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RESONANCE IONIZATION MASS SPECTROSCOPY FOR
NUCLEAR RESEARCH AND TRACE ANALYSIS

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Abstract: Resonance ionization mass spectroscopy (RIMS) has been applied, firstly, to a study of the nuclear properties of short-lived Au isotopes at the on-line isotope separator ISOLDE at CERN and, secondly, to a trace-analysis experiment of Pu.

1. INTRODUCTION

Recent advances in laser technology and experimental techniques led to a variety of spectroscopic schemes which allow the study of optical resonances with high sensitivity, selectivity and resolution. This report concentrates on the resonance ionization spectroscopy (RIS) which was pioneered by Letokhov [1] and Hurst [2] and is described in detail in recent reviews and conference proceedings [3]. The selectivity of this technique can be increased by mass spectroscopy of the ions created in the optical resonance. This so-called resonance ionization mass spectroscopy (RIMS) offers a great potential as a general tool for optical spectroscopy and has a variety of important specific applications such as isotope separation and trace analysis.

Two recent experiments will be described in this contribution: the first one concerns the study of the nuclear properties of unstable Au isotopes; the other one is a trace-analysis experiment on plutonium. Both are based on the RIMS technique. The ISOLDE experiment at CERN was performed by an Atlanta-CERN-Mainz-Madrid Collaboration, composed of G. Bollen, M.J.G. Borge, J. Campos, A. Dohn, P. Egelhof, J. Grüner, H.-J. Kluge, U. Krönert, F. Lindenlauf, R.B. Moore, A. Rodriguez Yunta, A. Venugopalan, K. Wallmeroth and J.L. Wood. More detailed results of this experiment will be published elsewhere [4]. The trace-analysis experiment was performed by a collaboration between the Institut für Physik, Mainz, and the Institut für Kernchemie, Mainz, composed of F. Ames, J. Bonn, G. Herrmann, H.-J. Kluge, U. Krönert, E.W. Otten, P. Peuser, H. Rimke, W. Ruster, P. Sattelberger, N. Trautmann and K. Wallmeroth [5-7].

2. THE MOTIVATION TO STUDY SHORT-LIVED Au ISOTOPES

The nuclei near $Z = 80$ and $N = 104$ have attracted considerable attention following the discovery by Bonn et al. [8,9] that there is a big change in the ground-state mean-square charge radius between ^{187}Hg and ^{185}Hg , which was interpreted as an onset of strong deformation at $A \leq 185$. This observation, 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 with $Z = 80$, 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 nuclear shapes occur widely throughout the region below $Z = 80$.

A measurement of the isotope and/or isomer shift represents the most straightforward and model-independent signature for shape transitions or shape coexistence. In recent experiments, the ISs in the isotopic chains of Tl [10], and Au [11] have been determined. No indication of a shape transition or coexistence was observed because the experiments were restricted to the study of heavier isotopes ($N \geq 108$), whereas the shape effects are expected to occur in nuclei with a nearly half-filled neutron shell.

3. EXPERIMENTAL TECHNIQUE AND PERFORMANCE OF THE Au EXPERIMENT

The experiment was designed to investigate shorter-lived Au isotopes and to determine their ISs. As a by-product the magnetic hyperfine structure (HFS) splitting is obtained. The Au isotopes were produced at the ISOLDE mass separator at CERN. Since no target and ion-source system exists for a direct production of Au isotopes, these nuclei have at present to be obtained as daughters of Hg isotopes, which can be produced with yields up to 10^{10} atoms per second and mass number by a $\text{Pb}(p,3pxn)\text{Hg}$ spallation reaction [12]. Many different techniques were tested [13]. Finally, the RIS technique was chosen and combined with time-of-flight (TOF) mass spectroscopy. Because of the short half-lives and the small production rates of the Au isotopes far from stability, the investigations had to be performed on line with the ISOLDE facility. Recently the first application of RIS to a study of short-lived isotopes was reported [14-16]. This contribution reports the first application of RIMS to an on-line study of short-lived isotopes.

Figure 1 shows the experimental set-up. The ISOLDE beam is focused into an atomic beam oven. After a suitable time has elapsed, during which the Hg nuclei decay to the daughter isotope, a thermal atomic beam is formed by evaporating the radioactive sample. The Au atoms are excited and photo-ionized via a three-step resonant process: by the light from two tuneable dye lasers pumped by a Nd: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 indicated in fig. 2.

In order to increase the signal-to-background ratio, the photo-ions are detected and mass-selected by a TOF spectrometer. It consists of a two-stage acceleration region, a field-free drift tube, and a microchannel plate (MCP) detector. The spectrometer is of the same type as the one described in detail by Krönert et al. [5]. Figure 3 shows the HFS pattern and fig. 4 is the TOF spectrum in the case of the measurement of ^{186}Au . A background rate as low as 1 event per 1000 laser shots was obtained with a mass resolution of the TOF set-up of 250 and a time window of 70 ns for gating on the mass of the isotope under investigation.

The overall detection efficiency ϵ of the RIMS technique for Au was determined to be $\epsilon = 10^{-8}$ (defined as the ratio of Au ions detected by the MCP to the number of atoms collected in the oven). The efficiency is mainly limited by the low repetition rate of the laser system and the small cross-section of the third excitation step, which cannot be saturated even with laser powers up to 5 MW/cm^2 .

4. RESULTS FOR THE SHORT-LIVED Au ISOTOPES

Measurements were performed on Au isotopes in the mass range $190 \geq A \geq 185$. The shortest half-life of the isotopes investigated was $T_{1/2} = 4.2$ min (^{185}Au) where $\sim 10^9$ atoms could be collected in the oven. Similar spectra, as shown in fig. 3 for the case of ^{188}Au , were obtained for the low-intensity cases ^{185}Au , ^{186}Au and the isomeric state of ^{189}Au . For the more abundant isotopes $^{188-190}\text{Au}$ much better signal-to-noise ratios were obtained.

Nuclear magnetic moments and changes of mean-square charge radii ($\delta\langle r^2 \rangle$) were deduced from the HFS splitting and the IS in the D_1 line. The $\delta\langle r^2 \rangle$ values are plotted in fig. 5 relative to ^{197}Au . In addition, the $\delta\langle r^2 \rangle$ values of Hg are shown [17]. The slope of the mean-square charge radii is very monotonic and is similar to those of Hg down to ^{187}Au . Then, between ^{187}Au and ^{186}Au , a drastic change occurs, similar to the one in the Hg isotopic chain except that, firstly, it takes place two neutron numbers earlier and, secondly, there is a much less pronounced odd-even staggering than for the Hg isotopes.

As in the case of Hg, the break in the IS can be related to a change in the nuclear quadrupole deformation β . These results confirm earlier suggestions [18,19] that the ground states of these nuclei might be strongly deformed. Clear evidence for a shape transition from weak ($A \geq 188$) to strong ($A < 187$) deformation in the neutron-deficient Au isotopes has now been observed by optical spectroscopy, providing a sensitive test of nuclear model calculations. More details can be found elsewhere [4].

5. THE MOTIVATION FOR A TRACE-ANALYSIS EXPERIMENT ON Pu

Plutonium is one of the most serious radioactive poisons in the environment. Almost all of it is made by mankind and stems from nuclear bomb tests, accidents, and nuclear power plants. Very sensitive and selective methods are required to study the ecological behaviour and migration in the environment. The detection technique actually used is α spectroscopy, which has, however, some severe drawbacks: the sensitivity is limited (4×10^8 atoms for ^{239}Pu), the element and isotope selectivity is restricted (other isotopes emit α particles with similar energies), the measurements take a long time, and ^{241}Pu can not be detected at all because of the absence of α emission.

An alternative, promising, detection scheme is RIMS. Its detection limit does not depend on the nuclear properties of the isotope under investigation such as the half-life and decay mode. It takes advantage of the large cross-section for the absorption of an optical photon. The first optical Pu experiments for trace analysis were performed by Donohue et al. [20,21]. At Mainz, very similar experiments were performed by Krönert et al. [5], the main difference being the use of other optical transitions and the use of a magnetic or a TOF mass spectrometer. In all these experiments, however, the detection limit was of the order of only 10^{11} Pu atoms. This was due, firstly, to the low repetition rate of the pump laser used (1 - 30 Hz) and, secondly, to the use of a single-colour excitation and ionization. Clearly, a much higher sensitivity and also an increased selectivity can be achieved by applying a high-repetition laser and resonant excitation in each step. Since a thermal ion traverses the region of interaction with the laser

light within $\sim 10^{-4}$ s (if this interaction region has a diameter of 1 cm) a repetition rate of the pump laser as high as 10 kHz is needed to reach a duty cycle of about unity. The only laser available with such a high rate is the Cu-vapour laser. This laser delivers laser light at two wavelengths, namely 510.6 nm and 578.2 nm, with about equal intensity. These beams can be used to pump tuneable dye lasers. Hence tuneable laser light is available (without frequency doubling) in the visible region with wavelengths above 530 nm.

An excitation scheme for Pu is shown in fig. 6. The two first steps are resonant, the last one excites the Pu atom into the continuum or an auto-ionizing state. The cross-sections for photon absorption are high (10^{-10} - 10^{-12} cm²) for the first two excitations. Hence, it is easy to saturate these transitions even with moderate laser power. The limiting transition is usually the last one, which can either lead into the continuum ($\leq 10^{-17}$ cm²) or to auto-ionizing or Rydberg states ($\sim 10^{-15}$ cm²).

6. THE EXPERIMENTAL SET-UP FOR Pu TRACE ANALYSIS

Figure 7 shows the experimental set-up. One or two Cu-vapour lasers (Oxford Lasers, Mod. CU25 and CU40) pump two or three dye lasers (Lambda Physics, Mod. 2001E) with a maximum average power of 20 and 30 W at a repetition rate of 6.5 kHz and a pulse length of 30 ns. The conversion efficiency is as high as 10-25% and the bandwidth of the dye laser is about 6 GHz. The three laser beams needed for ionization are coupled into an optical fibre to assure an optimum spatial overlap when interacting with the atoms. As in the Au experiment, this interaction takes place in the acceleration region of a TOF spectrometer [5,6]. The mass resolution is 500 and goes up to 2,500 in the case where a HV pulse is applied to the grids in the acceleration region of the TOF spectrometer after laser excitation and ionization.

The detection limit is very much affected by the efficiency with which the Pu sample is converted into free thermal atoms taking part in the RIMS process. For this, the Pu sample is electrolytically deposited on a Re filament [22] and covered again with Re in order to obtain a slow evaporation in a neutral, non-molecular form at filament temperatures of about 1800 K.

7. RESULTS OF THE TRACE-ANALYSIS EXPERIMENT

Plutonium samples containing between 10^{10} and 10^{12} atoms of ^{239}Pu were measured by scanning the dye lasers for the first two steps over wavelength intervals of 0.05 nm while the third step was made with part of the yellow pump beam ($\lambda = 578.2$ nm). Strong resonance signals were found for plutonium (fig. 8) at an extremely low background. The laser powers available for the three steps were 780 mW, 320 mW, and 3200 mW, respectively.

A TOF mass spectrum from a sample which contained about 10^{11} atoms of ^{239}Pu is shown in fig. 9. This spectrum was recorded for 60 s with the TOF spectrometer having a drift length of 1.0 m when the wavelengths of the two dye lasers were in resonance. From these experiments an overall detection efficiency of 10^{-7} has been determined, corresponding to a detection limit of about 10^8 plutonium atoms in the sample. This value includes the losses in generating atoms from the sample, the ground-state population, the solid angle, unsaturated excitation and ionization, the transmission of the two acceleration grids, and the ion counting efficiency of the MCP detector.

In test experiments with gadolinium, when photo-ionization by a three-resonant step was performed via auto-ionizing states, saturation could be achieved for all three excitation steps with the laser powers available yielding an overall detection efficiency of 10^{-5} (fig. 10). In a very recent experiment on Pu the third dye laser was scanned while the wavelengths of the first and second dye laser were in resonance. Two Cu-vapour lasers

were used to pump the three dye lasers. An overall detection efficiency of 10^{-6} was obtained with diameters of the laser beam of 2 mm in the interaction region and the third dye laser tuned to a strong transition to an auto-ionizing state. Hence, a detection limit of 10^7 Pu atoms is obtained, 40 times more sensitive than α spectroscopy.

8. FUTURE DEVELOPMENTS

As can be seen from table 1, real one-atom detection is still far in the future. For the time being there is no chance of introducing one atom into an apparatus and detecting it there with 100% probability. The detection efficiencies of the present experiments are given in the 7th column of table 1. They range from 10^{-11} to 10^{-5} . Only in the case of noble gases does the efficiency reach higher values, because these atoms are not absorbed on the walls of the vacuum container and are for RIS available in a sealed-off resonance vessel until they are photo-ionized [3]. The detection efficiencies achieved (table 1) reflect, of course, the different repetition rates of the pulsed laser system used, as well as the excitation scheme and the available laser power, especially for the last crucial step.

There is still much room for improvements. For example, in case of the measurements of short-lived Au isotopes, the drawback of the 10 Hz repetition rate of the Nd:yag laser can be compensated by producing a pulsed atomic beam in order to obtain a better use of the available number of atoms. In a recent laser desorption experiment a second Nd:yag laser was

used to heat the foil containing a Au sample [23]. A 30 times higher overall detection efficiency was observed (table 1) when surface-adsorbed Au atoms were used. In a further investigation, an ion beam of radioactive ^{195}Au was implanted into a graphite foil. About 40% of the implanted radioactivity was found on a catcher foil after laser desorption. Still it has to be proved in what form the radioactivity is released: as neutrals atoms, as needed for RIMS, or, unusable for RIMS: as ions or molecules; or even as Au nuclei embedded in graphite clusters? The fraction of atomic evaporation can be determined by the RIMS technique as discussed above.

In the case of experiments using Cu-vapour lasers the main losses in detection efficiency are due to the distance between the filament which evaporates the Pu atoms into 2π and the laser-atom interaction region. It is difficult to diminish this distance because a very homogeneous electric field is needed for a good time resolution in between the acceleration electrodes of the TOF spectrometer.

A completely different approach is to confine the atoms under investigation in a container and to extract the ions when they are ionized by the RIS process. Such a high-efficiency device has first been proposed by Letokhov et al. [24] and in a modified version by Kluge et al. [25]. First results using a laser-ion source for Fr were presented by Mishin during this workshop [16]. Our apparatus, which has now just been built at Mainz, is shown in fig. 11. The ion source consists of a cylinder with isolated end plates, a hole as entrance opening for the laser beams and for extraction of the ions, and a surrounding oven for keeping the atoms under investigation in the vapour phase. In the case of a cubic ionization

chamber with length L and an extraction hole of radius r , and neglecting surface ionization, the rate of escape as neutral atoms is proportional to the number of atoms N in the chamber, the bouncing rate of the atoms against the walls, and the probability of finding the extraction hole. The rate of the wall collisions is given by $v/\lambda \approx v/L$, where λ is the mean free path in the ionization chamber, and v the mean velocity. The probability of escaping is determined by the ratio of the area of the hole and the total surface area of the ionization chamber $\pi r^2/6L^2$. Hence this escape rate is given by

$$R_{\text{escape}} = (Nv/L) \times (\pi r^2/6L^2) \quad . \quad (1)$$

The rate of the photo-ionized ions is determined again by the number N of atoms in the chamber, the repetition rate of the laser v_{rep} , the ionization efficiency η , and by the ratio of the laser interaction volume $\pi r^2 L$ and the total volume L^3 as

$$R_{\text{photo-ion}} = N v_{\text{rep}} \eta \pi r^2 / L^2 \quad . \quad (2)$$

Hence the ratio of the rates reduces to the simple expression

$$R_{\text{photo-ion}}/R_{\text{escape}} = (v_{\text{rep}} \eta) / (v/6L) \quad , \quad (3)$$

which no longer depends on the radius of the extraction hole. With $\eta \approx 1$ as achieved for the case of Gd (see fig. 10) the photo-ionization efficiency is given by

$$\epsilon = R_{\text{photo-ion}} / (R_{\text{escape}} + R_{\text{photo-ion}}) = 6v_{\text{rep}} / (v/L + 6v_{\text{rep}}) \quad , \quad (4)$$

which reaches values as high as 26% for $\nu_{\text{rep}} = 6$ kHz, $L = 1$ cm, and $v = 10^5$ cm/s.

Such a laser-ion source allows an increase of the detection efficiency by several orders of magnitude as already demonstrated [16]. Furthermore, it can be used to produce ion beams of high isotopic purity as needed, for example, in accelerator mass spectroscopy. It will be ideal for on-line mass separators where isobaric contamination is sometimes a big problem for special target-ion source combinations. As an example, fig. 12 shows the yield of a Ta foil target irradiated with 1 μ A of 600 MeV protons. More or less all alkaline earth nuclei are produced by a spallation reaction and are ionized by a W ionizer. Hence this target is almost unusable for nuclear spectroscopy. With a laser-ion source instead of the conventional W surface ion source, ion beams could be obtained free of isobaric contamination. Hence the combination of a Ta foil target with a laser-ion source tuned to the excitation steps of an individual rare earth element would yield quite a number of highly-efficient and very selective ion-beam sources.

For many experiments, the pulse structure of the laser-generated ion beams (6 kHz repetition rate, 30 ns pulse width) will be advantageous. If, for example, nuclear spectroscopy or collinear laser experiments [26-28] are background limited, the signal-to-background ratio is enhanced by a factor of almost 10^4 , the inverse of the duty cycle of the pulsed laser system. The pulse structure will also allow very efficient (almost 100%) in-flight capture of ions into a Penning trap [29]. This is essential for mass measurements of short-lived isotopes [30] or measurements of the hyperfine anomalies [31] of isotopes of elements which are not surface-ionizable.

Finally, an ultra-sensitive and ultra-selective detection scheme should be discussed, a combination of a laser-ion source and a Penning trap. Figure 13 shows the proposed set-up. A sample is introduced into a laser-ion source as discussed above. Only the ions with the correct mass pass a kicker device and are captured in the Penning trap which is opened at the right TOF. All still remaining contaminating ions are driven to the electrodes by applying a broad-band RF excitation with the exception of the frequency of the cyclotron resonance of the mass under investigation. In this way extreme selectivity and high sensitivity would be achieved. The numbers are given in fig. 13. An efficiency in the percent region seems quite realistic, at least for elements with simple level schemes, transitions in the visible part of the spectrum, and low chemical reactivity. However, it is quite unsafe if the selectivity in each step can be simply multiplied.

9. CONCLUSION

Resonance ionization mass spectroscopy will, in the future, play a major role in the determination of nuclear properties of unstable nuclei, in isotope separation, and in trace analysis. We have applied RIMS to determine the isotope shift and the HFS of short-lived Au isotopes. A nuclear shape transition was found which takes place between ^{187}Au and ^{186}Au . The detection efficiency of 10^{-8} was enhanced by using pulsed laser desorption. A dye-laser system pumped by Cu-vapour lasers was used for a trace-analysis experiment on plutonium. A detection limit of 10^7 Pu atoms was deter-

mined. Hence this method is 40 times more sensitive than the conventionally used α spectroscopy. RIMS has proved to be a very sensitive and selective technique for the investigation of small samples. Important improvements of the this technique still seem feasible.

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TABLE 1. Detection efficiencies of RIMS in different experiments.^{a)}

INVEST- IGATED ELEMENT	EXCI- TATION SCHEME	SOURCE	LASER SYSTEM [Hz]	REPETITION RATE BEAMS	DIAMETER OF LASER [cm]	EFFI- CIENCY
Pu	1-colour/ 2-photon/ 1-step resonant	Filament	Nd:yag pumped DL	10	0.01	5×10^{-11}
Pu	3-colour/ 2-step resonant	Filament	CVL pumped DLs	6×10^3	0.1	10^{-7}
Pu	3-colour/ 3-step resonant	Filament	DLs pumped by 2 CVLs	6×10^3	0.2	10^{-6}
Gd	3-colour/ 3-step resonant	Oven	CVL pumped DLs	6×10^3	1.0	10^{-5}
Au	3-colour/ 2-step resonant	Oven	Nd:yag pumped DLs	10	0.3	10^{-8}
Au	3-colour/ 2-step resonant	Oven/ pulsed beam	Nd:yag pumped DLs	10	0.3	3×10^{-7}

a) The efficiency is defined as the ratio of ions detected by the MCP detector in the appropriate window of the TOF spectrum to the number of atoms put onto the filament or into the oven. CVL = Cu-vapour laser, DL = dye laser.

Figure captions

Fig. 1. Set-up for on-line RIMS on short-lived Au isotopes.

Fig. 2. Atomic level scheme of Au. The excitations used for RIS are indicated by arrows.

Fig. 3. Simultaneously-recorded frequency-dependent signals i) of the reference atomic beam containing ^{197}Au , ii) of ^{186}Au in the RIMS spectrometer recorded by an analog-to-digital converter, iii) as before but recorded by single-event counting, and iv) of the reference absorption spectrum of molecular iodine (from top to bottom).

Fig. 4. Time-of-flight mass spectrum of ^{186}Au . Stable ^{197}Au was added as mass marker. The RIMS signal shown in fig. 3 was obtained by setting a 70 ns gate on the ^{186}Au mass peak.

Fig. 5. Changes of the mean-square charge radii of Au (dots) and of Hg (squares [17]) isotopes. ^{198}Hg and ^{197}Au are used as reference isotopes.

Fig. 6. Excitation scheme for RIS of Pu.

Fig. 7. Experimental set-up for RIMS of Pu with a dye laser system pumped by a Cu-vapour laser.

Fig. 8. Optical resonance of Pu measured by scanning the dye lasers for the first two excitation steps while λ^- is fixed.

Fig. 9. Time-of-flight mass spectrum of ^{239}Pu with λ_1 and λ_2 in resonance. No molecular Pu is observed.

Fig. 10. Excitation scheme (right) and RIMS signals for gadolinium as a function of the power of the laser light used for the different excitation steps (left). All transitions can be saturated with the available laser power of the Cu-vapour/dye-laser system.

Fig. 11. Layout of the laser-ion source.

Fig. 12. Yield of the Ta foil target with W surface ionizer as used at the on-line isotope separator ISOLDE at CERN. Owing to the similar chemical properties, a large number of alkaline earth isotopes are produced simultaneously by a 600 MeV, 1 μA proton beam.

Fig. 13. Combination of laser-ion source, TOF spectrometer, and Penning trap-mass spectrometer for ultra-selective and ultra-sensitive detection.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

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Fig. 8

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Fig. 13