 

*LASER PHOTOIONISATION* DOI: 10.1070/QEL16021

# Effect of amplified spontaneous emission on selectivity of laser photoionisation of the $^{177}$Lu radioisotope

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***Abstract.*** **A significant deselecting effect of amplified spontaneous emission has been observed in the experiments on selective laser photoionisation of the $^{177}$Lu radioisotope according to the scheme $5d6s^2\,{}^2D_{3/2} \rightarrow 5d6s6p\,{}^4F^g_{5/2}$ (18505 cm$^{-1}$) → $5d6s7s\,{}^4D_{3/2}$(37194 cm$^{-1}$) → autoionisation state (53375 cm$^{-1}$). The effect is conditioned by involvement of non-target isotopes from the lower metastable level $5d6s^2\,{}^2D_{5/2}$(1994 cm$^{-1}$) into the ionisation process. Spectral filtering of spontaneous emission has allowed us to significantly increase the selectivity of the photoionisation process of the radioisotope and to attain a selectivity value of $10^5$ when using saturating light intensities.**

***Keywords:*** *selectivity, laser photoionisation, $^{177}$Lu isotope, dye laser, amplified spontaneous emission, metastable state, mass spectrum.*

## 1. Introduction

Emission of pulsed dye lasers (DLs), which are hitherto considered as the most effective for selective photoionisation of isotopes in the atomic vapour, always contains frequency-nonselective amplified spontaneous emission (ASE). Alongside the frequency stability and spectral linewidth, ASE also affects the process of stepwise ionisation, and thus the efforts of laser system developers are always directed towards attainment of a low intensity of this background component. For its suppression, spectral and spatial filtering is applied [1, 2], and precise correlation of durations of the lasing and pump pulses, together with synchronisation of the pulse arrival times in the dye laser amplifiers, is carried out [3]. A typical relative intensity of ASE for 'master oscillator power amplifier' chains constitutes a few percents [4, 5], and, despite the fact that spontaneous emission can be displayed in a wide spectral range of luminescence of a laser dye (tens of nanometres), the deselecting effect of the background component may be significant.

In the case of laser isotope separation, the selectivity $S$ is determined by the ratio of photoionisation probabilities of the target ($W_p$) and non-target ($W_{np}$) isotopes, and can be found experimentally from the content $C_p$ of the target isotope's photoions, provided that its initial content is $C_f$:

$$S = \frac{W_p}{W_{np}} = \frac{C_p(1 - C_f)}{(1 - C_p)C_f}. \quad (1)$$

The deselecting effect of ASE is associated with the excitation of non-target isotopes by raiation from the spontaneous background spectrum and their subsequent ionisation. It is easy to estimate the selectivity $S_{ASE}$ of a single-stage process (or, for example, for ionisation in accordance with a single-colour two-stage scheme), which is only restricted by the background component, through the laser emission parameters:

$$S_{ASE} \approx \frac{P/\Delta\nu}{P\alpha_{ASE}/\Delta\nu_{ASE}} = \frac{\Delta\nu_{ASE}}{\alpha_{ASE}\Delta\nu}, \quad (2)$$

where $\Delta\nu$ is the lasing linewidth; $\alpha_{ASE}$ is the proportion of non-selective background emission in the laser beam; $\Delta\nu_{ASE} = c\Delta\lambda_{ASE}/\lambda^2$; $\Delta\lambda_{ASE}$ is the ASE spectral width; $\lambda$ is the wavelength; and $c$ is the speed of light.

At typical values $\Delta\nu$ = 100–120 MHz, $\Delta\lambda_{ASE}$ = 20–40 nm and $\alpha_{ASE}$ = 1%–2%, the selectivity is greater than $10^7$. This exceeds substantially the selectivity defined by the spectral contrast of the laser line contour. At the laser contour width of 100 MHz (FWHM, Gaussian distribution), the spectral light intensity at the spectral distance corresponding to a typical isotope shift of 1 GHz constitutes $\sim 10^{-4}$ of the intensity at the line centre, and, consequently, the selectivity is limited to the value of $\sim 10^4$. Thus, in a single-stage process, the background component of laser radiation does not virtually affect the selectivity. This may explain the fact that we were unable to find the works dedicated to studying the effect of ASE on selectivity of laser photoionisation.

In practice, however, most photoionisation schemes have two or three excitation stages, depending on the ionisation potential of the separated element. Even if the selectivity is only attained in a single transition, it is necessary to take into account the possibility of exciting the non-target isotopes by background emission from other non-selective stages:

$$S_{ASE} \approx \frac{P_l/\Delta\nu_l}{\sum_i \alpha_{ASE\,i} P_i/\Delta\nu_{ASE\,i}}, \quad (3)$$

where $i$ is the excitation stage number, and $P_i$ is the power of $i$th laser. When deriving this formula, it has been assumed that the selectivity of the photoionisation process is mainly attained within a single excitation stage, the background emission has a uniform distribution of intensity with respect to wavelengths, the absorption profile in atomic vapour corresponds to the lasing linewidth, and the summation is performed over the stages for which the background emission is

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capable of carrying the atoms to the upper level of the selective transition.

Typically, the resonant excitation cross section $\sigma_{exc} = 10^{-12}-10^{-13}$ cm$^2$ of the selective transition in the first or second stage is several orders greater than the ionisation cross section $\sigma_{ion} = 10^{-15}-10^{-18}$ cm$^2$. Accordingly, in order to obtain efficient photoionisation, the emission intensity in the last stage should significantly exceed that in the first stages, which, according to (3), leads to a proportional selectivity reduction. In addition, the losses in selectivity are increased if the ionising radiation is generated at the edge of the laser dye's contour gain, where the value of $\alpha_{ASE}$ is sharply increased and can reach tens of percent [6]. As a result, for example, at $P_3 = 1000P_1$ and $\alpha_{ASE\,3} = 10\%-20\%$, we have $S_{ASE} \approx 10^3$, which immediately eliminates the possibility of achieving high enrichment by an isotope with the initial content of $C_f < 10^{-3}$.

We have observed a strong nonselective impact of background emission in the ionising beam in the course of experiments on laser ionisation of the $^{177}$Lu radioisotope [7] which represents a great interest for nuclear medicine as a promising therapeutic radionuclide [8–10]. The present work is dedicated to experimental investigation of this effect and to a search for the techniques of dealing with it in order to increase the photoionisation selectivity.

## 2. Experimental technique

Natural lutetium consists of two isotopes, $^{175}$Lu and $^{176}$Lu, with a concentration of 97.4 % and 2.6%, respectively. In order to obtain the $^{177}$Lu isotope ( $T_{1/2}$ = 6.74 days), metallic lutetium was irradiated within 24 hours by a flux of thermal neutrons of $1.5\times10^{13}$ cm$^{-2}$ s$^{-1}$ [neutron capture reaction $^{176}$Lu (n,$\gamma$) $^{177}$Lu]. The initial content $C_f$ of radioactive atoms, having been determined by means of the gamma-spectrometric analysis, was $10^{-5}$. Photoionisation of lutetium atoms was conducted using a three-stage scheme through the auto-ionisation state (AIS, Figure 1). This scheme had been previously studied in [7, 11].

Excitation of the $^{177}$Lu isotope (nuclear spin $I$ = 7/2) was performed through the hyperfine structure levels with the following values of the total atomic momentum $F$: 5 → 6 → 5 → 4. The lines of the non-target isotopes $^{175}$Lu ($I$ = 7/2) and $^{176}$Lu ($I$ = 7), being the nearest to the line of the hyperfine structure $^{177}$Lu (transition 5 → 6), are separated from that line by –1.16 and 0.93 GHz, respectively [7]. Photoionisation was performed by means of emission from a DL system consisting of three single-mode oscillators with wavelength stabilisation [12] and a third-stage emission amplifier [13]. The system was pumped by copper vapour lasers. Main parameters of the laser system are listed in Table 1.

Laser beams are telescoped up to the beams with a diameter of 12 mm and collimated; their spatial convergence into a single beam is performed using translucent and dichroic mirrors. The output power of the beams corresponds to the previously measured saturation intensities of relevant transitions 10 mW cm$^{-2}$, 10 mW cm$^{-2}$ and 3 W cm$^{-2}$ [7]. Next the radiation is directed into a vacuum chamber with a high-temperature evaporator and an MS-7302 quadrupole mass spectrometer. The diaphragm system above the evaporator restricts the full expansion angle of the atomic beam to 3° (the Doppler width of the absorption contour is ~100 MHz). The laser beam crosses the atomic beam immediately in the ion source ionisation chamber of the quadrupole mass spectrometer. The directions of atomic and laser beams, and also of the mass spectrometer's optical axis, are mutually perpendicular. Recording of ions is conducted using a secondary electron multiplier.

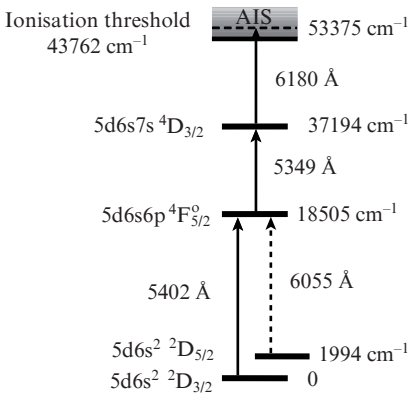


**Figure 1.** Three-stage photoionisation scheme for LuI.

## 3. Results and discussion

The mass spectrum of photons, produced in the initial experiment on selective photoionisation of $^{177}$Lu, is shown in Fig. 2. Maintaining a significant background photoionisation signal from $^{175}$Lu when locking the first-stage beam suggests the presence of the excitation channel of non-target isotopes ($^{175}$Lu and $^{176}$Lu) to the first level by amplified spontaneous emission of the second and/or third stages. At $\alpha_{ASE\,2} \approx 0.3\%$ and $P_2$ = 10 mW, the non-selective impact of this beam,

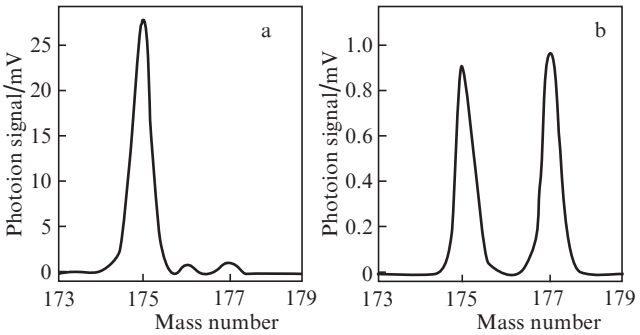


**Figure 2.** Photocurrent mass spectrograms for lutetium isotopes (a) without and (b) with spectral filtering of ASE in the ionising beam. Initial content of the $^{177}$Lu isotope is $10^{-5}$.

**Table 1.** Main parameters of the laser system.

| Excitation stage number | Centre wavelength/Å (DYE) | Pump wavelength/nm | Average output power/W | $\alpha_{ASE}$ (%) | Spectral width (FWHM)/MHz | Pulse duration (FWHM)/ns | Repetition rate/kHz |
|---|---|---|---|---|---|---|---|
| 1 | 5402.6 (PM556) | 510 | 0.1 – 0.3 | 0.3 | | | |
| 2 | 5349.1 (PM556) | 510 | 0.1 – 0.3 | 0.3 | 100 – 120 | 15 – 20 | 10 |
| 3 | 6180.1 (SR640) | 578 | 3 – 4 | 15 | | | |

according to (3), is exceedingly small. On the other hand, the background emission and beam power on the third stage are significantly higher ($\alpha_{ASE\,3} = 15\%$, $P_3 = 3$ W). Despite the fact that the ASE spectrum on this stage is shifted from the first transition wavelength (540 nm), it contains the emission transforming the atoms from the lower metastable level $5d6s^2\ ^2D_{5/2}$ to the first excited state (see Fig. 1), the population of which at the lutetium evaporation temperature of 1600 °C is rather high – about 0.2. Thus, the transition wavelength $5d6s^2\ ^2D_{5/2} \rightarrow 5d6s6p\ ^4F^o_{5/2}$ (605.5 nm) is close to the wavelength of the luminescence maximum for the SR640 dye we have used. Suppression of the background component of the third-stage laser emission by means of an external diffraction grating has led to virtually complete disappearance of the background photoion signal of $^{175}$Lu.

To determine the causes of appearance of a high background component, we studied spectral dependences of average emission power and ASE for the third-stage laser (Fig. 3). The minimum value $\alpha_{ASE\,3} \approx 0.4\%$ was observed at the maximum lasing wavelength for the SR640 dye (607 nm). Its growth up to 15% occurred at the generation bandwidth edge (618 nm), where, due to the reduced cross section of stimulated (induced) transitions of the laser dye, a slowdown of the rate of induced inversion removal in the active medium and, as a consequence, an increase in the spontaneous component intensity, were observed. The sharp rise in $\alpha_{ASE\,3}$ was facilitated by a decrease in the lasing power (Fig. 3) and, accordingly, in the emission intensity at the amplifier input, and also by a decrease in the laser pulse duration.

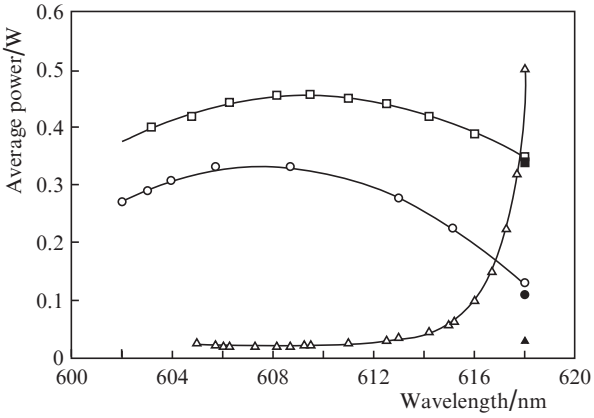


**Figure 3.** Average emission power of the third-stage laser (○ – generator, □ – generator + amplifier) and ASE (△ – generator + amplifier) vs. lasing wavelength. The points ●, ■, ▲ correspond to the case of a dichroic mirror placed between the generator and amplifier. The pump power of the master oscillator and amplifier is 3 and 9 W, respectively; the dye is SR640 (0.25 mmol $L^{-1}$); the solvent is ethanol–water (1/1).

Background emission at the output of the generator – DL amplifier system is formed by four main channels schematically shown in Fig. 4. The first two channels are weak. Spontaneous emission in these channels undergoes one- and two-fold gain in the active medium. The ASE light power measured for channel 1 (in the case of screening the generator) and channels 1 + 2 (in the case of screening the highly reflecting resonator mirror), even in the absence of generation, turned out small and did not exceed 10 mW. When connecting channels 3 and 4 (in the case of screening the folding resonator mirror), the ASE power increases up to 2.5 W. Despite the fact that 'separation' of channels 3 and 4 is rather complicated, the largest contribution to ASE is made obviously by channel 4, where spontaneous emission in the active medium is maximally amplified. In this case, spontaneous emission 'starts' from the amplifier 'cuvette' in the master oscillator direction, passes through the master oscillator cuvette, is reflected from the highly reflecting resonator mirror (a scheme with a diffraction grating in the grazing incidence regime) and returns to the amplifier, thereby being increased during four passes. Obviously, if generation occurs at the tuning curve's edge, introduction of the emission losses between the master oscillator and amplifier in the dye luminescence peak region leads to a decrease in the background signal through channels 2, 3 and 4. A multilayer dielectric mirror coating has been used for this purpose, ensuring high transmittance in the auto-ionisation transition ($T \approx 0.9$) at a wavelength of 618 nm, and high reflectance in the wavelength range of 603–610 nm, which corresponds to the peak of luminescence for the SR640 dye ($R > 0.8$) (Fig. 5).

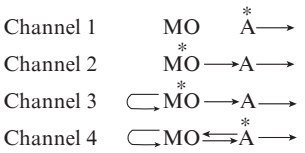


**Figure 4.** Four-channel scheme to form the background emission: (MO) master oscillator; (A) amplifier; asterisks and arrows indicate the start and the propagation direction of spontaneous emission.

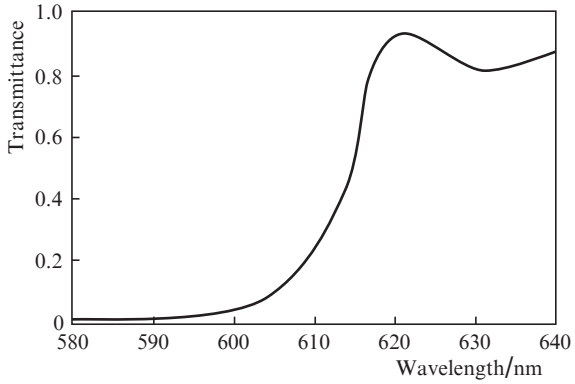


**Figure 5.** Mirror transmittance vs. emission wavelength (at normal incidence).

As a result, the $\alpha_{ASE\,3}$ value for the third-stage laser beam has decreased by 25 times (to 0.6%) without a noticeable decrease in the light output power $P_3$ (Fig. 3). The mass spectrum of photons recorded in the experiment on selective photoionisation for the $^{177}$Lu isotope with the use of such spectral filtering is presented in Fig. 2b. The current amplitude for the non-target $^{175}$Lu isotope has decreased by more than 20 times. Thus, the content of photoions in the $^{177}$Lu isotope constitutes 0.5, which corresponds to the selectivity of $S = 10^5$ at the initial radioisotope content of $C_f = 10^{-5}$. Additional purification of the background component in the ionising beam, performed by means of an external diffraction grating, has only twice reduced the current signal for the $^{175}$Lu isotope, which indicates the efficiency of the ASE spectral filtering.

It should be noted that the formation of the background component on channel 4 can be effectively suppressed by

increasing the distance between the amplifier and oscillator, so that the background radiation from the amplifier would arrive to the generator by the end of its pump pulse. At the pump pulse duration of 25 ns, this distance should exceed 4 m, which is sometimes difficult to implement in practice.

The cross impact of ASE on selectivity of multistage laser photoionisation, when background radiation on one stage affects the transition selectivity on the other, may occur for a variety of the widespread schemes used for selective photoionisation of isotopes of neodymium, ytterbium, etc. For example, in the neodymium photoionisation scheme (Fig. 6), a background component of the third stage laser emission may result in excitation of non-target isotopes from the metastable state with an energy of 1128 $cm^{-1}$ (the transition wavelength is 639 nm).

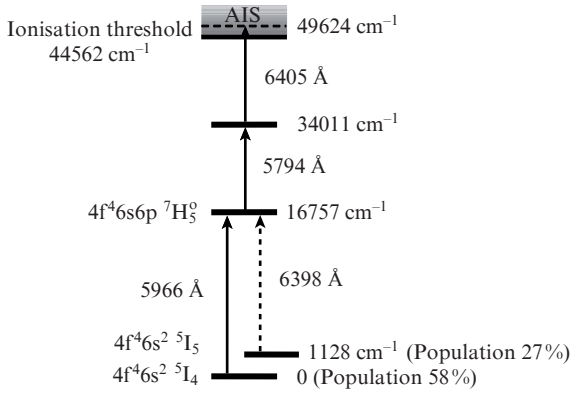


**Figure 6.** Three-stage photoionisation scheme for NdI.

In order to achieve a high photoionisation efficiency, the third-stage emission intensity should exceed by two orders the emission intensity on the first and second stages [14], which at $\alpha_{ASE} \approx 10\%$ restricts the selectivity to $\sim 10^4$.

In photoionisation of the $^{168}$Yb isotope according to the scheme 556 nm → 581 nm → 583 nm [15], the third-stage background component may cause excitation of non-target isotopes on the second transition stage (581nm) possessing a large isotopic shift, on which main selectivity of the photoionisation process has been achieved.

## 4. Conclusions

The experiments on selective laser photoionisation of the $^{177}$Lu radioisotope have shown that ASE may produce a significant effect on the selectivity of a multistage process. The selectivity reduction occurs if the transitions from the metastable or lower atomic levels to excited levels fall into background emission spectrum of a certain stage (commonly, photoionisation stage), which leads to involvement of non-target isotopes into the ionisation process. Evaluation and experimental results show that, in some cases, the selectivity of ionisation can be limited to the value of about $10^3$, which excludes the possibility of obtaining high enrichment by the isotope with an initial content of less than $10^{-3}$. To achieve a higher selectivity, it is necessary to pay special attention to the selection of the photoionisation scheme, taking into account the spectral lasing characteristics of the last-stage laser dye. The above-described approach to spectral filtering of background radiation may become efficient in that case.

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