

Sudden Change in the Nuclear Charge Distribution of Very Light Gold Isotopes

K. Wallmeroth, G. Bollen, A. Dohn,^(a) P. Egelhof, J. Grüner, F. Lindenlauf, and U. Krönert
Institut für Physik, Universität Mainz, D-6500 Mainz, Federal Republic of Germany

J. Campos and A. Rodriguez Yunta
Universidad Complutense and Junta de Energía Nuclear, Madrid, Spain

M. J. G. Borge and A. Venugopalan^(b)
ISOLDE Collaboration, CERN, CH-1211 Geneva, Switzerland

J. L. Wood
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332

R. B. Moore
McGill University, Montreal, Canada H3A2B2

and

H.-J. Kluge
*Institut für Physik, Universität Mainz, D-6500 Mainz, Federal Republic of Germany, and
 ISOLDE Collaboration, CERN, CH-1211 Geneva, Switzerland*
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On-line resonance-ionization mass spectrometry has been applied to determine the isotope shift and hyperfine structure of $^{185-189}\text{Au}$ and $^{189}\text{Au}^m$ in the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2}$ ($\lambda = 268\text{ nm}$) transition. The Au atoms were obtained as daughters of mass-separated Hg isotopes produced at the ISOLDE facility at CERN ionized by a three-color, two-step resonant photoionization process, detected and mass selected by time of flight. A drastic change of the nuclear charge radius was observed between ^{187}Au and ^{186}Au , which is interpreted as an onset of strong deformation of $\beta \approx 0.25$ in ^{186}Au and ^{185}Au .

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The nuclei near $Z=80$ and $N=104$ have attracted considerable attention following the discovery by Bonn *et al.*¹ that there is a big change in the ground-state mean-square charge radius between ^{187}Hg and ^{185}Hg , and its interpretation as an onset of strong deformation at $A \leq 185$. This result, obtained from a measurement of the optical isotope shift (IS), was completely unexpected since at that time the unanimous view was that nuclei near closed shells, such as the isotopes of Hg ($Z=80$), are only weakly deformed. This view and our knowledge of the region around ^{185}Hg have since changed dramatically.

It is becoming evident that coexisting shapes occur widely throughout the Hg region. This is manifested in the Hg isotopic chain by a large and systematic odd-even staggering in the root-mean-square charge radii of the very neutron-deficient Hg isotopes,^{2,3} a huge isomer shift⁴ in ^{185}Hg , and strongly deformed bands coexisting at low excitation energy in the even-mass isotopes.^{5,6} In Pt, a similar (but more complicated) situation has been deduced in the even-mass light isotopes⁷ and a large odd-even staggering has been found in the rotational spacings of the ground-state bands of the very neutron-deficient Pt isotopes.⁸ In the neighboring odd-proton nuclei (Au and Tl isotopes) the shape coexistence is as-

sociated with shell-model intruder states.⁵ In particular, the $\pi h_{9/2}$ intruder orbital becomes the ground-state configuration in ^{185}Au (see Desthulliers *et al.*⁹ and

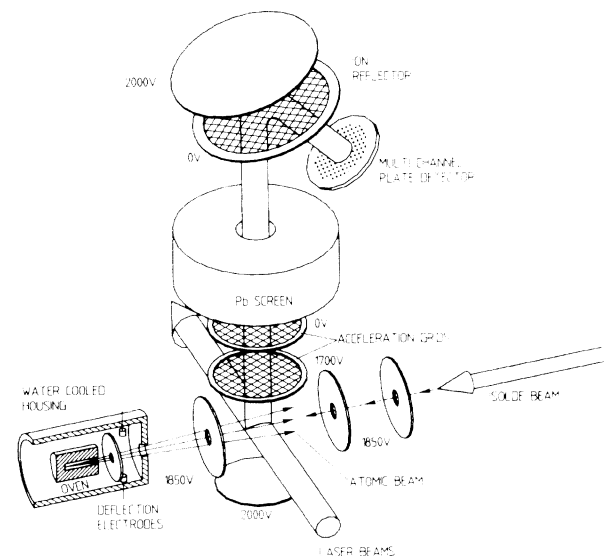


FIG. 1. Setup for on-line resonance ionization mass spectrometry on short-lived Au isotopes.

TABLE I. Optical transitions used for resonance ionization spectroscopy on short-lived Au isotopes, and data of the different lasers and laser beams (SH=second harmonic).

Excitation step	Optical transition	Wave-length (nm)	Light source	Dye	Pulse energy (μJ)	Pulse duration (ns)	Bandwidth (GHz)
No. 1	$6s^2S_{1/2} \rightarrow 6p^2P_{1/2}$	535	Molelectron DL16P ^a	C540A	10 ³	6	≤ 0.2
No. 2	$6p^2P_{1/2} \rightarrow 6d^2D_{3/2}$	268	SH of 535 nm	...	25	4	≤ 0.4
No. 3	$6d^2D_{3/2} \rightarrow \text{continuum}$	407	Lambda FL2001	DPS	150	6	6
		532	SH of Nd:YAG	...	4×10^4	15	15

^aModified as described by Bollen *et al.* (Ref. 17).

Ekström *et al.*¹⁰) whereas in the odd-mass Tl isotopes the $\pi h_{9/2}$ intruder state remains an excited state. For the latter case, isomerism results from this $\frac{9}{2}$ state, which has been studied recently by laser spectroscopy.¹¹ It thus became interesting to extend the IS studies to neutron-deficient gold isotopes. In an earlier experiment¹² the IS's of ¹⁹⁷⁻¹⁹⁰Au were determined by use of semi-on-line laser-induced fluorescence spectroscopy.

The present experiment was designed to investigate still shorter-lived Au isotopes and to determine their IS's, which represent the most straightforward and model-independent signature for shape transitions or coexistence. The Au isotopes were obtained at the ISOLDE mass separator at CERN, as daughters of Hg isotopes which could be produced with yields up to 10^{10} atoms per second per mass number by a $\text{Pb}(p,3pxn)\text{Hg}$ spallation reaction.¹³ The technique of resonance ionization spectroscopy^{14,15} was used and combined with time-of-flight mass spectroscopy. Alkhazov *et al.*¹⁶ performed the first on-line resonance-ionization-spectroscopy experiment on short-lived isotopes. The present Letter reports the first on-line application of resonance-ionization-mass spectroscopy.

Figure 1 shows the experimental setup. The ISOLDE beam was focused into an atomic beam oven. After a suitable time had elapsed during which the Hg nuclei decayed to the daughter isotope, a thermal atomic beam was formed by evaporating the radioactive sample. The Au atoms were excited and photoionized via a three-

color, two-step resonant process by the light from two tunable dye lasers pumped by a Nd-doped: yttrium aluminum garnet (YAG) laser and the frequency-doubled output of the same Nd:YAG laser (repetition rate = 10 Hz). The optical transitions used for this experiment are shown in Table I. In order to increase the signal-to-background ratio, the photoions were detected and mass selected by a time-of-flight spectrometer.¹⁸ An ion reflector was installed to allow an efficient shielding of the multichannel plate detector against nuclear radiation from the oven/acceleration region. A background rate as low as 1 event per 1000 laser shots was obtained with a time-of-flight mass resolution of 250 and a gate of 70-ns width on the mass of the isotope under investigation. The overall detection efficiency of our resonance-ionization-mass-spectrometry technique for Au (defined as the ratio of Au ions detected by the multichannel plate to the number of atoms collected in the oven) was about 10^{-8} , mainly limited by the low repetition rate of the laser system.

Data on the hyperfine structure (hfs) and IS in the D_1 line of ¹⁸⁵⁻¹⁸⁹Au and ¹⁸⁹Au^m were obtained. The IS of ¹⁹⁰Au was remeasured and agrees within error bars with the value cited in the earlier study.¹² The magnetic moments deduced are compiled in Table II. The agreement with earlier measurements^{10,20} is good, except for ¹⁸⁷Au. The reason for this discrepancy is not understood.

Table III gives the results obtained from the determination of the IS in the D_1 line. The third column

TABLE II. Spins, ground-state hyperfine constants, and magnetic moments of short-lived Au isotopes.

<i>A</i>	<i>I</i>	This work ^a		Literature	
		<i>A</i> ($6s^2S_{1/2}$) (MHz)	μ_I/μ_N	μ_I/μ_N	Reference
189	$\frac{1}{2}$	+28 632(128)	+0.49(3)
189 ^m	$\frac{11}{2}$	+32 625(42)	+6.19(3)	$\pm 6.19(12)$	19
188	1	-1940(127)	-0.07(3)	$\pm 0.069(24)$	18
187	$\frac{1}{2}$	+31 032(168)	+0.54(3)	+0.72(7)	18
186	3	-12 207(48)	-1.26(3)	$\pm 1.07(13)$	19
185	$\frac{5}{2}$	+25 176(56)	+2.17(3)	$\pm 2.22(14)$	19

^aCalculated from the magnetic HFS splitting in the D_1 line with the relation (Ref. 10) $\mu_I = (AI/29005)\mu_N$. The experimental error accounts for the uncertainty in the HFS anomaly.

TABLE III. Isotope shift and deduced data of the charge radii and deformations as obtained in the D_1 line of the short-lived Au isotopes. ^{197}Au is taken as the reference isotope. The errors given are purely experimental and do not account for the (common scaling) uncertainty of F_{268} .

A	$T_{1/2}$ (min)	$\delta\nu_{\text{expt}}^{197,A}$ (GHz)	$\lambda^{197,A}$ (fm ²)	$\delta\langle r^2 \rangle^{197,A}$ (fm ²)	$\delta\langle \beta^2 \rangle^{197,A}$	$\langle \beta^2 \rangle^{1/2}$
190	42.8	11.39(16)	-0.267(6)	-0.285(6)	0.007	0.14
189	28.3	12.35(18)	-0.290(7)	-0.309(7)	0.010	0.15
189 m	4.6	11.36(22)	-0.267(8)	-0.283(9)	0.012	0.15
188	8.8	13.91(23)	-0.327(8)	-0.347(9)	0.011	0.15
187	8.0	15.40(23)	-0.362(9)	-0.385(9)	0.012	0.16
186	10.7	0.84(17)	-0.025(8)	-0.014(8)	0.048	0.25
185	4.2	3.26(18)	-0.081(8)	-0.075(9)	0.048	0.24

shows the experimental IS's relative to ^{197}Au . After correction for the normal mass shift and taking into account an estimate²¹ for the specific mass shift, the remaining field shift $\delta\nu_{\text{FS}}^{A,A'}$ were related by $\delta\nu_{\text{FS}}^{A,A'} = F_{268}\lambda^{A,A'}$ to the nuclear parameter

$$\lambda^{A,A'} = \sum (C_n/C_1) \delta\langle r^{2n} \rangle^{A,A'},$$

where F_{268} is the electronic factor for the D_1 transition. The parameters $F_{268} \equiv C_1 = -43.07$ GHz/fm² and C_n/C_1 were calculated with multiconfigurational Dirac-Fock theory.²² The resulting model-independent λ parameters are shown in the fourth column of Table III. Column five of Table III and Fig. 2 show the $\delta\langle r^2 \rangle$ values, calculated as in the case of Hg (Ref. 3) and Eu (see Ahmad *et al.*²³). The slope of the mean square charge radii is monotonic down to ^{187}Au and is similar to the one of the Hg isotopes. Between ^{187}Au and ^{186}Au , a drastic change occurs, similar to the Hg isotopes but two neutron numbers earlier. There is a much less pronounced odd-even staggering for the Au isotopes than for the Hg isotopes. As in the Hg isotopes, the break in the IS can be related to a change in nuclear quadrupole de-

formation β by

$$\delta\langle r^2 \rangle = \delta\langle r^2 \rangle_{\text{sph}} + \delta\langle r^2 \rangle_{\text{def}}$$

and

$$\delta\langle r^2 \rangle_{\text{def}} = (5/4\pi) \langle r^2 \rangle_{\text{sph}} \delta\langle \beta^2 \rangle.$$

The values $\delta\langle r^2 \rangle_{\text{sph}}$ are taken from the droplet model,²⁴ which is known to describe well the spherical part of the change of the nuclear charge radius in the mass region under consideration. The resulting changes in the rms deformation parameters are compiled in Table III. Absolute deformation parameters can be evaluated if β is known for at least one isotope. As in the previous publication,¹² we take $|\beta(^{197}\text{Au})| = 0.11$, which leads to the values given in the last column of Table III.

The values of $\langle \beta^2 \rangle^{1/2}$ in Table III clearly imply the sudden onset of deformation in going from ^{187}Au to ^{186}Au , with its persistence of ^{185}Au . The change between ^{187}Au and ^{186}Au is also reflected in $B(E2)$ values for transitions feeding the ground states (see Berg and co-workers^{25,26} and Abreu *et al.*²⁷). The deduced deformation of ^{185}Au (see Table III) confirms the recent prediction²⁰ ($\beta = 0.25$). The observed shape change between ^{187}Au and ^{185}Au can also be understood simply in the intruder state picture⁵ as the difference between the core-particle couplings $\pi^{-1} \otimes ^{188}\text{Hg}$ and $\pi^{+1} \otimes ^{184}\text{Pt}$, respectively. In the heavy gold isotopes the ground-state configuration is $\pi d_{3/2}^{-1}$; however, between ^{187}Au and ^{185}Au the $\pi h_{9/2}^{+1}$ configuration intrudes⁵ to become the ground state. In ^{187}Au the $d_{3/2}$ proton hole couples to a ^{188}Hg core for which a small deformation $\beta = (-)0.15$ has been determined,³ in ^{185}Au the $\pi h_{9/2}^{+1}$ proton particle couples to a ^{184}Pt core for which $\beta = (+)0.24$ can be deduced from the $B(E2; 2^+ \rightarrow 0^+)$ value.²⁸ The absence of a strong odd-even staggering between $^{185}\text{Au}/^{186}\text{Au}$ as it has been observed in the chain of the Hg isotopes starting with ^{185}Hg (see Fig. 2) can be qualitatively understood as the result of the $\pi h_{9/2}^{+1}$ intruder configuration being the ground-state proton configuration in both ^{185}Au and ^{186}Au . This confirms the conclusion of Porquet *et al.*²⁹ based on a study of excited states in ^{186}Au and on a comparison with the excited-states systematics

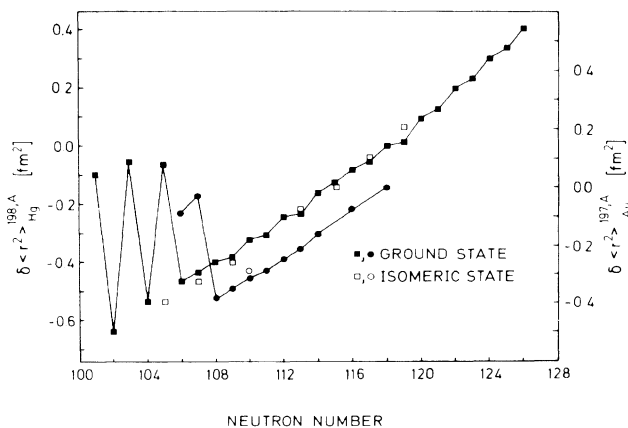


FIG. 2. Changes of the mean square charge radii of Au (circles: this work and Ref. 21) and of Hg (squares: Ref. 11) isotopes.

of the heavier odd-odd gold isotopes.

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^(a)Present address: W. C. Heraeus GmbH, D-6450 Hanau, Federal Republic of Germany.

^(b)On leave from the Bhabha Atomic Research Centre, Bombay 400085, India.

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