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ON-LINE LASER SPECTROSCOPY BY RESONANCE IONIZATION OF LASER- DESORBED, REFRACTORY ELEMENTS⁺

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Abstract: Resonance ionization mass spectroscopy was applied at the on-line isotope separator ISOLDE-3/CERN, Geneva to the refractory elements Au and Pt. The hyperfine structures and isotope shifts of gold and platinum isotopes were studied in long isotopic chains in the range from the stable isotopes down to the neutron-deficient ones with $A = 183$. Spins, magnetic moments, quadrupole moments and the differences in mean-square nuclear charge radii were determined. A mass-selected Hg ion beam was implanted into a graphite target. After the decay to Au or Pt, a pulsed atomic beam was formed by pulsed laser desorption. The atoms were ionized by three-colour, three-step resonant excitation. The photo ions were detected mass-selectively by a time-of-flight mass spectrometer. An overall efficiency of $\epsilon \simeq 10^{-5}$ was achieved defined as the number of atoms detected in the optical resonance to the number of Hg atoms implanted into the target. The background was as low as 1 event per 1000 laser shots. The publication gives a detailed description of the applied method, the experimental set-up and its performance.

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1 Introduction

In 1972 the first systematic investigation of a long isotopic series by atomic spectroscopy led to a completely unexpected result: Between ^{185}Hg and ^{187}Hg a sudden change in the isotope shift (IS) and hence in the mean-square nuclear charge radius was observed corresponding to the addition of eleven neutrons [1]. Such a drastic break in the development of the charge radius was never observed before and only later explained as due to a sharp nuclear-shape transition [2, 3, 4, 5, 6, 7]: The Hg nuclei with $A \geq 187$ are slightly oblate-deformed whereas the nuclei ^{181}Hg , ^{183}Hg , and ^{185}Hg are strongly prolate-deformed with a deformation parameter of $\beta \geq +0.25$. The reason for this shape transition was found, (i) in the strong polarizing power of the $\nu^{1/2}$ [521]-Nilsson state in the mid neutron shell which steeply slopes down with increasing prolate deformation, and (ii) in the nearly magic ($Z = 80$) proton configuration which favours almost spherical shape.

This discovery offered the key for understanding the complicated rotational band structures observed in the neighbouring even-even Hg nuclei by γ -spectroscopy [8, 9, 10]: A new phenomenon in nuclear physics, namely nuclear-shape coexistence leads to two bands, one built on the slightly deformed oblate nuclear ground state, the other one on a strongly deformed state above the ground state. Reviews on shape coexistence have been published recently [11, 12]. These even-even isotopes were not accessible by nuclear radiation detected optical pumping (β - or γ -RADOP [13, 14]) used for investigation of the odd Hg isotopes. Only with the availability of tunable dye lasers the IS's of the even-even Hg isotopes could be determined down to ^{184}Hg [15] and those of the $I = 13/2$ isomers including their hyperfine structure (HFS) down to ^{185m}Hg [16]. These experiments were performed by resonance fluorescence and quantum beat spectroscopy [17]. Again a surprise came up: In addition to the prolate-deformed nuclear ground state ($I = 1/2$) observed by β -RADOP, an $I = 13/2$ state was found which is only slightly deformed. Since both states are nearly degenerated in energy, the ^{185}Hg isotope is a textbook nucleus for shape coexistence.

Later on, also the IS of ^{182}Hg could be measured, this time by collinear spectroscopy [18]. It turned out that also this lightest even-even Hg isotope investigated optically is slightly oblate deformed in contrast to its odd neighbours. Hence, a drastic odd-even staggering of the IS's, of the charge radii or the deformation is observed below $A = 186$ which cannot be found of an only approximately similar size and sharpness in any known region of the chart of nuclei [19].

This stimulated the investigation of the isotopes in the same mass region ($A \simeq 185$) of the neighbouring elements gold and platinum. Both are refractory elements which are not or only very recently (Au) available at the ISOLDE facility/CERN, Geneva as on-line mass-separated ion beams. For this reason, collinear laser spectroscopy could not be used. New spectroscopic methods had to be applied. Non-linear techniques like polarization [20] or two-photon spectroscopy [21] were shown to fail in test experiments due to their limited sensitivity. Also fluorescence spectroscopy in a high-temperature cell [22, 23] suffered from the strong background light scattered off the walls of a sapphire cell containing the Au sample. Nevertheless, this method allowed to study the HFS and IS of the Au isotopes in the mass region $197 \geq A \geq 190$ [24].

A breakthrough was achieved by the application of resonance ionization mass spectro-

scopy (RIMS) which allowed the investigation of the Au isotopes down to ^{185}Au [25, 26]. In this experiment, the mass-separated Hg ion beam was collected in an oven. After the decay to the corresponding Au daughter isotope, the oven was heated to produce a thermal atomic Au beam which was interrogated by the light of two dye lasers pumped by a Nd:YAG laser. In resonance, the Au atoms were stepwise excited and finally ionized. The photo-ions created in resonance were detected mass-selectively by use of a time-of-flight (TOF) spectrometer. Due to the pulse repetition rate of the laser ($\nu_{rep} = 10\text{ Hz}$) the efficiency was limited: Only one Au photo ion was detected in resonance for 10^8 Hg ions implanted in the oven.

An increase in sensitivity by three orders of magnitude was achieved by the production of a pulsed atomic beam instead of a continuous one as before which is synchronized with the dye lasers pulses used for resonance ionization spectroscopy (RIS). Such a pulsed atomic beam can be obtained by pulsed-laser induced desorption (PLID) [27, 28].

The RIMS/PLID technique enabled the optical investigation of two more neutron-deficient isotopes, ^{184}Au ($T_{1/2} = 53\text{ s}$) and ^{183}Au ($T_{1/2} = 42\text{ s}$) [29], and the determination of the nuclear properties of the Pt isotopes in the mass range $198 \geq A \geq 183$ [30, 31, 32].

Fig. 1 shows the compiled results of all the efforts described above for the case of the changes of mean-square charge radii $\delta \langle r^2 \rangle^{AA'}$ in the Hg-Au-Pt region. The Hg chain shows the discussed features of nuclear shape transition and coexistence. In Au, only one drastic change in deformation is observed due to the stabilizing power of the unpaired proton, and in the isotopic chain of Pt no indication of a sharp break is obtained. Nevertheless, an analysis of the IS of Pt shows that a similar deformation ($\beta = 0.24$) is observed for ^{183}Pt as in the isobaric and isotonic Au and Pt nuclei.

This publication describes the experimental set-up used for RIMS/PLID of short-lived Au and Pt isotopes and reports on its performance. Furthermore possible applications to the investigation of isotopes of refractory elements at on-line isotope separators will be briefly discussed as well as its use for trace analysis or as pulsed ion source for injection of isobaric-pure ion beams into storage devices like ion traps.

A similar set-up as described here was developed by an Orsay-Montreal collaboration and installed at the ISOCELE facility/Orsay [33, 34]. Measurements were performed on Au and Pt for isotopes with mass number $A \geq 185$ [35, 36].

2 Resonance Ionization Mass Spectroscopy (RIMS) of Au and Pt

The scheme of resonance ionization spectroscopy has been proposed and for the first time realized by Ambartzumian, Kalinin and Letokhov [37, 38] as early as 1971. In the meantime it has developed to be a powerful tool in many disciplines of physics and chemistry because of the availability of suited laser systems [39, 40, 41, 42, 43, 44]. It was first introduced to laser spectroscopy of radioactive isotopes at an on-line mass separator by Alkhazov et al. [45, 46].

In a RIS experiment, the atoms are stepwise excited and finally ionized by use of resonant laser light. The resonance condition for each excitation step ensures selectivity. The linear power dependence (until saturation sets in) and the detection of ions instead of the usually detected fluorescence photons result in high sensitivity. If a mass analyzer is added for the ions created in resonance (RIMS) the method is almost background-free.

Fig. 2 shows the excitation schemes used for RIMS of Au and Pt. Since the last excitation step above the ionization limit into the continuum has a very low cross section ($\sigma \simeq 10^{-17} - 10^{-19} \text{ cm}^2$) as compared to transitions between bound atomic states, this last step represents generally the bottleneck of the excitation path. A higher efficiency for photoionization can be obtained by exciting the atom to an autoionizing state. A careful search for such states in Au yielded an enhancement by a factor of 20 as compared to the non-resonant ionization into the continuum [28]. Such an enhancement is similar in magnitude to the loss due to the conversion efficiency of the light of the pump laser into tunable dye laser light. Nevertheless, excitation to an autoionizing state was used since less laser power had to be sent into the interaction region with the Au atoms resulting in a lower production of residual gas ions. In the case of Pt, two transitions to autoionizing states were found at $\lambda_3 = 602.7 \text{ nm}$ and $\lambda_3 = 593.1 \text{ nm}$ within the tuning range of the used dye ($580 \text{ nm} \leq \lambda_3 \leq 610 \text{ nm}$). However, the enhancement was only a factor of three as compared to non-resonant ionization. Therefore, the frequency-doubled output of a Nd:YAG laser was used for the last non-resonant step into the continuum.

3 Pulsed-laser induced desorption (PLID)

The interaction of laser light with surfaces of bulk material has attracted much attention during the last years because of its wide range for technical applications like hole drilling, cutting, welding, surface etching, ablation, annealing and so on. Especially the effects of short laser pulses are nowadays intensively studied due to the extreme temperature changes ($\geq 10^{11}$ K/s) which are achievable. PLID in combination with a TOF spectrometer is used for micro analysis of surfaces as the commercially available in the LAMMA 1000¹ instrument. Here, the PLID process is simultaneously used for creation of a micro plasma in order to ionize the sample atoms. In contrast, a beam of thermal, neutral atoms is required for RIS. This restricts the maximum applicable laser power density to about $10^7 - 10^8$ W/cm² as found experimentally for the case of a graphite surface.

In this case, the absorbed light from a short laser pulse (10 ns) leads to a fast heating process of the surface which is described by the equation of heat conduction. Carslaw and Jeager [47] have calculated the heating process. At the surface, during a laser pulse of rectangular shape, the temperature T increase is given by

$$T(t) = T_0 + \frac{2P_0}{K} \sqrt{\frac{\kappa t}{\pi}} \quad (1)$$

as a function of the time t . T_0 is the temperature of the surface before the laser pulse is fired at $t = 0$, P_0 is the energy density of the absorbed laser light, κ the thermal diffusivity and K the thermal conductivity.

The resulting variation of the temperature at the surface of graphite ($\kappa = 0.25$ cm²/s, $K = 0.5$ W cm⁻¹K⁻¹) is shown in fig. 3 simulated for a Gaussian laser pulse of $t_{1/2}$ (FWHM) = 10 ns and $P_0 = 200$ mJ/cm². A reflectivity of graphite of $R \simeq 0.35$ is assumed for a wavelength of $\lambda = 532$ nm of the desorption laser. A maximum temperature of $T = 1650$ K is obtained which depends linearly on the laser power (see eq. (1)) and is reached very fast within 13 ns after the switch-on of the laser pulse. The inset of fig. 3 shows the temperature variation as a function of the depth in the bulk material calculated with the same parameters as given above. Also indicated is the implantation depth of Hg ions of an energy of 60 keV as obtained at ISOLDE. In earlier experiments [25, 26] it was found that a temperature of $T \simeq 1600$ K is required for evaporation of Au implanted into an oven made of graphite. As seen from fig. 3 this temperature can well be reached by PLID.

The energy of atoms evaporated out of a matrix will follow a Boltzmann distribution. Assuming an activation energy E_a required for the diffusion in the bulk material to the surface one obtains an Arrhenius-like equation for the number of desorbed atoms N_{des} given by

$$\frac{N_{des}}{N_0} \sim e^{-E_a/kT} (1 + E_a/kT). \quad (2)$$

N_0 is the number of implanted atoms. Setting the activation energy to a reasonable value of $E_a = 1$ eV, one calculates for the temperature profile of fig. 3 the time dependence of the desorption yield as shown in fig. 4. Hence, a sharply defined desorption pulse ($t_{1/2}$ (FWHM) $\simeq 5$ ns) is obtained for a Gaussian laser pulse of $t_{1/2}$ (FWHM) = 10 ns.

¹LAMMA 1000 is a trade mark of Leybold.

4 The experimental set-up

Fig. 5 shows an overview of the apparatus used for RIMS/PLID of Au and Pt isotopes at the on-line isotope separator ISOLDE-3 [48] at CERN/Geneva. It consists of an implantation/desorption part where the mass-separated Hg^+ beam is implanted with an energy of 60 keV and, after the nuclear decay, a pulsed atomic beam of Au or Pt is produced by means of laser desorption, a TOF spectrometer for mass-selective detection of the ions created by RIS, the tunable pulsed dye laser system, and the data acquisition system. These parts will be described in the following. Their performance will be discussed in section 5.

4.1 Pulsed atomic beam source for refractory elements

The choice of the target material used for implantation is of crucial importance. It should have low reflectivity for efficient heating by the pulse of the desorption laser. In case of high reflectivity, higher desorption power is required leading to more background ions created by the reflected laser light at the inner surfaces of the vacuum chamber or even to a laser generated-plasma. Furthermore the target material should release the refractory elements Au and Pt at a temperature as low as possible and nearly quantitatively. Finally highest purity and a high melting point are required so that only the atoms under investigation are desorbed but not easily ionizable impurities or the target material itself. A series of test experiments with high-temperature materials (Ta, W, C, Ir, tungsten carbide (WC)) showed that high-grade pyrolytic graphite (Ringsdorf EK 986) is best suited for efficient desorption at a low rate of background ions [28]. Tests were performed with blank target materials as well as with implanted radioactive samples. Optimum power density of the desorption laser was determined to be about 20 MW/cm^2 . It should be mentioned that all materials investigated showed impurities of stable Au and Pt isotopes at the level of sensitivity achieved in this work. Hence, efficiency measurements of the RIMS/PLID technique should only be performed by use of radioactive isotopes.

A target wheel of cylindrical shape (diameter: $d = 40 \text{ mm}$, length: $l = 20 \text{ mm}$) was machined from the high-grade graphite. This wheel was mounted turnable (fig. 6) on insulated supports in order to enable a measurement of the current of the 60 keV ion beam. For proper alignment of this beam, an insulated diaphragm was mounted in front of the wheel with a hole size of $3 \text{ mm} \times 5 \text{ mm}$, adapted to the ISOLDE beam focus. The wheel could be turned by a stepping motor and its position was controlled by an incremental angle decoder. An accuracy and reproducibility of 0.5° could be obtained for the angular position of the wheel corresponding to 0.2 mm at its circumference. Before a measurement was started, the wheel was continuously turned, and the desorption laser operated at about three times higher power density as used in the RIMS experiment. A low background was obtained in this way during the actual experiment.

Two modes of operation were used during the measurements: In case of sufficiently abundant isotopes (e.g. $A \geq 184$ for Au) the wheel was turned continuously. The time for a turn by 180° was adjusted to the half-lives of the mother and daughter nuclei. Such a continuous mode ensures constant density of the implanted sample and hence constant source strength. It has, however, the drawback that the sample is distributed

over the whole circumference of the wheel resulting in a poorer signal-to-background ratio. Therefore the ion beam was implanted at one spot for the less abundant ^{183}Au isotope ($T_{1/2} = 42$ s). After implantation of ^{183}Hg ($T_{1/2} = 8.8$ s) for a time of 20 s the wheel was turned by 180° within 2 s. Then the desorption was started. This half-turn mode increased the signal-to-background ratio by a factor of 20. The problem of varying source strength during the wavelength scan was solved by shifting the start time of the turn of the wheel by a constant time interval at each start of a scan of the laser wavelength for RIS. A wavelength scan lasted about 60 s. Since 50-100 scans had to be taken in case of ^{183}Au for reasonable statistics, a constant source strength for each wavelength point was guaranteed. This half-turn mode was also used in case of the investigation of the Pt isotopes. For example an implantation time of 15 min was used for ^{183}Pt ($T_{1/2} = 6.5$ min).

A Faraday cup and several insulated diaphragms mounted in the beam tube before the target wheel allowed a measurement of the current of the ion beam delivered by ISOLDE and hence its alignment by the help of x- and y-deflectors. A small NaJ detector was mounted near the target wheel for a measurement of the amount of implanted or desorbed radioactivity.

4.2 The vacuum system

An ultra-high vacuum is essential for low background in the case of the RIMS/PLID technique. Test measurements resulted in a re-covering time of 5 min after cleaning the surface by laser desorption at a pressure of $p \leq 10^{-7}$ mbar. Several measures were taken to keep the vacuum pressure in the wheel chamber below this value, and that of the RIS interaction zone and of the spectrometer below 10^{-8} mbar.

1. A differential pumping unit was installed between the beam line of ISOLDE-3 ($p \simeq 10^{-6}$ mbar) and the wheel chamber. A turbomolecular pump (360 l/s) served for a pressure of $p = 5 \cdot 10^{-8}$ mbar (see fig. 7).
2. A non-evaporable getter was installed around the target wheel (fig. 6). It was a folded getter strip (length: 1 m, width: 37 mm) of the type St-707/CTAM/30D (SAES Getters SpA, Milano) which has a pumping speed of about 100 l/s at room temperature after activation by heating.
3. The chamber containing the target wheel and the getter pump was mounted on a flange. It was constructed as a closed cylinder ($d = 148$ mm) with an insulated endplate. This endplate had a rectangular cut-out for the target wheel (see fig. 6) and was made of stainless steel with an inlet of high-grade graphite to avoid the desorption beam hitting a steel surface. Hence, the vacuum of the wheel chamber was quite well separated from that of the copper-sealed interaction zone and of the TOF spectrometer.
4. Copper-sealed UHV flanges, stainless steel as construction material, and alumina for insulation were used throughout except for those parts mentioned explicitly. The vacuum chamber for the interaction zone and the grids of the TOF spectrometer (see below) had a cubic shape ($31 \text{ cm} \times 31 \text{ cm} \times 31 \text{ cm}$) and was pumped by a turbomolecular pump (200 l/s) to a pressure below 10^{-8} mbar. The composition of the residual gas could be checked by a quadrupole mass analyzer. The drift tube of the TOF spectrometer and the chamber containing the ion reflector (see below) were pumped by small turbomolecular

pumps (50 l/s). Here, a vacuum better than 10^{-8} mbar was obtained also.

4.3 The time-of-flight mass spectrometer

A time-focusing TOF mass spectrometer [49] was used to discriminate wanted Au or Pt photoions against unwanted background ions. It is shown in fig. 7 and its front part in fig. 8. Similar devices were described in detail earlier [50, 51]. It consists of a two-stage acceleration region, a drift tube of a total length of $l = 1.3$ m, a 40° ion reflector and a multichannel plate (MCP) detector (VUW 8920 ES, Varian, $d = 42$ mm) in tandem geometry. The ion deflector was installed for shielding the MCP detector by lead bricks against the radioactivity emanating from the collection region. An einzel lens could be used for imaging the interaction region onto the surface of the MCP detector. The acceleration region consisted of the insulated endcup of the cylindrical enclosure of the target wheel region (see figs. 6 and 8) and the visible part of the target wheel itself ($U_1 = 2600$ V), a first grid (diameter $d = 50$ mm, $U_2 = 2250$ V) installed 22 mm apart from the desorption spot on the target wheel, and a second grid on ground potential another 20 mm away from the first grid. Both grids were made of Mo-wire with a mesh size of 1 mm x 1 mm, resulting in a transmission as high as 95% (Eyring, Karlsruhe). The grids as well as the einzel lens and an electrode (for corrections of aberrations of the electric field due to the holders of the meshes) were mounted on a four-rod ion optical bench centered in a tube with an inner diameter of 100 mm. This tube (multiply perforated for efficient pumping) was welded into a CF150-flange which itself was mounted at the cubic vacuum chamber opposite to the flange holding the target wheel chamber (fig. 7). For highest efficiency the photo ions were created as near as possible to the desorption spot in an interacting region with largest diameter. The optimum at the available laser powers was found at a diameter of the RIS laser beams of about $d = 10$ mm centered 6 mm apart from the surface of the target wheel.

4.3.1 Suppression of background ions

The most essential task in RIMS/PLID-experiments with minute samples — decisive for the success of the experiment — is the suppression of background ions. There are manifold sources of background events seen by the MCP detector:

1. Primary ions produced in the desorption process.
2. Secondary ions produced by electrons which themselves are created in the desorption process. Secondary ions can be generated from residual gas, evaporated target material and target impurities, or desorbed from surfaces hit by an electron.
3. Ions produced at surfaces by light of the desorption laser scattered off the target. Again, these ions might be generated directly or via an intermediate electron.
4. Same as above but produced by the light of the RIMS laser beams scattered off, e.g., by reflection at the surfaces of the windows in the vacuum chamber.
5. Ions produced by the RIS laser beams from residual gas or from evaporated target material or target impurities.
6. Secondary ions produced at surfaces hit by an ion. This might happen directly or via an intermediate electron.

Intensive tests were performed to achieve optimum conditions. Background sources 1-3 can be tackled with the help of the short delay (several μs) between the pulse of the desorption laser and those of the RIS lasers. Sources 4 and 5 are mainly excluded by good vacuum conditions, imaging the interaction region onto the detector and good mass resolving power of the TOF spectrometer. Source 6 is of major concern since those background ions are related timewise neither to the desorption nor to the RIS lasers. They might be avoided by appropriate deflectors and diaphragms.

The final arrangement used for the on-line experiment is shown in fig. 8. The potentials applied to the different electrodes are compiled in tab. 1. Electrode FK turned out to be most efficient, as it tackles ions and electrons nearest to the point of creation by the desorption laser. It was placed 5 mm below the interaction region and switched to +500V with respect to the potential of the target wheel during the desorption pulse. Hence, electrons were collected and ions deflected in such a way that they did not reach the MCP detector. Before the thermal, neutral Au or Pt atoms were interrogated by the RIS laser beams, the electrode was switched with a rise time of $\tau = 50$ ns to 2490 V as required for a homogenous electric field between the end plate and the first grid in a time-focussing TOF spectrometer. The deflectors behind the einzel lens and before the reflector were pulsed with appropriate delays, so that only the isotope under investigation could reach the MCP detector unhindered. Four diaphragms (opening: 22 mm) installed between the deflectors, and the ring electrodes at the entrance and exit of the deflectors themselves (opening: 20 mm) served to suppress time-non-correlated background events (source 6). Pulsing of the reflector turned out to be inefficient since this device was found to be itself a source of background in a pulsed mode.

4.4 Laser system

4.4.1 Desorption laser

A home-made Nd:YAG-laser [52] was used for desorption. No significant difference was found for desorption by the infrared, fundamental or by the green, frequency-doubled ($\lambda = 532$ nm) laser light. Hence, for sake of safety, the visible output was used for desorption. Typical operating conditions during the tests and the on-line experiments were a repetition rate of $\nu_{rep} = 10$ Hz, a pulse width of $T = 10$ ns, and an output pulse energy for PLID of $P = 6$ mJ. This corresponds to a power density of 20 MW/cm² since the light spot on the target wheel had a size of 1 mm $\times$ 3 mm due to a focussing lens and the tilted incidence of the laser beam. Pulse energies of around 20 mJ were used for cleaning the target before a measurement was started.

4.4.2 Dye laser system for resonant ionization

A Nd:YAG-laser-pumped dye laser system was used for RIS, synchronized with the Nd:YAG laser for desorption. Three (two) dye lasers (DL1, DL2, DL3) delivered the light as required for resonant ionization of Au (Pt) according to the excitation scheme shown in fig. 2. The relevant data of the laser system are compiled in tab. 2.

Nd:YAG pump laser: The second and third harmonic output of a Nd:YAG laser (Molelectron MY32-10) were used for pumping the dye lasers. The output was boosted by a

home-made amplifier [53].

DL1: The optical transition induced by this laser yielded the information on HFS and IS of the isotope under investigation. Hence it should have a linewidth similar to the Doppler width of the desorbed atoms and allow an accurate scanning of the laser light. A modified Molelectron dye laser (DL16P) was used with three additional amplifier stages and an etalon reducing the bandwidth to about 400 MHz [21]. Tuning was accomplished by pressure change in the resonator cavity. The output was frequency-doubled in a KDP crystal. Frequency calibration was performed as in the earlier experiment [25, 26] by using part of the laser output in the visible spectral region for absorption spectroscopy in a cell filled with molecular iodine [54] and part of the ultraviolet output for resonance fluorescence spectroscopy on an auxiliary atomic beam of stable Au or Pt isotopes. The signal of the reference atomic beam served also as diagnostics for the dye laser performance.

DL2 and DL3: These lasers (FL2001, Lambda Physik) were operated without etalon and hence with a broad linewidth which was larger than the HFS's and the IS's of the different isotopes. Therefore these lasers could be fixed in wavelength.

The dye laser system was installed in a laser hut some 17 m away from the RIMS/PLID apparatus. The output of DL1 was transmitted by a telescope/mirror system to the experimental set-up with an efficiency of about 60%, since second harmonic generation had to be performed near the entrance window of the vacuum chamber. The light of DL2 was transmitted by an optical fiber (Fluosil SX, core diameter: 0.2 mm, Schott) with a transmission of about 40%. Laser light for the ionization step was again transported via a telescope/mirror system with an efficiency of about 60%. The three laser beams used for RIS were superimposed by means of dichroic mirrors and sent through vacuum windows made of sapphire into the interacting region. The optimum delay between the pulses of the lasers were found experimentally and are given in tab. 2.

4.5 Data acquisition

A Computer (LSI 11/23, DEC) was used via CAMAC to control the pressure tuning of the dye laser DL1, the turning of the target wheel, and the activation of a shutter for the ISOLDE beam and of another one used for blocking the desorption laser beam during turning of the wheel. During a laser scan the following data were recorded for each laser pulse and used for on-line data analysis:

- The signal of the MCP-detector via a linear fan-out as analog signal by use of a gated ADC as well as digital signal by use of a gated discriminator. Both units were gated by a rectangular pulse with a width of 70 ns with a delay with respect to the RIS laser pulse as determined by the time of flight of the isotope under investigation.
- An analog signal of the MCP-detector gated with a different delay for observation of the resonance of another isotope for reference purposes.
- As above but digital signal recording for an additional isotope.
- The signal of the MCP-detector in digital mode with a gate set to a TOF region where neither Au nor Pt ions are counted. This signal served to determine the rate of background events.
- The absorption signal from the I₂-cell for frequency calibration.
- The fluorescence signal from the reference atomic beam of Au or Pt for frequency

calibration.

- The pressure in the resonator cavity of DL1 which determines the wavelengths of the laser light.

The recording of the MCP-signals in five different TOF windows enabled to identify events where the desorption led to a burst of background ions. Those events were excluded from the data analysis. Fig. 9 shows the recorded signals for the case of a measurement of ^{191}Pt . In addition to the wave length dependent spectra the simultaneously observed TOF spectra were recorded by a transient recorder (TR8818, Le Croy) with a time resolution of 10 ns. An example of such a spectrum is displayed in fig. 10.

5 Performance of the RIMS/PLID apparatus and results

5.1 Pulsed laser-induced desorption of implanted refractory elements

Target material: In addition to tantalum and tungsten tested in an earlier experiment [28] tungsten carbide and iridium were tested as target material. According to a calculation made by Eichler [55] both materials should have a heat of absorption for Pt smaller than graphite. Tungsten carbide has a similar reflectivity R for green light as graphite, but Ir has $R(\lambda = 532 \text{ nm}) = 0.7$ [56] resulting in a higher desorption power required. The background observed for both targets by the MCP-detector with all deflectors in operation was similar to that for a C-target. However, no significant photo-ion yield could be observed during a short on-line tests with radioactive Pt isotopes. Two explanations are possible: Either the desorption efficiency is much lower than for graphite, or the desorption was so fast that the signal appeared only in the first laser shots where the background is still large. Hence the graphite target was used for the Au as well as for the Pt experiments. The good performance of graphite is in agreement with earlier observations [28] and was also found by Lee et al. [34].

Velocity distribution in the desorbed atomic beam: The velocity distribution was measured at a distance of 6 mm from the target surface with RIS laser beams of a diameter of 1 mm. Fig. 11 shows the calculated evolution of the pulsed atomic beam (fig. 4) in the interacting region with the RIS laser beams. Varying the delay between desorption pulse and RIS pulses one obtains a distribution as shown in fig. 12. The experimental data are fitted by the theoretical signal (as the function of the delay time) given by [29]

$$S(t_D) \propto \int_{z_1}^{z_2} v^3 \cdot e^{-v^2/v^2} dz \quad (3)$$

with z in the direction of the beam axis perpendicular to the target surface. The mean velocity is $\bar{v} = \sqrt{2kT/m}$ and z_1 and z_2 are the boundaries of the RIS lasers in z -direction. The solid line in fig. 11 represents a fit by (3) with $z_1 = 0.5 \text{ cm}$ and $z_2 = 0.7 \text{ cm}$. Free parameters of the fit are the temperature T , the maximum signal and a constant background. The agreement is very good. The temperature determined by the fit is indicated. Fig. 13 shows the temperatures determined in this way as a function of the energy density of the desorption laser. The dashed line is a fit of the data points by a straight line resulting in

$$T = 500(200) + 6.5(8) \cdot E_{\text{des}}^{\text{exp}} \quad (4)$$

$E_{\text{des}}^{\text{exp}}$ is the measured energy density of the laser pulse in mJ/cm^2 and T is given in Kelvin. Correcting for the reflectivity of graphite ($R = 0.35$) one obtains for the temperature as a function of the energy density E_{des} deposited in the target material

$$T = 500(200) + 10.0(1.2) \cdot E_{\text{des}} \quad (5)$$

This has to be compared with the result according to (1) for a pulse with $t_{1/2}$ (FWHM) = 10 ns. The calculation gives

$$T = T_0 + 11.3 \cdot E_{\text{des}} \quad (6)$$

and is shown in fig. 3 as solid line. The excellent agreement between (5) and (6) should not be taken too seriously since the beam diameter of the desorption laser on the target surface cannot be determined exactly. Furthermore, there are inhomogeneities in the beam profile. Nevertheless, the linear dependence of the surface temperature on the energy deposited by the desorption laser is obvious.

Time constant of the desorption process: The number of desorbed Au or Pt atoms decreases exponentially with time after switching on the desorption laser. In the on-line experiment, immediately after the implantation of ^{190}Hg and the decay to the daughter, a $1/e$ - decrease was observed after about 550 laser shots corresponding to a time constant of $\tau = 55$ s at $\nu_{\text{rep}} = 10$ Hz. No significant correlation with the power density deposited by the desorption laser was observed when varying the power from 120 mJ/cm² up to 200 mJ/cm². This is in agreement with earlier measurements [28] performed with ^{195}Au a long time after implantation. These tests, however, resulted in a somewhat shorter time constant of $\tau = 20$ -30 s at the same repetition rate. They also yielded no significant dependence on the implantation energy between 6 keV and 60 keV.

Efficiency: About 50% to 60% of the implanted radioactivity could be released by PLID from the target if the beam diameter of the desorption laser was larger than that of the implantation spot. The temporal overlap between atomic and RIS laser beams (fig. 11, $z_1 = 1$ mm, $z_2 = 11$ mm) is calculated to be 80% for a delay of 10 μs between the desorption and the RIS pulses. Assuming a $\cos^2$ distribution of the desorbed atoms and that only those atoms with a Doppler shift of $\Delta\nu_D \leq 1$ GHz can interact with the RIS laser light, one obtains a spatial overlap of the atomic and the laser beams of 60%. Hence, the total efficiency of the PLID process is

$$\epsilon_{\text{PLID}} \simeq 0.2 \quad (7)$$

In the actual on-line experiment at ISOLDE-3 the transmission of the beam line in front of the target wheel was measured to be

$$\epsilon_{\text{BL}} = 0.3 \quad (8)$$

This was mainly due to the limited aperture of the differential pumping unit. Because of the length of this unit the quadrupole lens used for focussing the ISOLDE beam on the target wheel had to be placed 140 cm away from the target. Hence, the ISOLDE-3 beam could only be focussed to a diameter of about 5 mm on the target wheel. Since it was more important to avoid background than to optimize the countrate of the Au or Pt photo ions, the focal spot of the desorption laser had to be kept well inside the implantation area. With an area of 1 mm $\times$ 3 mm for the focal spot an overlap of

$$\epsilon_{\text{focus}} \simeq 0.2 \quad (9)$$

was obtained.

5.2 Resonance ionization

Delay between lasers: The optimum delay Δt_{01} between the pulse of the desorption laser and the pulse of the dye laser DL1 was determined to be $\Delta t_{01} = 11 \mu\text{s}$ (fig. 12). Also the pulses of the three dye lasers had to be delayed with respect to each other for optimum efficiency. For example, in case of Au the RIS ion signal was highest with a delay of $\Delta t_{12} = 2.5 \text{ ns}$ between DL1 and DL2. The optimum delay between DL2 and DL3 was determined earlier to be $\Delta t_{23} = 7.5 \text{ ns}$ [28]. The population of the different states can be calculated as a function of the delays Δt_{12} , Δt_{23} by rate equations, neglecting coherence effects. Fig. 14 shows the result of such a calculation. The statistical weights are neglected as well as the HFS splitting of the ground state. The following parameters were used: The lifetimes $\tau_1 = 6.0 \text{ ns}$ [57], $\tau_2 = 10.7 \text{ ns}$ [28], the dye laser pulse widths $\Delta t_1 = \Delta t_2 = 6 \text{ ns}$, $\Delta t_3 = 10 \text{ ns}$, the delays $\Delta t_{12} = 2.5 \text{ ns}$, $\Delta t_{23} = 5 \text{ ns}$, and the products of cross section times laser flux density $\sigma_{01} \cdot \phi_1 = \sigma_{12} \cdot \phi_2 = 1$ and $\sigma_{23} \cdot \phi_3 = 0.04$.

Line width in the tuned, first transition: If one aims for highest efficiency of RIS, one has to illuminate as many atoms as possible which are desorbed from the target. Hence one has to accept sacrifices with respect to Doppler broadening and hence resolution. With a diameter of the laser beam of 1 cm centered 6 mm apart from the desorption spot (fig. 11) one calculates a Doppler broadening $\Delta\nu_{\text{Doppler}} = 2.3 \text{ GHz}$ for a target temperature of 1200 K which increases to about 3.0 GHz for 2000 K. The experimentally observed width $\Delta\nu_{\text{exp}}$ was of this order as shown in fig. 15 for the example of ^{184}Au . Since the HFS splittings are large in the light Au isotopes, they can be determined with the sufficient accuracy of 10^{-3} . Hence the linewidth is of no concern. In case of the platinum experiments, a line width of $\Delta\nu_{\text{exp}} = 3 - 4.5 \text{ GHz}$ was observed caused at least partially by saturation broadening in the first two transitions. Since the HFS splitting is rather small for some isotopes, the laser power was diminished and the contribution to the line width by Doppler broadening was reduced by only imaging part of the ions leaving the interaction zone onto the MCP detector. This was achieved by switching off the voltages of the einzel lens and the correction electrode. In this way, an experimental linewidth of $\Delta\nu \simeq 1.3 \text{ GHz}$ was obtained (fig. 16).

Efficiency: The efficiency of those atoms illuminated by the laser beams is determined by the cross section for the different excitation steps, the lifetime of the levels involved, and the power densities and line widths of the RIS laser beams. The latter quantities are given in tab. 2. A calculation can only be done for Au where all quantities were determined. The oscillator strength for the first excitation at $\lambda = 268 \text{ nm}$ has been determined to $f = 0.176(3)$ [58], resulting in an effective cross section of $\sigma_{01}^{\text{eff}} = 2 \cdot 10^{-11} \text{ cm}^2$ for a laser width of $\Delta\nu_{\text{DL1}} = 0.86 \text{ GHz}$. The effective cross section of the second excitation step is $\sigma_{12} = 1.8(4) \cdot 10^{-12} \text{ cm}^2$ [59] ($\Delta\nu_{\text{DL2}} = 6 \text{ GHz}$). Both transitions can be saturated by the available laser power. Hence the population of the $^2\text{D}_{3/2}$ state is $1/3$ (disregarding the statistical weights) and finally 10%, if $\Delta\nu_{\text{Doppler}} \simeq 3 \cdot \Delta\nu_{\text{DL1}}$ is taken into account. The cross section to the ionizing state located at $\lambda = 592 \text{ nm}$ above the $^2\text{D}_{3/2}$ state was determined in an earlier experiment [60] to be $\sigma_{23} = 6(2) \cdot 10^{-17} \text{ cm}^2$. Only 2 mJ/cm^2 were available in the test experiments resulting in an efficiency of this last step of about 10%. Taking the two HFS levels of the ground state into account, one obtains a

total efficiency of the RIS process of the order of

$$\epsilon_{\text{RIS}} \simeq 5 \cdot 10^{-3} \quad (10)$$

which is in rather good agreement with the results of the calculations by rate equations shown in fig. 14.

5.3 Time-of-flight mass spectrometer

The imaging properties of the TOF spectrometer were studied by installing a phosphor screen behind the MCP detector. For high sensitivity, the desorbed Au or Pt atoms were ionized by the RIS laser beams in a large solid angle of the divergent ion beam. This beam was focussed with a voltage applied to the einzel lens and the correction electrode as given in tab. 1. The optimum focal length was found to be $f \simeq 0.7$ m.

Mass resolving power: The resolving power of the TOF spectrometer was determined to be $M/\Delta M(\text{FWHM}) = 260$ at $A \simeq 185$ from a time of flight of $31 \mu\text{s}$ and $t_{1/2}(\text{FWHM}) \simeq 60$ ns of the TOF signal. The dependence of the ion signal of the isotope under investigation on the wavelength of DL1 was recorded in a time window of $\Delta t = 70$ ns centered to the peak in the TOF spectrum. The moderate but sufficient resolving power of the TOF spectrometer was mainly caused by the large interaction volume (laser/atomic beam) as well as the velocity spread of the desorbed atoms, and only partly due to the einzel lens in the drift tube for focussing also diverging ions onto the MCP detector.

Background: As in the earlier work with a continuous atomic Au beam evaporated from an oven [25, 26], a background could be achieved as low as one event per 1000 laser shots in a time window of $\Delta t = 100$ ns. In case of PLID, however, it was essential to use the pulsed deflectors and the diaphragms installed in the drift tube. A peak at $A = 181$ was observed all the time in case of the Au investigations. Probably it is caused by ^{181}Ta which can be ionized by the laser light of DL3 ($\lambda = 592$ nm). The low background resulted in a signal-to-background ratio of $S/B = 20$ in case of ^{183}Au and ^{184}Au and $S/B = 6$ for ^{183}Pt and ^{184}Pt .

Efficiency: The transmission of the TOF spectrometer is estimated to be $\epsilon_{\text{trans}} \simeq 0.3$ due to the three grids and incomplete imaging of all photo ions onto the MCP detector. The detection efficiency of a MCP detector in tandem geometry is determined to $\epsilon_{\text{MCP}} \simeq 0.3$ - 0.4 [61]. Hence, the total efficiency of the TOF spectrometer is of the order of

$$\epsilon_{\text{TOF}} \simeq 0.1 \quad (11)$$

5.4 Overall efficiency of the RIMS/PLID experiment

The theoretical overall efficiency is obtained from (7), (10) and (11) as

$$\epsilon_{\text{tot}}^{\text{theo}} = \epsilon_{\text{PLID}} \cdot \epsilon_{\text{RIS}} \cdot \epsilon_{\text{TOF}} \simeq 10^{-4} \quad (12)$$

for a sample implanted in the target wheel in an area covered by the desorption laser. Test experiments with implanted, long-lived ^{195}Au gave an overall efficiency for Au of

$$\epsilon_{\text{tot}}^{\text{exp}} = 10^{-5} \quad (13)$$

for the described setup as well as for an earlier one with slightly different geometry [28]. The missing order of magnitude in detection efficiency can have many reasons: Overestimating the partial efficiencies (7), (10), (11), evaporation of Au as molecule or attached to a laser-desorbed carbon cluster (which have been observed by us by ionization by means of electron bombardment), ionization of Au in a laser-produced microplasma during desorption, inhomogeneities of the intensities of the three dye lasers in the beam profile and so on. Nevertheless, an enhancement by three orders of magnitude was achieved in comparison to the continuous evaporation of Au out of an oven. The experimental overall efficiency for Pt was determined to be $\varepsilon_{\text{tot}}^{\text{exp}} \simeq 3 \cdot 10^{-8}$ for 10^{10} and 10^{11} atoms of stable Pt brought as platinum chloride onto the target wheel. The slightly reduced efficiency as compared to Au might be due to the higher heat of adsorption of Pt on graphite [55].

In the on-line run, the efficiency was reduced to $\varepsilon_{\text{tot}}^{\text{exp}} = 5 \cdot 10^{-8}$ for the case of the measurement of the shortest-lived Au isotope investigated, namely ^{183}Au ($T_{1/2} = 42$ s). The reasons were the transmission of the differential pumping unit between ISOLDE beam line and target wheel ($\varepsilon_{\text{PL}} = 0.3$, see eq. (8)), the overlap between the area of the implantation spot and that of the laser beams ($\varepsilon_{\text{focus}} = 0.2$, see eq. (9)), the losses by radioactive decay during collection, turning of the wheel and laser desorption ($\varepsilon_{T_{1/2}} = 0.5$) and a lower performance of the dye laser used for the last excitation step (DL3) during the beamtime. In case of the on-line measurements of platinum, the overall efficiency was determined to $\varepsilon_{\text{tot}}^{\text{exp}} = 1 \cdot 10^{-8}$ for the shortest-lived isotope investigated, namely ^{183}Pt ($T_{1/2} = 6.5$ min). A difference in desorption yield between surface-adsorbed and implanted Pt might play a role beside the losses in transmission, overlap, and radioactive decay as already discussed for Au.

6 Results and outlook

The RIMS/PLID apparatus described above enabled us to extend the measurements (done by RIMS on a Au beam evaporated from an oven [25, 26]) to the shorter-lived isotopes ^{184}Au and ^{183}Au [62]. The number of ^{183}Au atoms accumulated in the decay of the ^{183}Hg was more than two orders of magnitude smaller than that in case of ^{185}Au , the most neutron-deficient isotope accessible earlier [25]. The smaller number is due to the shorter half lives and the rapidly decreasing yields for nuclei further away from stability. Platinum isotopes could be measured [30, 32] in the series from the stable ^{198}Pt isotope down to ^{183}Pt ($T_{1/2} = 6.5$ min). Hence, the interesting mid-neutron region could be investigated between the shell closures at $N = 126$ and $N = 82$. Data on the shape transition are now available for three neighbouring isotopic chains. The information obtained on the change of the mean-square charge radii are shown in Fig. 1. The results for ^{183}Au and ^{184}Au are published [62]. The HFS data of the platinum isotopes will be published elsewhere [32, 31].

The RIMS/PLID technique enables to investigate also those isotopes which are not available directly as on-line ion beams but are only produced in the decay of the primary beams delivered by an ISOL facility. This mainly concerns refractory elements where the atoms do not diffuse out of the matrix of the ISOL-target system because of their low vapor pressure. Since collinear spectroscopy works excellently on the direct ion beam but not in case of daughter isotopes, RIMS is complementary to this technique. It is even comparable to collinear spectroscopy regarding sensitivity since the sample can be accumulated before the measurement takes place. RIMS is also beam-time efficient due to the low background rate when suitable measures are taken as described above. For example, the whole measurements on Au and Pt were performed at the ISOLDE-3 facility during a beamtime of a total of six days divided into two parts, where, in addition, we were faced with the many problems associated with a just completed new on-line separator. The only disadvantages of the RIMS/PLID method as compared to collinear spectroscopy are the achievable resolution and the noisy RIS ion signal due to fluctuations in desorption yield. As shown in Ref. [36] higher resolution can be obtained by a sacrifice in sensitivity. Still, the accuracy of the data could not be increased significantly by use of dye lasers with small bandwidth due to strong fluctuations in the intensity of the desorbed atomic beam.

The RIMS/PLID technique opens many new experimental possibilities besides being an efficient tool for the determination of nuclear ground state properties as shown above. It might be used for ultra-sensitive trace analysis of surfaces with high spatial resolution. A possible application could be the quantitative determination of Pt in the environment and the measurement of its distribution in, e.g, organic material. It is known that platinum is released by cars with catalytic converters in amounts of about $1\text{ }\mu\text{g}$ per 100 km. Little is known about its distribution and possible enrichment in the environment. Almost nothing is known about the risks of microscopically distributed catalytic platinum.

RIMS/PLID could also be used as isobaric-pure ion source for experiments which suffer from contamination. It could even be a very fast ion source, e.g., when a foil is placed behind a target collecting the recoil ions in a heavy ion reaction. Since RIMS yields a pulsed ion beam, the pulse structure can be used efficiently for injection into storage rings or ion

traps. Presently, only RIMS/PLID provides a feasible injection technique to measure the mass of the isotopes of refractory elements by a Penning trap mass spectrometer [63].

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Table 1: Voltages applied to the electrodes of the TOF spectrometer and the timing of the pulses. Δt is the delay of the electrical pulse with respect to the start of the desorption laser whereas T is the length of the pulse.

Ion optical element	U_{DC} [V]	U_{pulse} [V]	Δt [μs]	T [μs]
Target	2600	—	—	—
Electrode (FK)	2990	−500	8.0	13.0
First grid	2250	—	—	—
Correction electrode	1065	—	—	—
Electrostatic lense	1300	—	—	—
Y-deflector 1	400	−400	14.5	11.5
Y-deflector 2	500	−500	$\approx 30^*$	1.2
Reflector	2460	—	—	—
MCP detector	− 2000	—	—	—

* To be adjusted for the isotope under investigation.

Table 2: Properties of the lasers used for RIMS/PLID. The repetition rate of all lasers is fixed to 10 Hz. The dye laser used for the spectroscopy in the first excitation step (modified Molelectron laser DL 16 P) is described in detail in [21]. An amplifier stage is added to the Nd:YAG laser (model Molelectron MY 32-10). (C: Coumarin; SR: Sulforhodamin)

	1. step		2. step		3. step	
	Au	Pt	Au	Pt	Au	Pt
Dye	C 540 A		DPS	C 460	SR B	—
Pump wavelength [nm]	355		355		532	—
Pump energy [mJ]	20		8		15	—
Wavelength [nm]	535 (268)	532 (266)	407	451	592	532
Pulse energy [mJ]	1 (≥ 0.01)		0.15		0.27	10
Pulse duration [ns]	6 (4)		10		10	10
Delay [ns] (rel. to 1. step)	0		2.5	4	10	13
Bandwidth [GHz]	≤ 0.4 (≤ 0.8)		≈ 35	10	6	15

Figure Captions

Figure 1: Changes of the mean-square charge radii in Hg (top), Au (middle) and Pt (bottom) as obtained by measurement of the isotope shift. The prediction of the spherical droplet model is indicated by a dashed line. The experimental errors are smaller than the size of the symbols (except for ^{184}Au where the spin is not known), but there is a common scaling uncertainty in each isotopic chain of the order of 10 %. Isomeric states are indicated by asterisks. Data are taken from ref. [18] for Hg. The Au data are cited from ref. [25, 26, 62] and from ref. [35] for $^{194,196}\text{Au}$. The $\delta \langle r^2 \rangle$ values of Pt were determined in this work [30] and that of $^{185\text{m}}\text{Pt}$ by ref. [35].

Figure 2: Partial atomic level scheme of Au (left) and Pt (right). The transitions used for resonance ionization are indicated. The last step in case of Au leads to an autoionizing state (AI) [28], the last one for Pt into the continuum by use of frequency-doubled Nd:YAG laser light.

Figure 3: Temperature at the surface of a graphite target as a function of time during and after irradiation with a Gaussian laser pulse (pulse length $t_{1/2}(\text{FWHM}) = 10 \text{ ns}$, energy density $P_0 = 200 \text{ mJ/cm}^2$). The curve is calculated by means of eq. (1). Inset: Temperature profile inside the graphite immediately after the laser pulse ($z = 0$: surface). The implantation depth is indicated for ^{197}Au ions which had been implanted with 60 keV into the graphite target.

Figure 4: Desorption rate as a function of the time during and after irradiating a graphite target with a Gaussian laser pulse (pulse length $t_{1/2}(\text{FWHM}) = 10 \text{ ns}$, energy density $P_0 = 200 \text{ mJ/cm}^2$) The curve is calculated by means of eq. (2) and the temperatur curve given by eq. (1).

Figure 5: Experimental set-up used for the on-line resonance ionization mass spectroscopy of short-lived Au and Pt isotopes with pulsed-laser induced desorption. Laser wavelengths and signals (hyperfinestructure splitting of an isotope and time-of-flight spectrum) are indicated for the case of platinum.

Figure 6: Target wheel chamber for collecting the 60 keV ion beam delivered by the on-line mass separator (coming through the beam tube from the bottom). The wheel is turned by a stepping motor placed outside the vacuum. A pulsed thermal beam is formed when the sample implanted into the graphite wheel is rotated to the top and desorbed by pulsed laser light.

Figure 7: Set-up and vacuum system used for on-line resonance ionization mass spectroscopy of short-lived Au and Pt isotopes with pulsed-laser induced desorption (not to scale). The pumping speed of the turbomolecular pumps (TP) is indicated. MCP: multichannel plate detector, VG: vacuum gauge, QMF: quadrupol mass filterfor residual gas analysis, DEF: y - deflector.

Figure 8: Schematics of the target wheel and interaction region of the desorbed atoms with the laser beams for resonant ionization and the front part of the time-of-flight spectrometer for mass-selective detection of the photoions.

Figure 9: Signals taken during the run for the example of ^{191}Pt ($T_{1/2} = 2.8 \text{ d}$). From top to bottom: Single-ion counting signal of ^{191}Pt , analog ion signal of ^{191}Pt , the background event rate in the mass range $A < 191$, and the absorption signal of the I_2 cell. The background gate is ten times longer than the other gates. Therefore the background event rate must be divided by 10.

Figure 10: Time-of-flight spectrum of $^{186,190}\text{Pt}$ and of the stable isotopes $^{194,195,196,198}\text{Pt}$ found as impurities in the high-grade graphite target.

Figure 11: The calculated spatial evolution of the pulsed atomic beam $5 \mu\text{s}$, $10 \mu\text{s}$ and $15 \mu\text{s}$ after the desorption. The distance z from the target surface is indicated as well as the boundaries of the laser beams for resonance ionization ($z_1 = 1 \text{ mm}$, $z_2 = 11 \text{ mm}$ for the on-line experiments. The time behaviour of desorbed atoms was determined (fig.12) by laser beams of smaller diameter ($z'_1 = 5.5 \text{ mm}$, $z'_2 = 6.5 \text{ mm}$).

Figure 12: Photo ion signal of gold as a function of drift time after desorption. Top: For an energy density of the desorption pulse of 240 mJ/cm^2 . Bottom: Same but for 320 mJ/cm^2 . The diameter of the laser beams for the resonance ionization were narrowed to 1 mm centered 5 mm apart from the desorption target (fig. 11). The solid line represents a fit yielding a maximum temperature of the surface of the desorption target as indicated.

Figure 13: Maximum surface temperature of the desorption target as a function of the energy density of the desorption laser. The temperatures were determined from the fitted velocity distributions like those shown in fig. 12. The dashed line is the result of a linear regression fit, the solid line was calculated by means of eq. (1).

Figure 14: Three-colour, three-step resonant ionization of gold. The populations of the ground, the first and second excited state, and that of the continuum (via an autoionizing state) were calculated by rate equations. Gaussian temporal beam profiles of the dye lasers were assumed with appropriate delays as indicated by the dashed lines. The input parameters of this calculation are given in the text.

Figure 15: Photo ion signal of ^{184}Au taken in the mode for optimum sensitivity (imaging the maximum number of photo ions onto the ion detector). The solid line represents a fit by a Gaussian with a line width of $\Delta\nu_{1/2}(\text{FWHM}) = 2.9 \text{ GHz}$. The fit yields an accuracy for the hyperfine splitting in the ground state of 43 MHz and 120 MHz for the isotope shift.

Figure 16: Photo ion signal of ^{191}Pt taken in the mode for maximum resolution (imaging only a part of the photo ions onto the ion detector). The solid line represents a fit by a Gaussian with a line width (FWHM) of $\Delta\nu_{1/2} = 1.3 \text{ GHz}$. The fit yields an accuracy for the hyperfine splitting of 20 MHz; for the isotope shift an accuracy of 110 MHz was determined.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

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Fig. 10

Fig. 11

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Fig. 13

Fig. 14

Fig. 15

Fig. 16