

Fifty Years of Nuclear Quadrupole Moments*

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On March 2, 1935 Hermann Schüler and Theodor Schmidt reported the first experimental evidence of non-spherical nuclei. From careful hyperfine structure studies of several Eu I-lines, they had shown that the hyperfine components of ^{151}Eu and ^{153}Eu did not follow the Landé interval rule exactly. Since the deviations were larger for ^{153}Eu with the smaller magnetic moment, level perturbations were ruled out. This led to the conclusion of nuclear quadrupole moments. The theory was published June 1, 1935 by Hendrik B. G. Casimir. Nuclear deformations are playing a decisive role in modern nuclear structure physics. For solid state physics, spectroscopic quadrupole moments are very useful, since they probe the electric field gradient at the nuclei.

This review presents the discovery of 1935 in historical context: 1. Early measurements of nuclear radii. 2. Discovery of nuclear quadrupole moments. 3. Spectroscopic quadrupole moments (absolute measurements; relative hyperfine data, europium revisited). 4. Intrinsic quadrupole moments (discovery from isotope shifts; present status, samarium revisited). 5. Charge distribution of deformed nuclei.

1. Early Measurements of Nuclear Radii

75 years ago, on December 14, 1910, Ernest Rutherford wrote to a friend "I think I can devise an atom ... [which] will account for the reflected α particles observed by Geiger" [1]. One of the most important experiments of this century had been known since May 1909, and was not understood at all. But that found little attention outside Manchester. There, in Rutherford's laboratory, Hans Geiger and Ernest Marsden had discovered and convincingly confirmed that α rays were back-scattered from very thin metal foils [2]. Nearly two years after this surprising discovery, on March 7, 1911, Rutherford presented his nuclear atom to the Manchester Literary and Philosophical Society [3] and Geiger stated at the same meeting [4] how he had experimentally verified the $1/\sin^4(\theta/2)$ dependence of large angle scattering calculated by Rutherford.

Geiger and Marsden then started with a new series of careful measurements which proved Rutherford's scattering law to be valid with respect to all parameters. Their results, published in July 1912 [5],

gave the first experimental number for the extremely small radius of the nucleus: less than 30 fm in the case of gold.

Niels Bohr, whose 100th birthday the scientific world is celebrating this year, had moved to Manchester early in 1912. The first of his three famous papers "On the Constitution of Atoms and Molecules" is dated April 5, 1913. With the nuclear atom a new era for physics, chemistry, and all natural sciences had begun.

Already between 1915 and 1919, at least 12 geometric nuclear models – of course purely speculative – were proposed [6], all of them assuming electrons as constituents of the nucleus. Rutherford's disintegration of the nitrogen nucleus by α rays in 1919 gave the first evidence of nuclear structure [7]. In the same year he observed "anomalous results" for α -p-scattering and concluded that the α particle was a disc with about 3 fm radius [8]. In 1924 Bieler [9] measured the deviation from point-nucleus scattering as a function of angle in ring geometry for aluminium and obtained 3.44 fm for its nuclear radius. But different analyses yielded substantially different values [10]. The sizes of the heaviest nuclei were evaluated in 1928 by Gamow and Houtermans [11]. They applied the theory of α decay to the Geiger-Nuttall-relation of 1911 [12].

In 1930, the well known $A^{1/3}$ rule was written down (for light nuclei) by Rutherford, Chadwick, and Ellis [13]: "For a rough approximation we may

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Preface

Since their inception in 1972 the International Symposia on Nuclear Quadrupole Resonance Spectroscopy have been organised in a truly interdisciplinary spirit with equal representation of the physical and chemical aspects of the field: The present symposium was no exception to this rule, the subjects ranging from the application of NQR to the identification of polymorphic forms of pharmaceutical compounds to the observation of the quadrupole coupling of muonium in crystals.

1985 was the 50th anniversary of the first observation of a nuclear quadrupole effect, and the Symposium opened with a most interesting survey of the first fifty years by Professor P. BRIX. The following papers illustrate the wide variety of fields in which measurements of nuclear quadrupole coupling constants yield important information: the structure of metals and, especially interesting, of metallic surfaces; the electronic structure of molecules and complexes; the detection of second-order phase transitions and of the formation of incommensurate phases; the study of internal motions in crystalline solids; etc. Even today the nuclear quadrupole moments are seldom known with much precision since usually the field gradient in a simple system must be calculated in order to extract the nuclear moment from the coupling constant. This has implied a continuous effort on the part of theoreticians to judge precisely the large effects on field gradients of very small – from the energy point of view – modifications in the theoretical wave-functions. This theoretical effort is also well represented and includes a survey by a pioneer in the field: R. M. STERNHEIMER.

The papers have been grouped under the following headings:

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The final paper, by J. A. S. SMITH, reviews the present situation in NQR spectroscopy and indicates the directions in which new developments are to be expected.

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suppose that the volume of a nucleus is proportional to its weight A and we may write for the value of r_0 , the distance at which the potential energy of the α particle is a maximum, $r_0 = \text{ca. } 1.2 \times 10^{-13} A^{1/3} [\text{cm}]$. At that time, this was a daring generalization because data were scarce [10].

In 1932, Chadwick discovered the neutron [14] and found its collision radius for lead in agreement with the radii of the α -radioactive nuclei, about 7 fm. The neutron was now recognized as a nuclear constituent. Although speculations flourished [6] on what nuclei might look in detail, experiments up to 1935 had given no evidence beyond the picture of very tiny spheres with vaguely defined but measurable radii.

2. Discovery of Nuclear Quadrupole Moments

50 years ago, Hermann Schüller and Theodor Schmidt reported conclusive evidence of non spherical nuclei*.

Schüller, then 40 years old, was a well known spectroscopist. He worked at the astrophysical observatory in Potsdam. Schmidt (born 1908) had joined him in 1934, coming from Leipzig where he had done postdoctoral work with Heisenberg. At Potsdam there existed a collection of rare earth samples; these had been used for studies of the line spectra of stars. Schüller and Schmidt started a program to study hyperfine structures (hfs) of rare earth elements in order to measure nuclear spins. In the spring of 1935 they put europium into their hollow cathode. The hfs of three resonance lines revealed that both isotopes 151 and 153 had spins $I = 5/2$, and that the ratio of the nuclear magnetic moments μ was 2.2:1. But the hyperfine components did not follow the Landé interval rule exactly (Figure 1). Such deviations had been seen before. They could arise from level perturbations if the magnetic hfs was not small compared to the fine structure. But these should be proportional to μ^2 . However, ^{153}Eu with the smaller μ had the larger effect. Schüller and Schmidt therefore concluded that they had found a new nuclear property: a deviation from spherical symmetry. Their publication in *Zeitschrift für Physik* is dated March 2, 1935

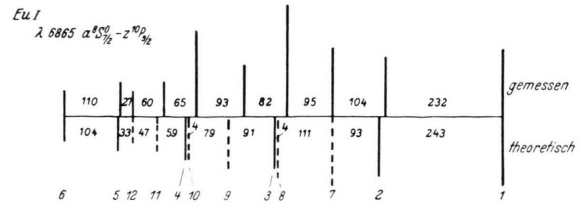


Fig. 1. Comparison of measured hfs (in 10^{-3} cm^{-1}) with that calculated ("theoretisch") without quadrupole interaction (with the same total splitting). Solid lines: ^{151}Eu , broken lines: ^{153}Eu (the isotopes were identified by the sign of the isotope shift). Since the ground state splitting was smaller than the line width, the pattern is mainly due to the upper P-state. From Schüller and Schmidt 1935 [17].

[17]. Another paper from April 28 with similar effects for ^{175}Lu [18], brings the word "electric quadrupole", which was brought to the authors attention by Delbrück.

Already on June 1, 1935, the dutch journal *Physica* received a manuscript from Hendrik B. G. Casimir with the correct quantum mechanical interpretation of the Eu hfs [19]. This paper gives the well known definition of the (spectroscopic) quadrupole moment Q and the formula for its interaction energy with the electron core. In his famous prize essay of 1936 "On the Interaction between Atomic Nuclei and Electrons" [20], Casimir elaborated the subject further. He obtained $Q(^{151}\text{Eu}) = +150 \text{ fm}^2$, $Q(^{153}\text{Eu}) = +320 \text{ fm}^2$, and $Q(^{175}\text{Lu}) = +560 \text{ fm}^2$. Assuming a nuclear radius of 7 fm, Casimir came "to the conclusion that the quadrupole moment cannot possibly be due to one proton. Thus it is necessary to assume that it is caused by a group of particles. It might even be that the nucleus as a whole has a prolate shape and that the nucleus as a whole is rotating about its major axis".

Schmidt had shown in 1937 [21] that the systematic dependence of μ as a function of I ("Schmidt lines") led to a single particle model of mechanical and magnetic nuclear moments. He applied his model to nuclear quadrupole moments three years later [22]. After subtracting the negative contribution of an odd proton, he plotted a deformation parameter α for the nuclear core. This plot is shown in Figure 2. It shows the striking predominance of large positive deformations and the now well known influence of shell structure. At that time (1940) it seemed to support an α particle model of the nucleus [24]. (See [25] for a simple theoretical proof that most nuclei must have positive Q .)

* Personal recollections about these years have been written by Schmidt [15] and Casimir [16].

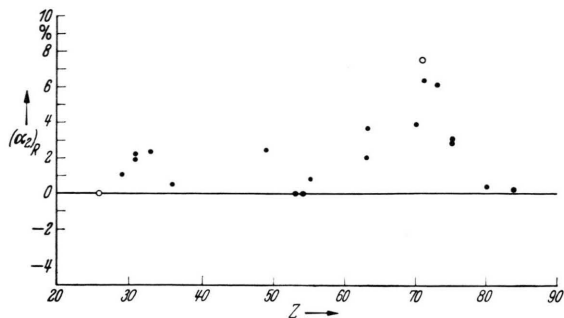


Fig. 2. Nuclear deformation parameter α (different from the presently used β) plotted against atomic number Z . The negative contribution of an odd proton has been removed; this has to be noted in comparing with later presentations, e.g. of Townes, Foley, and Low [23]. From Schmidt 1940 [22].

3. Spectroscopic Quadrupole Moments

3.1. Absolute Measurements

In atomic hfs, the measured electric quadrupole interaction constant called B (by Casimir) contains the product $V_{zz}Q$. In order to evaluate Q , the electric field gradient V_{zz} at the nucleus has to be known. Casimir showed how V_{zz} could be calculated for the p, d... valence electrons. But he made clear that “it is very difficult to estimate the accuracy” of that value. The perturbations of the electron core by the quadrupole moment give rise to shielding and antishielding effects. These were first calculated by Sternheimer [26], the respective correction factors bear his name. Since there will be a contribution by Sternheimer, no further explanation is necessary.

The spectroscopic quadrupole moment Q is one of the few model-independent well defined properties of nuclear states; absolute values are, therefore, very important for nuclear physics. They can be used to calibrate field gradients and thus to determine empirical Sternheimer factors.

The simplest well known electric field gradient is that of the nuclear Coulomb field outside of the nuclear forces. It has only recently been possible to use it for an absolute determination of Q . The experiment was based on the measurements of angular distributions for the Coulomb scattering of aligned ^7Li ions with Ni and Sn targets. The only available beams of polarized or aligned heavy ions are those installed by a Marburg-Heidelberg co-

operation at our Heidelberg tandem van-de-Graaff accelerators. From the tensor analysing powers, which require only the ratios of differential cross sections for unpolarized and aligned ^7Li beams, $Q(^7\text{Li}) = -3.70(8) \text{ fm}^2$ was obtained [27]. This is in excellent agreement with $-3.66(3) \text{ fm}^2$ as deduced [28] from ^7LiH , but not with a very recent re-evaluation of the same data [29] which favours -4.06 fm^2 . With the new absolute Li quadrupole moment [27], V_{zz} can be reliably determined in the bulk [30] and on the surface [31] of materials.

For nuclei heavier than Li, this method will not be easily applicable. Fortunately, a universal possibility exists for measuring Q without “Sternheimer uncertainties”, namely by utilizing the field gradients of those states in muonic atoms where the orbits are outside of the nuclear volume. Great progress has recently been achieved by Leisi and collaborators through their installation of a curved-crystal spectrometer at the muon channel of the SIN at Villigen, Switzerland [32]. Figure 3 shows the amazing accuracy that has been reached in resolving the hfs of the $2p_{3/2}$ level in the muonic ^{23}Na atom. It should be noted that one bin (0.95 eV) corresponds to an instrumental angle of 0.9 arcsec; this is the angle under which Jupiter’s moon Io is seen from the earth. A value $Q(^{23}\text{Na}) = +10.06(20) \text{ fm}^2$ has been determined [33], and by the same method $Q(^{25}\text{Mg}) = +20.1(3) \text{ fm}^2$ and $Q(^{27}\text{Al}) = +15.0(6) \text{ fm}^2$ [34].

Precise spectroscopic ground-state quadrupole moments for eleven rare earth nuclides were determined recently with an uncertainty of less than one percent from the hfs of muonic M X-rays measured with a Ge(Li) spectrometer [35]. They include $Q(^{151}\text{Eu}) = 90.3(10) \text{ fm}^2$ and $Q(^{153}\text{Eu}) = 241.2(21) \text{ fm}^2$. Other new absolute data are, e.g., $Q(^{233}\text{U}) = 366.3(8) \text{ fm}^2$ and $Q(^{235}\text{U}) = 493.6(6) \text{ fm}^2$ from muonic M and N X-rays [36].

3.2. Relative Hyperfine Data. Europium Revisited

25 years ago, the laser was invented. In the last, say, 10 years, with the development of tunable dye lasers and the availability of on-line mass separators at particle accelerators, optical hfs studies have come back into full bloom (see, e.g. [37]). This is a good example for the “golden rule of experimental physics”: “One has to make something new in order

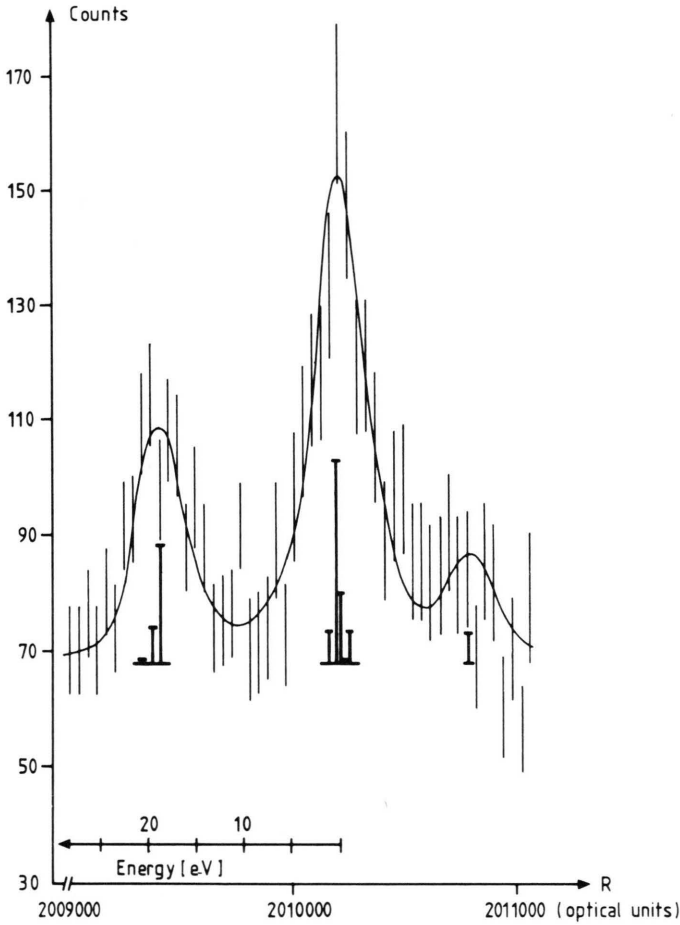


Fig. 3. Measured and fitted hfs of the $3d_{5/2}-2p_{3/2}$ X-ray line in muonic ^{23}Na . The quadrupole interaction in the p-state is dominant. The peaks may, therefore, be identified with transitions to the hfs levels with F equal 2, 1 + 3, 0 (from left). One bin corresponds to 0.95 eV. From Jeckelmann et al. 1983 [33].

to see something new” (this remark is from the greatest German physicist of the 18th century, G. C. Lichtenberg, Professor at Göttingen, who lived in Darmstadt from the age of three to twenty-one). Another application of this “golden rule” is collinear laser spectroscopy.

The limited resolution of hyperfine structures due to the Doppler broadening has been the central problem for generations of atomic spectroscopists. Schüler started to fight it in 1930 with his liquid-air cooled hollow cathode [38]. 50 years ago (1935) Jackson and Kuhn [39] and Minkowski [40] were the first to use a “one dimensionally cold gas” by producing atomic beams and observing the light vertical to the beam direction. Kaufman [41] found the important fact that the simple relation

$$\delta E = m v \delta v \quad \text{from} \quad E = m v^2/2 \quad (1)$$

shows how the velocity spread δv in an atomic or ion beam can be highly reduced by *acceleration*. The Doppler broadening due to the thermal kinetic-energy spread δE of the ion source is thus reduced immensely (of the order of 1000) by observing *in* the beam direction. The overlap of isotopic hfs is eliminated. Beams can be prepared in a variety of states (see, e.g. [42], [43]). More than about 200 nuclides have already been investigated with this technique.

Only two new examples relevant to quadrupole moments can be presented. Figure 4 is from the first investigation of radium hfs. The on-line collinear fast beam laser spectroscopy was carried out at the ISOLDE mass separator at CERN. It has enabled the determination of I , μ and Q for the isotopes 211, 213, 221, 223, 225, 227 and 229, in addition to isotope shifts in the mass range 208 to 232, with

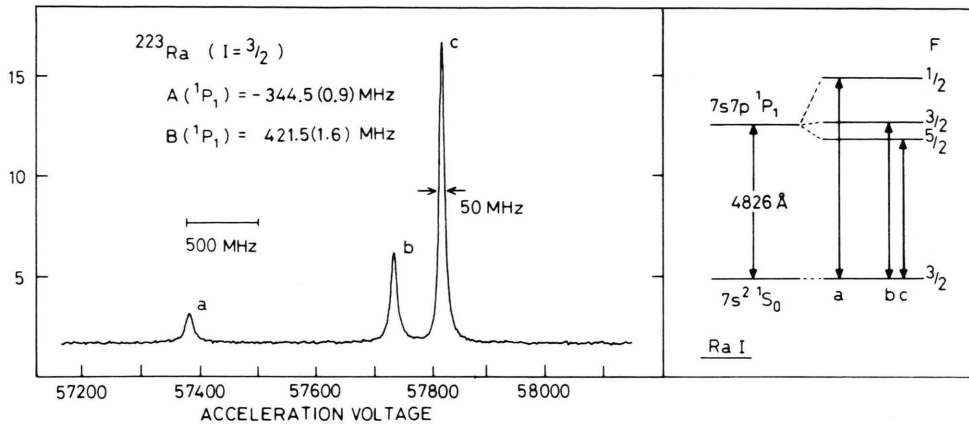


Fig. 4. Hfs in the atomic (Ra I) resonance line for ^{223}Ra . The fluorescence signal is shown as a function of the acceleration voltage ("Doppler scanning"); the frequency of the laser stayed fixed. From Ahmad et al. 1983 [44].

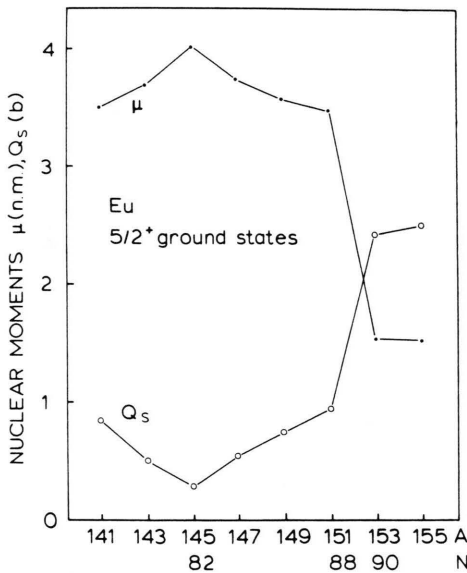


Fig. 5. Plot of magnetic and spectroscopic quadrupole moments ($1b = 100 \text{ fm}^2$) for the $5/2^+$ ground states of odd Eu isotopes against neutron number N . The values of ^{155}Eu are from [45]. From Ahmad et al. 1985 [46].

beam intensities as low as 10^4 Atoms/s. The figure shows a strong deviation from the interval rule. The precise interaction constant B of the 1P_1 state for ^{223}Ra and four other Ra isotopes (those with $I > 1/2$) lead to relative Q values of comparable accuracy. It is interesting to note that the spectroscopic quadrupole moments, e.g. $Q(^{223}\text{Ra}) = 120 \text{ fm}^2$, have been derived [44] via the intrinsic quadrupole

moment Q_0 of the strongly deformed isotope ^{229}Ra , obtained from purely nuclear experiments.

We return to europium with Figure 5. It presents the present knowledge of nuclear magnetic and quadrupole moments for the ground states of odd Eu isotopes. The values for Q show a minimum at the neutron shell closure ($N = 82$) and the big jump between $N = 88$ and 90 . The unique situation that Eu has two stable isotopes, that a drastic shape transition just happens for them, and that it is correlated with an opposite change in magnetic moments, was Schüler and Schmidt's good fortune. But "fortune favours the prepared mind"*. As a matter of fact, Racah had shown in 1931 [48] that a nucleus prolonged by one tenth (1 fm) of its radius would cause deviations from the Landé interval rule of the order of the magnetic hfs in $^2P_{3/2}$ levels. At that time, such deviations had not been observed. When they were found, perturbations between close lying levels (see [20]) were assumed as a plausible reason.

4. Intrinsic Quadrupole Moments

4.1. Discovery from Isotope Shifts

The unusual nuclear structure change between neutron numbers 88 and 90 (Fig. 5) has also led to

* I learnt this rule from the great spectroscopist Gerhard Herzberg who studied, worked, and taught at the Technische Hochschule Darmstadt until he had to leave in 1935 – fifty years ago [47].

the discovery of intrinsic quadrupole moments. Schüler and Schmidt's interferometric studies of the Potsdam rare earth samples again marked the beginning. In 1934 they reported on non-equidistant isotope shifts (IS) for the even samarium isotopes [49]: The ^{150}Sm – ^{152}Sm shift was nearly twice that between ^{152}Sm and ^{154}Sm . The reason for the “*einschneidende Strukturänderung*” (sincere change of structure) between ^{150}Sm and ^{152}Sm was brought into connection with the α activity of Sm and later with an α particle model [24].

A quantitative connection with the differently deformed Eu isotopes of the same neutron numbers was made 1946/47 at Göttingen [50] by the following idea: If one assumed that the necessarily spherical symmetric $I=0$ nuclei ^{150}Sm and ^{152}Sm had about the same *intrinsic* deformation as ^{151}Eu and ^{153}Eu , then the increase in deformation should yield a deformation contribution to the volume IS calculable from the Eu quadrupole moments. In modern presentation and slightly simplified*, the total IS δE is

$$\delta E = (2\pi/3) Z e^2 \Delta L(0) \delta \langle r^2 \rangle + c \delta A + \delta E_{\text{pol}}. \quad (2)$$

Here $\Delta L(0)$ is the change of electron density at the nucleus in the respective atomic transition, the last two terms are the mass and nuclear polarization contributions. With [50]

$$\delta \langle r^2 \rangle = \delta \langle r^2 \rangle_{\text{vol}} + \delta \langle r^2 \rangle_{\text{def}}, \quad (3)$$

the IS of heavy elements should thus be a nuclear *radius* effect, not a simple *volume* effect: The rms radius depends upon the deformation as well as the volume.

The quantitative test [54] of this hypothesis was rather complicated. The last two terms in (2) could be neglected for Sm by reasonable arguments. In order to determine $\delta \langle r^2 \rangle$, the ratio $\delta E / \Delta L(0)$ had to be evaluated from the hyperfine structures. But Schüler and Schmidt [49] had only given the hfs of a single line ($\lambda = 5321 \text{ \AA}$) which was not uniquely classified and, amazingly, showed the “wrong” sign of the IS. New measurements had thus to be made for reliably known transitions.

* See, e.g., [51], [52] for finer details and for the relation to the formerly used IS constants C . An elementary derivation of the first term in Eq. (2) may be found in [53].

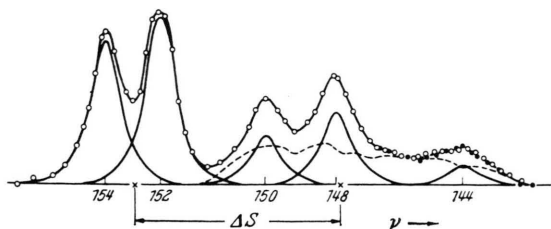


Fig. 6. Intensity distribution of $\lambda 6589 \text{ \AA}$ (Sm I) hfs from a photometer registration of the Fabry Perot photograph. The intensity below the broken curve is due to the odd isotopes. ΔS is the distance (0.153 cm^{-1}) between the two centers of gravity (see text). From Brix and Kopfermann 1949 [54].

Figure 6 presents the then best resolved hfs (see [55] for presently obtainable resolution!). In other lines, the odd isotopes 147 and 149 complicated the structure even more. For 5 favourable lines, the relative IS was measured and related to the splitting ΔS (Figure 6). This distance between the centers of gravity of the two groups ($^{154}\text{Sm} + ^{152}\text{Sm}$ and the rest) was primitively determined: Intensity curves registered by a photometer were enlarged, copied on thick paper, cut out and the centers of gravity of the two parts found mechanically. Using ΔS had the advantage that many lines with a badly resolved hfs could be included in the analysis: The distance between the “sharper” component ($152 + 154$) and the “diffuse” rest was easier to measure. Furthermore, available grating measurements for Sm II [56], where the two complexes were resolved, could also be utilized.

Altogether, 87 lines of Sm I and 18 of Sm II were used to determine the IS of levels with known electron configuration [54], [57]. The “wrong” sign of $\lambda 5321 \text{ \AA}$ (and other lines) was “explained away” by the assumption of a transition $4f^6 6s^2 - 4f^5 5d 6s^2$ with corresponding screening effects on s^2 electrons. This interpretation was later confirmed.

Using for $\Delta L(0)$ the Goudsmit-Fermi-Segrè formula, $\delta \langle r^2 \rangle$ from Sm I and Sm II agreed well. The result is presented in Figure 7. Also shown is the large ^{151}Eu – ^{153}Eu radius change, calculated from the data of [17], which confirmed the postulated quadrupole effect. The deformation contribution to the IS of ^{150}Sm – ^{152}Sm was found to be about twice that calculated from the ^{151}Eu and ^{153}Eu quadrupole moments (using 120 fm^2 and 250 fm^2 from [59]). In view of the various uncertainties and

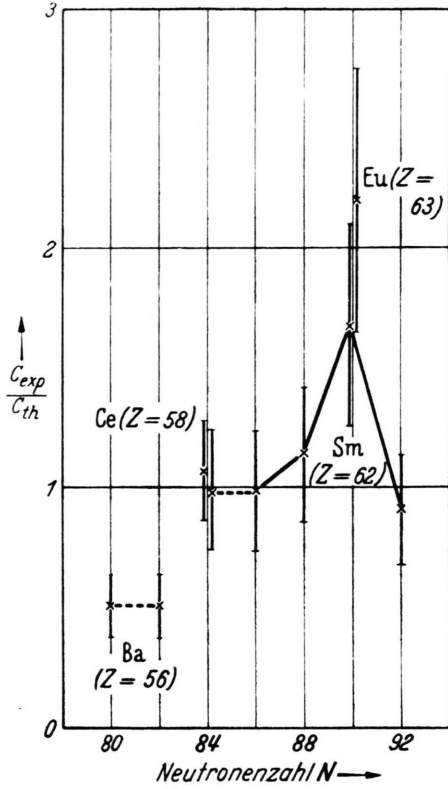


Fig. 7. The first diagram of radius change for isotopic pairs against neutron number N (of heavier isotope). The ordinate corresponds to $\delta\langle r^2 \rangle$ for $\Delta N = 2$ in units of a “standard” value which is here 0.29 fm^2 . From Brix and Frank 1950 [58].

assumptions, this was considered a “surprisingly good” agreement [54].

The idea of intrinsic quadrupole moments for $I = 0$ nuclei was at first not generally accepted. It was said that they cannot be measured and therefore do not exist. I remember that C. F. von Weizsäcker at that time made the encouraging remark that “an elephant in a s-state is spherically symmetric but still remains an elephant”. When I met von Weizsäcker recently, he recalled at once that he had said this, but added that the example came from Heisenberg.

The IS between ^{140}Ce and ^{142}Ce was studied next [58] (Figure 7). It confirmed that $\delta\langle r^2 \rangle$ increases abruptly beyond the “magic” neutron number 82, as had been suspected from the Sm data.

A severe uncertainty remained, however, because so far it had always been assumed that only the valence s-electrons contributed to $\Delta L(0)$. The

possible contribution of deeper s^2 shells due to a change of screening in the transition was first evaluated [60] for Eu by comparing IS and magnetic hfs carefully; luckily, it turned out to be only a few percent. These measurements yielded an improved radius change $\delta\langle r^2 \rangle$ ($151-153$) = $0.67(9) \text{ fm}^2$ (from $C_{exp}/C_{th} = 2.29(29)$, see Figure 7). The most recent value is $0.602(33) \text{ fm}^2$ [46] (see also [61]).

Age Bohr was the first theoretician who took the consequences of intrinsic quadrupole moments (Q_0) seriously. He also pointed out [62] that (for nuclei with $I > 1/2$) the spectroscopic quadrupole moment Q is quite different from Q_0 by the relation

$$Q_0/Q = (I+1)(2I+3)/I(2I-1). \quad (4)$$

This holds for strong coupling, i.e. strongly deformed nuclei.

Both the Eu nuclei have $I = 5/2$; thus $Q_0/Q = 14/5$. This enormous factor enters quadratically into the deformation effect of the IS. So, in retrospect, the anomalous shift between ^{150}Sm and ^{152}Sm was not *larger* than inferred from Eu [54], but actually *smaller*. “Fortunately”, the big error in using Q instead of Q_0 was partly compensated by other uncertainties. For instance, the volume part $\delta\langle r^2 \rangle_{vol}$ (Eq. (3)), taken [54] from the 144-148-150 IS was overestimated in the light of our present knowledge (compare Figure 7 with Figure 10).

4.2. Present Status. Samarium Revisited

After 1950, nuclear physics experiments brought convincing evidence of intrinsic quadrupole moments, above all through the rotational motion of strongly deformed nuclei. This development is well known and shall not be discussed. It is presented, e.g., in the Nobel lecture of A. Bohr [63] and in the monograph of Bohr and Mottelson [64]. An early review (as of 1957) was given by Temmer [65].

A striking demonstration of large Q_0 is the splitting of the nuclear giant electric dipole resonance [66], [67]. In Fig. 8 it is seen that this indeed happens from ^{150}Sm to ^{152}Sm , as inferred from the isotope shifts. The lower energy component corresponds to a vibrational absorption of the prolate nucleus along its symmetry axis, the higher in the perpendicular direction. The measured energy splitting yields $Q_0(^{152}\text{Sm}) = 590(40) \text{ fm}^2$ [68], slightly smaller than $Q_0(^{153}\text{Eu}) = (Q_0/Q) \cdot 241 \text{ fm}^2 = 675 \text{ fm}^2$ [35].

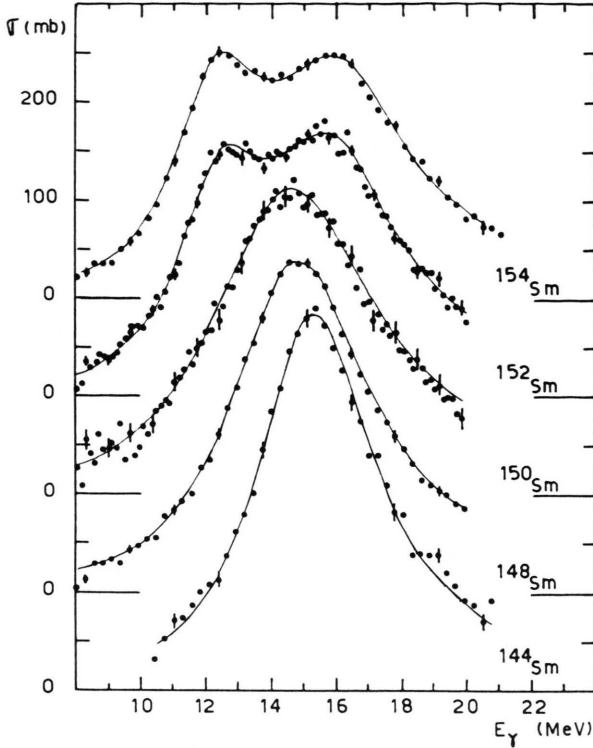


Fig. 8. Total photoneutron cross sections of even Sm isotopes. Single Lorentz fits are shown for 144, 148, 150; two Lorentz lines have been fitted for 152 and 154. From Carlos et al. 1974 [68].

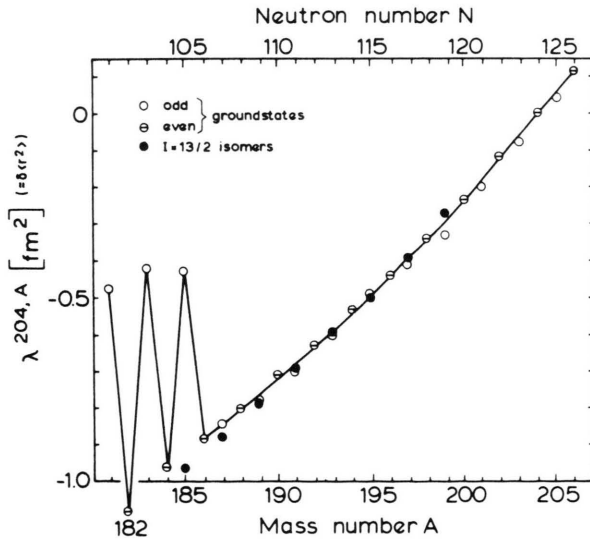


Fig. 9. Hg isotope shifts including results from radioactive detection of optical pumping (1972), laser fluorescence spectroscopy (1977), and collinear laser spectroscopy (1983). The ordinate is practically $\delta\langle r^2 \rangle$, referred to ^{204}Hg . From Otten 1985 [71].

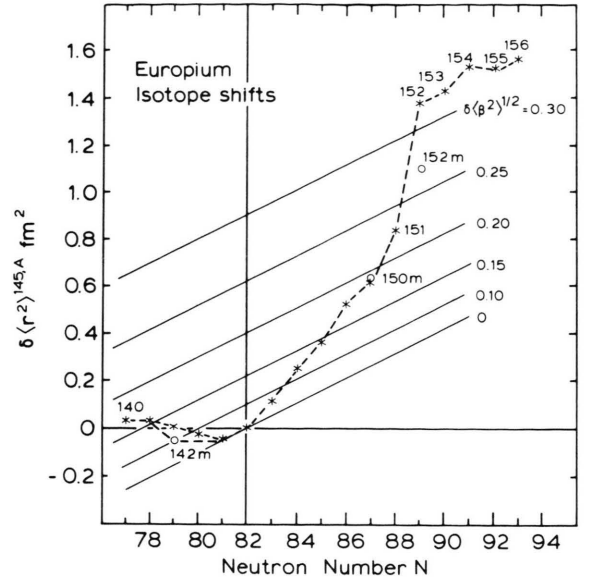


Fig. 10. Eu isotope shifts from collinear fast-beam laser spectroscopy [73]. The ordinate is $\delta\langle r^2 \rangle$, referred to ^{145}Eu which is assumed to be spherical. The deformation can be read from the parallel lines, the slope of which is given by the spherical droplet model. Data from Ahmad et al. 1985 [46] and Dörschel et al. 1984 [45]. Corresponding plots for Sm are presented by [74], [75].

A surprising effect was found about a decade ago by Otten and his collaborators when they studied the isotope shifts of a long chain of radioactive neutron deficient mercury isotopes [69], [70]: the addition of a single neutron may change the nuclear shape drastically. The present available data in Fig. 9 show that the odd isotopes $^{181}, ^{183}, ^{185}\text{Hg}$ have much larger radii than the adjacent even ones. This is interpreted in the following way [72]: The nuclei with even A are nearly spherical (slightly oblate), the odd ones are strongly prolate ($\beta = 0.25$). In retrospect, therefore, it was by no means trivial that the “removal” of the odd protons from $^{151}, ^{153}\text{Eu}$ should lead to similarly deformed $^{150}, ^{152}\text{Sm}$ nuclei!

How well a radius change can be interpreted nowadays as a sum of volume and deformation changes (Eq. (3)) is illustrated for Eu by Figure 10. The spherical droplet model of Myers and Schmidt [76] was used to calculate $\delta\langle r^2 \rangle_{\text{vol}}$, the deformation parameter β is then obtained by

$$\begin{aligned} \delta\langle r^2 \rangle - \delta\langle r^2 \rangle_{\text{vol}} &= \delta\langle r^2 \rangle_{\text{def}} \\ &= (5/4\pi) \langle r^2 \rangle \delta\langle \beta^2 \rangle. \end{aligned} \quad (5)$$

For a spheroidal nucleus, β is related to the radius $R(\theta)$ by

$$R(\theta) = R_0(1 + \beta Y_2^0(\theta)), \quad (6)$$

where R_0 is the radius of a sphere with equal volume. The value $\beta = 0.32$ for ^{153}Eu (and $^{152}\text{Eu}!$) from Fig. 10 agrees exactly with $\beta = 0.32$ from $Q_0 = 675 \text{ fm}^2$ (see above) and also with $\beta = 0.31$ from $B(E2)$ values [46]. The analyses as well as the model of a static deformation of ^{153}Eu are, therefore, consistent.

Returning to Sm and Eq. (2), the mass term $c\delta A$ had been neglected by [54] with the argument that the non-equidistant relative IS seemed to be the same for all lines. Modern laser spectroscopy has improved the accuracy immensely. Differences in relative shifts are now well measurable and can be used to eliminate the mass dependent shifts, as shown by [55]. The methods for evaluating $\Delta L(0)$ have also been much refined during the last decades. Recent values for $\delta\langle r^2 \rangle$ are given in Table 1. (They are well within the 25% uncertainty estimated for the old values [54], see Figure 7.)

The ignorance about the contribution δE_{pol} in (2) caused, or should have caused, a permanent bad conscience. Fortunately, polarizabilities of some Sm isotopes have at last been calculated [77] in 1984. Table 1 shows that the differences δE_{pol} are not negligible, not even comfortably small, compared with the present experimental precision.

The correct interpretation of the IS for stable isotopes is finding renewed interest with respect to the measurements on numerous radioactive isotopes. Such data for Sm were presented by two groups

Table 1. Radius and polarization effects in the IS of stable even Sm isotopes. Column 2 gives $\delta\langle r^2 \rangle$ evaluated under the usual assumption of a negligible polarization shift (from laser-atomic-beam spectroscopy on 15 Sm I lines by Brand, Seibert and Steudel [55]). The contribution δE_{pol} due to different quadrupole polarizability, calculated by Hoffmann, Baur and Speth [77] is formally expressed as $\delta_{\text{pol}}\langle r^2 \rangle$ (column 3), it has to be subtracted from column 2.

Isotopes	measured $\delta\langle r^2 \rangle$ [fm ²]	calculated $\delta_{\text{pol}}\langle r^2 \rangle$ [fm ²]
144–148	0.517(27)	
148–150	0.303(16)	–0.006
150–152	0.423(22)	–0.027
152–154	0.230(12)	–0.014

[74], [75] at the 7th International Conference on Atomic Masses and Fundamental Constants in Darmstadt-Seeheim 1984.

5. Charge Distribution of Deformed Nuclei

Whereas the spectroscopic quadrupole moment Q is uniquely defined, the intrinsic moment Q_0 and the angular shape parameter β can only be derived with additional nuclear information and model assumptions. But one would like to “see” the intrinsic nuclear charge distribution which gives rise to Q_0 or Q , and such pictures may be useful.

Holmium with $I = 7/2$ was the first strongly deformed nucleus for which the intrinsic shape became well known. Figure 11 is the spectrum of the muonic K_α line of ^{165}Ho , measured with a large Ge(Li) spectrometer. In these low states, the muon moves partly within the nucleus and thus probes the spacial distribution of charge and quadrupole moment. An analysis of the transitions between the levels 4f, 3d, 3p, 2s, 2p and 1s led to five parameters for the intrinsic charge distribution which is drawn in Figure 12. For the ground state, $Q = 349(3) \text{ fm}^2$ and $Q_0 = 747(7) \text{ fm}^2$ were evaluated; the first excited state has nearly the same Q_0 [78]. The charge distribution for ^{181}Ta in Fig. 12 was obtained in a similar way [79].

Strongly deformed $I = 0$ nuclei have a well developed ground-state rotational band. The form factors

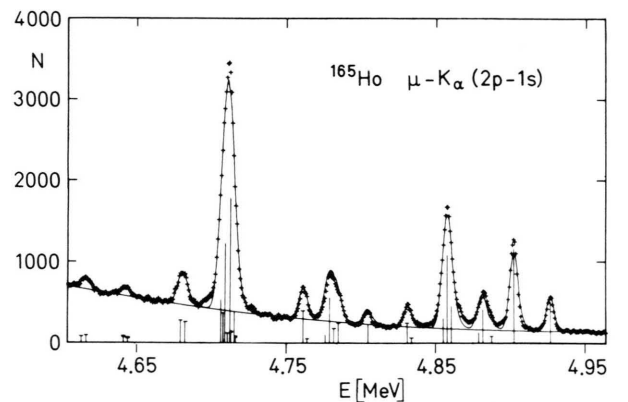


Fig. 11. Muonic 2p–1s transition for ^{165}Ho . The vertical lines indicate the hfs components that have been fitted to the measured spectrum. One or two horizontal bars are for transitions where the Ho nucleus was left in the first or second excited state. From Powers et al. 1976 [78].

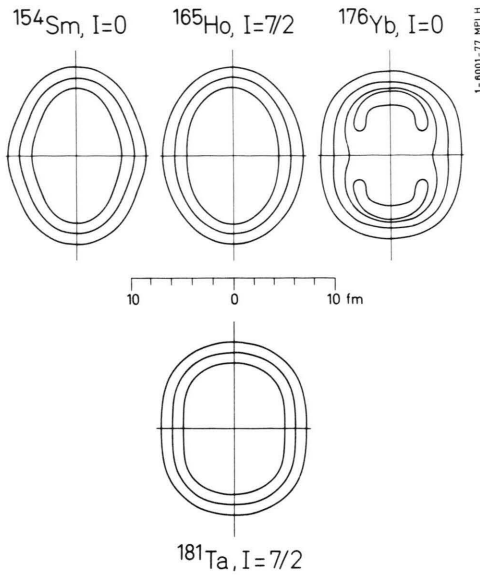


Fig. 12. Intrinsic charge distributions of permanently deformed nuclei. Lines of constant charge densities of 0.1, 0.5, and 0.9 (for Yb also 1.05) times the central density are drawn in a cross section of the nuclei. There is rotational symmetry around the long (vertical) axis. For Ho and Ta from muonic X-rays alone, for Sm and Yb mainly from electron scattering (see text).

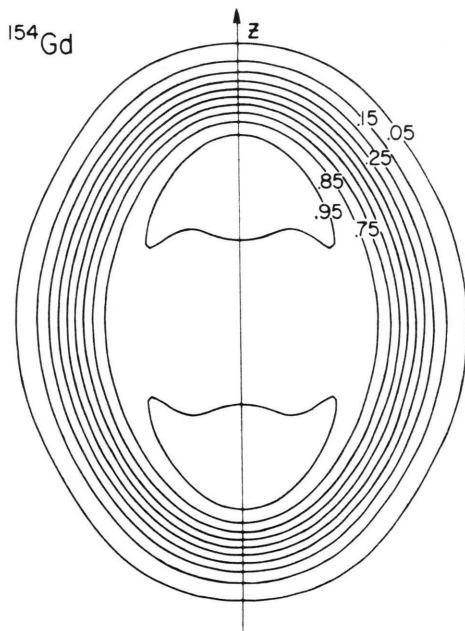


Fig. 13. Intrinsic charge distribution of ^{154}Gd , similar to Figure 12. From Heisenberg and Blok 1983 [81].

for the excitation of these states by inelastic scattering of high energy (several 100 MeV) electrons, combined with elastic electron scattering from the ground state as well as muonic X-ray data, give enough information to draw similar intrinsic charge distributions for $I=0$ nuclei. Results [80] for ^{154}Sm and ^{176}Yb have been added in Figure 12. The similar shapes of the odd and even nuclei determined in completely different ways, is impressive. A newer contour plot which gives an even more suggestive picture of the $N=90$ nucleus ^{154}Gd is presented in Figure 13. The ground state and rotational states up to 6^+ have been utilized in the analysis.

The nuclear contour lines in Figs. 12 and 13 are not elliptical: ^{154}Sm , ^{154}Gd and ^{165}Ho have a positive, ^{176}Yb and ^{181}Ta a negative hexadecapole (E4) moment. Precise E4 moments have recently been measured by muonic X-ray spectroscopy for uranium isotopes [36]. The wealth of information on E4 moments from Coulomb excitation and other nuclear methods must be omitted, as has been done for Q_0 values.

In his review on angular shapes of nuclei, Temmer wrote that in the vicinity of radium an octupole (pear-shaped) deformation seemed to occur: "Whether this β_3 deformation represents an actual equilibrium shape or merely a time-dependent octupole vibration ... is susceptible of experimental test" [65]. Figure 14 is from his paper of

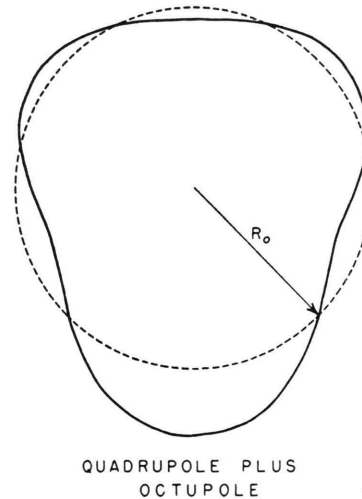


Fig. 14. Nuclear octupole deformation ($\beta_3=0.30$) superimposed upon a quadrupole deformation ($\beta=0.25$). R_0 gives the radius of the sphere of equal volume. From Temmer 1958 [65].

1958. As was mentioned, the hfs and IS of a series of Ra isotopes (with half lives between 23 ms and 1600 yr) was measured 25 years later [44], 85 years after the discovery of Ra by Mme Curie. The IS is larger than that inferred from the quite reliably calculable quadrupole deformation effect [82]. Again it is concluded – as earlier for Sm – that this could be due to an additional higher order deformation which increases $\delta\langle r^2 \rangle$ further. A deformation parameter $\beta_3 \cong 0.1$ is inferred; this means an intrinsic pear-shape, less pronounced than Figure 14. The IS further indicates that some odd-A isotopes of Ra have a larger β_3 than neighbouring even ones. All this fits in nicely with evidence for stable octupole deformations in the Ra-Th-region from nuclear spectroscopy [83], [84], [85]. The new radioactive decay mode, where a complete ^{14}C nucleus is emitted [86], [87] from $^{222,223,224}\text{Ra}$ may be connected with this unsymmetric nuclear shape.

Another hot topic in these days is the question whether triaxially deformed nuclear ground states occur. One manifestation of this could be a splitting

of the *magnetic* dipole giant resonance. It has been observed for ^{164}Dy and ^{174}Yb in the Institut für Kernphysik of the TH Darmstadt [88] and may be taken as an indication, though not by itself as a proof, of triaxial deformation [89].

6. Conclusion

Fifty years of nuclear quadrupole moments: they were an exciting chapter in the studies of matter, looking at the atomic nuclei in enormous magnification. It even seems that the field has only now been really opened. Admittedly, this was a rather hurried “european” tour, therefore apologies to all those whose important work has been omitted.

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