EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-EP/87-229
23.12.1987

MEAN SQUARE CHARGE RADII OF RADIUM ISOTOPES
AND OCTUPOLE DEFORMATION IN THE $^{220-228}\text{Ra}$ REGION

S.A. Ahmad^{1*}, W. Klempt¹, R. Neugart², E.W. Otten²,
P.-G. Reinhard³, G. Ulm¹⁺ and K. Wendt²

1 CERN, Geneva, Switzerland

2 Institut für Physik, Universität Mainz,
Mainz, Federal Republic of Germany

3 Institut für Theoretische Physik, Universität Erlangen
Erlangen, Federal Republic of Germany

and the ISOLDE Collaboration
CERN, Geneva, Switzerland

Abstract

The first investigation of isotope shifts in both the atomic and ionic resonance lines of radium has been carried out using the technique of on-line collinear fast-beam laser spectroscopy at the ISOLDE facility at CERN. The measurements cover 19 isotopes in the mass range $208 \leq A \leq 232$ with half-lives between 23 ms and 1600 y. The differences in the nuclear mean square charge radii $\delta\langle r^2 \rangle$ have been evaluated and - together with earlier published spins and moments from the hyperfine structure - related to nuclear deformation. In particular the inversion of the odd-even staggering effect for the isotopes ^{221}Ra , ^{223}Ra and ^{225}Ra can be interpreted by the presence of octupole instability and adds weight to the concept of near-stable octupole deformation in the odd-A isotopes which already explained their spins and magnetic moments.

(Submitted to Nuclear Physics A)

* Present address: Bhabha Atomic Research Centre, Bombay-400 005, India

+ Present address: Physikalisch-Technische Bundesanstalt, Institut Berlin, D-1000 Berlin 10, Germany

Keywords:

NUCLEAR STRUCTURE $^{208-214,220-230,232}\text{Ra}$;
measured isotope shifts, hfs; deduced mean square charge
radii, spins, magnetic dipole moments, electric quadru-
pole moments, deformation β , odd-even staggering;
collinear fast-beam laser spectroscopy

1. Introduction

The optical isotope shift (IS) probes sensitively the changes in the nuclear charge distribution between the isotopes of an element, thus providing systematic information about nuclear volume and shape within an isotopic chain. This approach has gained particular importance in the regions of unstable nuclides, owing to the high sensitivity and spectral resolution of new laser-spectroscopic techniques. We report here the first measurement of IS in the atomic (Ra I) and ionic (Ra II) spectra of radium involving 19 isotopes in the mass range $208 \leq A \leq 232$. The lighter isotopes lie in the spherical region of the neutron shell closure at $N = 126$, whereas the neutron-rich, heavier ones lie in an interesting region of deformation the character of which is being very much discussed. There are theoretical predictions as well as spectroscopic evidence for the existence of sizeable and possibly stable octupole deformation in this region of nuclei [refs.¹⁻³) and references therein].

The IS essentially yield $\delta\langle r^2 \rangle$, i.e. the changes in the nuclear mean square charge radius as a function of neutron number N . Since the quadrupole deformation parameter β_2 is known from $B(E2)$ values for a few doubly-even isotopes⁴), one can attempt to study the influence of higher-order deformations on $\delta\langle r^2 \rangle$. This can be done by analyzing deviations of the experimental results from the standard geometrical two-parameter model⁵) in which one parameter corresponds to the change in nuclear volume, and the other takes into account the effect of nuclear shape which is usually represented by the difference $\delta\langle \beta_2^2 \rangle$ in the quadrupole deformation only. We investigate qualitatively the influence of octupole deformations in the neutron-rich isotopes by comparing their mean square radii to the one of the spherical, magic nucleus ^{214}Ra .

Out of the 19 isotopes for which the IS has been measured, 11 are even-even and 8 are even-odd. We have previously reported the nuclear spins and moments from the hyperfine structure (hfs) of the odd- A isotopes⁶⁻⁸). Together with data from nuclear spec-

troscopy, they have supported the theoretical expectation of near-stable octupole deformation in the region around $A = 224$ [refs.^{2,9}]. A regular feature of the IS is the odd-even staggering (OES) which usually indicates a slightly smaller mean square radius of the odd- N isotopes compared to the average of their even- N neighbours. This is called the normal OES. We observe, however, an inversion of the OES for the sequence ^{221}Ra , ^{223}Ra and ^{225}Ra , and we find arguments for this unusual behaviour being also due to octupole instability.

2. Experimental Details

2.1 Production of the Radium Isotopes

The experiment was carried out at the ISOLDE mass separator at CERN¹⁰). Spallation of ^{238}U , induced by 600 MeV protons from the CERN synchro-cyclotron, leads to Ra isotopes either as primary reaction products or as decay products from isotopes of the neighbouring elements Th and Ac. The target consists of UC_2 (13 g/cm²) formed on the surface of a graphite cloth. During operation it is heated to 2200°C in order to release the volatile spallation products. Typical release times are of the order of 1 sec. Therefore, the very short-lived α -emitters $^{215-220}\text{Ra}$ decay mostly in the target. The atoms are ionized on the hot (2400°C) tungsten orifice of the target container. The ions are then accelerated to 60 keV, mass separated and transmitted to the setup for collinear fast-beam laser spectroscopy. The yield of even- A Ra isotopes, normalized to a proton beam intensity of 1 μA , is shown in Fig. 1. During the course of our experiment, typical intensities were about 2 μA . The relative yields have been determined from the intensity of fluorescence signals and calibrated with an absolute yield measurement of ^{222}Ra by α -counting. The fluorescence signals could be observed down to yields of 10^4 atoms/sec, requiring a total integration time of about 1 hour (see the top part of Fig. 1). The heaviest isotope ^{232}Ra was discovered in the present study by laser spectroscopy⁶). Recently, its half-life has been reported¹¹) to be 250(50)s.

2.2 High Resolution Spectroscopy of Radium

There is only limited knowledge available on the atomic energy levels of radium which mainly stems from just two publications on the spectra of Ra I¹²⁾ and Ra II¹³⁾; they concern only the long-lived ²²⁶Ra. Our laser spectroscopy measurements have been carried out in the Ra I line at 4826 Å ($7s^2\ ^1S_0 - 7s7p\ ^1P_1$) and the ionic Ra II line at 4682 Å ($7s\ ^2S_{1/2} - 7p\ ^2P_{1/2}$). Both these lines are the strongest of the respective system suitable for cw laser spectroscopy. The hfs and IS in a few additional Ra I lines have been studied for the analysis of the semi-empirical hfs parameters. This is described and discussed separately⁷⁾, with emphasis on the atomic physics aspects of the high-resolution spectroscopy in Ra I and Ra II.

The experimental setup used for the atomic 4826 Å line is essentially the same as described in our publication on Ba isotopes¹⁴⁾. In order to investigate the ionic 4682 Å line, this setup has been slightly modified. The charge-exchange cell has been removed and the observation region has been electrically insulated from the ground to form a Faraday cage (Fig. 2). The Doppler tuning could then be performed by applying to this cage a variable voltage up to ± 10 kV, corresponding to a frequency span of about 80 GHz.

A commercial single-frequency stabilized cw dye laser, operated with the dye Coumarin-102 and pumped with the violet output of a krypton laser, was used for the experiment. The detection and the data acquisition system are the same as described in ref.¹⁴⁾.

3. Data Taking and Results

For data taking the resonances of three different isotopes are recorded in a computer-controlled fast-cycle measuring sequence. Of the three recorded spectra, two are on fixed reference isotopes; the IS between them provides an on-line test of the stability of the high voltage and laser frequency. Examples of scans are given at the top of Fig. 1. The relative positions of all the

resonances observed in both the $4826 \text{ \AA}$ and $4682 \text{ \AA}$ lines have been evaluated with an experimental error of the order of 10 MHz. The measured IS [see ref.⁷] are presented in Table 1 for the Ra I line ($4826 \text{ \AA}$). The observed resonance positions are plotted in Fig. 3, taking as reference the isotope ^{214}Ra with the closed neutron shell at $N = 126$; this figure also provides the details of the hyperfine splittings of the odd-A isotopes and the relative intensities of the hfs components. The centres of gravity for the odd-A isotopes have been evaluated from their hfs analyses as reported in refs.^{6,7}). A corresponding plot of the resonance line in Ra II can be found in ref.⁷).

4. Evaluation of the Nuclear Parameters

4.1 Spins and Moments

The analysis of the hfs, yielding spins, magnetic dipole and electric quadrupole moments, has been described elsewhere⁶⁻⁸). These results are compiled in Table 2. The spins were derived from a consistent parametrization of the hfs in the Ra I transition $7s^2 \text{ } ^1S_0 - 7s7p \text{ } ^1P_1$ and the Ra II transition $7s \text{ } ^2S_{1/2} - 7p \text{ } ^2P_{3/2}$. Initially the magnetic moments had to be related to the magnetic interaction constants (A-factors) of the ionic $7s \text{ } ^2S_{1/2}$ ground state by a semi-empirical evaluation of the hyperfine field^{6,7}), or by ab-initio calculations using relativistic many-body perturbation theory (RMBPT)¹⁵⁻¹⁷). The remaining 5 % uncertainty on the magnetic moments was removed recently by a direct g_I -factor measurement from the free Larmor precession of ^{213}Ra and ^{225}Ra nuclei⁸) of which the results are quoted in Table 2. The spectroscopic quadrupole moments are still somewhat preliminary. With the B-factors taken from the hfs of the $7s7p \text{ } ^1P_1$ state, they were calibrated with a reference value for ^{223}Ra obtained from a hfs measurement and semi-empirical analysis of the electric field gradient in the $7s7p \text{ } ^3P_2$ state⁷). The final analysis will include a recent measurement on the $7p \text{ } ^2P_{3/2}$ state performed in the UV resonance line ($3814 \text{ \AA}$) of the alkali-like Ra II¹⁸).

4.2 Extraction of the Field Shift

The observed IS between two isotopes A,A' in a transition i, $\delta v_{i,A,A'} = v_{i,A'} - v_{i,A}$, is the sum of the field shift (FS) $\delta v_{f,i}^{A,A'}$ and the mass shift (MS) $\delta v_{m,i}^{A,A'}$, and we have

$$\delta v_{i,A,A'} = \delta v_{f,i}^{A,A'} + \delta v_{m,i}^{A,A'} = \delta v_{f,i}^{A,A'} + M_i (m_{A'} - m_A) / m_A m_{A'}, \quad (1)$$

where m_A and $m_{A'}$ are the atomic masses. The MS is composed of the normal (NMS) and the specific mass shift (SMS). In s^2 -sp and s-p transitions the SMS is supposed to be small¹⁹). Conventionally one considers in these spectra only the NMS by taking $M_i = v_i / 1836.1$ where v_i is the transition frequency of the spectral line. In the Ra resonance lines the NMS is about 15 MHz for a pair of isotopes with $A' - A = 2$ and causes a contribution of a few times 10^{-3} to the total IS.

The field shift in each of the transitions is related to the nuclear parameter λ by the expression

$$\delta v_{f,i}^{A,A'} = F_i \lambda^{A,A'} \quad (2)$$

where F_i is the electronic factor and $\lambda^{A,A'}$ gives the change of the radial nuclear charge parameter written as

$$\lambda^{A,A'} = \sum_n \frac{C_n}{C_1} \delta \langle r^{2n} \rangle^{A,A'}. \quad (3)$$

The coefficients C_n have been calculated by Seltzer²⁰) for K x-ray transitions using a Fermi nuclear charge distribution. The ratios C_n/C_1 are almost independent of the principal quantum number and can be used also for optical transitions. For the heavy element Ra ($Z = 88$) the radial moments higher than $\delta \langle r^2 \rangle$ contribute almost - 10 %.

4.3 Evaluation of Electronic Factor F and Nuclear Parameter λ

The field shift of eq. (2) may be factorized further following the established semi-empirical procedure¹⁹⁾

$$\delta v_{f,i}^{A,A'} = E_i f(Z)^{A,A'} \lambda^{A,A'}. \quad (4)$$

The electronic factor E_i is proportional to the change of the total non-relativistic electron charge density $\Delta|\psi(0)|_i^2$ at the point nucleus,

$$E_i = a_0^3 \Delta|\psi(0)|_i^2 / Z = a_0^3 \beta_i |\psi_s(0)|^2 / Z, \quad (5)$$

where a_0 is the first Bohr radius and Z is the nuclear charge, and the shielding factor β_i is introduced to relate the IS to the dominating s-electron density. The factor $f(Z)$ includes the details of the Dirac wave function for a finite nuclear size. It is derived from the isotope shift constant $C_{\text{unif}} = \delta v_{\text{unif},i} / E_i$, calculated for the model nucleus of a uniformly charged sphere with radius $R = R_0 A^{1/3}$ and $R_0 = 1.2$ fm. Recently tabulated C_{unif} values²¹⁾ have removed some inadequate approximations of the earlier approaches. As pointed out by Zimmermann²²⁾, the improved expressions include the contributions of higher-order radial moments of the nuclear charge distribution at least within 1 % of C_{unif} . Therefore we have*)

$$f(Z) = C_{\text{unif}} / \lambda_{\text{unif}} \quad (6)$$

or, by insertion of (3) with $\langle r^{2n} \rangle_{\text{unif}} = \frac{3}{2n+3} R^{2n}$,

$$f(Z) = C_{\text{unif}} / \sum_n \frac{C_n}{C_1} \frac{2n}{2n+3} R_0^{2n} \bar{A}^{(2n/3)-1} (A'-A), \quad (7)$$

where $\bar{A} = (A+A')/2$. These relations form the basis of the semi-empirical evaluation of λ . We note that eqs. (6) and (7) differ from the conventional definition of $f(Z)$ which is made under the approximate assumption of $\lambda \approx \delta \langle r^2 \rangle$.

*) For the sake of convenience we have omitted in the following the superscripts A, A' on λ , $\delta \langle r^2 \rangle$, C_{unif} , etc., unless the context requires specification of the isotopes.

The normalization factor E_i is obtained most reliably for s-p transitions in the alkali-like spectra. Details of this evaluation for the ionic (Ra II) resonance line $7s\ ^2S_{1/2} - 7p\ ^2P_{1/2}$ at 4682 Å are discussed in the paper⁷⁾ dealing with the atomic physics aspects of the radium isotope shift and hyperfine structure. For the isotope pair 224,225 we obtain $F_{SE} = -49.6\text{ GHz/fm}^2$ by assuming the non-relativistic density $|\psi_{7s}(0)|^2 = 10.2\text{ a}_0^{-3}$ from the Goudsmit-Fermi-Segrè formula for the term energies. A similar value from the $7s\ ^2S_{1/2}$ hyperfine structure and the recently determined magnetic moments of $^{213,225}\text{Ra}$ [ref.⁸⁾] is subject to some uncertainty in the large relativistic corrections. With $\beta = 1.039$ [ref.²³⁾] this gives $E(7s-7p_{1/2}) = 0.378$. Insertion of $C_{unif}^{224,225}$ from ref.²¹⁾ into eq. (6) gives $f(88) = -131.1\text{ GHz/fm}^2$, and the weak A dependence of this function is taken care of by the expression given by Blundell et al.²¹⁾ together with (6). Ab-initio Dirac-Fock calculations by Torbohm et al.²³⁾ yield a considerable smaller value of $F_{DF} = -39.46\text{ GHz/fm}^2$. The 20 % difference between F_{SE} and F_{DF} follows the usual trend of semi-empirical and ab-initio values of the electronic factors^{14,23)}. It has not been removed by a careful reconsideration²⁴⁾ of possible insufficiencies of the semi-empirical procedure. As the electron densities derived by application of the Goudsmit-Fermi-Segrè formula generally agree within a few percent with those from the analyses of the contact hyperfine interaction, the reason for the discrepancy may be sought in an insufficient account for far configuration mixing effects by the Hartree (Dirac)-Fock procedure. On the other hand, the recently discussed correction²⁵⁻²⁷⁾ of about 10 % for spin-exchange core polarization indicates that $|\psi_{ns}(0)|^2$ may be overestimated from the hyperfine structure. Taking into account such a correction would require a more comprehensive knowledge of the situation in the heavier alkaline earth elements. It is also not obvious why the value based on term energies and the Goudsmit-Fermi-Segrè formula should require a similar correction. Therefore, in accordance with the arguments of King and Palmer²⁸⁾, we prefer to account for these uncertainties by assuming a 10 % error in the direction of a smaller value of F_{SE} .

The $\lambda^{A,A'}$ values of Table 1 have been obtained from the field shifts in the Ra I transition $7s^2 \ ^1S_0 - 7s7p \ ^1P_1$ at 4826 Å, assuming that specific mass shifts are negligible, i.e. (0 ± 0.5) NMS¹⁹). For this transition, the electronic factors have been related to those discussed above by a King plot⁷) which yields the ratio $F/F' = 1.4062(2)$ between the Ra II line (4682 Å) and the Ra I line (4826 Å). This is in excellent agreement with a multi-configuration Dirac-Fock calculation²³) yielding $F_{DF}/F'_{MCDF} = 1.403$, and confirms the general observation that field shift ratios are very well reproduced by theory.

From the discussion above, we adopt the λ_{SE} values from the semi-empirical evaluation with a 10 % calibration error towards λ_{DF} . A preference for these results will be suggested also by the nuclear model description of Section 6.2. Apart from this, we have to use them for consistency reasons in comparisons with $\delta\langle r^2 \rangle$ data on the neighbouring elements for which no ab-initio calculations have been performed so far.

4.4 Transformation from λ to $\delta\langle r^2 \rangle$ and Decomposition into Volume and Shape Contribution

As pointed out already, the radial charge moments higher than $\delta\langle r^2 \rangle$ contribute significantly to the nuclear parameter λ (see eq. (3)) and have to be considered for Ra ($Z = 88$). They can be expressed with sufficient accuracy in terms of standard models of the nuclear charge distribution. For the model of a uniformly charged, quadrupole-deformed nucleus we can calculate the quantities

$$\delta\langle r^{2n} \rangle = \int \delta\varrho \ r^{2n} \ dV / \int \varrho \ dV, \quad (8)$$

where the charge density $\varrho(r, \theta)$ is assumed to be constant within the sharp nuclear surface described by

$$r_1(\theta) = R_1 [1 + \beta_2 Y_{20}(\theta)]. \quad (9)$$

Then compared to a sphere with radius R the parameter

$$R_1 = R \left(1 + \frac{1}{4\pi} \beta_2^2 \right)^{-1} \quad (10)$$

is chosen to preserve the nuclear volume.

We then have

$$\delta \langle r^{2n} \rangle = \frac{3}{2n+3} \delta(R^{2n}) + \frac{3n}{4\pi} R^{2n} \delta(\beta_2^2) \quad (11)$$

which is written in the form of the usual separation

$$\delta \langle r^{2n} \rangle = \delta \langle r^{2n} \rangle_{\text{sph}} + \delta \langle r^{2n} \rangle_{\text{def}} \quad (12)$$

into volume (sph) and shape (def) dependent terms for all orders $2n$. To account for the contribution of higher ($n > 1$) radial moments, we introduce an expansion in terms of the second moments $\delta \langle r^2 \rangle_{\text{sph}}$ and $\delta \langle r^2 \rangle_{\text{def}}$ and obtain

$$\delta \langle r^{2n} \rangle = \frac{5n}{2n+3} R^{2n-2} \delta \langle r^2 \rangle_{\text{sph}} + \frac{3n}{4\pi} R^{2n} \delta \langle \beta^2 \rangle. \quad (13)$$

Thus the experimental observable λ can be expressed in terms of two variables representing the spherical equivalent radius and the deformation, corresponding to the two-parameter model of describing $\delta \langle r^2 \rangle$. Although dynamic deformations and higher-order multipoles have not been included explicitly, these and the leading contributions of $\beta_3, \beta_4, \dots$ can be subsumed under the deformation term in which $\delta(\beta_2^2)$ is replaced by $\sum_i \delta \langle \beta_i^2 \rangle$, written briefly $\delta \langle \beta^2 \rangle$.

Up to order $n = 3$, eq. (3) reads then explicitly:

$$\lambda = (1+x) \delta \langle r^2 \rangle_{\text{sph}} + (1+y) \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} \delta \langle \beta^2 \rangle, \quad (14)$$

$$\text{where} \quad x = \frac{C_2}{C_1} \frac{10}{7} R^2 + \frac{C_3}{C_1} \frac{5}{3} R^4$$

$$y = \frac{C_2}{C_1} 2 R^2 + \frac{C_3}{C_1} 3 R^4.$$

Using Seltzer's values of C_1, C_2, C_3 we obtain for Ra ($Z = 88$)
 $x = -0.077$; $y = -0.101$. In order to retain a maximum of model

independency for the two leading terms, R^2 has been replaced by $5/3 \langle r^2 \rangle_{\text{sph}}$ in the second expression of eq. (14). We can now retransform these two terms into $\delta \langle r^2 \rangle$, and obtain a relation between the experimental observable λ and $\delta \langle r^2 \rangle$

$$\delta \langle r^2 \rangle = \lambda - x \delta \langle r^2 \rangle_{\text{sph}} - y \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} \delta \langle \beta^2 \rangle. \quad (15)$$

Using eq. (14) we may further eliminate $\delta \langle \beta^2 \rangle$ from eq. (15) which then reads

$$\delta \langle r^2 \rangle = [\lambda - (x-y) \delta \langle r^2 \rangle_{\text{sph}}] / (1+y). \quad (16)$$

This latter relation facilitates the evaluation of $\delta \langle r^2 \rangle$, as it does not a priori require an approximate knowledge of deformation effects. On the other hand, droplet model²⁹⁾ values of $\delta \langle r^2 \rangle_{\text{sph}}$ are considered to be reliable enough for the described correction procedure. In calculating the final results for $\delta \langle r^2 \rangle$, given in the last two columns of Table 1, we have used the revised droplet model parameters³⁰⁾ which are shown below to permit a consistent interpretation of the data.

Of course, the interpretation would start most naturally from the experimental observable λ . However, it is convenient and has become common to use $\delta \langle r^2 \rangle$ for a comparison between the results of different experimental methods (e.g. electronic and muonic IS) and theory. Both procedures are equivalent as long as one stays within the same model as we shall do below. Fortunately, this is no stringent necessity: The factor $(x-y)/(1+y)$ limits the second, model-dependent term of eq. (16) to the insignificant level of about 2 %.

4.5 General Trends in $\delta\langle r^2 \rangle$

In the following we shall examine the consistency of our $\delta\langle r^2 \rangle$ data with the systematics of nuclear radii for the heavy elements. A satisfactory description of the overall trends is provided by the droplet model²⁹⁾, if deformation effects are included from empirical information, or from Strutinsky type calculations of deformed equilibrium shapes. Recently, there has been some criticism about the adequacy of conventional droplet model parameters³⁰⁾, and the finite range droplet model has been developed³¹⁾.

Here we take a pragmatical choice by comparing the predictions of refs.²⁹⁻³¹⁾ for spherical nuclei with the experimental $\delta\langle r^2 \rangle$ data for the Pb isotopes³²⁾. $\sum \langle \beta_i^2 \rangle = 0$ should be a good assumption for the doubly magic nucleus $^{1208}_{82}\text{Pb}$. The deviation of the experimental $\delta\langle r^2 \rangle$ curve from the straight lines produced by spherical gross models should reflect the increasing dynamic collective motion on either side of the magic neutron number. Considering the nearly symmetric lowering of the 2^+ excitation energies in the doubly-even Pb isotopes with respect to $N = 126$, this deviation should be symmetric as well. We thus conclude that the droplet parameters of Berdichevsky and Tondeur³⁰⁾ give the most realistic description of $\delta\langle r^2 \rangle_{\text{sph}}$ in this region. Therefore, we have chosen them for the discussion of the radium data in Fig. 4, where they are represented by the droplet line $\langle \beta^2 \rangle = 0$. The two different experimental curves correspond to the semi-empirical (SE) and ab-initio (DF) evaluations of the electronic factor. Deviations from the $\langle \beta^2 \rangle = 0$ line are attributed to deformation effects. To include these in the droplet model predictions we have used three different sets of deformation parameters: a) The β_2 and β_4 values from the Tables of Möller and Nix³³⁾ correspond to the calculated minima of potential energy surfaces and thus do not allow for dynamical contributions to $\langle \beta^2 \rangle$. b) The same applies to the β_2 , β_3 and β_4 values calculated by Nazarewicz et al.³⁾ with inclusion of the octupole degree of freedom. c) Empirical $\langle \beta_2^2 \rangle$ values are available for $^{222,224,226,228}\text{Ra}$, where $B(E2; 0^+ \rightarrow 2^+)$ values have been extracted from lifetime measurements⁴⁾. To these we have added the non-negligible corrections for β_4 from theory³³⁾. For the reference isotope ^{214}Ra , it seems justified to

use $\langle \beta_2^2 \rangle^{1/2} \approx 0.06$ from the systematics of $B(E2)$ values in singly magic nuclei and the empirical rule³⁴⁾ for the product $B(E2; 0^+ \rightarrow 2^+) \times E(2^+)$.

In all three cases the deformation terms in $\langle r^2 \rangle$ have been calculated according to the prescriptions of Myers and Schmidt²⁹⁾ which contain a consistent set of formulas for the radii and moments. This procedure is basically identical with the two-parameter description of eq. (13) which for $n = 1$ reads

$$\delta \langle r^2 \rangle = \delta \langle r^2 \rangle_{\text{sph}} + \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} \delta \langle \beta_2^2 \rangle, \quad (17)$$

except for the redistribution term for the density and the higher-order corrections in β_i . It has been shown to reproduce nearly quantitatively the mean square radii of strongly-deformed nuclei with respect to spherical ones, particularly in the rare-earth region³⁵⁻³⁷⁾.

From Fig. 4 it is obvious that a droplet model interpretation for radium can only be based on the $\delta \langle r^2 \rangle$ curve obtained from the semi-empirical evaluation of the IS data.* As expected, the experimental and theoretical curves, each related to the nearly spherical ^{214}Ra , converge towards strong deformation ($A \geq 228$), and the differences between theoretical static deformation values β_2 and experimental $\langle \beta_2^2 \rangle^{1/2}$ become less significant. It is mainly the transitional region ($220 \leq A \leq 226$) that has to be discussed later in the context of a possible manifestation of octupole-deformed shapes.

The droplet model analysis has been performed with the assumption of a constant intrinsic skin diffuseness²⁹⁾. This seems to be justified by the overall agreement and in particular the consistency with the development of the radii below $N = 126$. A decrease of the skin thickness has occasionally been postulated for other major shell closures⁴⁰⁾ and discussed also in connection with mercury²⁷⁾.

* In using these results we also remain consistent with $\delta \langle r^2 \rangle$ analyses for the neighbouring elements, e. g. francium and radon^{38,39)}, for which no alternative ab-initio values of the electronic factor are available.

However, this constancy is not in contradiction to a pronounced shell effect of the skin thickness parameter σ from monopole form factors in electron scattering⁴¹⁾, because the latter contain collective motion of all multipole orders as $\delta\langle r^2 \rangle$ does.

5. The Light Isotopes $^{208-214}\text{Ra}$

The radii of the light isotopes below the $N = 126$ neutron shell closure indicate near-spherical shapes for a rather extended range of nuclei. It is remarkable that for $Z = 88$ the isotope shifts still follow the features observed for the neighbourhood of the magic proton number $Z = 82$: There, the long sequences $^{196-208}\text{Pb}$ ³²⁾, $^{186-206}\text{Hg}$ ²⁷⁾ and $^{187-197}\text{Au}$ ⁴²⁾ are described essentially by the spherical droplet model, and only beyond ^{186}Hg and ^{187}Au there are sudden changes in $\delta\langle r^2 \rangle$ associated with the co-existence of near-spherical and strongly-deformed shapes.

The small influence of collective effects is consistent with the spins and moments (Table 2) of ^{213}Ra ($I = 1/2$) and $^{211,209}\text{Ra}$ ($I = 5/2$) which can be assigned to rather pure $p_{1/2}$ and $f_{5/2}$ shell-model states [see ref.⁶⁾]. A more extended discussion of the nuclei in this region will be presented³⁹⁾ in connection with the recent results on the radon isotopes below $N = 126$ which are preliminarily reported in ref.⁴³⁾.

6. Octupole Deformation of the Heavier Ra Isotopes

Sizeable effects of strong octupole correlation have been predicted theoretically and found experimentally in the spectra of nuclei in the Ra-Th region with neutron numbers around $N = 136$ [see references quoted by Leander and Sheline²⁾ and Nazarewicz et al.³⁾]. The tendency to develop reflection-asymmetric shapes is associated with the near degeneracy of positive- and negative-parity orbitals to the same spin for both protons and neutrons. Several authors^{1-3,44-49)} have investigated the nature of this octupole instability and the possible signatures for stable octupole deformation of the ground states.

6.1 Spins and Moments

Of the ground-state properties the spins and moments⁶⁾ from the hyperfine structure of the odd-A isotopes $^{221-229}\text{Ra}$ have provided an essential piece of information for this discussion. With increasing quadrupole deformation the spins follow exactly the sequential filling of neutron orbitals calculated on the basis of an axially symmetric but reflection-asymmetric folded Yukawa potential²⁾, or Woods-Saxon potential⁴⁶⁾ which tends to give more shallow minima of the potential energy surfaces at $\beta_3 \neq 0$. Also an octupole-deformed Nilsson potential with readjusted parameters⁵⁰⁾ has been successful in reproducing the whole spin sequence⁴⁷⁾. Recently, Sheline⁵¹⁾ has proposed to define the region of octupole deformation on the basis of a mapping of ground-state spins which are compared with the predictions of different models.

It has been pointed out by Ragnarsson⁹⁾ and by Leander and Sheline²⁾ that the magnetic moments provide a crucial test of the parity mixing of single-particle orbitals. They have explained the moments of $^{221,223,225}\text{Ra}$ with ϵ_3 (or $\beta_3 \approx 0.75 \epsilon_3$) close to the calculated equilibrium values of about 0.1. The octupole effects seem to diminish definitely in ^{229}Ra , and ^{227}Ra plays an intermediate role. The experimental results of Table 2 have now been based on the directly measured reference moments of ^{213}Ra and ^{225}Ra ⁸⁾. As they are consistent with the estimated 5 % calibration uncertainty on the earlier values of refs.^{6,7)}, it is evident that all conclusions for the nuclear structure remain valid. Details of the comparison with theory for the magnetic moments of $^{221-229}\text{Ra}$ may be found in Table 3 of ref.²⁾.

The quadrupole moments are only indirectly affected by the octupole degree of freedom, and they have been shown⁶⁾ to follow roughly the particle-rotor predictions assuming a reflection-symmetric Nilsson potential with neutron orbitals suggested by the measured spin. For increasing β_2 they approach the strong-coupling limit. This has been used to calibrate the Q_s values

from the hyperfine structures with the reference moment $Q_s = 2.9$ b, corresponding to $Q_0 = 8.0$ b, for ^{229}Ra which was obtained from the systematics of the intrinsic quadrupole moments⁴). These preliminary values agree very well with the results of a semi-empirical analysis of the electric field gradient at the nucleus performed in the $7s7p$ states of the neutral atom⁷). As mentioned in Section 4.1, this analysis is now being completed by an investigation of hyperfine structures in the $7p\ ^2P_{3/2}$ state of the alkali-like Ra^+ ion¹⁸).

6.2 Mean Square Charge Radii

In order to examine how octupole deformation effects may be reflected in the development of the radii, we include β_3 and describe the nuclear surface by

$$r_1(\theta) = R_1 (1 + \beta_2 Y_{20} + \beta_3 Y_{30} + \beta_4 Y_{40}). \quad (18)$$

Maintaining volume conservation yields the expression for the deformation part of $\langle r^2 \rangle$, with $\langle r^2 \rangle_{\text{sph}} = 3/5 R^2$ (see section 4.4)

$$\begin{aligned} \langle r^2 \rangle_{\text{def}} = \langle r^2 \rangle_{\text{sph}} \{ & \frac{5}{4\pi} (\beta_2^2 + \beta_3^2 + \beta_4^2) \\ & + \frac{5}{84} \left(\frac{5}{\pi}\right)^{3/2} \beta_2^3 + \frac{1}{6} \left(\frac{5}{\pi}\right)^{3/2} \beta_2 \beta_3^2 \\ & + \frac{75}{28} \left(\frac{1}{\pi}\right)^{3/2} \beta_2^2 \beta_4 \} \end{aligned} \quad (19)$$

For practical purposes only the first four terms of this expansion have to be taken into account, and they can be identified with the respective expectation values $\langle \beta_2^2 \rangle$, ... or their differences $\delta \langle \beta_2^2 \rangle$, The leading three are of second order in β_2 , β_3 , β_4 each contributing with the same fore factor $5/4\pi$.

Hints from theory and nuclear spectroscopy indicate that the octupole term of eq. (19) should be appreciable only within the range $220 \leq A \leq 226$. On the other hand, the convergence of all curves for $A > 228$ proves the consistency of the droplet model

parametrization with the difference in $\langle r^2 \rangle$ between spherical and well-deformed reflection-symmetric nuclei. This leads us to a qualitative discussion of the nuclei in the intermediate range.

In Fig. 4 there is no significant difference between the theoretical curves including and not including the octupole degree of freedom. Only some trends seem to be better reproduced by the curve based on a reflection-asymmetric Woods-Saxon potential³⁾ which is representative for a number of similar calculations allowing for octupole-deformed shapes^{45,52)}. Recently we have learned of a new approach⁵³⁾ indicating not only appreciable contributions of odd multipoles up to β_7 , but also a more pronounced reflection-asymmetric potential minimum. For the lighter transitional nuclei either description in terms of static deformation parameters is expected to fail: The calculated equilibrium deformations become rather sensitive to details of the assumed nuclear potential, and the expectation values $\langle \beta_i^2 \rangle$ are dominated by zero-point motion in the soft potential. Therefore, we resume the semi-empirical approach using experimental $B(E2)$ values, since they essentially contain $\langle \beta_2^2 \rangle$: In the relation

$$\sum_i B(E2; 0^+ \rightarrow 2_i^+) = \left(\frac{3}{4\pi} ZR^2 \right)^2 \langle \beta_2^2 \rangle, \quad (20)$$

usually the first term corresponding to excitation of the first 2^+ state gives the dominant contribution⁵⁴⁾.

To illustrate the situation more clearly we have plotted in Fig. 5 only the deformation part $\delta \langle r^2 \rangle_{\text{def}}^{228,A}$, representing the change in $\langle \beta^2 \rangle = \sum_i \langle \beta_i^2 \rangle$, for the heavier isotopes $A \geq 220$. ^{228}Ra is chosen as reference isotope in order to minimize experimental and systematic errors. It is the heaviest isotope for which a $B(E2)$ value is known, indicating a large and probably static quadrupole deformation, and it lies outside the octupole region. For the odd- A isotopes the β_2 contribution to $\delta \langle r^2 \rangle_{\text{def}}$ has been calculated from the spectroscopic quadrupole moments Q_s assuming strong coupling for the projection of Q_0 . According to the droplet model description^{29,30)} they are consistently converted into $\langle \beta_2 \rangle$ and $\delta \langle r^2 \rangle_{\text{def}}$

which both include the theoretical corrections for β_4 values. ^{229}Ra is chosen as reference for the odd-A isotopes, and the consistency with the data on even-A isotopes is achieved by a 3 % readjustment of the $\langle\beta_2\rangle$ values. Although the strong-coupling scheme is hardly justified for the lighter transitional nuclei, the calculated values of $\langle\beta_2\rangle^2$ fit well into the trend of $\langle\beta_2^2\rangle$ for the even isotopes, if ^{222}Ra is disregarded. Here, the exceptionally large $B(E2)$ value is doubtful, because it is not reflected in the trend of 2^+ excitation energies.

Apart from these insufficiencies, the plot of Fig. 5 suggests a gap between the development of the radii and the even multipole deformations which is increasing from ^{228}Ra towards the lighter isotopes. This gap can be closed by the inclusion of the octupole mode with β_3 values of the predicted order. For instance, this requires $\langle\beta_3^2\rangle^{1/2} = 0.07$ in the case of ^{224}Ra . The parameters underlying Fig. 5 are compiled in Table 3.

In general it seems that the inclusion of octupole deformation effects improves the consistency of $\delta\langle r^2\rangle$ at least with the semi-empirical description. However, conclusions can be drawn only with reservations as for the model dependence of the analysis.

6.3 Differential Isotope Shift and Odd-Even Staggering

The accuracy of the IS measurements can reveal details of the $\delta\langle r^2\rangle$ curve which are much smaller than the general calibration error. These details are accentuated in a plot of the derivative of $\langle r^2\rangle$ which for isotope pairs $A-2, A$ is known as the Brix-Kopfermann diagram. This is shown in Fig. 6 for the doubly-even isotopes (open squares). The spherical droplet line gives the reference for no change of deformation. It clearly divides the data into two classes: below $N = 126$ the deformation decreases as a function of A and above $N = 126$ it increases. The trend between $A = 220$ and 228 reflects a gradual drop in the increase of the overall $\langle\beta^2\rangle$, and this may be partly due to the dissipation of the β_3 mode. The distinct peak at $A = 230$ can be attributed to the closure of a purely β_2 -deformed subshell at $N = 142$ [ref.²] which

gives rise to an extra large step towards stronger deformation.

The lower curve of Fig. 6 corresponds to the changes in $\langle r^2 \rangle$ between neighbouring isotopes $A-1$ and A (open circles for odd- A and full circles for even- A). It exhibits a pronounced odd-even staggering (OES) which in the radium case shows a very unusual phenomenon: Between $A = 220$ and 226 the increase in $\langle r^2 \rangle$ is larger from even to odd than from odd to even nuclei, opposite to the "normal" behaviour which is found all over the chart of nuclides with very few exceptions. We have put forward the idea of connecting this inversion of the staggering with the octupole degree of freedom⁵⁵), and we shall now discuss the arguments for such an interpretation in more detail.

The OES is a general feature observed in nuclear radii, and it consists in a reduction of $\langle r^2 \rangle$ for the odd- N isotopes, compared to the average of their even- N neighbours. A measure for this effect is provided by the difference

$$D = \langle r^2 \rangle^A - \frac{1}{2}(\langle r^2 \rangle^{A-1} + \langle r^2 \rangle^{A+1}) = \delta \langle r^2 \rangle^{A-1,A} - \frac{1}{2} \delta \langle r^2 \rangle^{A-1,A+1}, \quad (21)$$

where A is the isotope with odd neutron number. This is essentially the same as the definition used in ref.³⁸), except for the sign which is chosen in analogy to the conventional staggering parameter

$$\gamma = 2 \delta \langle r^2 \rangle^{A-1,A} / \delta \langle r^2 \rangle^{A-1,A+1}. \quad (22)$$

This latter is less directly related to the physics of the effect, because the normalization factor $(\delta \langle r^2 \rangle^{A-1,A+1})^{-1}$ introduces artificial trends depending on the general slope of the $\delta \langle r^2 \rangle$ curve. In Table 4 we have listed both D and γ for the odd- A isotopes of radium. Normal staggering is represented by $D < 0$ or $\gamma < 1$, whereas inverted staggering is represented by $D > 0$ or $\gamma > 1$.

The OES is understood qualitatively as an effect of pairing on the collective potential, which gives a tendency towards larger $\langle \beta_2^2 \rangle$ for even-N systems of vibrational character. Recently, it has been shown by Zawischa⁵⁶⁾ that three- and four-body correlations may be essential in reproducing this staggering quantitatively. The influence of deformation will be considered later, but we can state here that the experimental phenomenon is similar for nearly all nuclei. Inverted OES occurs only in a few cases most of which are explained by exceptional situations of shape instability. Pertinent examples are the very neutron-deficient Hg isotopes²⁷⁾ or the N = 89 isotones in a few rare-earth elements around Z = 64. So far no explanation has been found for the continuation of an inverted staggering beyond N = 90 in $^{153-156}\text{Eu}$ ⁵⁷⁾ and for the neutron-deficient Sr isotopes^{58,59)}.

In the heavy Ra region, there is no indication of an instability in β_2 , and the observed inversion of the OES is likely to be connected with the octupole degree of freedom. There is a trivial argument for this: The inversion occurs exactly between A = 220 and 226 where reflection-asymmetric shapes are predicted and suggested also by experimental evidence. The normal staggering is restored towards the heavier isotopes which are supposed to be reflection-symmetric.

In trying to corroborate this conjecture, we consider the $\langle \beta_3^2 \rangle$ term of eq. (19) responsible for an over-compensation of the normal staggering in $\langle \beta_2^2 \rangle$ which should correspond to about 0.04 fm². Comparing this with the experimental data (Table 4) suggests a $\langle \beta_3^2 \rangle$ staggering of 0.07 fm² which seems reasonable compared to a total contribution of 0.13 fm² from the generally adopted value of $\beta_3 \approx 0.1$.

Most of the quantitative calculations of octupole deformation are based on the Strutinsky method⁶⁰⁾. The physical mechanism is briefly as follows^{61,2)}: First the spherical symmetry is broken by the

residual quadrupole interaction between the valence nucleons themselves and the core. At a certain deformation β_2 , Nilsson levels to the same Ω and different intrinsic parity are crossing and, at that point, trigger a further breakdown of symmetry to reflection-asymmetric shapes by the octupole interaction. Thereby they remove their degeneracy and lower the total energy of the system by a certain amount, called the shell correction energy. In the language of molecular spectroscopy this mechanism of symmetry breaking is known as Jahn-Teller effect^{62,63}). It has been calculated^{44,45,64}) that in odd nuclei the unpaired particle polarizes the soft quadrupole-octupole potential energy surface in such a way that the octupole is more stabilized than in the neighbouring even nuclei. This is reflected e.g. in the parity doublets (a necessary signature of reflection asymmetry) which are experimentally found to be more closely spaced in the odd nuclei than in their even neighbours^{64,65}).

We thus have a natural explanation for the observed odd-even effect, provided that the dominating quadrupole deformation develops more or less independently of the octupole mode and behaves as in other regions of nuclei. Leander et al.⁴⁵) have found from their calculated potential energy surfaces, that the β_2 and β_3 equilibrium parameters are only weakly coupled. Nevertheless, a staggering in β_3 may retroact slightly on β_2 in a competitive or promotive sense.

The octupole deformation should go along with a non-vanishing intrinsic dipole moment⁶⁶) of the order $p = 1 \text{ e} \cdot \text{fm}$. From this one can estimate an effect on the mean square radius

$$\delta \langle r^2 \rangle_{\text{dip}} = \frac{2}{5} \frac{p}{Ze} R \quad (23)$$

which amounts to 0.03 fm^2 and is negligible at the present accuracy level of the interpretation.

Irrespective of core polarization arguments for an enhancement of the octupole deformation by the unpaired neutron another qualitative explanation will be outlined in the

following. The essence is an interpretation of normal and inverted OES for deformed nuclei within the same theoretical picture that has been mentioned earlier for vibrational nuclei. The nuclear shape is governed by a collective potential whose depth is determined by shell effects and reduced by pairing. A typical potential is drawn in Fig. 7 as a function of some multipole deformation parameter β , and the effect of pairing is indicated by the difference between the full and the dashed curve. For odd nuclei compared to even ones the steeper outer and inner walls have counteracting effects on the wave function: The higher barrier at $\beta = 0$ pushes it towards larger deformation and thus $\langle \beta^2 \rangle$ becomes larger. On the other hand, the higher outer barrier pushes it inside and thus $\langle \beta^2 \rangle$ becomes smaller. It depends on the details of the collective dynamics which of the trends dominates. For the quadrupole mode with axial symmetry, one has a three-dimensional collective dynamics in β_2 and two of the Euler angles. Accordingly, the volume element $\beta_2^2 d\beta_2$ puts the weight β_2^2 on the probability density (see dashed line in the lower part of Fig. 7). Thus $\langle \beta_2^2 \rangle$ is more sensitive to the outer barrier, i.e. reduced for odd-N nuclei. This leads to the observed normal OES. In a few of the Ra isotopes we encounter a different situation, namely an octupole mode which develops on top of a well-established quadrupole deformation. The dynamics in β_3 with $\beta_2 \neq 0$ is only one-dimensional with a simple volume element $d\beta_3$, and thus $\langle \beta_3^2 \rangle$ is fully sensitive to the barrier at $\beta_3 = 0$. In addition, this barrier is much smaller than it is for the quadrupole mode [see e.g. the potential energy surfaces displayed in refs.^{3,45}]. Both effects together are likely to overcompensate the normal and lead to the observed inverted OES in the region of octupole instability. This has been figured out in quantitative comparisons of either collective dynamics which also hint the delicate balance of counteracting effects producing the staggering.

In addition to correlation effects⁵⁶), this balance may be an important reason why quantitative theoretical calculations have not yet been very successful in reproducing at least partly the rich experimental data on the odd-even effect of mean square charge radii. Here, the qualitative arguments are convincing enough for

the close connection between octupole deformation and the inverted OES of $^{220-226}\text{Ra}$. A similar effect has also been found in $^{220-225}\text{Fr}^{38)}$ and $^{218-222}\text{Rn}^{39,43)}$ by recent IS studies. In this context we note that also a core polarization model by Talmi⁶⁷⁾, which operates in a single j^n shell, infers normal OES for even and inverted OES for odd multipoles. Although such a description seems inadequate for deformed nuclei far from closed shells and the theories to be compared have not very much in common, their agreement in that point is not necessarily accidental.

7. Conclusion

The present study of isotope shifts in the long chain of Ra isotopes has provided the first evaluation of the variation of mean square nuclear charge radii between the neutron numbers $N = 120$ and 144 . A complementary study of the Fr isotopes between $N = 120$ and 141 has been performed by the Orsay group³⁸⁾. Owing to the vital interest in experimental clues to octupole-deformed nuclear shapes, the discussion has been concentrated on the heavy isotopes beyond $N = 132$, where such shapes are predicted by theory and supported by strong indications from nuclear spectroscopy. This subject was first raised by the apparent non-existence of two-phonon octupole vibrations⁶⁸⁾ and later established by the observation of (i) closely spaced parity doublets, (ii) unusual decoupling factors in odd-A rotational bands, (iii) large $B(E1)$ values, and (iv) unusually large α branches in the decay [see refs.^{1,2)} and references therein].

From the rather clear evidence of octupole-deformed excited states the question arises whether stable reflection asymmetry is a good description for the ground state. Here the experimental evidence can only be indirect, because reflection-asymmetric charge distributions are not reflected in an observable spectroscopic moment. Of the ground-state properties, mainly the sequence of spins and magnetic moments have provided evidence for reflection asymmetry in the odd-A isotopes $^{221,223,225,227}\text{Ra}$ [refs.^{2,9,46,47)}]. We have shown that for overall features of the radii neither model description is sensitive enough to dis-

tinguish between different shapes. On the other hand, some details are consistent with the assumption of β_3 deformation values of the predicted order of 0.1. In particular the inversion of the odd-even staggering effect between ^{220}Ra and ^{226}Ra indicates that octupole deformation in the ground state may be well developed for the odd-N isotopes.

Reflection-symmetric strong deformation is restored beyond $N = 138$, and describes well the radii and moments of $^{228-232}\text{Ra}$ of which the heaviest isotope, ^{232}Ra has been discovered in this experiment by laser spectroscopy.

Our results have also shown that the standard droplet-model parameters²⁹⁾ seem to underestimate the dependence of $\langle r^2 \rangle$ on the neutron number for elements around and above $Z = 82$. Modifications in the parameters³⁰⁾ have largely removed this problem. The radii below the $N = 126$ shell closure follow rather well the predictions for spherical nuclei and indicate little contribution from collective motion. This is in contrast with the pronounced effect of the $N = 50$ and $N = 82$ shell closures on the nuclear radii. On the other hand, also the moments of $^{209,211,213}\text{Ra}$ correspond to spherical configurations.

We acknowledge fruitful discussions and correspondence with T.P. Das and B. Fricke on questions of atomic theory, and with C. Ekström, G.A. Leander, I. Ragnarsson, and I. Talmi on nuclear theory. This work has been funded by the German Federal Minister for Research and Technology (BMFT) under the contract number 06 MZ 458 I.

References

- 1) R.R. Chasman, J. Phys. (Paris) 45 (1984) C6-167
- 2) G.A. Leander and R.K. Sheline, Nucl. Phys. A413 (1984) 375
- 3) W. Nazarewicz, P. Olanders, I. Ragnarsson, J. Dudek, G.A. Leander, P. Möller and E. Ruchowska, Nucl. Phys. A429 (1984) 269
- 4) S. Raman, C.H. Malarkey, W.T. Milner, C.W. Nestor, jr. and P.H. Stelson, At. Data Nucl. Data Tables 36 (1987) 1
- 5) S. Ullrich and E.W. Otten, Nucl. Phys. A248 (1975) 173
- 6) S.A. Ahmad, W. Klempt, R. Neugart, E.W. Otten, K. Wendt and C. Ekström, Phys. Lett. 133B (1983) 47
- 7) K. Wendt, S.A. Ahmad, W. Klempt, R. Neugart, E.W. Otten and H.H. Stroke, Z. Phys. D4 (1987) 227
- 8) E. Arnold, W. Borchers, M. Carré, H.T. Duong, P. Juncar, J. Lermé, S. Liberman, W. Neu, R. Neugart, E.W. Otten, M. Pellarin, J. Pinard, G. Ulm, J.L. Vialle and K. Wendt, Phys. Rev. Lett. 59 (1987) 771
- 9) I. Ragnarsson, Phys. Lett. 130B (1983) 353
- 10) H.L. Ravn, Phys. Rep. 54 (1979) 201
- 11) K.-L. Gippert, E. Runte, W.-D. Schmidt-Ott, P. Tidemand-Petersson, N. Kaffrell, P. Peuser, R. Kirchner, O. Klepper, W. Kurcewicz, P.O. Larsson, E. Roeckl, D. Schardt and K. Rykaczewski, Nucl. Phys. A453 (1986) 1
- 12) E. Rasmussen, Z. Phys. 87 (1934) 607
- 13) E. Rasmussen, Z. Phys. 86 (1933) 24
- 14) A.C. Mueller, F. Buchinger, W. Klempt, E.W. Otten, R. Neugart, C. Ekström and J. Heinemeier, Nucl. Phys. A403 (1983) 234
- 15) T.P. Das, New York, private communication
- 16) J.L. Heully and A.M. Martensson-Pendrill, Phys. Scr. 31 (1985) 169
- 17) V.A. Dzuba, V.V. Flambaum and O.P. Sushkov, Phys. Scr. 32 (1985) 507
- 18) W. Neu, E. Arnold, R. Neugart, E.W. Otten, G. Passler, K. Wendt, B. Fricke and H.-J. Kluge, to be published
- 19) K. Heilig and A. Steudel, At. Data Nucl. Data Tables 14 (1974) 613
- 20) E.C. Seltzer, Phys. Rev. 188 (1969) 1916
- 21) S.A. Blundell, P.E.G. Baird, C.W.P. Palmer, D.N. Stacey, G.K. Woodgate and D. Zimmermann, Z. Phys. A321 (1985) 31

- 22) D. Zimmermann, Z. Phys. A321 (1985) 23
- 23) G. Torbohm, B. Fricke and A. Rosén, Phys. Rev. A31 (1985) 2038
- 24) S.A. Blundell, P.E.G. Baird, C.P. Botham, C.W.P. Palmer,
D.N. Stacey and G.K. Woodgate, J. Phys. B17 (1984) 53
- 25) J. Bauche, H. Hühnermann, D.N. Stacey, V. Stacey and M. Wilson,
Z. Phys. A320 (1985) 157
- 26) F. Buchinger, P. Dabkiewicz, H.-J. Kluge, A.C. Mueller and
E.W. Otten, Nucl. Phys. A462 (1987) 305
- 27) G. Ulm, S.K. Bhattacharjee, P. Dabkiewicz, G. Huber, H.-J.
Kluge, T. Kühl, H. Lochmann, E.W. Otten, K. Wendt, S.A. Ahmad,
W. Klempt and R. Neugart, Z. Phys. A325 (1986) 247
- 28) W.H. King and C.W.P. Palmer, Comments At. Mol. Phys 18 (1986) 11
- 29) W.D. Myers and K.-H. Schmidt, Nucl. Phys. A410 (1983) 61
- 30) D. Berdichevsky and F. Tondeur, Z. Phys. A322 (1985) 141
- 31) P. Möller, W.D. Myers, W.J. Swiatecki and J. Treiner, Proc.
7th Int. Conf. on At. Masses and Fund. Const., AMCO-7 (O. Klep-
per, ed.), Darmstadt-Seeheim (1984), p. 457
- 32) M. Anselment, W. Faubel, S. Göring, A. Hanser, G. Meisel,
H. Rebel and G. Schatz, Nucl. Phys. A451 (1986) 471
- 33) P. Möller and J.R. Nix, At. Data Nucl. Data Tables 26 (1981) 165
- 34) L. Grodzins, Phys. Lett. 2 (1962) 88
- 35) R. Neugart, in Lasers in Nucl. Physics (C.E. Bemis, jr. and
H.K. Carter, ed.) Harwood Academic Publishers, New York (1982),
p. 231
- 36) S.A. Ahmad, W. Klempt, C. Ekström, R. Neugart and K. Wendt,
Z. Phys. A321 (1985) 35
- 37) S.A. Ahmad, W. Klempt, R. Neugart, K. Wendt and C. Ekström,
to be published
- 38) A. Coc, C. Thibault, F. Touchard, H.T. Duong, P. Juncar,
S. Liberman, J. Pinard, M. Carré, J. Lermé, J.L. Vialle,
S. Büttgenbach, A.C. Mueller and A. Pesnelle, Nucl. Phys.
A468 (1987) 1
- 39) G. Ulm, H.T. Duong, K. Wendt, W. Borchers, R. Neugart and
E.W. Otten, to be published
- 40) H. Gerhardt, E. Matthias, H. Rinneberg, F. Schneider, A. Tim-
mermann, R. Wenz and P.J. West, Z. Phys. A292 (1979) 7
- 41) J. Friedrich and N. Voegler, Nucl. Phys. A373 (1982) 192

- 42) K. Wallmeroth, G. Bollen, A. Dohn, P. Egelhof, J. Grüner, J. Campos, A. Rodriguez, M.J.G. Borge, A. Venugopalan, J.L. Wood, R.B. Moore and H.-J. Kluge, Phys. Rev. Lett. 58 (1987) 1516
- 43) W. Borchers, R. Neugart, E.W. Otten, H.T. Duong, G. Ulm and K. Wendt, Hyp. Int. 34 (1987) 25
- 44) R.R. Chasman, Phys. Lett. 96B (1980) 7
- 45) G.A. Leander, R.K. Sheline, P. Möller, P. Olanders, I. Ragnarsson and A.J. Sierk, Nucl. Phys. A388 (1982) 452
- 46) W. Nazarewicz and P. Olanders, Nucl. Phys. A441 (1985) 420
- 47) P. Rozmej, K. Boening and A. Sobiczewski, Proc. XXIV Int. Winter Meeting on Nuclear Physics, Bormio, Italy (1986) - GSI-86-8 preprint
- 48) R. Piepenbring, J. Physique (Paris) Lett. 45 (1984) L-1023
- 49) R. Piepenbring, Z. Phys. A323 (1986) 341
- 50) P. Rozmej, GSI-85-41 preprint
- 51) R.K. Sheline, Phys. Lett. B197 (1987) 500
- 52) R.R. Chasman, Phys. Lett. B175 (1986) 254
- 53) P. Rozmej, Ch. Briancon, S. Cwiok and A. Sobiczewski, 5th Int. Conf. on Nuclei far from Stability, Rosseau Lake, Canada (1987) and to be published
- 54) K. Kumar, Phys. Rev. Lett. 28 (1972) 249
- 55) S.A. Ahmad, C. Ekström, W. Klempt, R. Neugart, E.W. Otten and K. Wendt, Proc. 7th Int. Conf. on At. Masses and Fund. Const., AMCO-7 (O. Klepper, ed.), Darmstadt-Seeheim (1984), p. 361
- 56) D. Zawischa, Phys. Lett. 155B (1985) 309
- 57) K. Dörschel, W. Heddrich, H. Hühnermann, E.W. Peau, H. Wagner, G.D. Alkhazov, E.Ye. Berlovich, V.P. Denisov, V.N. Panteleev and A.G. Polyakov, Z. Phys. A317 (1984) 233
- 58) M. Anselment, K. Bekk, S. Chongkum, S. Göring, A. Hanser, H. Hoeffgen, W. Kälber, G. Meisel and H. Rebel, Z. Phys. A326 (1987) 493
- 59) D.A. Eastham, P.M. Walker, J.R.H. Smith, D.D. Warner, J.A.R. Griffith, D.E. Evans, S.A. Wells, M.J. Fawcett and I.S. Grant, Phys. Rev. C36 (1987) 1583
- 60) V.M. Strutinsky, Nucl. Phys. A95 (1967) 420
- 61) I. Ragnarsson, S.G. Nilsson and R.K. Sheline, Phys. Rep. 45 (1978) 1

- 62) H.A. Jahn and E. Teller, Proc. Roy. Soc. A161 (1937) 220
- 63) P.-G. Reinhard and E.W. Otten, Nucl. Phys. A420 (1984) 173
- 64) R.K. Sheline, D. Decman, K. Nybø, T.F. Thorsteinsen, G. Løv-
høiden, E.R. Flynn, J.A. Cizewski, D.K. Burke, G. Sletten,
P. Hill, N. Kaffrell, W. Kurcewicz, G. Nyman and G. Leander,
Phys. Lett. 133B (1983) 13
- 65) I. Ahmad, J.E. Gindler, R.R. Betts, R.R. Chasman and A.M.
Friedman, Phys. Rev. Lett. 49 (1982) 1758
- 66) G.A. Leander, W. Nazarewicz, G.F. Bertsch and J. Dudek, Nucl.
Phys. A453 (1986) 58
- 67) I. Talmi, Nucl. Phys. A423 (1984) 189
- 68) W. Kurcewicz, E. Ruchowska, J. Zylicz, N. Kaffrell and N. Traut-
mann, Nucl. Phys. A304 (1978) 77

Table 1. Isotope shifts in the transition $7s^2 \ ^1S_0 - 7s7p \ ^1P_1$ and deduced nuclear parameters $\lambda^{214,A}$ and $\delta\langle r^2 \rangle^{214,A}$.

A	$\delta\nu$ (MHz)	$\delta\nu_{\pi}$ (MHz)	λ (fm ²)	$\delta\langle r^2 \rangle$ (fm ²)
208	8485(8)	8530(31)	-0.240[25]	-0.256[27]
209	8313(8)	8351(27)	-0.235[25]	-0.253[25]
210	6003(6)	6033(21)	-0.170[18]	-0.182[19]
211	5522(4)	5544(15)	-0.156[16]	-0.168[17]
212	3256(3)	3271(11)	-0.092[10]	-0.099[10]
213	2167(3)	2175(7)	-0.061[7]	-0.066[7]
214	0	0	0	0
220	-21904(8)	-21947(30)	0.620[63]	0.679[69]
221	-25945(8)	-25995(33)	0.734[75]	0.805[82]
222	-28827(8)	-28884(37)	0.816[83]	0.894[91]
223	-32453(9)	-32517(41)	0.919[93]	1.007[102]
224	-35123(9)	-35194(45)	0.995[101]	1.090[110]
225	-38888(10)	-38965(49)	1.102[112]	1.208[122]
226	-41124(11)	-41208(53)	1.166[119]	1.277[129]
227	-43937(12)	-44028(58)	1.246[127]	1.356[138]
228	-46950(12)	-47047(61)	1.332[135]	1.459[148]
229	-50066(15)	-50170(67)	1.421[144]	1.556[158]
230	-53634(15)	-53744(70)	1.522[154]	1.667[169]
232	-59585(17)	-59708(79)	1.692[172]	1.854[188]

For λ and $\delta\langle r^2 \rangle$ we have assumed a calibration error of 10 % towards larger absolute values. The experimental errors, to be used in relative comparisons, are less than one unit in the last digit. In addition to the experimental errors of $\delta\nu$ [see ref.¹⁴], the errors of $\delta\nu_{\pi}$ account for the specific mass shift (see 4.3).

Table 2. Nuclear spins and moments of odd-A Ra isotopes from the hyperfine structure of $7s\ ^2S_{1/2}$, $7p\ ^2P_{1/2}$ in RaII and $7s7p\ ^1P_1$ in RaI.

A	I	μ_I/μ_N	Q_e (barn)
209	5/2	0.865(13)	0.38(4)
211	5/2	0.8780(38)	0.46(5)
213	1/2	0.6133(18)	-
221	5/2	-0.1799(17)	1.90(20)
223	223	0.2705(19)	1.19(12)
225	1/2	-0.7338(15)	-
227	3/2	-0.4038(24)	1.50(15)
229	5/2	0.5025(27)	2.96(30)

(a) Calibrated with the directly measured magnetic moments of ^{213}Ra and ^{225}Ra [ref.⁸].

(b) Calibrated with electric field gradients from a semi-empirical analysis for $7s7p\ ^3P_2$ [ref.⁷] and $7p\ ^2P_{3/2}$ [ref.¹⁸].

Table 3. Deformation parameters with respect to ^{228}Ra .

A	Isotope Shift		B(E2)		Q_s	
	$\delta\langle r^2 \rangle_{\text{def}}$	$\langle \beta^2 \rangle^{1/2}$	$\langle \beta_2^2 \rangle^{1/2}$	$(\langle \beta_2^2 \rangle + \langle \beta_4^2 \rangle)^{1/2}$	$\langle \beta_2 \rangle$	$(\langle \beta_2 \rangle^2 + \langle \beta_4 \rangle^2)^{1/2}$
	(fm ²)		(a)		(b)	
220	-0.290	0.173				
221	-0.226	0.186			0.142	0.158
222	-0.198	0.191	0.183(8)	0.195		
223	-0.146	0.200			0.155	0.179
224	-0.124	0.204	0.169(4)	0.191		
225	-0.068	0.213				
226	-0.060	0.214	0.188(5)	0.212		
227	-0.033	0.223			0.190	0.214
228	0	0.223	0.203(8)	0.223		
229	0.037	0.229			0.209	0.229
230	0.087	0.237				
232	0.153	0.246				

(a) Errors are given only for the parameters which are directly related to experimental quantities.

(b) The β values from Q_s are adjusted to the isotope shift data of ^{229}Ra .

Table 4. Odd-even staggering parameters for the odd-A with respect to their even-A neighbours.

A	$D(10^{-2}\text{fm}^2)$	γ
209	-3.35[34]	0.10(3)
211	-2.79[28]	0.33(3)
213	-1.69[17]	0.66(1)
221	1.82[18]	1.169(2)
223	1.50[15]	1.153(3)
225	2.40[24]	1.257(3)
227	-0.31[3]	0.966(1)
229	-0.72[7]	0.931(1)

For the errors of absolute D values see Table 2. The errors of γ are determined by our Seltzer correction procedure.

Figure Captions

Fig. 1: Production yield of even Ra isotopes from ISOLDE, normalized to a proton current of 1 μA (target material UC_2 , thickness 13 g/cm^2). Examples of the resonance $7s^2\ ^1S_0 - 7s7p\ ^1P_1$ (4826 $\text{\AA}$) are shown at the top for the most and least abundant isotopes.

Fig. 2: Experimental setup used for spectroscopy on ions and on neutral atoms: The Doppler-tuning potential is alternatively applied to the observation chamber (full line) or the charge-exchange cell (dashed line).

Fig. 3: Position of the observed resonances (IS and hfs) in the 4826 $\text{\AA}$ line, relative to ^{214}Ra . The horizontal bars indicate the positions and intensities of individual hfs components, and the dots represent the centres of gravity.

Fig. 4: Experimental $\delta\langle r^2 \rangle^{214,A}$ for the Ra isotopes ($208 \leq A \leq 232$) compared to droplet model³⁰⁾ calculations (thin lines) including deformation: a) spherical reference line; b) theoretical deformation values β_2, β_4 taken from ref.³³⁾ - dotted; c) theoretical deformation values $\beta_2, \beta_3, \beta_4$ from ref.³⁾ - dashed; d) experimental $\langle \beta_2^2 \rangle$ from B(E2) values with correction for theoretical β_4 - open circles with error bars. SE and DF refer to results of using semi-empirical and Dirac-Fock calibration factors, respectively.

Fig. 5: Comparison of $\delta\langle r^2 \rangle_{\text{def}}^{228,A}$ from the isotope shifts with droplet calculations from a) B(E2) values - open circles with error bars; b) spectroscopic quadrupole moments - open squares.

Fig. 6: Brix-Kopfermann plot of $\delta\langle r^2 \rangle^{A-2,A}$ for Ra (upper curve) and odd-even staggering of $\delta\langle r^2 \rangle^{A-1,A}$ (lower curve). Full dots represent steps from odd to even and open circles represent steps from even to odd isotopes.

Fig. 7: Schematic view of the odd-even effect on $\langle \beta^2 \rangle$: a) Collective potential $V(\beta)$ as a function of the deformation β . The curve is combined from a smooth liquid-drop background and oscillating shell corrections which have a larger amplitude for odd nuclei (dashed line). b) Typical probability distribution $|\psi(\beta)|^2$ which is relevant for one-dimensional collective dynamics. For three-dimensional dynamics $\beta^2 |\psi(\beta)|^2$ is the appropriate density.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7