Detection of Trace Amounts of Actinides by Resonance Ionization Mass Spectrometry*

H. RIMKE†, G. HERRMANN, C. MÜHLECK, P. SATTELBERGER, N. TRAUTMANN

Institut für Kernchemie, Universität Mainz, D-6500 Mainz, F.R.G.

F. AMES, H.-J. KLUGE, E.-W. OTTEN, D. REHKLAU and W. RUSTER

Institut für Physik, Universität Mainz, D-6500 Mainz, F.R.G.

For studies of the ecological behaviour of actinides very sensitive detection methods are necessary. To ascertain the origin of the samples the isotopic composition should also be known. Conventional detection methods have some disadvantages, *i.e.*, in mass spectrometry isobaric interferences may occur, or in α-spectrometry some isotopes cannot be resolved because of small differences in their α-energies and furthermore the detection sensitivity depends on the half-life of the isotope to be measured. In the experiments described here, laser resonant photoionization in combination with time-of-flight measurement has been used to detect trace amounts of plutonium and uranium. The actinide atoms are excited from the ground state by absorption of two resonant photons and ionized by a third photon. Mass analysis of the generated ions is performed by time-of-flight spectrometry.

Experimental

The experimental set-up [1] is shown in Fig. 1. The laser system consists of a pulsed copper vapour laser (Oxford Lasers, Mod. Cu-40) which pumps simultaneously three dye lasers (Lambda Physik, Mod. 2001 E). With a pump laser power of about 30 W at a pulse repetition rate of 6.5 kHz and 30 ns pulse-width dye laser conversion efficiencies between 10 and 25% have been reached in a wavelength range between 520 and 850 nm. The dye laser beams are deflected by means of glass fibres (500 μm diameter, 70% transmission) into the interaction zone with the atomic beam located in the time-of-flight spectrometer. With the optical fibres a homogeneous spatial profile and a good overlap of the three dye laser beams are achieved. For the generation of neutral atoms, the samples, deposited by an electrolytical procedure on a rhenium foil and covered with a thin rhenium layer, are heated to 1500–1700 °C. The ions are accelerated by two grids and (after passing a drift length of 2 m) are detected with channel plates. Mass analysis is performed by time to amplitude conversion and pulse height multichannel analysis. Ion count rates as a function of the wavelengths of the dye lasers are recorded with a LSI-11 microcomputer which also controls the dye laser wavelengths.

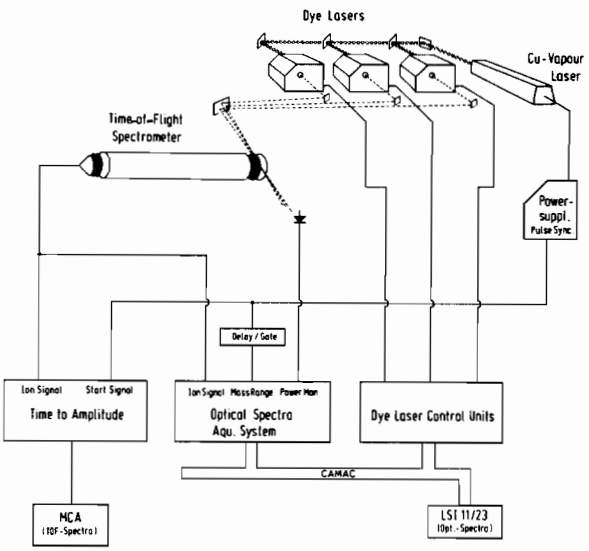

Fig. 1. Experimental set-up.

Results and Discussion

The excitation schemes investigated for the ionization of plutonium [2] and uranium [3] are shown in Figs. 2 and 3. The atoms are excited in two steps via resonant atomic transitions to high lying atomic levels and then ionized by absorption of a third photon, whose energy must be sufficient to exceed the ionization limit. High ion count rates are obtained by ionization via autoionizing states. These states

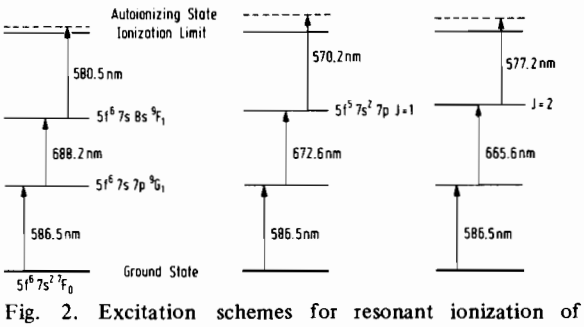

Fig. 2. Excitation schemes for resonant ionization of plutonium-239.

*Paper presented at the Second International Conference on the Basic and Applied Chemistry of f-Transition (Lanthanide and Actinide) and Related Elements (2nd ICLA), Lisbon, Portugal, April 6–10, 1987.

†Author to whom correspondence should be addressed.

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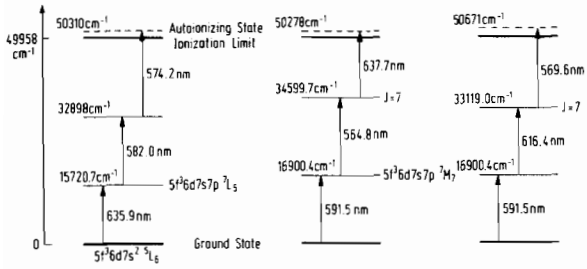

Fig. 3. Excitation schemes for resonant ionization of uranium-238.

can be found by tuning the wavelength of the dye laser used for the third step. The other wavelengths must fulfill the resonance condition. For uranium, many autoionizing resonances were found (Fig. 4), whereas in the case of plutonium only a few autoionizing states could be observed (Fig. 5).

With laser beam diameters of 2–3 mm the excitation steps including the ionization can be saturated in the schemes studied. The detection efficiency of the

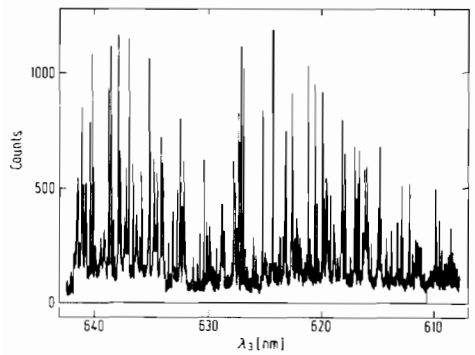

Fig. 4. Autoionizing states of uranium-238. λ_1 and λ_2 in resonance at 591.5 and 564.8 nm.

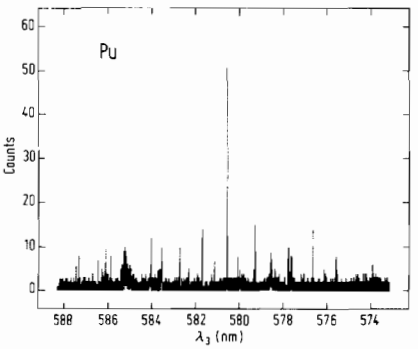

Fig. 5. Autoionizing states of plutonium-239. λ_1 and λ_2 in resonance at 586.5 and 688.2 nm.

system was determined with plutonium-239 samples to be 4×10^{-6}. With a vanishing background a detection limit of less than 10^7 atoms in the sample can be given for plutonium-239. The isotopic composition is measured with the time-of-flight spectrometer, which has a mass resolution of $\geqslant 2500$ as determined with gadolinium samples. The isotopic resolution can be improved by using excitation transitions with large isotope shifts and by scanning the three dye lasers.

The experiments have shown that laser resonant ionization mass spectrometry is a valuable method for the detection of trace amounts of actinides with unambiguous element and isotope assignment.

References

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