

## Selective photoionisation of lutetium isotopes

A.B. D'yachkov, S.K. Kovalevich, A.V. Labozin, V.P. Labozin, S.M. Mironov,  
V.Ya. Panchenko, V.A. Firsov, G.O. Tsvetkov, G.G. Shatalova

**Abstract.** A three-stage laser photoionisation scheme intended for enriching the  $^{176}\text{Lu}$  isotope from natural lutetium was considered. An investigation was made of the hyperfine structure of the second excited state  $5d\ 6s\ 7s \rightarrow ^4D_{3/2}$  with an energy of  $37\ 194\ \text{cm}^{-1}$  and the autoionisation state with an energy of  $53\ 375\ \text{cm}^{-1}$  of the  $^{176}\text{Lu}$  and  $^{175}\text{Lu}$  isotopes. The total electron momentum of the autoionisation level and the constant  $A$  of hyperfine magnetic interaction were determined. Due to a small value of the isotopic shift between  $^{176}\text{Lu}$  and  $^{175}\text{Lu}$ , appreciable selectivity of their separation may be achieved with individual hyperfine structure components. The first tentative enrichment of the  $^{176}\text{Lu}$  isotope was performed to a concentration of 60%–70%.

**Keywords:** lutetium,  $^{177}\text{Lu}$  isotope, laser-induced photoionisation, hyperfine structure, autoionisation, AVLIS technology.

### 8. Introduction

Recent years have seen active introduction of atomic vapour laser isotope separation (AVLIS) technique intended for separation of stable isotopes. The first commercial isotope separation realised by the AVLIS technique was reported in Ref. [1] concerned with ytterbium isotope separation. A technology for enriching the  $^{150}\text{Nd}$  isotope [2] was developed at the National Research Centre 'Kurchatov Institute'; this isotope is required as a source in the quest of the double neutrinoless beta decay and the neutrino mass. In progress at the present time is the engineering of an industrial facility capable of producing, in three years, the requisite 100 kg of neodymium with a  $^{150}\text{Nd}$  concentration of about 60%. This facility is diagrammed in Fig. 1. The facility comprises a complex of copper vapour lasers (CVLs), a complex of dye lasers, and a separator complex. The CVL complex consists of a master amplifier and amplifiers, the laser pulse duration is equal to 30 ns, and the pulse repetition frequency is 10 kHz. The total average output power of the laser radiation complex amounts to 1000 W. This radiation is employed to pump wavelength-tunable dye lasers. The dye laser complex comprises four oscillator–amplifier chains in accordance with the number of photoionisation stages.

A.B. D'yachkov, S.K. Kovalevich, A.V. Labozin, V.P. Labozin,  
S.M. Mironov, V.Ya. Panchenko, V.A. Firsov, G.O. Tsvetkov,  
G.G. Shatalova National Research Centre 'Kurchatov Institute',  
pl. Akad. Kurchatova 1, 123182 Moscow, Russia;  
e-mail: tsvetkov@imp.kiae.ru

Received 28 June 2012  
Kvantovaya Elektronika 42 (10) 953–956 (2012)  
Translated by E.N. Ragozin

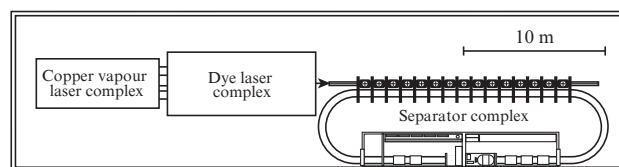
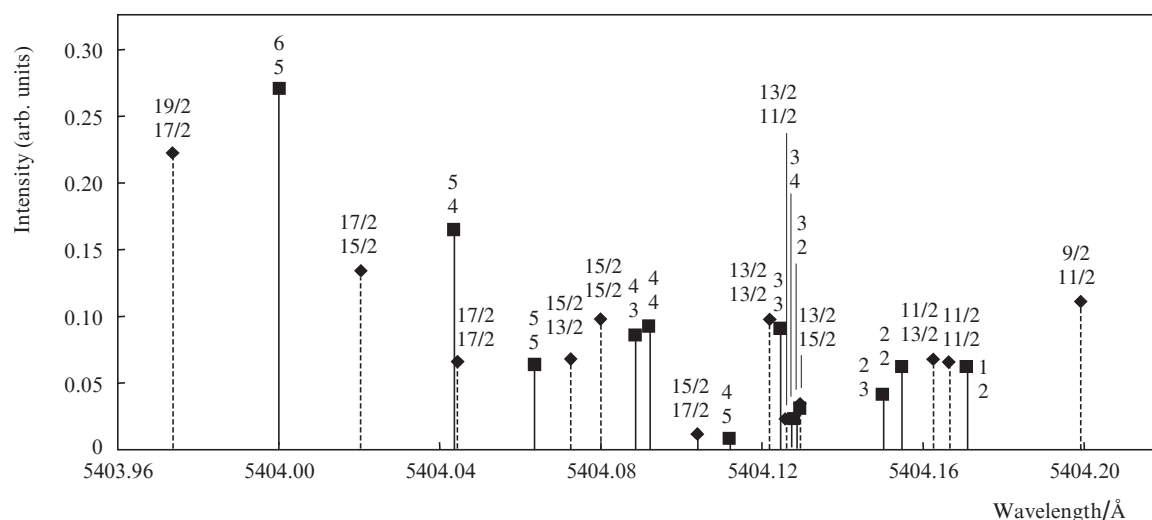


Figure 1. Schematic of the AVLIS facility.

The oscillators produce wavelength-stabilised laser radiation with a spectral width of 100 MHz in the visible range, which is directed to the separator chamber on passing through the amplifiers. The total average output power of the dye laser complex amounts to 450 W. The separator complex consists of 30 separator cells accommodated in 15 vacuum modules. The atomic stream formed in the evaporator of a separator cell passes through the working volume filled with laser radiation and experiences selective photoionisation. The resultant ions of the desired isotope are extracted by the electric field to the product collector from the main stream, while the atoms of other isotopes remain neutral and arrive at the waste collector. The separator cell was described at length in Ref. [2]. The production capacity of the facility is equal to 100 g per day of neodymium with a  $^{150}\text{Nd}$  isotope concentration of 60%.

However, the neodymium enrichment is not the only task which may be solved using the facility under design. Recently, the  $^{176}\text{Lu}$  isotope with a concentration of 60%–70% has come into high demand on the market [3]. This isotope is employed for the production of the radionuclide  $^{177}\text{Lu}$ , which is used in nuclear medicine. Lutetium has no volatile compounds, and therefore its isotopes cannot be separated by the centrifugal technique, which has been well elaborated in Russia. The efficiency of electromagnetic method of  $^{176}\text{Lu}$  isotope separation from the natural mixture, which contains 2.59% of  $^{176}\text{Lu}$  and 97.41% of  $^{175}\text{Lu}$ , is also low due to its low initial concentration. The present work is concerned with the development of the AVLIS technology for producing enriched  $^{176}\text{Lu}$ .

The optical spectrum of lutetium is not as rich as with other rare-earth elements. Few absorption lines may be used for the first excitation stage in the separation technology. The authors of Refs [4, 5] came up with several schemes of two-stage lutetium photoionisation with sufficiently large cross sections; however, they make use of transitions in the 400–500-nm wavelength range, where the use of CVL-pumped dye lasers is inefficient. That is why there is good reason to consider another excitation scheme for the AVLIS technology, specifically, a three-stage photoionisation scheme. From the standpoint of oscillator strengths, for the first two excitation stages



**Figure 2.** HFS of the  $5d6s^2D_{3/2} (0 \text{ cm}^{-1}) \rightarrow 5d6s6p \ ^4F_{5/2} (18505 \text{ cm}^{-1})$  transition. The solid lines stand for the  $^{175}\text{Lu}$  components and the dotted lines for the  $^{176}\text{Lu}$  components. The figures above the peaks indicate the quantum numbers of the HFS components of the upper (the upper figure) and lower (the lower one) levels. From here on we specify the wavelengths in a vacuum.

advantage can be taken of the  $5d6s^2D_{3/2} (0 \text{ cm}^{-1}) \rightarrow 5d6s6p \ ^4F_{5/2} (18505 \text{ cm}^{-1}) \rightarrow 5d6s7s \ ^4D_{3/2} (37194 \text{ cm}^{-1})$  transition [6]. Also important is the magnitude of the isotopic shift between  $^{175}\text{Lu}$  and  $^{176}\text{Lu}$ , especially due to the fact that these isotopes have mutually overlapping level hyperfine structures (HFSs). Figure 2 shows the hyperfine isotopic structure of the  $5d6s^2D_{3/2} (0 \text{ cm}^{-1}) \rightarrow 5d6s6p \ ^4F_{5/2} (18505 \text{ cm}^{-1})$  transition. The relative intensities of the peaks were calculated according to Ref. [7] and are given without regard for isotope densities.

The hyperfine splitting  $\Delta E_F$  relative to the level's centre of gravity for both isotopes was calculated from the constants  $A$  and  $B$  of the magnetic dipole and electric quadrupole interactions known for this transition [8, 9];

$$\Delta E_F = \frac{A}{2} K + \frac{B}{8} \frac{3K(K+1) - 4I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$

for  $J \geq 1, I \geq 1$ ,

where  $I$  is the nuclear spin ( $I = 7/2$  ( $^{175}\text{Lu}$ ),  $I = 7$  ( $^{176}\text{Lu}$ ));  $J$  is the total angular momentum of the electron shell;  $K = F(F+1) - J(J+1) - I(I+1)$ ;  $F$  is the total angular momentum of the atom ( $F = I+J, I+J-1, \dots, |I-J|$ ); when  $J = 1/2, B = 0$ . The level's centre of gravity  $E_{cg}$  is determined from the relation

$$\sum_F (E_F - E_{cg})(2F+1) = 0.$$

Despite the small isotopic shift (388 MHz [10]), several components of  $^{175}\text{Lu}$  and  $^{176}\text{Lu}$  are sufficiently widely spaced, which makes it possible to excite  $^{176}\text{Lu}$  with a high selectivity. These transitions may be employed for the first excitation stage in the three-stage lutetium photoionisation scheme.

## 9. Measurement techniques

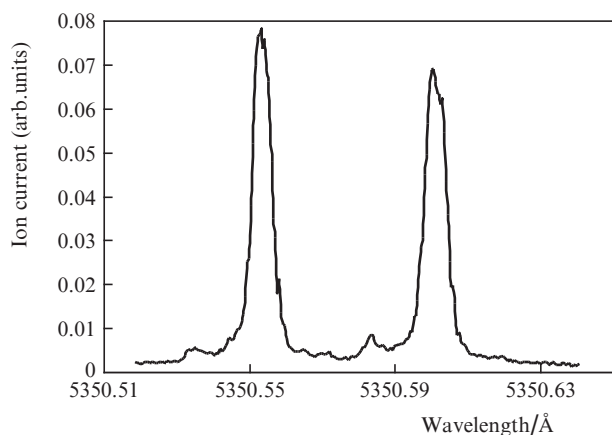
The optical lutetium photoionisation scheme was investigated on a facility intended for spectroscopic experiments with narrow collimated atomic beams as well as for determining the isotopic composition of the ions produced under selective laser-induced photoionisation. The facility consists of a vacuum chamber with an evacuation system and an evaporator, a quadrupole MS-7302 mass spectrometer, and a system for

luminescence detection. The radiation of single-mode CVL-pumped dye lasers is used to excite the atoms. The laser linewidth is  $\Delta\nu = 100$  MHz. The dye lasers can be scanned in wavelength by changing the control voltage at the dispersion elements of laser resonators [11]. To monitor the radiation wavelength, use was made of a precision meter with a measurement uncertainty of  $\pm 0.0002 \text{ \AA}$ . In the vacuum chamber, the laser beam intersects the atomic beam directly in the ionisation chamber of the ion source of the mass spectrometer. The directions of the atomic and laser beams and the ion optical axis of the mass spectrometer are mutually perpendicular. The ions in the mass spectrometer were recorded with a secondary electron multiplier (SEM). The facility is equipped with a system for luminescence detection with a photomultiplier; its special feature consists in that the spontaneous emission is recorded in the ion optical axis of the mass spectrometer in the opposite direction to the ion stream. The facility was controlled and data were recorded in the on-line regime. The software, which was realised in the LabView (National Instruments) environment, performed the necessary operations: received data from the wavelength meter and the signals from the photomultiplier and the SEM, and changed the control voltage with an interface block, thereby tuning or stabilising the lasers in wavelength. The acquired data were stored, processed, and promptly estimated.

## 10. Experimental results

The spectroscopic investigation of the HFS of the second transition  $18505 \text{ cm}^{-1} \rightarrow 37194 \text{ cm}^{-1}$ , for which we failed to find literature data, was supposedly carried out for the first time. In doing this, the laser was tuned to the HFS components of the first transition. The laser of the second stage was tuned in the neighbourhood of the line with  $\lambda = 5350 \text{ \AA}$  and excited atoms to the sublevels of the  $5d6s7s \ ^4D_{3/2}$  state, while the CVL radiation ionised the atoms to the continuum. The photoionisation spectrum of the  $^{176}\text{Lu}$  isotope is exemplified in Fig. 3.

Table 1 shows the experimentally derived structure of the second excited state of lutetium. It is noteworthy that we discovered no noticeable isotopic shift in comparison with  $^{175}\text{Lu}$



**Figure 3.** Ion current of  $^{176}\text{Lu}$  as a function of second-stage laser wavelength. The left peak: the  $11/2 \rightarrow 13/2$  transition; the right peak: the  $11/2 \rightarrow 11/2$  transition (the first stage:  $11/2 \rightarrow 11/2$ ,  $\lambda = 5404.162 \text{ \AA}$ ).

**Table 1.**

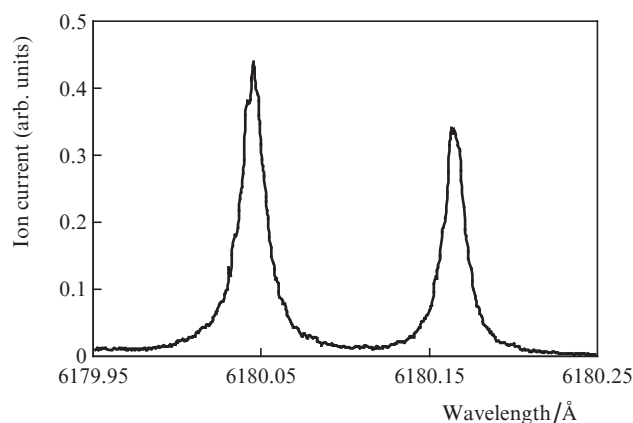
Isotope	$F$	$E/\text{cm}^{-1}$	$\Delta E_F/\text{cm}^{-1}$	$E_{\text{cg}}/\text{cm}^{-1}$
$^{176}\text{Lu}$	11/2	37 193.67	-0.32	37 193.99
	13/2	37 193.84	-0.15	
	15/2	37 194.04	0.05	
	17/2	37 194.27	0.28	
$^{175}\text{Lu}$	2	37 193.74	-0.25	37 193.99
	3	37 193.85	-0.14	
	4	37 194.00	0.01	
	5	37 194.19	0.20	

at this stage, and therefore the separation selectivity may be achieved, like at the first stage, using separate HFS components.

The use of autoionisation transitions at the photoionisation stage significantly relaxes the requirement on the power of the laser system, which has a highly favourable effect on the cost of the AVLIS technique. The search for autoionisation transitions was performed by scanning the wavelength of the third-stage laser for stabilised wavelengths of the first- and second-stage lasers tuned to the corresponding HFS components. In our experiments the laser was tuned over the 6000–6300 Å range. Among the several autoionisation transitions discovered experimentally, the transition with  $\lambda = 6180 \text{ \AA}$  proved to be the brightest one. Transitions to the autoionisation state took place from four HFS levels of the second excited state  $5d6s7s \ ^4D_{3/2}$ : for instance, from levels with  $F = 11/2, 13/2, 15/2$ , and  $17/2$  for  $^{176}\text{Lu}$ . One resonance peak was observed for components with  $F = 11/2$  and  $17/2$ ; there were two peaks for levels with  $F = 13/2$ . In view of selection rules, this signifies that the autoionisation state is split into two HFS levels with  $F = 13/2$  and  $15/2$  (Fig. 4). A similar situation was observed for the  $^{175}\text{Lu}$  isotope, whose autoionisation state is split into levels with  $F = 3$  and  $4$ .

Since transitions were observed to only two HFS components, the electron angular momentum of the level is  $J_{\text{ai}} = 1/2$ . Table 2 gives the structure of the autoionisation level and the hyperfine interaction constant  $A$ . Since  $J_{\text{ai}} = 1/2$ , the constant  $B = 0$ .

We estimate the uncertainty in determining the HFS energy levels (Tables 1 and 2) at  $0.01 \text{ cm}^{-1}$ . They arise primarily from: (i) the uncertainty in determining the resonance peak positions in our experimental spectra, as well as from the uncertainty in the calibration of the wavelength meter, which was



**Figure 4.** Ion current of  $^{176}\text{Lu}$  as a function of third-stage laser wavelength. The left peak: the  $15/2 \rightarrow 15/2$  transition; the right peak: the  $15/2 \rightarrow 13/2$  transition (the first stage:  $\lambda = 5404.017 \text{ \AA}$ ; the second stage:  $\lambda = 5350.655 \text{ \AA}$ ).

**Table 2.**

Isotope	$F$	$E/\text{cm}^{-1}$	$\Delta E_F/\text{cm}^{-1}$	$E_{\text{cg}}/\text{cm}^{-1}$	$A/\text{GHz}$
$^{175}\text{Lu}$	3	53 374.89	-0.13	53 375.02	1.73
	4	53 375.12	0.10		
$^{176}\text{Lu}$	13/2	53 374.84	-0.16	53 375.00	1.24
	15/2	53 375.15	0.15		

performed using the known wavelengths in the luminescence spectra; (ii) the instability of the laser wavelength, which stemmed from the fact that the laser wavelengths were performed sequentially with the use of one meter.

## 11. Isotope separation

The lutetium evaporation temperature is only slightly higher than the neodymium evaporation temperature and, according to our practical experience, the separation cell designed for the separation of neodymium isotopes can be employed for lutetium without any modifications. Table 3 shows the results of several test separation runs.

**Table 3.**

Productivity/mg h <sup>-1</sup>	22.5	22.6	2.1	2.2	4.3	3.5	3.9
$^{176}\text{Lu}$ isotope concentration (%)	29.3	32.1	45.3	47.7	60.5	67.9	68.9

For a  $^{176}\text{Lu}$  fraction of 60%–70% in the product, the specific productivity of the separation cell turns out to be approximately 10 times lower than for  $^{150}\text{Nd}$ . This is due to the fact that the initial concentration of the desired isotope  $^{176}\text{Lu}$  is two times lower than that of  $^{150}\text{Nd}$ . Furthermore, with the use of the ground state and the first metastable state in neodymium, respectively 56% and 25% of the number of desired isotope atoms are involved in the photoionisation process. In the separation of the  $^{176}\text{Lu}$  isotope, use was made of only one HFS component ( $17/2 \rightarrow 19/2$ ) of the first transition (see Fig. 2). As a result, the fraction of irradiated  $^{176}\text{Lu}$  atoms involved in photoionisation is equal to 22%.

The productivity rises with increasing input atomic flux, but this is attended with a lowering of the desired isotope

concentration in the final product, which is due to the scattering of the dense atomic stream in the working volume.

The AVLIS facility productivity in lutetium can be estimated at 10 g per day<sup>-1</sup>; considering the operating costs and the present-day state of the market, this gives us hope for successful commercialisation of the projected facility.

## 12. Conclusions

The HFS of the excited and autoionisation states of the stable isotopes <sup>175</sup>Lu and <sup>176</sup>Lu was investigated for the first time. We developed a scheme for the photoionisation of atomic lutetium suitable for isotope separation by the AVLIS technique with the use of CVL-pumped dye lasers. Test separation runs for enriching the <sup>176</sup>Lu isotope were made. We showed that the projected industrial AVLIS facility intended for obtaining enriched <sup>150</sup>Nd may be employed without significant modifications for producing enriched <sup>176</sup>Lu, which is in stable demand on the market.

**Acknowledgements.** The authors express their appreciation to A.P. Babichev and M.A. Burlak for the mass spectrometric analysis of enriched lutetium samples as well as to A.V. Bobkov and A.E. Sereda for the quantitative chemical analysis.

This work was supported by the Russian Foundation for Basic Research (Grant Nos 11-02-120034-ofi-m-2011 and 12-08-00561-a-2012).

## References

1. Tkachev A.N., Yakovlenko S.I. *Kvantovaya Elektron.*, **25** (11), 971 (1998) [*Quantum Electron.*, **28** (11), 945 (1998)].
2. Babichev A.P. et al. *Kvantovaya Elektron.*, **35** (10), 879 (2005) [*Quantum Electron.*, **35** (10), 879 (2005)].
3. Knapp F.F., Mirzadeh S., et al. *Proc. 5th Int. Conf. on Isotopes* (Brussels, 2005).
4. Bekov G.I., Vidolova-Angelova E.P. *Kvantovaya Elektron.*, **8** (1), 227 (1981) [*Sov. J. Quantum Electron.*, **11** (1), 137 (1981)].
5. Xu C.B., Xu X.Y., Ma H., Li L.Q. *J. Phys. B: At. Mol. Opt. Phys.*, **26**, 2827 (1993).
6. Corliss C.H., Bozeman W.R. *Experimental Transition Probabilities for Spectral Lines of Seventy Elements* (Washington, DC: US Government Printing Office, 1962; Moscow: Mir, 1968).
7. Axner O. et al. *Spectrochim. Acta, Part B*, **59**, 1 (2004).
8. Zimmermann D., Zimmermann P., Aepfelbach G., Kuhnert A. *Z. Physik. A*, **295**, 307 (1980).
9. Nunneman A., Zimmermann D., Zimmermann P. *Z. Physik. A*, **290**, 123 (1979).
10. Jin W.G., Wakasugi M., et al. *Phys. Rev. A*, **49** (2), 762 (1994).
11. Grigoriev I.S., Dyachkov A.B., et al. *Techn. Dig. Conf. on Laser Application and Technologies (LAT)* (Moscow, 2002) p. 296.