ISOTOPE SHIFTS OF NEUTRON-DEFICIENT GOLD ISOTOPESWITH $193 \geq A \geq 190$ §)J. Streib^{*)}, H.-J. Kluge^{**)}, H. Kremmling^{***)}, R.B. Moore^{****)},H.W. Schaaf^{*****)}, and K. Wallmeroth

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ABSTRACT

The isotope shift between ^{197}Au (stable) and the radioactive Au isotopes ^{190}Au , ^{191}Au , ^{192}Au and ^{193}Au were determined by resonance fluorescence spectroscopy in the $6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$, $\lambda = 267,6\text{ nm}$ line. The unstable Au isotopes were produced at the ISOLDE mass separator at CERN. The nuclei were investigated semi on-line in a resonance vessels, heated to 1400°C . The results are $\delta\nu^{190,197} = -11,12(39)\text{ GHz}$, $\delta\nu^{191,197} = -9,67(12)\text{ GHz}$, $\delta\nu^{192,197} = -8,32(15)\text{ GHz}$, $\delta\nu^{193,197} = -6,29(11)\text{ GHz}$, corresponding to a change of the mean-square charge radius by $\delta\langle r^2 \rangle^{190,197} = 0,261(12)\text{ fm}$, $\delta\langle r^2 \rangle^{191,197} = 0,227(5)\text{ fm}$, $\delta\langle r^2 \rangle^{192,197} = 0,195(5)\text{ fm}$, $\delta\langle r^2 \rangle^{193,197} = 0,148(4)\text{ fm}$.

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1. INTRODUCTION

The measurements reported in this paper are a continuation of our efforts to determine nuclear ground state properties of short-lived isotopes in the Hg region. In a preceding paper [1], we reported on a first determination of an isotope shift (IS) in Au between the only stable isotope ^{197}Au and the long-lived one ^{195}Au ($T_{1/2} = 183\text{ d}$). In this paper, we report on an extension of these measurements to the short-lived isotopes $^{190}\text{Au} - ^{193}\text{Au}$ performed at the on-line mass separator ISOLDE/CERN, Geneva. The motivation for this work will be recapitulated briefly in the following.

Systematic investigations of the hyperfine structure (HFS) and the IS of neutron-deficient Hg isotopes ($Z=80$) [2-5] have yielded clear evidence for a nuclear shape transition from slightly oblate ($A > 186$) to strongly prolate deformation ($A=185, 183, 181$), a shape coexistence in ^{185}Hg , and a huge odd-even staggering in the region $183 \leq A \leq 186$. A drastically large odd-even effect has also been found [6] for the 2^+ energies of the neutron-deficient isotopes of Pt which contain, with $Z=78$, two protons less than Hg. Furthermore, the observed spectroscopic quadrupole moments of the Hg isomers could be explained as being due to a change in the $i_{1/2}$ neutron-core coupling [5,7], which represented the first quantitative interpretation of the spectroscopic quadrupole moments of transitional nuclei [8].

Since these effects critically depend on the shell occupation and on the shell and pairing energies, it is interesting to perform a similar systematic study in the element Au ($Z=79$), which is located between Pt and Hg.

In the mass region of special interest ($A \approx 185$), the Au nuclei are short-lived with half lives in the range of minutes or seconds. Therefore, their investigation has to be performed on-line with the production of the isotopes. Furthermore, a mass separation beforehand is needed, since

the nuclear production processes which have high yields (e.g. spallation by medium-energy protons), are isotopically unselective. At present, there are no efficient target-ion-source systems for on-line production and mass separation of gold isotopes. However, the Au nuclei can be obtained as daughters in the decay of Hg isotopes, which are produced with high efficiency at on-line mass separators. For example, maximum yields of more than 10^{10} ions per second per mass number are routinely achieved at the ISOLDE facility at CERN [9].

To make efficient use of these nuclei, the Hg mother isotope is confined in a resonance vessel until it decays to the daughter Au isotope of interest. The optical investigation of the Au atoms has then to be performed in the same cell.

The magnetic ground state HFS and the nuclear spins of a number of radioactive Au isotopes have been determined by the Rabi technique [10-12]. No information exists on the quadrupole moments, with the exception of the only stable isotope ^{197}Au , which was investigated by optical techniques [13]. In addition, nuclear spins have been measured for a number of excited nuclear levels in the Au-chain and nuclear magnetic moments of some Au isomers containing unpaired $h\mu/2$ protons or $i43/2$ neutrons have been determined by NMR on oriented nuclei [14-18]. The results obtained have been explained in the frame of the rotational-aligned coupling scheme [19-22] and reflect the features of transitional nuclei in the region between the spherical Pb nuclei and the strongly deformed rare earths. The data resulting from this work complete the picture.

2. MAGNETIC HYPERFINE STRUCTURE AND ISOTOPE SHIFT IN THE D1-LINE OF GOLD

Fig.1 shows the atomic level scheme of Au. It is an alkaline-like spectrum with a $6s^2S_{1/2}$ ground state. For the experiments reported here, the D1-resonance line ($6s^2S_{1/2} - 6p^2P_{1/2}$, $\lambda = 267.6$ nm) was used.

Because $J=1/2$ in both states, only the magnetic HFS has to be considered, leading to a HFS splitting of $\Delta E=2A$ where A represents the HFS interaction factor in the state under consideration. In the case of ^{197}Au , $A(^2S_{1/2})=3049.660092(7)$ MHz has been obtained by atomic-beam-magnetic-resonance (ABMR) [23] and $A(^2P_{1/2})=337.5(7.5)$ MHz has been obtained by optical spectroscopy [13].

The IS between isotopes with mass number A and A' is given by [24] as

$$\delta \nu^{A,A'} = \nu^{A'} - \nu^A = \delta \nu_f^{A,A'} - \delta \nu_m^{A,A'} \quad (1)$$

where the first term represents the field shift and the latter is the sum of the normal and specific mass shift;

$$\delta \nu_m^{A,A'} = (\gamma_0/1836)(A'-A)/A'A + M_{\text{spez}}(A'-A)/A'A \quad (2)$$

As in the preceding paper [1], we use

$$\delta \nu_m^{A,197} = -(4.0 \pm 2.8)(1-197/A) \text{ [GHz]} \quad (3)$$

where the error is due to the uncertainty of the contribution by the specific mass effect [24].

From [24] the field shift is connected to the change in the nuclear charge radius between isotopes with mass number A and A' by;

$$\delta \nu_f^{A,A'} = c_1^{A,A'} \cdot \lambda^{A,A'} \quad (4)$$

with

$$\lambda^{A,A'} = \delta \langle r^2 \rangle^{A,A'} + (c_2/c_1) \delta \langle r^4 \rangle^{A,A'} + (c_3/c_1) \delta \langle r^6 \rangle^{A,A'} + \dots \quad (5)$$

In the preceding paper [1], we used $c_1^{195,197} = -50(5)$ GHz/fm obtained by atomic spectroscopic data (HFS and the Fermi-Goudsmit-Segre formula). A recent calculation of the screening factor β by P. Aufmuth [65] yielded $\beta = 1.14$, which results in a reduction of c_1 to $c_1 = -47.5(5.0)$ GHz/fm². The ratios c_k/c_1 were taken from Selzer [25]. In the meantime, Fricke and Rosen [26] have performed relativistic single-configuration Dirac Fock calculations which yield

$$\begin{aligned} c_1 &= -45.14 \text{ GHz fm}^{-2} \\ c_2/c_1 &= -1.19 \cdot 10^{-3} \text{ fm}^{-2} \\ c_3/c_1 &= 3.66 \cdot 10^{-6} \text{ fm}^{-4} \end{aligned} \quad (6)$$

in agreement within the limits of error with the values used in the preceding paper. The SCDF values will be used in the following with an estimated uncertainty of 10%. The dependence of c_1 on the mass number is small. From [3] for different isotope pairs,

$$c_1^{A,A'} = \left[(A'+A)/(A'''+A''') \right]^{(2\epsilon-2)/3} c_1^{A''',A'''} \quad (7)$$

with $\epsilon = (1 - \alpha^2 Z^2)^{1/2}$ where α is the fine structure constant.

In order to isolate the change in the second moment from the power expansion (5), the contributions of the higher moments are calculated for a uniform charge distribution by

$$\delta \langle r^{2n} \rangle_{\text{unif}} = (2n/2n+3) r_o^{2n} \cdot \delta A/A \quad (8)$$

with $r_o = 1.2 A^{1/3} \text{ fm}$.

This leads to

$$\delta \nu_f^{A,197} = -42.0 \cdot \delta \langle r^2 \rangle^{A,197} \quad (9)$$

with $\delta \nu_f$ given in GHz and the mean-square radius in fm^2 .

3. EXPERIMENTAL METHOD

Different experimental techniques have been tested in the laboratory at Mainz in order to choose the most appropriate method for the investigation of short-lived Au isotopes which are produced in the decay of Hg. Important criteria were, (i) sensitivity, (ii) resolution, and (iii) easy applicability in view of the limited beam time available for an accelerator-based experiment.

In the preceding publication [1], we reported on a first determination of an IS in Au by polarization and resonance fluorescence spectroscopy. Polarization spectroscopy [27] has two important advantages: It is Doppler free and the photomultiplier monitoring the probe beam (which passes two crossed polarizers with the resonance cell in between) can be positioned far away from the resonance cell. By this, laser light scattered off the walls of the resonance cell can be suppressed very efficiently. However, this method was ruled out for on-line measurements of small quantities of radioactive material because no cell window material could be found which is transparent in the UV spectral region and does not become birefringent at temperatures above 1150°C [1].

An attractive alternative is two-photon spectroscopy [28] with detection of the optical resonance by fluorescence or by the ion created by the photoionization of the excited atom by a third photon. This method is also Doppler free and has the advantage that all atoms can be excited independently of their velocity. Furthermore, the optical observation of the resonance takes place at a different wavelength than the excitation. Hence, wall-scattered laser light can be suppressed efficiently in the observation of the resonance fluorescence.

For the observation of ions, a detection angle of 4π and a high detection efficiency for the Au ions can easily be obtained. The high laser power and narrow laser bandwidth needed for two-photon spectroscopy was achieved by modification of a commercial pulsed laser system [29]. For a test of this technique the $6s\ ^2S_{1/2} - 8s\ ^2S_{1/2}$ ($\lambda = 2 * 308.9\text{ nm}$) transition in Au was used. The resonance was observed optically in the $8s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$ ($\lambda = 424.2\text{ nm}$) transition (Fig.1). However, it was found that the two-photon signal is extremely sensitive to small misalignments of the laser system as well as of the overlap of the two counter-propagating beams [30]. In addition, high Au vapor pressures were needed in order to obtain reasonable signal-to-noise ratios. Details of the laser system and the two-photon experiments which yielded HFS, IS and lifetimes of excited S-states in Au and Hg will be published elsewhere [31].

As a consequence of these tests, we fall back on the straightforward scheme of resonance-fluorescence spectroscopy which has been applied already successfully in the case of the investigation of Hg and Cd isotopes [4,5,32,33].

In the experiment described below, the D1-line ($6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$, $\lambda = 267.6\text{ nm}$) was used for excitation and resonance detection. Because of the thermal velocity distribution of the Au atoms in the cell, the resonance fluorescence signal suffers from the Doppler width ($\delta\nu_D = 2.3\text{ GHz}$ at 1300°C). However, this is not a serious drawback in the case of Au where the IS's are large. More disadvantageous is the impossibility of efficient discrimination between the fluorescent photons and the laser light scattered off the walls of the resonance cell. This is because the wavelengths for excitation and observation are the same and because the magnitude of the laser pulse length ($\approx 3\text{ ns}$) and the lifetime of the $6p\ ^2P_{1/2}$ state (6 ns [34]) are similar.

Observation of the resonance via the transition to the low lying $^2D_{3/2}$ state ($\lambda = 627.8$ nm) is excluded due to the presence of the strong black-body radiation of the hot cell-oven system.

Test experiments with stable ^{197}Au proved that a minimum number of 10^{11} atoms are needed in the cell for the detection of a resonance fluorescence signal.

4. PREPARATION OF THE RADIOACTIVE AU SAMPLE IN THE CELL

The measurement of the radioactive Au isotopes were performed in a semi on-line mode at the ISOLDE mass separator at CERN [9]. The Au nuclei are obtained as daughters in the decay of mass-separated Hg isotopes. Mercury is produced in a spallation reaction by bombarding a Pb target with 600 MeV protons from the synchro-cyclotron at CERN (typical beam intensity $1\ \mu\text{A}$). The Hg isotopes diffuse out of the target into a plasma-ion source, are ionized, accelerated to 60 keV and mass separated.

The isotopically clean ion beam with a typical intensity of 10^{10} ions per second and mass number was implanted into a graphite foil (2.5 mm diameter, 0.4 mm thickness). After a collecting time of 10 to 60 minutes, the foil was withdrawn from the vacuum and manually put into the cell. When heating the cell to temperatures above 1300°C , 90 % of the radioactive atoms collected on the foil diffused out of the foil and were available for optical spectroscopy.

5. EXPERIMENTAL SET-UP

The experimental set-up is shown in Fig.2. Radioactive Au atoms confined in a high-temperature sapphire cell are excited by light pulses from a frequency-doubled dye laser pumped by a Nd:Yag laser. An atomic beam of stable ^{197}Au , an absorption cell containing $(^{130}\text{Te})_2$, a Fabry-Perot, and a pressure reading in the oscillator cavity of the dye laser serve for a frequency determination of the laser light. All signals are fed via CAMAC into a minicomputer which also controls the experiment. The details of the set-up are discussed in the following.

5.1. THE CELL-OVEN SYSTEM

The low vapor pressure of Au (10^{-6} mbar at 950°C) and the need of a short adsorption time of the atoms on the wall of the resonance cell require a high operating temperature of the resonance vessel. The last requirement is most important when only a limited number of radioactive atoms is available. The fraction of free atoms in the cell was measured with an optical absorption experiment with stable Au atoms. In the case of 10^{13} Au atoms in the sapphire cell ($T=1300^\circ\text{C}$) a vapor pressure was observed which was 100 times lower than expected from the saturated vapor pressure curve [35]. Different materials for the collecting foil were tested. Minimum reduction of the fraction of free atoms in the vapor phase was found for a graphite foil. The design of the resonance vessel was the central problem at the beginning of this work. A detailed description of the solution found is published elsewhere [36]. A schematic outline of the design of the high-temperature cell is given in Fig.3.

The resonance cell is made of sapphire. It consists of a tube which is closed by pressing two optically flat windows onto the ends. The inner diameter of the cell is 7 mm and the length is 20 mm. The resonance vessel is mounted inside a Mo tube which is heated to 1400°C by passing a current of up to 700 A through it. The entire cell-oven system is surrounded by heat shields and placed in a vacuum container which is evacuated to pressures below 10^{-5} mbar. The laser beam is sent through the cell coaxial to the Mo tube, while the resonance light is observed perpendicular to the laser beam through a hole of 5 mm diameter cut in the tube. The photomultiplier (Valvo 56DUVP) was shielded against the light of the oven by a color glass filter (Schott UC5, d=2 mm) and an additional interference filter centered at the D1 line of Au.

5.2. THE LASER

Laser light for exciting the ultraviolet D1 transition of Au is easily obtained by frequency doubling of the output of a pulsed dye laser. The low duty cycle of such a laser is not a major drawback because the Au atoms are confined in a resonance vessel. The laser used was a commercial system (Nd:Yag laser: model MY 32-10 , Dye laser: model DL 16P. , both manufactured by Molelectron Corp.), which was modified in order to satisfy the requirements for high-resolution two-photon spectroscopy.

The essential improvements consist of the replacement of the oscillator etalon of low finesse by one of high-finesse, synchronisation of the cavity and etalon modes by a galvano plate, additional frequency filtering of the laser light to a bandwidth of about 50 MHz by an external etalon and amplification of the laser light by three additional amplifier stages [31]. A simplified version (without external etalon)

was used in the experiment reported here.

Typical values of the laser output are given in Table 1 for $\lambda = 535.2$ nm and $\lambda = 267.6$ nm. The ultraviolet laser light is obtained by frequency doubling in an ADP crystal heated to 62.3°C to fulfill phase-matching condition. In order to avoid saturation of the optical transition and by this a decrease of the signal-to-background ratio, the laser light is strongly attenuated by neutral density filters.

The frequency tuning of the laser is accomplished by changing the pressure in the cavity of the dye laser containing the frequency selective grating and air-gap etalon. The scanning speed is variable by the use of different capillaries through which the pressure chamber is filled with dry nitrogen. Scanning speeds of typically 760 MHz/s were used during the measurements of unstable Au isotopes. The scanning is controlled by the minicomputer which, via a sensitive pressure transducer, reads the value of the pressure and stores it for each laser pulse.

5.3. FREQUENCY CALIBRATION

The intensity of the resonance light of the unstable Au isotope under investigation is simultaneously measured with, (i) the transmission of a Fabry-Perot etalon (free spectral range = 6 GHz), (ii) the resonance fluorescence of stable ^{197}Au in an atomic beam, (iii) the pressure in the oscillator chamber of the laser, and (iv) the absorption profile of $(^{130}\text{Te})_2$ in a heated cell ($T = 700^\circ\text{C}$).

With the exception of the pressure values, these data are shown in Fig. 4 for the case of a measurement of ^{192}Au . In addition, the intensity of the ultraviolet laser light is recorded in order to normalize the signals to constant laser intensity.

Since the IS of an unstable isotope is given relative to the center of gravity of the HFS of a reference isotope, it is very convenient and safe to measure the positions of the resonances relative to the signal of ^{197}Au in the atomic beam. This beam is collimated to such an extent that the reduced Doppler width ($\Delta\nu_D = 200\text{ MHz}$) enables not only the resolution of the ground state HFS but also the HFS of the excited $^2P_{1/2}$ state (see Fig. 1).

The well known ground state HFS of ^{197}Au in the atomic beam can be used to establish a relative frequency scale in order to determine the IS of the unstable isotopes. This calibration is very reasonable, provided the linearity of the whole scan is assured. To prove this, the frequency increment per channel is obtained by use of the tabulated absorption lines of molecular ^{130}Te [37] and of the pressure values in the oscillator chamber of the laser recorded for each laser pulse. The Te data have a typical accuracy of 30 MHz and, in addition, allow an absolute calibration of the wavelength.

If only terms linear and quadratic in p are considered, a scan of about 440 mbar with a scanning speed of 5 mbar/s results, for example, in a frequency-pressure relation of

$$\nu = \nu_0 + 152.04(40) * p - 0.008(10) * p^2 \quad (10)$$

with ν in MHz and p in mbar. As can be seen from (10), a satisfactory linearity of the scan is obtained.

A Fabry Perot etalon enables an additional check on the frequency calibration and is used as a diagnostic tool for the laser light in respect to bandwidth, purity of the laser modes, and intensity ratio of stimulated to superfluorescent radiation.

The signals of the photomultipliers and the photodiodes are digitized by gated analog-to-digital converters and transmitted via CAMAC to the computer. A maximum of 500 channels (i.e. double-precision memory locations) are available for each of a maximum of 6 simultaneously measured signals in one scan. In a typical scan, the signals are recorded for each laser pulse.

6. ISOTOPE SHIFT MEASUREMENTS OF NEUTRON-DEFICIENT AU ISOTOPES

Three days of beam time were used at the ISOLDE facility for investigation of the Au isotopes with mass numbers $187 \leq A \leq 193$. On the average, 10^{13} Hg atoms were collected for each sample.

About 20 min were required, (i), to transport the foil from the collection station at the ISOLDE beam line to the apparatus for optical spectroscopy, set up in a hut in the ISOLDE experimental hall, (ii), to put the foil into the sapphire cell, (iii), to pump down the vacuum container, (iv), to reach a temperature of more than 1350°C and (v), to detect the first signal.

With 10^{13} Au atoms in the cell, the resonance signal was observable for about 1 hour with steady decreasing intensity. Probably, this loss was due to small leaks between the faces of the sapphire tube and the windows but adsorption on the walls of the cell could also have contributed. The sensitivity of the method used for the investigation of radioactive Au isotopes depends critically on many parameters like cleanliness of the inner surface of the sapphire cell, surface quality of the windows, buffering of the surfaces by stable Au, diameter of the laser beam, laser intensity etc.. Generally, a signal-to-background ratio of 1 was achieved with 10^{11} Au atoms present in the cell where the background was almost entirely due to laser light scattered off the walls of the resonance cell.

In some of the measurements, 10^{12} or 10^{13} atoms of stable ^{197}Au were put into the resonance vessel in the form of AuCl_3 on an additional graphite foil. Since the evaporation of these stable atoms is much faster than the diffusion of the implanted radioactive isotope out of the collecting foil, a saturation of the walls of the resonance vessel by ^{197}Au can be expected during the heating-up phase. This assumption is supported by the ratio of the resonance fluorescence intensity of the radioactive Au to that of ^{197}Au for the same amount of each isotope being placed in the cell (

Fig.5).

Figure 5 shows the signals obtained for ^{193}Au - ^{190}Au during a single laser scan (one laser pulse per channel, scanning time about 30 s). On the average 20 scans were taken per sample. The spectra taken for ^{189}Au - ^{187}Au , which are produced with lower yields, showed contaminations by a number of lines which have a width similar to the expected lines but which are extremely temperature dependent in respect to height and center frequency. Since these lines partly overlap and mask the signals of the radioactive Au isotopes under investigation, the spectra could not be evaluated. During the limited beam time available, the source of contamination could not be completely identified. Most probably, the added graphite foil (prepared with a $^{197}\text{AuCl}_3$ solution in order to saturate the walls of the cell) is responsible for the additional (molecular?) lines.

7. FITTING PROCEDURE

The nuclear spin and HFS splitting (and from this the magnetic moment) of the atomic ground state of the isotopes investigated in this work have been measured by ABMR. In the cases of ^{190}Au and ^{192}Au the sign of the magnetic moment was unknown (see below). The HFS of the excited $^2P_{1/2}$ state can be calculated from $A(^2S_{1/2})$ neglecting the difference in the hyperfine anomalies in the ground and excited states. The HFS of the excited state was used as a fixed parameter in the fitting procedure because it has a very small effect on the line profile of the resonances, this line profile being governed primarily by the Doppler profile of the absorption and the spectral profile of the laser light. The latter profile was unknown a priori, but found experimentally to be primarily Lorentzian. Hence the profile of the resonance should be described by a Voigt profile.

However, in the case of the cell signal $\delta\nu_{\text{DOPPLER}}$ is much larger than $\delta\nu_{\text{LASER}}$ and therefore no significant difference was found when using a Voigt or Gaussian profile for the determination of the center of gravity of the HFS. In order to avoid the expenditure of time for an exact folding, a purely Gaussian profile with a halfwidth of $\delta\nu = (\delta\nu_D^2 + \delta\nu_L^2)^{1/2}$ was used to fit the experimental data by use of a least-square routine [38]. The program determined the center, the width and the height of the HFS components. As mentioned in the previous section, in the case of ^{190}Au and ^{192}Au , stable ^{197}Au was used to passivate the walls of the resonance vessel. Here, the positions of the HFS components of ^{197}Au were fixed to the positions known from the atomic beam spectra. During the measurement of ^{190}Au , the mass resolution of the separator was poor. As a result, the ^{190}Au signal was contaminated. Therefore, an admixture of another isotope was introduced in the fitting procedure of ^{190}Au . The additional components exhibited the HFS splitting and IS of ^{194}Au . Furthermore, the time-differential observation of the height of the signals

of ^{191}Au and ^{190}Au yielded, after correction for the holding time of the cell, the correct ratio for the nuclear half lives.

The previously unknown sign of the magnetic moment of ^{190}Au could be determined to be negative. Only this choice of sign enables a satisfactory description of the resonance pattern obtained for ^{180}Au . Due to the small HFS, the sign of the magnetic moment of ^{192}Au could not be determined.

On the average, 5 to 15 scans were analysed and accepted for final evaluation. From these data, the mean of the centers of gravity and its standard deviation were calculated. The resulting IS relative to ^{197}Au are listed in Table 2 which also gives the $\lambda_{A,A'}$ parameter and $\delta\langle\tau^2\rangle$ values calculated after correction for the mass shift (3) by (4) and (9). The already measured value of the IS of ^{195}Au [1] is reevaluated and included in Table 2.

8. INTERPRETATION OF THE RESULTS AND DISCUSSION

Figure 6 shows a plot of the available $\delta\langle r^2 \rangle^{A,A'}$ data in the Pb to Os region. Since the direct use of the results published until now (and obtained by optical spectroscopy, muonic x-rays and electron scattering experiments) leads to inconsistencies and unphysical results, we have used the following procedure:

The absolute value $\langle r^2 \rangle$ of one isotope of each element is obtained from the Droplet model [44-46] with a correction for deformation as calculated by Nix and Moeller [47,48]. These "reference" isotopes are indicated by a circle in Fig.6. The scale on the right side of Fig.6 gives directly $\langle r^2 \rangle$ as obtained with this Droplet normalization. Using the absolute muonic calibration of the "reference" isotope, the isotopic chains have to be shifted as indicated by the arrows and crosses. The values of $\delta\langle r^2 \rangle^{A,A'}$ with A' being the "reference" isotope are calculated by use of (4) and (5). In some cases, $c_A^{A,A'}$ is recalculated as detailed in the following:

Osmium: $\delta\langle r^2 \rangle^{A,A'}$ is taken from muonic x-ray data [50].

Platinum: $c_A^{136,198}$ is calculated by use of the Droplet model with a correction for deformation [46,47]. This procedure is justified because the Droplet model is known to describe well the $\delta\langle r^2 \rangle$ of the neutron-deficient isotopes in this region. Furthermore, the changes of the shape of the isotopes ^{196}Pt and ^{198}Pt are small. Hence, the c_A factors obtained in this way are expected to be reasonable and this can be checked for the cases of Pb, Hg, Au and Os. The experimental data are taken from ref. [24].

Gold: $c_A^{A,A'}$ is available from a recent Dirac-Fock calculation [26,49] as discussed in section 2. The experimental data are given in Tables 2 and 3.

Mercury: The plotted $\delta\langle r^2 \rangle^{A,A'}$ values are obtained with $c_A^{A,A'}$ obtained by a recent Dirac-Fock calculation [49]. The experimental data are taken from ref. [2-5,32,61].

Thallium: $c_A^{203,205}$ is calculated as described in the case of Pt. The experimental data are taken from ref. [24].

Lead: The $\delta\langle r^2 \rangle_{R,R'}$ data are taken from Thompson et al. [43] who determined $c_A^{R,R'}$ by means of a Kingplot of the optical data versus $\delta\langle r^2 \rangle$ values obtained by a recently published combined analysis of muonic x-ray and electron-scattering data [43].

The use of the Droplet model for calibration of absolute $\langle r^2 \rangle$ values has its limitations: the Droplet model shows deviations from the absolute experimental data just in the vicinity of magic numbers [46]. An analysis of the $\langle r^2 \rangle$ values in terms of isotonic shifts and their possible odd-even staggering is handicapped by these uncertainties and has to be waited for until all muonic data are reevaluated in a consistent manner. As a consequence, the following discussion is restricted to the relative IS of the individual elements.

With the exception of the nuclear shape transitions observed for the Hg isotopes below $N=106$, the nuclear charge radii shrink quite regularly with decreasing neutron number. A small kink of the average slopes around $N=118$ indicates a subshell closure or an onset of increasing or decreasing deformation at this neutron number. A pronounced odd-even staggering is superimposed on the general shrinking of the charge radii and the magnitude of the staggering effect is surprisingly independent on the proton number. Despite the uncertainties in c_A for the individual elements (estimated to less than 10%), a parallel course of the charge radii versus neutron number for different elements is interpreted as an identical change in nuclear shape for these elements. This can be seen by the simple two-parameter model

$$\delta\langle r^2 \rangle^{A,A'} = \delta\langle r^2 \rangle_{\text{volume}}^{A,A'} + \delta\langle r^2 \rangle_{\text{shape}}^{A,A'} \quad (11)$$

where the first part accounts for the change of the nuclear charge radius of the hypothetically spherical nucleus and the second part is due to a change in nuclear shape. The volume contribution is known to be well described by the Droplet model in the case of heavy nuclei [46].

Since $\delta\langle r^2 \rangle_{\text{DROPLET}}$ changes only by 1% between Pb and Os for isotope pairs with the same neutron number, the shape-dependent part of (11) is responsible for different average slopes of the individual isotopic chains. The prediction of the spherical Droplet model is plotted in Fig.6 for the case of Au. It is obvious, that the Au isotopes are shrinking less with decreasing neutron number than expected by a pure volume effect described by the Droplet model. In the frame of the two-parameter model (11), an increase of $\delta\langle r^2 \rangle_{\text{SHAPE}}$ is observed which is due to increasing static or dynamic nuclear deformation or increasing diffuseness of the nuclear surface. In heavier nuclei, the dominant contribution to the change of $\delta\langle r^2 \rangle_{\text{SHAPE}}$ stems from a change in static or dynamic quadrupole deformation $\langle \beta^2 \rangle$. Considering only these effects, the second term of (11) reduces to [56]

$$\delta\langle r_{\text{shape}}^2 \rangle^{A,A'} \approx \delta\langle r_{\beta}^2 \rangle^{A,A'} = 3/(4\pi) R_0^2 \delta\langle \beta^2 \rangle^{A,A'} \quad (12)$$

with $R_0 = 1.2 A^{1/3}$ fm and the quadrupole parameter β . If $\delta\langle r^2 \rangle_{\text{VOLUME}}$ is identified with $\delta\langle r^2 \rangle_{\text{DROPLET}}$ calculated with the help of the spherical Droplet model, this leads to

$$\delta\langle r^2 \rangle^{A,A'} = \delta\langle r_{\text{Droplet}}^2 \rangle^{A,A'} + 3/(4\pi) R_0^2 \delta\langle \beta^2 \rangle^{A,A'} \quad (13)$$

and allows a determination of the change of the quadrupole deformation along an isotopic chain relative to a reference isotope as, for example, the only stable isotope ^{197}Au . These data are given in the second column of Table 3.

For the calculation of absolute deformation parameters (third column), the theoretical value of $\beta_{\text{theor.}}^{197} = 0.113$ is used as calculated by Nix and Möller [47,48]. This value is in reasonable agreement with $\beta_{Q_s}^{197} = 0.102$ calculated from the spectroscopic quadrupole moment of the $^2P_{3/2}$ atomic state of ^{197}Au [13] under the assumption of strong coupling. It is also consistent with the deformation parameters of the neighbouring nuclei ^{198}Hg ($\beta = 0.107(3)$) and ^{198}Pt ($\beta = 0.110(3)$) calculated from $B(E2)$ values [57,58]. In addition, calculations of level schemes of decoupled bands and of transition rates support our choice of the calibration of the deformation parameter. Such calculations have been performed by Meyer-ter-Vehn [22,59] as well as by Toki and Faessler [60] in the frame of the particle-plus-rotor model, leading to $\beta = 0.14$ for the $h_{M/2}$ proton state in ^{195}Au . The trends of the deformation parameters of Au are consistent with the expectation that the deformation increases with the distance from the magic proton number $Z=82$. No drastic effect is observed down to ^{190}Au , the lightest Au isotope optically investigated. The data available do not allow any conclusion to be drawn as to whether the strong deformation expected around $N \simeq 105$ is reached in a smooth way or by a sudden onset of deformation caused by the nearby magic proton number. Recent investigations of ^{186}Au by nuclear spectroscopy indicate a similar shape coexistence in this nucleus [62] as found for ^{185}Hg [5]. Hence, a sudden change of the nuclear shape is also expected in the very neutron-deficient Au isotopes. The optical measurement of this effect via a determination of the IS or quadrupole moment would require spectroscopic methods of considerably improved sensitivity.

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wavelength	535.2 nm	267.6 nm

pulse energy	2 mJ	200 μ J
pulse length	5 ns	3 ns
bandwidth	200 MHz	420 MHz
repetition rate	10 Hz	10 Hz

Table 1:

Typical values of the laser output

Table 2:

Compilation of the half life, nuclear spin, HFS splitting in the $6s\ ^3S_{1/2}$ state, isotope shift in the D1-line and radial change of the Au isotopes investigated. The errors cited for λ and $\delta\langle r^2 \rangle$ are only due to the experimental errors of the IS and the uncertainty of the specific mass shift. The numbers have an additional common scaling uncertainty of 10% due to the estimated uncertainty of c_1 .

a) : [23], b) : [39], c) : [40], d) : [41], e) : [40],
f) : [42], g) : [1],

\$) the sign was determined in this work

*) the values of the magnetic moments are given without corrections for diamagnetism

A	$\delta \langle \beta^2 \rangle^{A, 197}$	$\langle \beta^2 \rangle^A$
197	-	-0.113 *)
195	-0.0029(5)	-0.125(2)
193	-0.0055(3)	-0.135(1)
192	-0.0059(4)	-0.137(2)
191	-0.0077(4)	-0.143(1)
190	-0.0093(10)	-0.148(3)

Table 3:

Changes of mean-squared and absolute rms quadrupole deformation parameters of Au isotopes

*) Taken from the calculation of Nix and Möller [47]

FIGURE CAPTION

Figure 1:

Atomic level diagram of Au. The D1-resonance transition ($6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$) at $\lambda = 267.6\text{ nm}$ was used for fluorescence spectroscopy. The hyperfine structure splittings are indicated for the case of stable ^{197}Au ($I=3/2$).

Figure 2:

Set-up for semi on-line resonance fluorescence spectroscopy of Au isotopes. The output of a dye laser is amplified in 3 amplifier stages (ampl.) and frequency doubled. The radioactive Au isotope is confined in a sapphire cell and heated to temperatures above 1300°C . The resonance fluorescence is detected by a photomultiplier (PM). An atomic beam of stable ^{197}Au , absorption of molecular ^{130}Te , and a Fabry-Perot etalon serve for wavelength calibration.

M = mirror, S = beam splitter, D = photodiode, MO = monitor for laser intensity, ADC = analog-to-digital converter.

Figure 3:

Schematic outline of the cell-oven system.

Figure 4:

Simultaneously recorded signals during a single laser scan of about 20 GHz corresponding to 40GHz in the frequency doubled spectral range. The scan duration is about 30s. The signals are (from the top to the bottom): Resonance fluorescence signal from 10^{13} atoms of ^{192}Au and an equal amount of ^{197}Au confined in the sapphire cell at 1350°C ; Resonance fluorescence of stable ^{197}Au in an atomic beam (the small hyperfine structure splitting in the excited state is partially resolved); Laser intensity in the UV spectral region; $(^{130}\text{Te})_2$ absorption signal; Transmission curves of a Fabry-Perot etalon (FSR =6 GHz).

Figure 5:

Resonance fluorescence signal of Au isotopes recorded in a single laser scan. Top: Signal of stable ^{197}Au in an atomic beam. Other: Signal of neutron-deficient isotopes. In each case, about 10^{13} atoms were confined in the sapphire cell at temperatures above 1300°C . The solid lines represent the result of a least-square fit. The F quantum number of the atomic ground state and the center of gravity are indicated. In the case of ^{192}Au and ^{190}Au , stable Au was put into the resonance vessel in order to passivate the walls of the cell.

Figure 6:

Mean squared charge radii in the Pb to Os region. For details see text.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6