Isotope Shift Measurements for Superdeformed Fission Isomeric States

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Optical isotope shift measurements have been performed for the $^{240,242}\mathrm{Am}^f$ fission isomers with low target production rates of $10\ \mathrm{s}^{-1}$ employing resonance ionization spectroscopy in a buffer gas cell. Isotope shift ratios $IS^{240f,241}/IS^{243,241} = 39.2(8)$ and $IS^{242f,241}/IS^{243,241} = 41.4(8)$ have been measured at the 500.02 nm transition. A difference in the nuclear mean charge radii $\delta\langle r^2\rangle_{\mathrm{opt}}^{242f,241} = 5.34(28)\ \mathrm{fm}^2$ and an intrinsic quadrupole moment $Q_{20}^{242f} = (35.5 \pm 1.0_{\mathrm{st}} \pm 1.2_{\mathrm{mod}})\ e\,\mathrm{b}$ have been deduced, neglecting the nuclear polarization correction. The small difference $\delta Q_{20}^{242f,240f} = 0.63(8)\ e\,\mathrm{b}$ demonstrates the stability of the deformation if two neutrons are removed. [S0031-9007(97)04957-0]

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It is well known that isomeric fission originates from a superdeformed nuclear shape with low excitation energy (2–3 MeV) and low spin ($I \leq 4$), (see, e.g., the review articles by Metag *et al.* [1] and Bjornholm *et al.* [2]). Fission isomers, therefore, offer the unique possibility to test calculations of shell corrections to the potential energy of the liquid nuclear drop as well as single-particle model calculations at this state of extreme deformation. For a rigorous and meaningful test a set of data as complete as possible should be available. Such data as the deformation parameter, the magnetic moment, and the nuclear spin can best be obtained by optical hyperfine spectroscopy [3]. In a first experimental step a measurement of the isotope shift would already allow one to determine the nuclear deformation parameter β_2. If, in addition, the isotope shift can be measured in chains of fission isomers, a detailed insight into the stability of the nuclear deformation as a function of the neutron number can be obtained.

A first optical spectroscopy experiment has been conducted by Bemis *et al.* [4] for the fission isomer $^{240}\mathrm{Am}^f$ with a half-life $T_{1/2} = 0.9$ ms. The isotope shift ratio $X_{\mathrm{exp}}^{240f} = IS^{240f,241}/IS^{243,241} = 26.8 \pm 2.0$ with respect to the reference isotopes $^{241,243}\mathrm{Am}$ was measured for the $^{10}P_{7/2} \rightarrow {}^{8}S_{7/2}$ (640.5 nm) optical transition with a laser-induced nuclear polarization method (LINUP). From this result an intrinsic quadrupole moment $Q_{20}^{240f} = (29.0 \pm 1.3)\ e\,\mathrm{b}$ was deduced in a later analysis [5]. Such optical experiments at nuclei in the second potential minimum are extremely difficult since the production rate of fission isomers is very low, typically only on the order of a few per second, and the half-lives of the fission isomers are very short. As a consequence, the measured signal of Ref. [4] had poor statistics, and this fact alone is already reason enough to verify the reported experimental results at least once in an independent experiment. Our first approach to reach this goal with a fission-detected optical pumping method failed [6].

In this Letter we report on isotope shift measurements at $^{240,242}\mathrm{Am}^f$ fission isomers to test the stability of nuclear deformation in the second potential minimum using the very sensitive fission-radiation detected resonance ionization spectroscopy (f-RADRIS) method in a buffer gas cell. With such a method, hyperfine spectroscopy will also be possible, as has been demonstrated at the β-active isotope $^{208}\mathrm{Tl}$ [7].

Two experimental setups have been used. The old setup is described in Ref. [8], the new, improved setup is shown in Fig. 1. The $^{242}\mathrm{Am}^f$ fission isomers with a half-life of 14 ms were produced through the $^{242}\mathrm{Pu}\ (d, 2n)^{242}\mathrm{Am}^f$ reaction by using a pulsed (5 ms on, 5 ms off) 12 MeV deuteron beam from the Emperor tandem Van de Graaff accelerator at the Max-Planck-Institut für Kernphysik in Heidelberg. The fission isomers left the target (50 $\mu\mathrm{g/cm}^2$ $^{242}\mathrm{Pu}F_3$ on C backing) with an energy of <100 keV. Conversion electron transitions and succeeding Auger cascades result in large nonequilibrium ionic charge states, typically between 10^+ and 35^+ [1]. After postacceleration in an appropriate electrical potential difference of 95 kV, the energy of the fission isomers was high enough to penetrate a 50 $\mu\mathrm{g/cm}^2$ thick entrance window of the optical cell. The fission isomer rate at the entrance of the optical cell amounted typically to $6\ \mathrm{s}^{-1}$ at a beam current of 5 μA.

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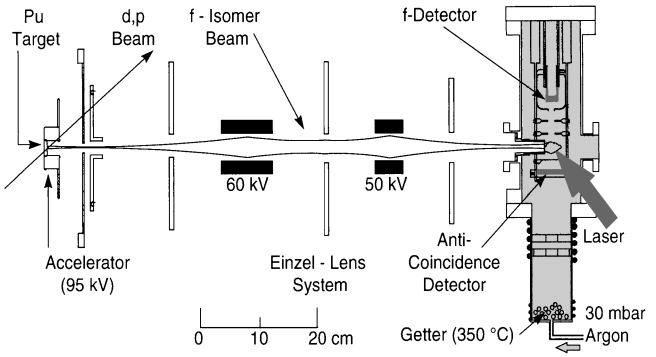

FIG. 1. Experimental setup. The primary beam hits the ^{242}Pu target under an angle of 45°. The fission isomers recoiling out of the target are accelerated and focused with a Einzel-lens system onto the optical buffer gas cell. The gas was purified using getter techniques to a 0.1 ppm level to avoid molecule formation of americium recoils with gas impurities.

The optical cell, filled with 30 mbar argon and 0.3 mbar nitrogen, the latter serving as a quenching gas, was loaded with fission isomers in the beam-on periods. A fraction of about 13% of the recoiling ions was neutralized during the slowing down process in the gas [6]. The remaining ions are transported with an appropriate electrical field onto a 250 μg/cm^2 electrode foil which was placed in front of the anticoincidence fission detector (PIN PD chip Siemens SFH 873, 30 × 30 mm^2). The gas acted at the same time as a storage medium for the neutral fission isomers. The diffusion time to the cell walls has been estimated to be of the order of 30 ms. Resonance ionization was performed in the beam-off periods by two-color laser beams via typical excitation schemes as shown in the inset in Fig. 2. To produce the required ionization energy of 5.97 eV [9] an excimer laser EMG 104 MSC from Lambda Physik, lasing with XeF at a wavelength of 351/353 nm for the second excitation step, and a dye laser FL 2001 for the first excitation step, was used. The excimer laser supplied, every 10 ms, two laser pulses with an interval of 2.2 ms. The resonantly ionized fission isomers were transported in the electric field of an ion electrode system to the fission detector (windowless PIN Photodiode Hamamatsu S3071, diameter = 5 mm). The average transportation time was measured to be 1.40(8) ms. A few fission events originated from isomers sticking on the foil in front of the anticoincidence detector or from neutrals remaining in the gas phase. However, these were completely rejected by the signal which the simultaneously emitted second fission fragment generated in the anticoincidence detector. Occasionally occurring electrical breakdown pulses could easily be identified by visual inspection of the signals stored with an oscilloscope.

Wavelength calibrations and measurements to optimize the laser system were performed in an off-line buffer gas cell at the α-active isotopes ^{243}Am ($T_{1/2}$ = 7370 yr) and ^{241}Am ($T_{1/2}$ = 432.6 yr) [10]. Typical spectra are shown in Fig. 3.

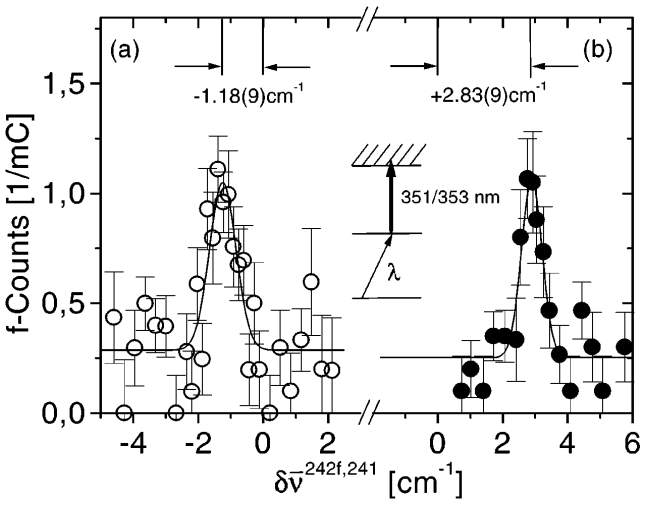

FIG. 2. Resonance ionization signals at the λ = 468.17 nm (a) and λ = 499.08 nm (b) transitions. The excitation ladder is shown in the inset of (b). The isotope shift is defined as $\delta\bar{\nu}^{242f,241} = \bar{\nu}^{242f} - \bar{\nu}^{241}$. Data acquisition time T, collected d-beam charge Q, and number of events N are T = 27 h, Q = 0.48 C, N = 286 (a); T = 26 h, Q = 0.28 C, N = 148 (b).

The isotope shift has been measured for four optical transitions for which the isotope shifts $IS^{243,241}$ between the ^{243}Am and ^{241}Am isotopes are known. First experiments have been performed with the old experimental setup [8] at the wavelength λ_1 = 466.28 nm for which the isotope shift is small in order to find a signal. The results have been reported in previous papers [8,10]. The first isotope shift measurement was performed at the λ_1 = 468.17 nm transition [see Fig. 2(a)]. An isotope shift $IS^{242f,241}(468\ \text{nm}) = -(1.18 \pm 0.09)\ \text{cm}^{-1}$ was measured, corresponding to an isotope shift ratio

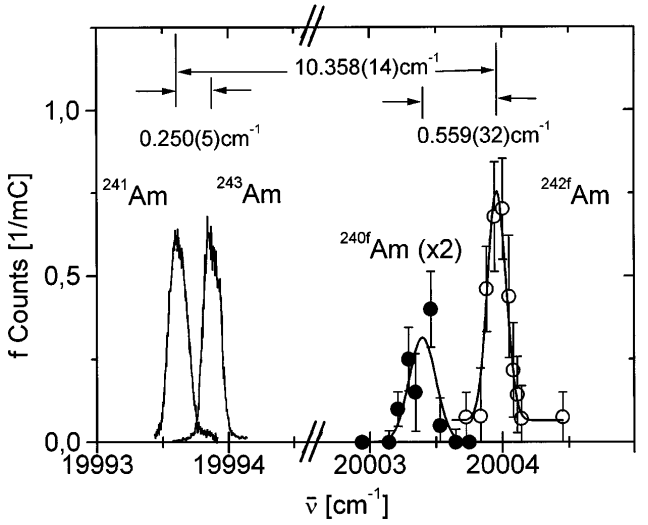

FIG. 3. Resonance ionization signals at the 500.02 nm transition of the fission isomers ^{240,242}Am^f and the long-living reference isotopes ^{241,243}Am. The count rate of ^{240}Am^f is multiplied by a factor of 2. Experiment parameters for ^{242}Am^f: T = 18 h, Q = 0.16 C, N = 62; for ^{240}Am^f: T = 25 h, Q = 0.28 C, N = 19.

$X_{\mathrm{exp}}^{242f}(468\ \mathrm{nm}) = IS^{242f,241}/IS^{243,241} = 24.6 \pm 2.4$ using $IS^{243,241} = 0.048(3)\ \mathrm{cm}^{-1}$, the weighted sum of three independent measurements $-0.052(6)\ \mathrm{cm}^{-1}$ [11], $-0.057(10)\ \mathrm{cm}^{-1}$ [12], and $-0.045(4)\ \mathrm{cm}^{-1}$ (this paper). A second measurement at a transition of $\lambda_1 = 499.08$ nm resulted in an isotope shift $IS^{242f,241}(499\ \mathrm{nm}) = +(2.83 \pm 0.09)\ \mathrm{cm}^{-1}$ [see Fig. 2(b)], corresponding to an isotope shift ratio $X_{\mathrm{exp}}^{242f}(499\ \mathrm{nm}) = 44.9 \pm 2.6$ with $IS^{243,241}(499\ \mathrm{nm}) = +0.063(3)\ \mathrm{cm}^{-1}$ determined from $0.064(4)\ \mathrm{cm}^{-1}$ [11], $0.048(10)\ \mathrm{cm}^{-1}$ [12], and $0.065(5)\ \mathrm{cm}^{-1}$ (this paper). The disagreement of the X factors originates from an anomaly in the $\lambda_1 = 468.17$ nm transition caused by configuration mixing. Close to the level at $21353.93\ \mathrm{cm}^{-1}$ another level with the same spin and parity can be found [13]. Both levels exhibit large isotope shifts but with opposite signs. The large nuclear deformation change of the fission isomeric state causes a reduction of the $114\ \mathrm{cm}^{-1}$ interval between the levels by about $7.8\ \mathrm{cm}^{-1}$. If the nondiagonal interaction matrix element between both levels is sufficiently large the energy perturbation behaves nonlinearly resulting in the above-mentioned anomaly of the isotope shift ratio. This interpretation has been corroborated by a third measurement at $\lambda_1 = 500.02$ nm of which preliminary results have been published in Ref. [14]. The isotope shift ratio of $X_{\mathrm{exp}}^{242f}(500\ \mathrm{nm}) = 41.7 \pm 0.9$ agrees well with the measurement at the unperturbed 499 nm transition.

For the $^{240}\mathrm{Am}^f$ fission isomer Bemis *et al.* [4] reported a ratio $X_{\mathrm{exp}}^{240f}(641\ \mathrm{nm}) = 26.8 \pm 2.0$, which would imply a drastic change of nuclear deformation between $^{240}\mathrm{Am}^f$ and $^{242}\mathrm{Am}^f$. In order to corroborate such an effect, which would indeed be a rather interesting one, an experiment has been performed for $^{240}\mathrm{Am}^f$ with the new setup shown in Fig. 1 at the 500.02 nm transition.

The isomers with a half-life of 0.9 ms were produced by the $^{242}\mathrm{Pu}(p,3n)^{240}\mathrm{Am}^f$ ($\sigma = 10\ \mu$b) with a pulsed (2 ms on, 2 ms off) proton beam of 23 MeV energy. Only one laser pulse in the beam-off period was supplied every 4 ms. Since the scanning interval was expected to be rather large, the laser bandwidth was chosen to be 6 GHz. Finally, the resonance ionization signal was found very close to the $^{242}\mathrm{Am}^f$ signal. The signal is shown in Fig. 3, together with a remeasurement of the $^{242}\mathrm{Am}^f$ resonance with a narrow dye laser bandwidth of 1.5 GHz. The isotope shifts $IS^{240f,241} = 9.799(28)\ \mathrm{cm}^{-1}$ and $IS^{242f,241} = 10.358(14)\ \mathrm{cm}^{-1}$ with respect to the $^{241}\mathrm{Am}$ correspond to isotope shift ratios $X_{\mathrm{exp}}^{240f}(500\ \mathrm{nm}) = 39.2(8)$ and $X_{\mathrm{exp}}^{242f}(500\ \mathrm{nm}) = 41.4(8)$, respectively. These ratios were determined with the isotope shift $IS^{243,241}(500\ \mathrm{nm}) = 0.250(5)\ \mathrm{cm}^{-1}$, the weighted sum of $0.244(4)\ \mathrm{cm}^{-1}$ [11], $0.253(10)\ \mathrm{cm}^{-1}$ [12], and $0.259(5)\ \mathrm{cm}^{-1}$ (this paper). The result $X_{\mathrm{exp}}^{240f}(500\ \mathrm{nm}) = 39.2(8)$ for $^{240}\mathrm{Am}^f$ is at variance with the value of $X_{\mathrm{exp}}^{240f}(640\ \mathrm{nm}) = 26.8 \pm 2.0$ reported by Bemis *et al.* [4] and does not support the above-mentioned conjecture of a drastic deformation change.

In the determination of the deformation of the fission isomeric states the mass shift contributions were neglected. This is justified because the normal mass shift of $3.8 \times 10^{-4}\ \mathrm{cm}^{-1}$ ($^{243,241}\mathrm{Am}$) is only 0.004% of the measured isotope shift of the fission isomers. The remaining field shift $IS^{A,A'} = F^{A'}\Lambda^{A,A'}$ was factorized, as usual, in an electronic factor $F^{A'}$ and a nuclear charge parameter $\Lambda^{A,A'} = \delta\langle r^2\rangle + S_4\delta\langle r^4\rangle + S_6\delta\langle r^6\rangle$ [15]. The $\delta\langle r^n\rangle = \langle r^n\rangle^A - \langle r^n\rangle^{A'}$ is the change of the nth moment $\langle r^n\rangle$ of the nuclear charge distribution for isotopes A and A', and S_n are the Seltzer coefficients ($S_2 = 1$, $S_4 = -1.365 \times 10^{-3}\ \mathrm{fm}^{-2}$, $S_6 = 3.155 \times 10^{-6}\ \mathrm{fm}^{-4}$). In this factorization the isotope shift ratio is given by $X = \Lambda^{242f,241}/\Lambda^{243,241}$. To determine the nuclear charge parameter $\Lambda^{242f,241}$ the corresponding parameter of the reference isotopes $\Lambda^{243,241}$ has to be known. Aufmuth *et al.* [16] quoted $\Lambda^{243,241} = 0.128(13)\ \mathrm{fm}^2$ which is in agreement with $\Lambda^{243,241} = 0.129\ \mathrm{fm}^2$ deduced from muonic atom measurements [5]. For further analysis this latter value has been used with an estimated error of $\Delta\Lambda^{243,241} = \pm 0.007\ \mathrm{fm}^2$, resulting in the nuclear charge parameter $\Lambda^{242f,241} = \delta\langle r^2\rangle_{\mathrm{opt}}^{242f,241} = 5.34(28)\ \mathrm{fm}^2$.

The deformation parameters β_2 and the intrinsic quadrupole moments Q_{20} have been deduced in a first analysis from the nuclear parameters, employing a charge distribution of a deformed Fermi model [17] and assuming that the isotope shift $^{243,241}\mathrm{Am}$ between the reference isotopes originates from a pure nuclear volume change. The measured quadrupole moment $Q_{20}^{243} = 12.02(9)\ e\,\mathrm{b}$ [18] was reproduced by numerical integration with $\beta_2^{243} = 0.284$, a hexadecapole parameter $\beta_4^{243} = 0$, a radius parameter $R_0^{243} = 7.2025$ fm, and a skin thickness parameter $a = 0.522$ fm. The radius parameter R_0^{243} has been adapted to reproduce $R_0^{241} = 7.1865$ fm [5] and the nuclear charge parameter $\Lambda^{243,241} = 0.129\ \mathrm{fm}^2$, assuming $\beta_2^{243} = \beta_2^{241}$. The calculated ratio of the $Q_{20}^{243}/Q^{241} = 1.004$ is in accord with the measured ratio $Q_{20}^{243}/Q_{20}^{241} = 1.00(1)$ [19], supporting the fact that the isotope shift of the reference isotopes is a pure nuclear volume effect. The required radii for the analysis of the fission isomer nuclear parameter are determined by linear extrapolation of the reference isotope values at $R_0^{240} = 7.1785$ fm and $R_0^{242} = 7.1945$ fm. For the nuclei in the second potential minimum, $\beta_4^{\mathrm{II}} = 0.08$ has been assumed as theoretically predicted by Howard and Möller [20].

The results for β_2^{242f} and Q_{20}^{242f} of this analysis can be approximated as expansions

$$\beta_2^{242f} = \beta_2 + a_1\Delta\Lambda + \sum_{n=1}^{2} b_n(\Delta\beta_4)^n, \qquad (1)$$

$$Q_{20}^{242f} = Q_{20} + p_1\Delta\Lambda + \sum_{n=1}^{2} q_n(\Delta\beta_4)^n, \qquad (2)$$

in which $\Delta\Lambda$ and $\Delta\beta_4$ are the deviations from $\Lambda^{242f,241} = 5.34\ \mathrm{fm}^2$ and $\beta_4 = 0.08$, respectively. The expansion parameters are summarized in Table I (column heading,

TABLE I. Expansion parameters and results for $^{242}Am^{f}$. For definitions see Eqs. (1) and (2). In the last row the average values for β_2 and Q_{20} are quoted, respectively, as obtained from the Fermi model (numbers with an asterisk) and the droplet model analysis, corrected for the deformation dependence of the electronic factor F and Seltzer coefficients S_i of 0.6%. Quoted are statistical (st) and model errors (mod).

	Fermi	Droplet
β_2	0.691	
	0.649*	0.699
a_1 [fm^{-2}]	0.057	0.062
b_1	−0.057	−0.445
b_2	−0.564	−0.306
	$0.678 \pm 0.016_{st} \pm 0.035_{mod}$	
Q_{20} [e b]	35.6	
	36.1*	34.5
p_1 [e b/fm^2]	3.40	3.25
q_1 [e b]	6.26	15.79
q_2 [e b]	−34.25	−34.60
	$(35.5 \pm 1.0_{st} \pm 1.2_{mod})$ e b	

"Fermi"). With the Eqs. (1) and (2), other assumptions on β_4 can simply be adapted, as well as improved values for $\Lambda^{242f,241}$ which may originate from new results for $\Lambda^{243,241}$. Model errors caused by variation within reasonable limits of the skin thickness parameter, $\Delta a = 0.02$ fm, and the radius parameter, $\Delta R_0 = 0.01$ fm, can be neglected.

In order to further study the model assumptions on the result for β_2^{242f} and Q_{20}^{242f}, a second analysis has been performed on the basis of the droplet model [21] (see column heading, "Droplet," in Table I). This model includes the compressibility of the nuclear matter which causes a redistribution correction of the nuclear charge. For a comparison with the Fermi-model results, a redistribution correction has been applied (see the numbers with an asterisk for β_2 and Q_{20} in Table I). From these and the corresponding droplet numbers the final results for β_2^{242f} and Q_{20}^{242f} have been deduced (see Table I). The surprising large nuclear polarization correction [22] $\delta\langle r^2\rangle_{pol} = 0.62$ fm$^2 \simeq \delta\Lambda_{pol}$, i.e., 11.6%, would result in an increase of β_2^{242f} and Q_{20}^{242f} of 5.5% and 5.8%, respectively. This correction has not been applied. Some details on the calculation of these corrections can be found in Ref. [14].

The small difference of the nuclear parameter $\Lambda^{242f,240f} = \delta\langle r^2\rangle_{opt}^{242f,240f} = 0.288(24)$ fm^2, for the $^{242}Am^{f}$ and $^{240}Am^{f}$ fission isomers results in changes $\delta\beta_2 = 0.0076(14)$ and $\delta Q_{20} = 0.63(8)$ e b (Fermi-model analysis), demonstrating the stability of the nuclear deformation in the second potential minimum. This result is in accord with the calculations of Howard and Möller ($\delta\beta_2 = 0.00$) [20]. An experiment with $^{244}Am^{f}$, for which a larger effect may be expected, is in preparation.

In conclusion, precise isotope shift measurements have been performed at the fission isomers $^{240,242}Am^{f}$ at very low target production rates of 10 s^{-1}. The laser spectroscopic method corroborates independently the charge plunger measurements [1], the extreme deformation of fission isomers which can also be expressed in the ratio of the nuclear axes q. The value $q = 1.88$ for $^{242}Am^{f}$ is actually very close to 2:1, a theoretical prediction value [2]. A hyperfine spectroscopy for determining the nuclear spin and g factor is in progress. Such an ultrasensitive method, based on resonance ionization spectroscopy in a buffer gas cell, is promising for investigating the atomic and nuclear properties of transeinsteinium elements [23].

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