.1-A.3 in *Ref.* [46]), including the spin-multiplicity according to Eq. 3. We achieved this comprehensive characterization of quadrupole deformation by carefully considering the single particle contributions to the strength of the E2 transitions, as well as M1 transitions, which in turn allowed us to isolate the contribution due to quadrupole deformation. The studies of single particle Weisskopf estimates were previously lacking in this mass range. Our work now paves the way and provides the much needed impetus to precisely measure the nuclear deformation in species like ^{223,225}Fr, ^{225,227}Ac and ^{229}Pa, where EDM measurements are foreseen [19, 20], but no measurement of nuclear deformations exists.

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