

ISOTOPE LASER EMISSION

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Experimental study of the degree of extraction of a target isotope in the process of laser photoionisation separation of lutetium isotopes

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Abstract. **The degree of target isotope extraction is studied in application to the problem of extracting the radioisotope ^{177}Lu by means of the laser photoionisation method using the developed high-efficiency three-stage scheme $5d6s^2\,{}^2D_{3/2}$–$5d6s6p\,{}^4F^o_{5/2}$–$5d6s7s\,{}^4D_{3/2}$–53375.049 cm^{-1}.**

Keywords: *laser isotope separation, laser photoionisation of atoms, ^{177}Lu radioisotope.*

1. Introduction

One of the most important parameters of any isotope separation process is the degree of extraction of the target isotope, i.e., its fraction extracted from the feed flux (initial raw material) into the product flux. In the laser photoionisation method the target isotope selectively ionised by laser radiation appears at the collector due to its extraction from the flux by the electric field. Simultaneously the deselecting process related to the scattering of neutral atoms in the working volume occurs. Due to rare collisions occurring in rarefied vapour, a small fraction of atoms is deflected from rectilinear motion and has a possibility to get to the product collector, too. As a result, the isotope-selective flux of photoions is mixed with the nonselective flux of scattered atoms, which leads to the reduction of the target isotope concentration in the product. With the increase in the degree of target isotope extraction at the expense of photoionisation not only the efficiency of the process, but also the concentration of the target isotope in the product grows, so that for a laser method this parameter is of primary importance [1].

The degree of target isotope extraction depends on the laser radiation intensity at every stage of the photoionisation scheme. With increasing intensity, the degree of extraction grows practically linearly at small intensities. A further increase in intensity leads to the deceleration of the growth and results in saturation. The physical cause of saturation is the depopulation of the atomic ground states. In this case, we assume that all atoms that occupied the ground level are converted into photoions. For example, in the photoionisation of the ^{177}Lu radioisotope from the sublevel $F = 5$ of the hyperfine structure ($F = 2, 3, 4, 5$) of the ground state $5d6s^2\,{}^2D_{3/2}$ the degree of extraction can attain $0.34 \cdot 0.7 = 0.24$. Here the factor 0.7 is the population of the ground state at the lutetium evaporation temperature of 1700 °C, and the factor 0.34 is the fraction of atoms that occupy the sublevel $F = 5$.

From the formal point of view, saturation of transitions is a necessary but not sufficient condition for full photoionisation. Strictly speaking, the situations are possible, in which saturation will not lead to complete extraction of atoms, occupying the ground state. For example, it is possible that in the photoionisation scheme at one of sequential stages the decay (deactivation) of an excited state occurs. The point is that in the search for a photoionisation scheme it is by far not always possible to use the levels known in advance from the literature. Often one has to search for new levels and transitions, as a rule, high-energy ones, by scanning the laser wavelength of the second or the third stage and fixing photoion resonances. In this case, the detection of a photoion resonance still does not mean that the atomic excitation occurs from the level, to which the atom is excited by the laser radiation at the previous stage. It is possible that the last level decays into a few lower lying states and the detected transition implements the excitation of atoms from one of these states. In this case, the fraction of atoms that will be converted into photoions is determined by the decay scheme even in the case of signal saturation, and this may lead to significant losses of target isotope atoms. Practically, to confirm the high efficiency of the chosen scheme of photoionisation a direct measurement of the extraction degree is used [2].

2. Experiment

The direct measurement of the degree of extraction by means of photoionisation is performed using the setup (Fig. 1) that consists of a vacuum chamber with evacuation system (*1*), a thermal evaporator (*2*), a quadrupole MS-7302 mass spectrometer (*3*), an optical multipass system, and an extraction system. A system of diaphragms installed above the evaporator forms a narrow atomic beam (*4*) (orthogonal to the laser beam) with an opening angle 3° (the Doppler width of the absorption line is ~100 MHz).

The multipass optical system is comprised of two vacuum flanges (*5*) with laser mirrors (*6*) installed on movable mounts implementing the mirror alignment from the atmosphere side. One of the flanges has a window (*7*) for coupling the laser radiation into the chamber. Due to multiple reflections of the laser beam with the diameter 1 cm, the multipass system forms the working volume with the cross section 1 × 8 cm. The atomic beam exiting the evaporator appears in an 8-cm-

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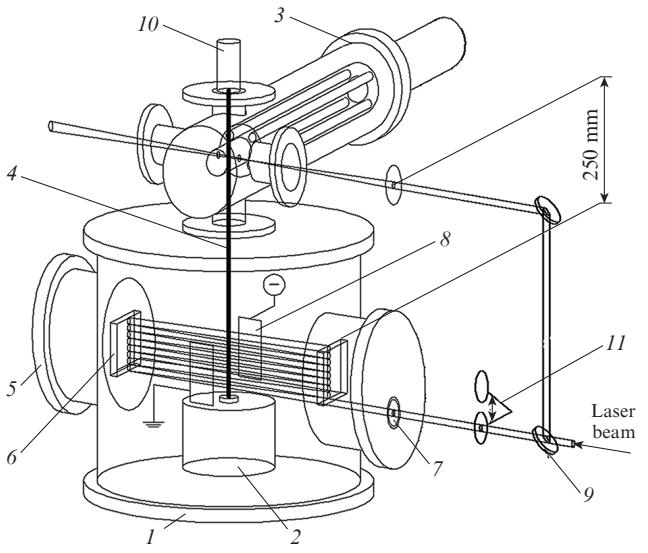

Figure 1. Setup for direct measurement of the degree of extraction due to photoionisation:
(*1*) vacuum chamber; (*2*) thermal evaporator; (*3*) quadrupole MS-7302 mass spectrometer; (*4*) atomic beam; (*5*) flange of the multipass system; (*6*) mirror of the multipass system; (*7*) entrance window for laser radiation; (*8*) extraction system; (*9*) semitransparent mirror; (*10*) deposition silica sensor; (*11*) shutter of the laser beam.

long illuminated zone along the direction of the beam propagation, which at the repetition rate of the laser pulses 10 kHz provides the probability 0.97 of irradiating each atom at least once. In the immediate proximity of the working volume there is an extracting system (*8*), consisting of the negatively charged collector of photoions and the grounded deflector intended to flatten the electric field strength. In the direction of the atomic beam propagation, at the distance 250 mm from the working volume a quadrupole mass spectrometer is installed. The atomic beam passing through the working volume arrives at the ionisation chamber of the mass spectrometer, where it crosses the testing laser beam, split from the main laser beam consisting of three coaxial beams of laser radiation having different wavelengths by means of a beam splitter (*9*). The zone of interaction between the testing laser beam and the atomic beam in the mass spectrometer has the shape of a cylinder 2 mm long and 2 mm in diameter (determined by the size of the diaphragm installed across the laser beam immediately before the interaction zone). The axis of the atomic beam, the laser beam axis, and the ion-optical axis of the mass spectrometer are mutually orthogonal. The registration of ions is implemented using a secondary electron multiplier (SEM).

Above the mass spectrometer a sensor of the silica meter of 'Micron-5' film thickness (OOO 'Izovak', Republic of Belarus) is mounted that allows one to measure the intensity of the atomic beam passed through the working volume and the mass spectrometer region of interaction.

The parameters of photoionisation efficiency are determined by the changes in mass spectrometer and silica sensor signals that occur when the laser beam shutter (*11*) is opened or closed. The change in the mass spectrometer signal recording the photoions produced by the testing beam determines the degree of depopulation of the initial atomic level. The decrease in the signal from the silica sensor allows one to measure a decrease in the atomic flux due to the photoionisation extraction of the target isotope.

For selective photoionisation, we used the radiation of single-mode pulsed dye lasers pumped by copper vapour lasers. The spectral width of the generated line was 100–120 MHz, the pulse duration was 20 ns, and the pulse repetition rate was 10 kHz. Detailed information about the laser system and its performance can be found in Refs [3, 4].

3. Results and discussion

The experimental studies were performed with lutetium having the natural isotope composition. To increase the sensitivity of measurements the laser radiation wavelengths were tuned to produce photoionisation of the isotope ^{175}Lu (its natural concentration is $C_{175} = 0.97$). The obtained values of the photoionisation efficiency parameters can be directly attributed to the radioisotope ^{177}Lu, since the atoms of these isotopes have similar nuclear spins (7/2) and, therefore, a similar hyperfine structure (Fig. 2). By analogy with the radioisotope ^{177}Lu, the ground state of ^{175}Lu consists of four sublevels with the quantum numbers $F = 2, 3, 4, 5$, which are populated proportionally to their statistical weight $2F + 1$. For example, in photoionisation from the sublevel $F = 5$ the maximal extraction degree amounts to $0.34 \times 0.7 = 0.238$, where the factor 0.7 is the population of the ground state at the lutetium evaporation temperature 1700 °C, and the factor 0.34 is the fraction of atoms populating the sublevel $F = 5$.

The extraction degree α was calculated using the formula

$$\alpha = \frac{\Phi_1 - \Phi_2}{C_{175}\Phi_1}, \tag{1}$$

where Φ_1 and Φ_2 are the indications of the silica sensor for the shutter (*11*) being open and closed, respectively. The ground state depopulation degree was determined as

$$\rho = \frac{I_1 - I_2}{I_1}, \tag{2}$$

where I_1 and I_2 are the photoion currents measured by the mass spectrometer, tuned to the mass 175, for the shutter (*11*) being open and closed, respectively. Table 1 presents the degrees of extraction and ground state depopulation obtained using the transitions between the hyperfine structure components of the levels $^2D_{3/2}$ ($F = 5$), $^4F^o_{5/2}$ ($F = 6$), $^4D_{3/2}$ ($F = 5$), and $X_{1/2}$ ($F = 4$) (the photoionisation scheme 5–6–5–4).

Table 1. Parameters of the photoionisation scheme 5–6–5–4.

Mean intensity of laser radiation/W cm^{-2}			Degree of ground state depopulation	Extraction degree
First stage	Second stage	Third stage		
0.025	0.025	2.5	0.88 ± 0.05	0.17 ± 0.03

Table 2 presents the results of measuring the degree of extraction using other possible photoionisation channels. From this table one easily sees that the extraction degree is in good correlation with the population of the initial level.

To increase the selectivity one can use a delay of laser pulses of the second and the third stages with respect to the pulse of the first stage, based on the fact that the lifetime of the first excited state is large enough and amounts to 472 ns [5]. However, at this delay the reduction of the extraction degree is unavoidable, since only part of atoms that have pop-

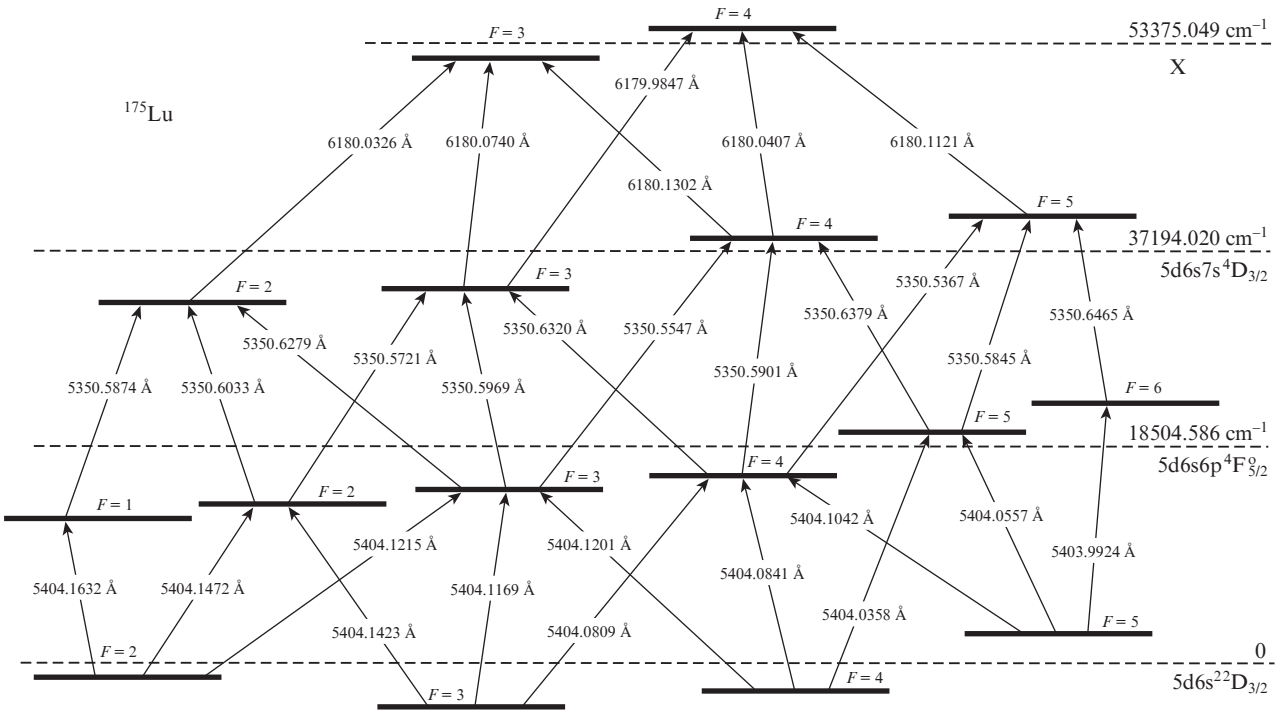

Figure 2. Schematic diagrams of possible channels of ^{175}Lu photoionisation. The symbol X denotes the level with the energy 53375.049 cm^{-1}, for which the electron configuration is unknown.

Table 2. Measured degrees of extraction using different photoionisation channels. The result for the 5–6–5–4 scheme presented in Table 1 is taken for a unit of measurement.

Photoionisation scheme	Relative population of the initial state	Relative extraction degree
5–6–5–4	1	1
4–4–5–4	0.81	0.75 ± 0.1
3–4–5–4	0.62	0.6 ± 0.1
2–1–2–3	0.46	0.5 ± 0.1

ulated the first excited state $F = 6$ after the end of the first-stage laser pulse, rather than all atoms that populated the initial level $F = 5$, are subjected to photoionisation. The fraction of these atoms in the saturation regime is equal to the ratio $g_2/(g_1 + g_2) = 0.54$, where g_1 and g_2 are the statistical weights $2F + 1$ of the ground and excited state, respectively. Direct measurements have shown that for the delay of the laser pulses of the second and the third stage equal to 18 ns, the extraction degree amounts to 0.6 ± 0.1 of the maximal value, measured under synchronous pulses, which is in good agreement with the presented estimate.

4. Lifetimes of energy levels

The maximal extraction degree of 0.17 attained in the experiments amounts to 0.7 of the theoretical limit. To clarify the cause of incomplete extraction we considered the lifetimes of the excited states involved in the photoionisation scheme.

The first excited state 5d6s6p^{4}F$^o_{5/2}$ is a long-lived one (the lifetime 472 ± 24 ns) [5]. No information about the lifetime of the second excited state 5d6s7s^{4}D$_{3/2}$ could be found in the scientific literature. According to Ref. [6], in the spectral region 5057–7758 Å seven channels of fast decay of this level due to dipole transitions exist; therefore, its lifetime is expected to be small, on the order of 1–10 ns [5].

The lifetime of the level 5d6s7s^{4}D$_{3/2}$ was determined experimentally. The laser pulses of the first and the second stage (coincident in time) with the mean intensity of the beams ~ 10 mW cm^{-2} essentially populated this level. After the end of the excitation pulses, the ionising pulse (the third stage) arrived at the working volume. The value of the photoion signal was proportional to the population of the level 5d6s7s^{4}D$_{3/2}$ at the time of the ionising pulse arrival. The photocurrent was measured at different delays of this pulse. For this purpose, a spatial delay line of variable length (up to 12 m, or 40 ns) in the optical path of the beam was used. Figure 3 presents the

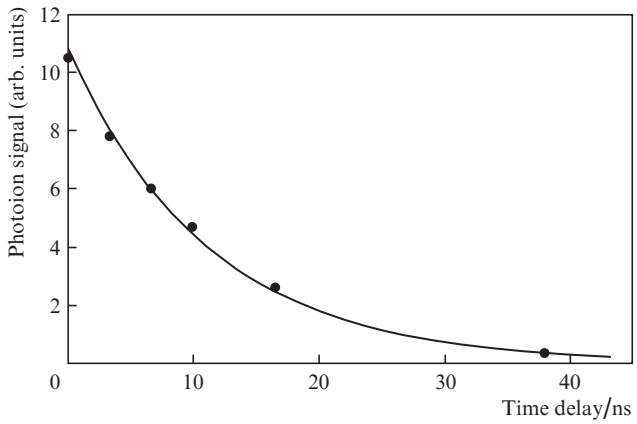

Figure 3. Dependence of the ^{175}Lu photoion signal on the time delay of the ionising laser pulse with respect to the laser pulse of the first stage (points) and the approximating curve $y = 10.8\exp(-x/11.5)$.

values of the photoion signal at different delays of the ionisation pulse relative to the laser pulse of the first stage. The experimental points are in good agreement with the exponential decay curve with the time constant 11.5 ns.

5. Effect of the second excited state decay on the extraction degree

The effect of decay of the second excited state $5d6s7s\,^4D_{3/2}$ on the extraction degree can be estimated by considering multistage photoionisation described by the equations of averaged balance of the particle number in different states, commonly referred to as rate equations [7]:

$$\frac{dN_1}{dt} = -N_1 w_{12} + N_2 w_{12}, \tag{3}$$

$$\frac{dN_2}{dt} = N_1 w_{12} - N_2(w_{12} + w_{23} + a_{2m}) + N_3 w_{23}, \tag{4}$$

$$\frac{dN_3}{dt} = N_2 w_{23} - N_3(w_{23} + w_{34} + a_{3m}) + N_4 w_{34}, \tag{5}$$

$$\frac{dN_4}{dt} = N_3 w_{34} - N_4 a_i, \tag{6}$$

$$\frac{dN_i}{dt} = N_4 a_i, \tag{7}$$

$$\frac{dN_m}{dt} = N_2 a_{2m} + N_3 a_{3m}, \tag{8}$$

where N_1, N_2, N_3, and N_4 are the numbers of particles in the ground state, the first and the second excited state, and in the autoionisation state, respectively; N_i is the number of photoions; a_i is the rate of photoionisation from the autoionisation state; N_m is the number of particles, lost by the system as a result of decay of the second and the third state; $w_{ij} = I_{ij}\sigma_{ij}/(h\nu_{ij})$ is the rate of the laser-induced transitions from state i to state j; I_{ij} is the intensity of laser radiation; σ_{ij} is the transition cross section; ν_{ij} is the transition frequency; and a_{im} is the decay rate of state i via transitions to metastable states. In these equations, we neglected the statistical weights of the quantum states, as well as the processes of spontaneous decay, opposite to the stimulated laser-induced transitions.

Generally, solving this system of equations is rather difficult; however, in a number of practically important cases simple solutions are possible that allow better understanding of the influence of different factors. Consider a particular case, when the rate of laser-induced transitions at the third stage w_{23} appears to be much smaller than those of the first and the second transitions, which, in turn, are much smaller than the rate of photoionisation a_i:

$$a_i \gg w_{12} \approx w_{23} \gg w_{34} \approx a_{3m}. \tag{9}$$

Let us also neglect the spontaneous decay of the first excited state, assuming the lifetime of this level to be much greater than the laser pulse duration, i.e.,

$$a_{2m} = 0. \tag{10}$$

Let the exciting laser pulses have a rectangular shape, the duration τ and be exactly synchronised in time with the onset at $t = 0$. In this case, two temporal scales appear in the system of rate equations. The fast time scale of the order of $1/w_{12} \approx 1/w_{23}$ characterises the establishment of particle distribution in the ground and excited states

$$N_1 = N_2 = N_3 = (N_1 + N_2 + N_3)/3, \tag{11}$$

$$N_4 = \frac{N_3 w_{34}}{a_i}. \tag{12}$$

When this distribution is established, the system of excited levels 'traces' quasi-stationary the population of the second excited state (N_3) that changes slowly in correspondence with the other time scale $1/w_{34}$. Then, considering the slow time scale, we add equations (3), (4), (5) and using Eqn (11) obtain the equation for the total population

$$\frac{d(N_1 + N_2 + N_3)}{dt} = -\frac{(N_1 + N_2 + N_3)}{3}(w_{34} + a_{3m}) \tag{13}$$

with the initial condition

$$(N_1 + N_2 + N_3)|_{t=0} = N_0. \tag{14}$$

The solution of this equation for N_3 has the form

$$N_3(t) = \frac{N_0}{3}\exp\left(-\frac{w_{34} + a_{3m}}{3}t\right). \tag{15}$$

Substituting Eqn (15) into Eqn (12) and then into Eqn (7) and integrating over time from zero to τ, we arrive at the expression for the fraction of atoms, photoionised to the moment of the laser pulse termination:

$$\frac{N_i}{N_0} = \frac{w_{34}}{w_{34} + a_{3m}}\left[1 - \exp\left(-\frac{w_{34} + a_{3m}}{3}\tau\right)\right]. \tag{16}$$

One can easily see that if the statistical weights of the states are taken into account, then the factor 1/3 in the exponent should be replaced with the ratio $g_3/(g_1 + g_2 + g_3)$.

The factor $w_{34}/(w_{34} + a_{3m})$ in Eqn (16) describes the competition of the processes of photoionisation and spontaneous decay of the second excited state. This factor relatively quickly (for $w_{34} = a_{3m}$) attains the values 0.5; however, its further

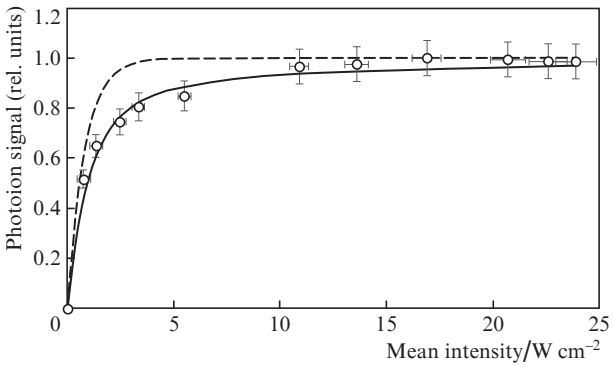

Figure 4. Dependence of the photoion signal on the mean intensity of laser radiation of the third stage (experimental points). The solid curve presents the calculation using Eqn (16) with the cross section $\sigma_{34} = 1.2 \times 10^{-14}$ cm^2 and the decay rate $a_{3m} = 87$ MHz, the dashed curve plots the same result but for $a_{3m} = 0$.

growth with the increase of w_{34} is strongly decelerated, and even at $w_{34} = 5a_{3m}$ the factor amounts only to 0.83. The achievement of the photoionisation degree above 0.9 requires increasing the rate w_{34} to values that are beyond the reasonable limits.

Figure 4 presents the experimental dependence of the photoion signal on the intensity of laser radiation at the third stage. It is seen that the signal continues to grow until the mean intensity reaches 15 W cm^{-2}. The dependence agrees well with the one calculated using Eqn (16) with the cross section $\sigma_{34} = 1.2 \times 10^{-14}$ cm^2 and the decay rate $a_{3m} = 87$ MHz, corresponding to the lifetime of the second excited state. For the mean intensity of laser radiation at the third stage, 2.5 W cm^{-2}, used in the experiments, the signal achieves ~ 0.7 of its maximal value, which corresponds to the results of measuring the extraction degree due to photoionisation.

6. Conclusions

Direct measurements of the degree of target isotope extraction have shown that in the developed three-stage scheme of lutetium photoionisation it is relatively easy to reach the extraction degree 0.7–0.8 of the theoretical limit, which with the population of the ground state taken into account amounts to 70%. A further essential increase in the extraction degree is hard because of the competition between the excitation of the atom from the second excited state and the decay of this state, for which the lifetime was measured to be 11.5 ± 0.5 ns. The increase in photoionisation selectivity at the expense of delay of the laser pulses of the second and third stages relative to the pulse of the first stage leads to the reduction of the extraction degree by 40%, which corresponds to the theoretical estimate. The results of our experiments provide a good ground for the development of laser photoionisation method of producing the radionuclide ^{177}Lu for application in medicine.

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