# Efficient three-step, two-color ionization of plutonium using a resonance enhanced 2-photon transition into an autoionizing state

P. Kunz[1], G. Huber[1], G. Passler[1] and N. Trautmann[2]
[1]Institut für Physik, Universität Mainz,[2]Institut für Kernchemie, Universität Mainz, D-55099 Mainz (Germany)

Resonance ionization mass spectrometry (RIMS) has proven to be a powerful method for isotope selective ultra-trace analysis of long-lived radioisotopes. For plutonium, detection limits of $10^6$ to $10^7$ atoms have been achieved for various types of samples. So far a three-step, three-color laser excitation scheme was applied for efficient ionization.

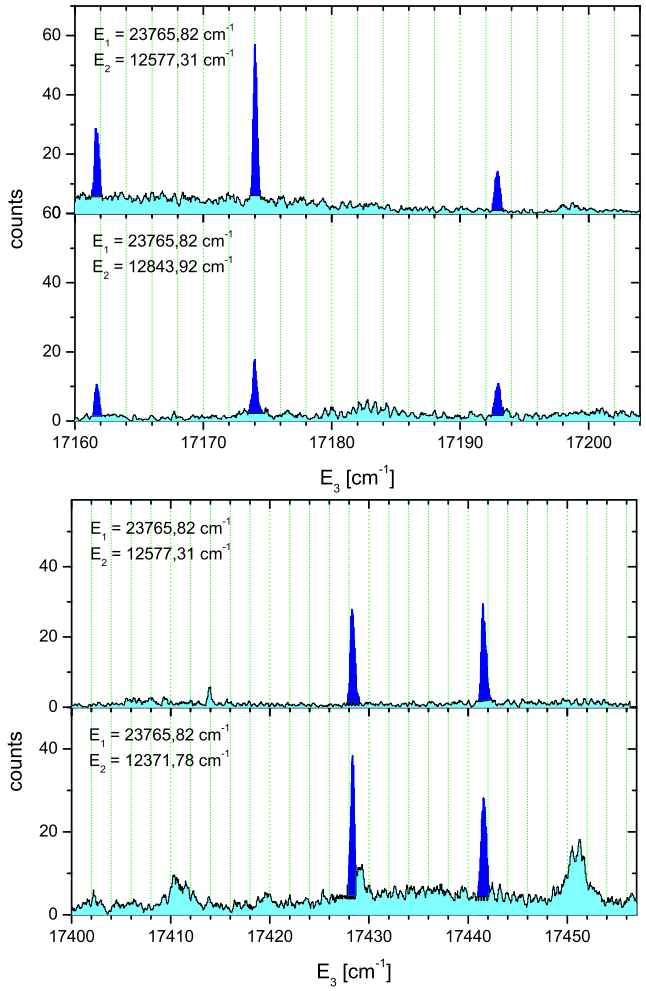


**Fig. 1.** Autoionizing states in plutonium. The scans of the ionization laser in different three-step excitation schemes show a number of resonances. However, some of those - indicated by a different color - are the result of two-photon transitions starting from the first exited state. Only the underlying broadband resonant structures are due to the three step ionization.

The RIMS apparatus used for the measurement of environmental samples as well as for spectroscopic studies on actinides [1] consists of three tunable titanium:sapphire lasers, a tunable dye laser and a reflectron type time-of-flight mass spectrometer (TOF) [2].

An extensive study of autoionizing states for the application in three-step, three-color excitation schemes [3] reveals several strong two-photon transitions into autoionizing states (Fig. 1), yielding ionization efficiencies of the same order of magnitude as those of three-step, three-color schemes as applied so far for routine measurements [4]. One three-step, two-color transition was investigated in detail by using the excitation scheme ($\lambda_1 = 420.77$ nm, $\lambda_{2p} = 582.27$ nm). It was directly compared with a three-step, three-color excitation scheme ($\lambda_1$=420.77 nm, $\lambda_2$= 847.26 nm, $\lambda_3$= 767.53 nm). An overall efficiency for the two-photon excitation scheme of $\varepsilon_{2p} > 10^{-5}$ has been determined. The line width of the two-photon transition is $\Delta\nu_{2p} = 7.7(3)$ GHz. The two-photon ionization efficiency as a function of laser intensity was investigated. This measurement can be compared with a saturation intensity measurement of a one-photon transition from a second excited state into an autoionizing state ($\lambda_1 = 420.77$ nm, $\lambda_2$= 808.29 nm, $\lambda_3 = 790.28$ nm) (Fig. 2). It reveals - as expected from the theory - a linear dependence on the laser intensity as long as the saturation effects at higher laser powers are not relevant. In contrast the two-photon transition (Fig. 2b) shows a linear dependence on the square of the intensity. This proves, that it is in fact a two-photon transition.

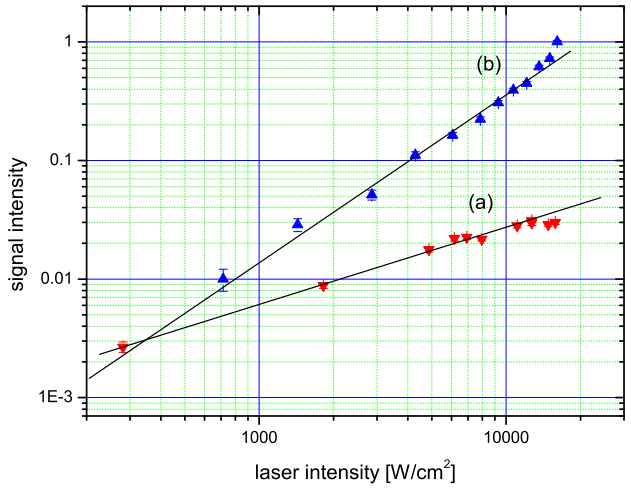


**Fig. 2.** Ion signal as a function of laser intensity. (a) The $\lambda_3 = 790.28$ nm single-photon transition into an autoionizing state shows a linear dependence on the laser intensity. (b) For a 2-photon transition the signal intensity is proportional to the square of the laser intensity, indicated by the higher slope in the logarithmic presentation.

While only two tunable lasers are required for this three-step, two-color excitation scheme, the overall ionization efficiency is sufficient for the application of this scheme in ultra-trace analysis of plutonium.

1. N. Erdmann et al., J. All. Comp. **271-273** (1998) 837
2. C. Grüning, Diss., Inst. f. Kernchemie, Univ. Mainz (2001)
3. P. Kunz, Diss., Inst. f. Physik, Univ. Mainz (2004)
4. P. Kunz et al., EPJ D (in press)