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HYPERFINE STRUCTURE AND ISOTOPE SHIFT OF THE NEUTRON-RICH BARIUM ISOTOPES $^{139-146}$Ba AND ^{148}Ba

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Abstract: The hyperfine structure and isotope shift in the 6s $^2S_{1/2}$ - 6p $^2P_{3/2}$ line of Ba II (455.4 nm) have been measured by collinear fast-beam laser spectroscopy for the neutron-rich isotopes $^{139-146}$Ba and ^{148}Ba. Nuclear moments and mean square charge radii of these isotopes have been recalculated. The isotope shift of the isotope ^{148}Ba ($T_{1/2}$ = 0.64 s) could be studied for the first time, yielding $\delta\langle r^2\rangle^{138,148} = 1.245(3)$ fm^2.

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1. Introduction

The high sensitivity and resolution of present laser spectroscopic techniques has enabled the study of isotope shifts (IS) and hyperfine structures (hfs) in the optical spectra of many elements for long isotopic sequences. Thus the knowledge of nuclear ground state properties could be extended far into the region of β- and α-instability.

One of the first examples is the chain of barium isotopes (Z=56), where IS and hfs of 30 isotopes and isomers in the range $122 \leq A \leq 146$ are known from two different experiments /1,2/. Both of them were carried out in the atomic transition $6s^2\ ^1S_0$ - $6s6p\ ^1P_1$ at 553.6 nm. The data yielded the changes in the mean square nuclear charge radii $\delta\langle r^2\rangle$, as well as the nuclear spins and moments on either side of the N=82 neutron shell closure. Extensive discussions of the nuclear structure aspects of these results are found in /1,2,3/.

Here we report on the extension of the IS measurements /2/ to ^{148}Ba, which is the heaviest known isotope in the barium chain. We have also performed complementary investigations of the hfs and IS in the other neutron-rich isotopes. For these we have chosen the ionic (Ba II) resonance line $6s\ ^2S_{1/2}$ - $6p\ ^2P_{3/2}$ at 455.4 nm, in order to avoid beam losses and line broadening due to neutralization of the beam in the charge-exchange cell. Thus we gained a slight increase in sensitivity as compared to the earlier experiment on Ba I. This gain enabled the study of ^{148}Ba, which is produced at ISOLDE with yields of about 10^4 ions/s. Moreover, the independent measurement of the hfs and IS in the Ba II transition provides an unambigious redetermination of nuclear spins and moments, which in the earlier work on Ba I had to be based partly on relative line intensities rather than on line splittings. The IS in both lines can be compared to each other by a King-plot, which also allows a calibration of $\delta<r^2>$ based on the IS in the simple alkali-like spectrum of Ba II.

The data are analyzed with special regard to recent ab-initio calculations for the atomic and ionic isotope shift parameters and hyperfine fields. Concerning the nuclear physics interpretation the discussion focusses on possible evidences supporting the prediction of an intrinsic reflection asymmetry for the nuclei around ^{145}Ba.

2. Experimental Setup and Results

We have used the setup for collinear fast beam laser spectroscopy /2/ at ISOLDE (CERN), in its simplified version for ion-spectroscopy /4/. The barium isotopes are produced by proton-induced fission in a UC_2 target and subsequent surface ionisation /5/. After mass separation the 60 keV ion beam is post-accelerated or -decelerated for Doppler-tuning and excited by a collinear laser beam. The optical resonances are detected by counting of fluorescence photons.

As an example the hfs of ^{145}Ba is shown in Fig. 1. The line widths are about 50 MHz. The determination of peak positions as a function of the acceleration voltage is performed by a computer fit, yielding the IS, the nuclear spin and the hfs parameters A and B. The results for the isotopes under study are given in Table 1. The quoted errors are the sum of systematic errors, stemming mainly from uncertainties of the acceleration voltage of the order 5 Volt, and of statistical errors as derived from the χ^2 of the fitting procedure. For the stable isotopes $^{134-138}$Ba the data from several previous measurements are included in the table for comparison.

3. Evaluation of Nuclear Moments and $\delta\langle r^2\rangle$ Values

Table 2 contains the spins, the nuclear moments μ_I and Q_s and the changes in mean square charge radii $\delta\langle r^2\rangle$ for the isotopes under study. The μ_I values have been determined from the experimental $A(^2S_{1/2})$ factors via the known reference values for $A(^2S_{1/2})$ and μ_I of ^{135}Ba and ^{137}Ba /6,10/. The influence of possible hfs anomalies can be estimated by comparing the ratios $A(^2P_{3/2})/A(^2S_{1/2})$ for the different isotopes, since only the $^2S_{1/2}$ state is subject to noticeable hfs anomaly. The isotopic variations of this ratio never exceed the experimental error which is dominated by the error of $A(^2P_{3/2})$. The latter is thus taken into account in the final error of μ_I.

The electric quadrupole moments Q_s have been determined from the experimental B factors via the relation $B = e \langle V_{zz} \rangle Q_s$. The electric field gradient has been taken from ab-initio calculations of Martensson-Pendrill and Heully to be $\langle V_{zz} \rangle = 378(4)$ MHz/ebarn /13/. The corresponding value of $Q_s^{137} = 0.245(4)$ barn is in very good agreement with the earlier semi-empirical results which include a Sternheimer anti-shielding correction of $R = -0.20(2)$ for Ba II. These values are also given in Table 2.

The $\delta\langle r^2 \rangle$ values are calculated from the field shifts in the one-electron s-p transition after subtraction of the mass shift (see below) using the electronic factor $F_{455} = -5.12$ GHz/fm² for the isotope pair 134,135. This factor is calculated semi-empirically according to /14/

$$F_i = (\pi a_o^3/Z) \beta |\Psi_{6s}(0)|^2 f(Z) K$$

using the electronic shielding constant $\beta = 1.087$ from /15/, and the electron density $|\Psi_{6s}(0)|^2 = 6.04\ a_o^{-3}$ which is the mean value evaluated from the Goudsmit-Fermi-Segre and the hfs formula. For the function f(Z) the most recent calculations from /16/ and the higher-order corrections K from /17/ have been used. The difference to the earlier semi-empirical values /2/ of 2% is well within the estimated accuracy for this evaluation of about 5%. The existing ab-initio value for the field shift factor $F_{455} = -3.827$ GHz/fm² /15/ deviates by more than 30% and is still believed to be ruled out by nuclear physics arguments as discussed already in /2/. A King plot of the data from the atomic resonance line /2/ versus those from the ionic one gives the ratio of electronic field shift factors as

$$\rho = F_{553} / F_{455} = 0.8079(16)$$

and the relation between specific mass shift parameters S

$$D = S_{553} - \rho S_{455} = -205(12) \text{ GHz amu}.$$

The former agrees perfectly with the ratio from a MCDF calculation, yielding $\rho_{MCDF} = 0.806$ /15/. The experimental value of D is much smaller than the normal mass shift constants and thus agrees with the standard assumption for the specific mass shift parameters for s-p and s^2-sp transitions /14/, which corresponds to D = -108(470) GHz amu. From an ab-initio calculation Pendrill estimated the specific mass shift of the atomic line to be $|S_{553}| < 100$ GHz amu /18/ in agreement with the results from King plots with muonic x-ray data /19,20/ which give $S_{553} \approx -240(140)$ GHz amu.

For the evaluation of $\delta\langle r^2\rangle$ values we have therefore assumed a vanishing specific mass effect $S_{553} = 0(160)$ GHz amu with an uncertainty of 50% of the normal mass shift, resulting in $S_{455} = 254(160)$ GHz amu. The complete set of $\delta\langle r^2\rangle$ values for heavy barium isotopes are included in Table 2. The errors in the circular brackets are the experimental uncertainties, while the additional error given in square brackets is ascribed to theory.

4. Discussion

The present measurements on the isotopes $^{139-146}$Ba have improved the accuracy of the nuclear moments by a factor of two to five. However, in view of the limited precision of present nuclear models this does not change the interpretations which were given in the earlier papers /1,2,3/. Concerning the nuclear spins our measurements have removed some remaining ambiguities of the earlier determination from relative line intensities /2/. As already noted in our "note added in proof" to ref. /2/, the independent measurement of the hfs for two states with different total angular momentum J unambigously determines the spin of ^{145}Ba to be I = 5/2. This spin value together with the corresponding magnetic dipole and electric quadrupole moments is in contradiction to any prediction from standard particle-rotor calculations /2/. While I = 5/2 and the moments of ^{143}Ba are well explained within that framework /2/, it seems that only an intrinsic field of reflection-asymmetric deformation can account for spin and moments of ^{145}Ba /21/. This is taken as an experimental hint of strong octupole correlation effects which are also suggested by Strutinski type calculations of equilibrium shapes /21/.

For the well investigated octupole-deformed region Ra-Th we have pointed out earlier /22/, that the observed inversion of the odd-even staggering of nuclear charge radii reflected in a staggering parameter $\gamma > 1$, may be understood as an effect arising from a stabilization of a reflection-asymmetric shape by the unpaired neutron. This polarisation effect has been discussed in several papers, e.g. /23,24/. In the heavy barium isotopes the staggering parameter comes remarkably close to $\gamma = 1$, reaching a maximum of $\gamma = 0.945$ for ^{143}Ba (N=87), but does not surpass $\gamma = 1$.

The complete series of mean square charge radii of barium isotopes, including the neutron-deficient ones, is shown in Fig. 2. The predictions of the spherical droplet model for two different sets of parameters as given by Myers and Schmidt /25/ and Berdichevsky and Tondeur /26/ are indicated by the solid and the dashed line, respectively. In addition droplet predictions including deformation from reduced B(E2) transition probabilities /27/ are shown for both models.

The new result for the heaviest isotope ^{148}Ba exactly follows the trend of the sequence of neutron-rich isotopes reaching a deformation value of $\beta_2(^{148}\mathrm{Ba}) \approx 0.26$. This deformation is significantly smaller than the one measured for the N = 92 isotones in the region of light rare-earth elements around Z = 64. In contrast to the sudden onset of deformation found there (comp. /28/ and references therein), the barium isotopes run smoothly into deformation.

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Table 1: Spin, A and B factors and IS values for all isotopes under study. The IS is given relative to ^{138}Ba.

A	I	$A(6s_{1/2})$ [MHz]	$A(6p_{3/2})$ [MHz]	$B(6p_{3/2})$ [MHz]	$\delta\nu^{138,A}$ [MHz]
134					234.6(4.1) 249(12)[d]
135	3/2	3593.3(2.2)[c] 3591.67011718(24)[a]	113.2(8)[c] 112.72(32)[b] 113.1(5)[e]	59.4(2.3)[c] 60.45(35)[b] 60.7(1.2)[e]	360.7(2.2)[c] 381(30)[d]
136					186.9(2.1)[c] 177(15)[d]
137	3/2	4020.3(3.2)[c] 4018.87083385(24)[a]	126.7(1.1)[c] 125.95(35)[b] 125.8(5)[d]	95.0(3.7)[c] 92.53(40)[b] 91.6(1.2)[e]	279.0(2.6)[c] 285(30)[d]
138					0
139	7/2	-1783.7(9)	-56.2(3)	-216.6(2.8)	-598.0(4.5)
140					-1355.2(6.4)
141	3/2	-1445.6(1.4)	-45.9(7)	171.7(1.9)	-1901.4(5.5)
142					-2554.6(7.1)
143	5/2	1139.4(1.8)	35.8(9)	-332.5(5.1)	-3153.2(7.3)
144					-3823.8(9.0)
145	5/2	-733.9(1.0)	-23.2(5)	462.4(2.6)	-4309.9(7.0)
146					-4915.8(9.8)
148					-6050.3(15.0)

a) ref. /6/, b) ref. /7/, c) ref. /4/, d) ref. /8/, e) ref. /9/

Table 2: Spins, nuclear moments μ_I and Q_s and changes of mean square charge radii $\delta<r^2>$. The Q_s moments include Sternheimer correction. For the $\delta<r^2>$ results the circular brackets give pure experimental errors, while square brackets include uncertainties in the evaluation.

A	I	μ_I [n.m.]	Q_s [barn]	$\delta<r^2>^{138,A}$ [fm^2]
134				-0.049(1)[12]
135	3/2	0.837943(17)[a]	0.160(3) 0.146(16)[b]	-0.074(1)[10]
136				-0.039(1)[7]
137	3/2	0.937365(20)[a]	0.245(4) 0.228(24)[b] 0.25(1)[d]	-0.057(1)[5]
138				0
139	7/2	-0.973(5) -0.975(17)[c]	-0.573(13) -0.50(4)[c]	0.123(1)[8]
140				0.278(1)[18]
141	3/2	-0.337(5) -0.346(16)[c]	0.454(10) 0.43(4)[c]	0.391(1)[25]
142				0.525(2)[36]
143	5/2	0.443(11) 0.454(20)[c]	-0.879(22) -0.81(7)[c]	0.648(2)[44]
144				0.786(2)[51]
145	5/2	-0.285(7) -0.272(36)[c]	1.224(21) 1.15(16)[c]	0.886(2)[58]
146				1.011(2)[66]
148				1.245(3)[82]

a) ref. /10/, b) ref. /11/ after application of Sternheimer correction, c) ref. /2/, d) ref. /12/

Figure Captions

Fig. 1.: Experimental resonance pattern of ^{145}Ba

Fig. 2.: Development of $\delta\langle r^2\rangle$ for $^{122-148}$Ba in comparison with the prediction from the spherical droplet model using the parameters of Myers and Schmidt (solid line) and Berdichevsky and Tondeur (dashed line). The full dots and open triangles include the deformation according to experimental B(E2) values.

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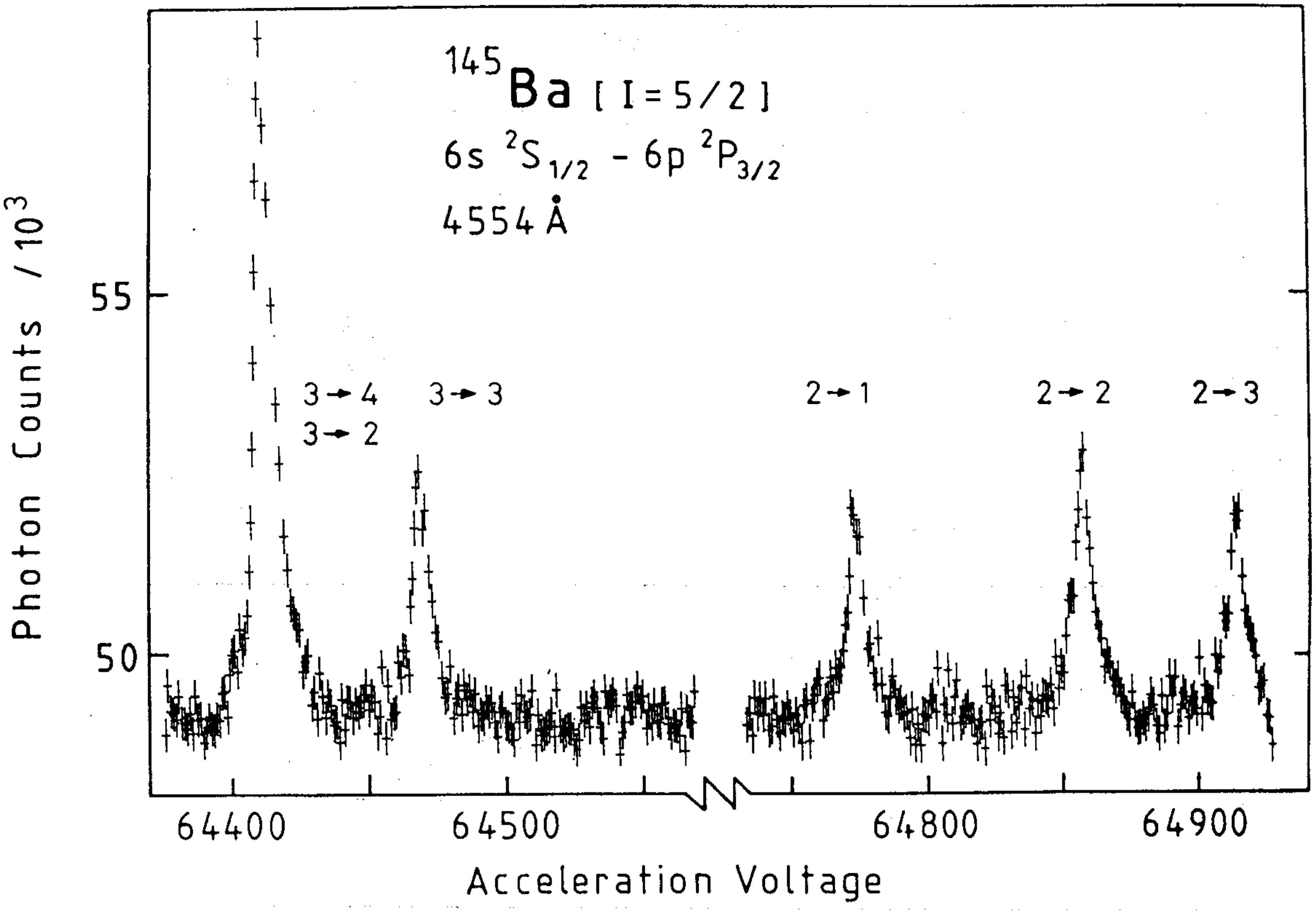
^{145}Ba [I = 5/2]
6s $^2S_{1/2}$ - 6p $^2P_{3/2}$
4554 Å
3 → 4
3 → 2
3 → 3
2 → 1
2 → 2
2 → 3
Photon Counts / 10^3
55
50
64400
64500
64800
64900
Acceleration Voltage

Fig. 1

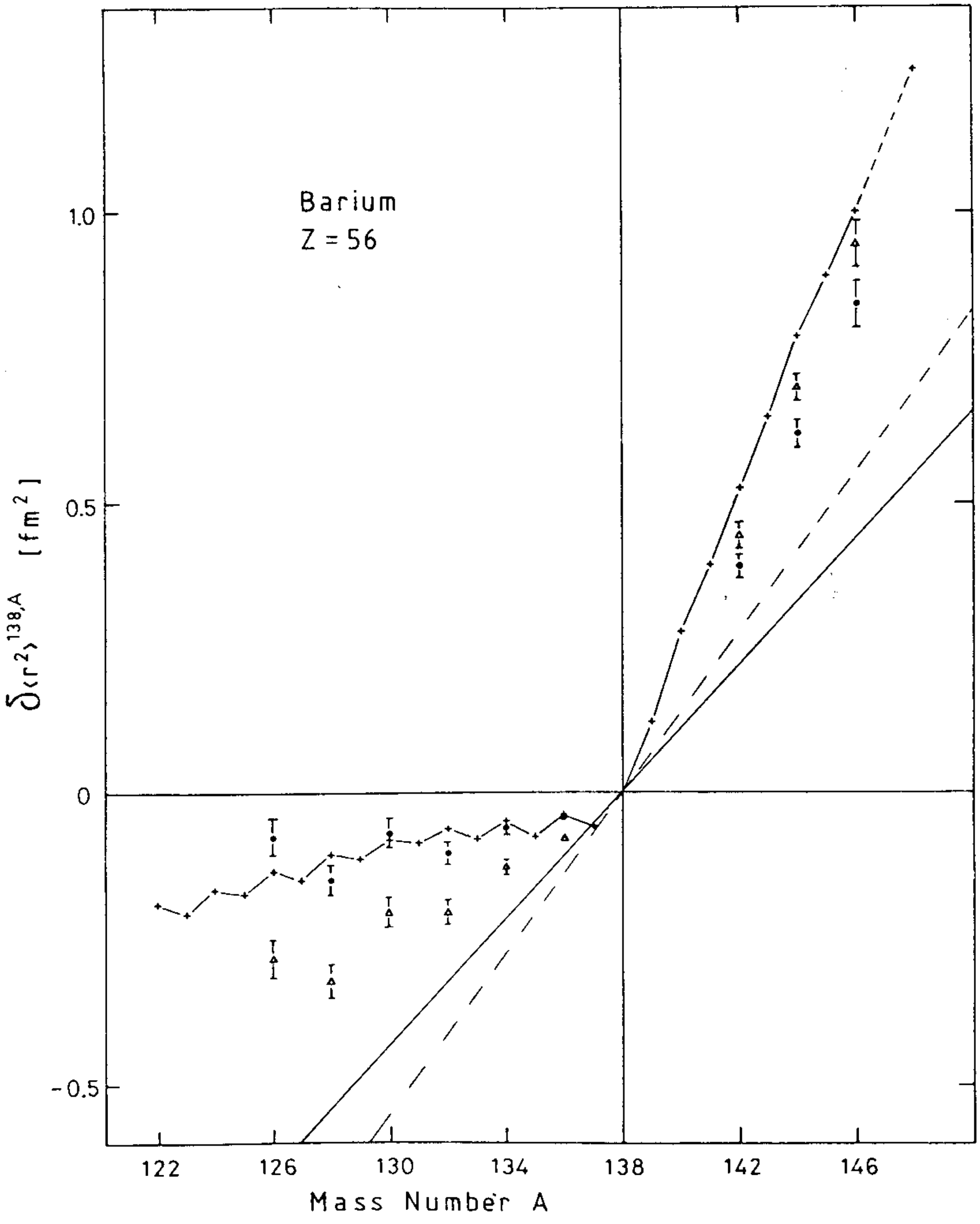
Barium
Z = 56
$\delta\langle r^2\rangle^{138,A}$ [fm^2]
1.0
0.5
0
-0.5
122
126
130
134
138
142
146
Mass Number A

Fig. 2