

Resonance Ionization Spectroscopy (RIS) on ^{237}Np

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Introduction: ^{237}Np ($T_{1/2} = 2.14 \cdot 10^6$ a) will be a main contributor to the radiotoxicity of spent nuclear fuel for periods exceeding one million years. Therefore, research on the geochemical properties and the migration behaviour of neptunium is important for the safety assessment of proposed nuclear waste repositories. Since the concentrations of neptunium in the aquifer in case of a leakage of a repository are expected to be less than $10^{-10} \text{ mol} \cdot \text{L}^{-1}$, very sensitive analytical methods are needed for detection and quantification of ^{237}Np . Previous studies by J. Riegel et al.¹ demonstrated the application of resonance ionization mass spectrometry (RIMS) with dye lasers for ultratrace analysis of neptunium. Meanwhile, this setup has been replaced by a more reliable all solid state laser system consisting of three titanium-sapphire (Ti:Sa) lasers pumped jointly by a Nd:YAG laser. Since these lasers are not capable of producing light of the wavelengths used previously, resonance ionization spectroscopy (RIS) was applied to identify high lying energy levels and autoionizing states which can be used for the development of three step excitation schemes for RIMS with Ti:Sa lasers.

Experimental: The laser setup used for RIS consisted of three Ti:Sa lasers. While two of these lasers contained lyot filters and an etalon and were operated at fixed wavelengths, the third Ti:Sa laser was equipped with an optical grating and was tuneable continuously over a broad wavelength range. Blue laser light, which is necessary for the first excitation step of the neptunium atoms into levels with an energy of about 25000 cm^{-1} , was produced by frequency doubling of the light of one Ti:Sa laser in a BBO crystal. Samples for RIS were prepared by applying about 10^{14} atoms of ^{237}Np as neptunium(V) nitrate solution onto a small piece of zirconium foil. After heating to dryness, the samples were placed in a graphite furnace. During resistive heating of the furnace, the neptunium ions were reduced by the zirconium and an atomic beam of neptunium was emitted. These neptunium atoms were subsequently ionized by light from the Ti:Sa lasers; the ions formed were detected with a quadrupole mass spectrometer. Since many suitable energy levels are known for the first excitation step², one laser was operated at different fixed wavelengths to populate several of these odd parity states. For excitation of unknown high lying energy levels, the continuously tuneable grating assisted laser was scanned and NIR light of the third Ti:Sa was used for non-resonant ionization of the highly excited atoms. Search for autoionizing states was performed similarly by populating several of the newly identified high lying levels with light from a laser operating at a fixed wavelength and scanning of the grating assisted Ti:Sa laser.

Results: Numerous high-lying energy levels and autoionizing states of neptunium have been identified (s. fig. 1 and 2). Also, several excitation schemes suitable for resonance ionization mass spectrometry with three Ti:Sa lasers have been developed. Two examples are shown in fig. 3. Saturation of the optical transitions is usually possible with moderate laser power for the first two excitation steps.

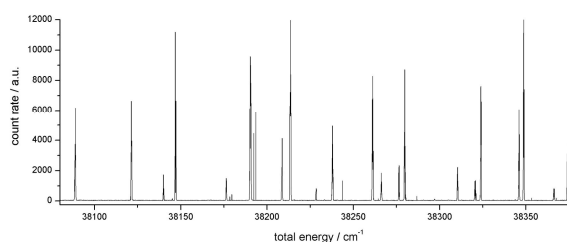


Figure 1. Spectrum of high lying levels of ^{237}Np accessible by resonant excitation from an energy level at 24798 cm^{-1}

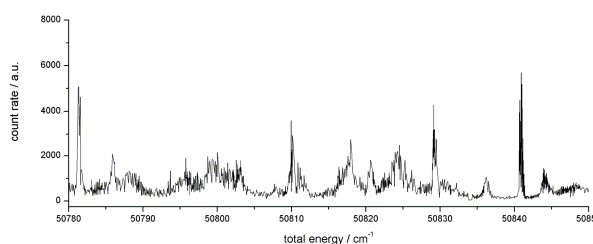


Figure 2. Spectrum of autoionizing states excited from a high lying energy level at 37789 cm^{-1}

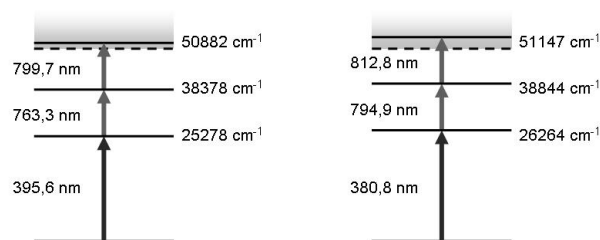


Figure 3. Two possible schemes for resonant excitation and ionization of ^{237}Np populating autoionizing states above the ionization potential at 50535 cm^{-1} .

References

- [1] J. Riegel et al., Appl. Phys. B 56 (1993), 275-280
- [2] J. Blaise, J.-F. Wyart, Selected Constants, Energy Levels and Atomic Spectra, Tables Internationales de Constantes, Université P. et M. Curie, Paris 1992

Acknowledgement

This work was financially supported by DFG graduate school 826 "Spurenanalytik von Elementspezies".