

## ТЕХНОЛОГІЯ ВИРОБНИЦТВА СЕНСОРІВ

## SENSORS PRODUCTION TECHNOLOGIES

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### NEW LASER PHOTOIONIZATION ISOTOPE SEPARATION SCHEME WITH AUTOIONIZATION SORTING OF HIGHLY EXCITED ATOMS FOR HIGHLY RADIOACTIVE ISOTOPES AND PRODUCTS OF ATOMIC ENERGETICS

*A. V. Glushkov<sup>1</sup>, G. P. Prepelitsa<sup>1</sup>, A. Yu. Pogosov<sup>1</sup>, V. G. Shevchuk<sup>2</sup>,  
A. A. Svinarenko<sup>1</sup>, A. V. Ignatenko<sup>3</sup> and E. V. Bakunina<sup>3</sup>*

<sup>1</sup>Odessa National Polytechnical University, Odessa, Ukraine

<sup>2</sup>I. I. Mechnikov Odessa National University, Odessa, Ukraine

<sup>3</sup>Odessa State Environmental University, Odessa, Ukraine

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**Abstract.** We present new optimal scheme of the separating highly radioactive isotopes and products of atomics energetics such as <sup>133,135,137</sup>Cs and others, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further autoionization or DC electric field pulse ionization.

**Key words:** laser photoionization method, highly radioactive isotopes, new scheme

### НОВА ЛАЗЕРНО-ФОТОІОНІЗАЦІЙНА СХЕМА ПОДІЛЕННЯ ІЗОТОПІВ З АВТОІОНІЗАЦІЙНОЮ СОРТИРОВКОЮ ВИСОКОЗБУДЖЕНИХ АТОМІВ ДЛЯ ВИСОКО РАДІОАКТИВНИХ ІЗОТОПІВ ТА ПРОДУКТІВ АТОМНОЇ ЕНЕРГЕТИКИ

*О. В. Глушков, Г. П. Препелица, О. Ю. Погосов, В. Г. Шевчук,  
А. А. Свиноренко, Г. В. Игнатенко, О. В. Бакунина*

**Анотація.** Представлена нова оптимальна схема лазерного поділення високо радіоактивних ізоотопів, продуктів атомної енергетики, зокрема, таких як <sup>133,135,137</sup>Cs та інші, яка базується на лазерному збудженні атомів ізоотопів у ридбергові стани та подальшій автоіонізації або іонізації імпульсом електричного поля.

**Ключові слова:** лазерний фотоіонізаційний метод, високо радіоактивні ізоотопи, нова схема

**НОВАЯ ЛАЗЕРНО-ФОТОИОНИЗАЦИОННАЯ СХЕМА РАЗДЕЛЕНИЯ ИЗОТОПОВ  
С АВТОИОНИЗАЦИОННОЙ СОРТИРОВКОЙ ВЫСОКО ВОЗБУЖДЕННЫХ АТОМОВ  
ДЛЯ ВЫСОКО РАДИОАКТИВНЫХ ИЗОТОПОВ И ПРОДУКТОВ АТОМНОЙ ЭНЕРГЕТИКИ**

*А. В. Глушков, Г. П. Препелица, А. Ю. Погосов, В. Г. Шевчук,  
А. А. Свинарченко, А. В. Игнатенко, О. В. Бакунина*

**Аннотация.** Представлена новая оптимальная схема лазерного разделения высоко радиоактивных изотопов, продуктов атомной энергетики таких как  $^{133,135,137}\text{Cs}$  и других, базирующаяся на лазерном возбуждении атомов изотопов в ридберговские состояния и дальнейшей автоионизации или ионизации импульсом электрического поля.

**Ключевые слова:** лазерный фотоионизационный метод, высоко радиоактивные изотопы, новая схема

To number of the very actual problem of modern nuclear technology, quantum and photoelectronics is related a search of the effective methods for isotopes and nuclear isomers separation and obtaining especially pure substances at atomic level [1–6]. The basis for its successful realization is, at first, carrying out the optimal multi stepped photo-ionization schemes for different elements and, at second, availability of enough effective UV and visible range lasers with high average power (Letokhov, 1977, 1979, 1983; etc) [1,2]. The standard laser photo-ionization scheme may be realized with using processes of the two-step excitation and ionization of atoms by laser pulse. The scheme of selective ionization of atoms, based on the selective resonance excitation of atoms by laser radiation into states near ionization boundary and further photo-ionization of the excited states by additional laser radiation, has been at first proposed and realized by Letokhov et al (Letokhov, 1969, 1977) [1,2]. It represents a great interest for laser separation of isotopes and nuclear isomers. The known disadvantage of two-step laser photoionization scheme a great difference between cross-sections of resonant excitation  $\sigma_{\text{exc}}$  and photo-ionization  $\sigma_{\text{ion}}$ . It requires using very intensive laser radiation for excited atom ionization. The same is arisen in a task of sorting the excited atoms and atoms with excited nuclei in problem of creation of  $\gamma$ -laser on quickly decayed nuclear isomers. Originally, Goldansky and Letokhov (1974) [7] (see also [5]) have considered a possibility of creating a  $\gamma$ -laser, based on a recoilless transition between lower nuclear levels and shown that a  $\gamma$ -laser of this type in the 20–60 keV region is feasible. A feature of design is operation based on relatively short-lived isomer nuclear states with lifetime of 0,1 to 10 sec. These authors has estimated the minimal number of excited nuclei required for

obtaining appreciable amplification and possibility of producing sufficient amounts of excited nuclei by irradiation of the target with a thermal neutron beam or by resonant  $\gamma$ -radiation. It is important that low-inertia laser selection of a relatively small friction of excited nuclei of a given composition from the target by the two-step method of selective laser photoionization of atoms with excited nuclei by the radiation from two lasers is principally possible. But, it is obvious that here there is a problem of significant disadvantage of the two-step selective ionization of atoms by laser radiation method. The situation is more simplified for autoionization resonance's in the atomic spectra, but detailed data about characteristics of these levels are often absent (Letokhov, 1977, 1983; Glushkov & Ivanov, 1986, 1992) [1–16]. The key problems here are connected with difficulties of theoretical studying and calculating the autoionization resonance characteristics. Several new optimal schemes for the laser photoionization sensors of separating heavy isotopes and nuclear isomers are proposed [4,12]. It is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further autoionization and DC electric field ionization mechanisms.

Let us remind that in a classic scheme the laser excitation of the isotopes and nuclear isomers separation is usually realized at several steps: atoms are resonantly excited by laser radiation and then it is realized photo ionization of excited atoms. In this case photo ionization process is characterized by relatively low cross section  $\sigma_{\text{ion}}=10^{-17}-10^{-18}\text{cm}^2$  and one could use the powerful laser radiation on the ionization step. This is not acceptable from the energetics point of view [1–8].

The alternative mechanism is a transition of atoms into Rydberg states and further ionization by electric field or electromagnetic pulse. As result,

requirements to energetic of the ionized pulse are decreased at several orders. The main feature and innovation of the presented scheme is connected with using the DC electric field (laser pulse) autoionization on the last ionization step of the laser photoionization technology. There is a principal difference of the simple ionization by DC electric field. The laser pulse ionization through the autoionized states decay channel has the advantages (more high accuracy, the better energetics, universality) especially for heavy elements and isotopes, where the DC electric field ionization from the low excited states has not to be high effective. This idea is a key one in the realization of sorting the definite excited atoms with necessary excited nuclei of the  $A^+$  kind, obtained by optimal method of selective photo-ionization of the A kind atoms at the first steps. The suitable objects for modeling laser photoionization separation technology are the isotopes of alkali element Cs, long-lived transuranium elements etc.

We considered the isotopes of  $^{133,135,137}\text{Cs}$ ,  $^{88,90}\text{Sr}$  and  $^{127,129}\text{I}$ . For example, the resonant excitation of the Cs atoms can be realized with using dye lasers with lamp pumping (two transitions wavelengths are:  $6^2\text{S}_{1/2} \rightarrow 7^2\text{P}_{3/2}$  4555Å and  $6^2\text{S}_{1/2} \rightarrow 7^2\text{P}_{1/2}$  4593Å). In table 1 there are listed the energy parameters for different states of the caesium, obtained in the different approximations (Derevianko & Porsev, 2003; Glushkov et al, 2008; Khetselius, 2009) (from ref. [19]). It is useful to remind the corresponding hyperfine splitting energy ( $6^2\text{S}_{1/2}$ , transition 4–3) of Cs: experimental data-  $\Delta\nu(\text{F},\text{F}') = 9192,64\text{MHz}$ ;  $\Delta\text{E}(\text{F},\text{F}') = 306,630 \cdot 10^{-3} \text{ cm}^{-1}$ ; theoretical data -  $\Delta\nu(\text{F},\text{F}') = 9177,80\text{MHz}$ ;  $\Delta\text{E}(\text{F},\text{F}') = 306,135 \cdot 10^{-3} \text{ cm}^{-1}$  (Khetselius, 2009). The next step is in the further excitation to the Rydberg S,P,D states with main quantum number  $n=31-37$  (the optimal value  $n=35$ ).

The final step is the autoionization of the Rydberg excited atoms by a laser pulse or DC electric field pulse ionization and output of the created ions. The scheme will be optimal if an atom is excited by laser radiation to state, which has the decay probability due to the autoionization (pulse ionization) higher than the radiation decay probability. So, one could guess that the accurate data on the autoionization states energies and widths and the same parameters for the DC Stark resonances are needed. The consistent and accurate theoretical approach to calculation of these characteristics is based on the operator perturbation theory formalism [10]

and corresponding advances relativistic version [23] with quantum defect approximation [17]. In table 2 we present the test results for the Stark resonances  $(n,n_1,n_2,m)$  energies in the Rb spectra for the electric field strength  $\varepsilon=2.189 \text{ kV/cm}$  [23–25].

Table 1

Valent electron ionization energies (in atom. units) of the  $^{133}\text{Cs}$ :  $\varepsilon^{\text{RHF}}$  –one-configuration Hartree-Fock data, relativistic Hartree-Fock (RHF);  $\varepsilon^{\text{RHF}} + \delta\varepsilon^{\text{RHF}}$  – the same data, but with account for the correlation corrections (Derevianko & Porsev, 2005;  $\varepsilon^{\text{QED}}$  – QED perturbation theory data (Khetselius, 2009);  $\varepsilon^{\text{Exp}}$ - experimental data (see text)

State	$\varepsilon^{\text{RHF}}$	$\varepsilon^{\text{RHF}} + \delta\varepsilon^{\text{RHF}}$	$\varepsilon^{\text{QED}}$	$\varepsilon^{\text{Exp}}$
$6s_{1/2}$	0,12737	0,14257	0,14295	0,14310
$6p_{1/2}$	0,08562	0,09198	0,09213	0,09217
$6p_{3/2}$	0,08379	0,08951	0,08960	0,08964
$7s_{1/2}$	0,05519	0,05845	0,05862	0,05865
$7p_{1/2}$	0,04202	0,04385	0,04391	0,04393
$7p_{3/2}$	0,04137	0,04303	0,04309	0,04310

Table 2

The test results for the Stark resonances  $(n,n_1,n_2,m)$  energies ( $\text{cm}^{-1}$ ) in the Rb spectra for the electric field strength  $\varepsilon=2.189 \text{ kV/cm}$

$n_1n_2m$	Exp. [25]	Theory [24] [23]	Present paper
23,0,0	133,1	132,8 132,9	133,0
22,0,0	157,0	157,1 157,2	157,1
21,1,0	161,1	159,5 160,6	160,9
20,2,0	163,9	163,2 163,7	163,9
21,0,0	185,2	184,2 184,8	185,1
20,1,0	186,3	185,4 185,8	186,2
20,0,0	217,2	214,6 214,9	216,9

Analysis of data of the table 2 shows that our theoretical approach provides physically reasonable agreement with the accurate experimental data by Kleppner et al [25]. The same data have been received for the caesium atom and other alkali atoms. In principle, the detailed analysis indicates on the non-regular features in the Stark resonances energies  $E_r$  and widths  $\Gamma^{(n,n_2,m)}$  behaviour. It can be easily explained if to compare the electric field strength  $\tilde{F} = \tilde{n}^4 \varepsilon$  with classic ionization threshold  $F_{\text{ion}}$ . In this case one could find sufficiently definite correlation between the values  $\Gamma/2$  and  $f = (F - F_{\text{ion}}) / F_{\text{ion}}$ . The latter characterizes nearness of the resonance energy to potential barrier top. Here one could obtain the following relations for states with  $n_1 \sim n \gg 1, n_2$  and  $m-1$  (look details in refs. [6,23,24], in particular, in the above-threshold region  $E > 0$ :

$$E_r^{(n,n_2,m)} = \varepsilon_{cl}(\tilde{n}^4 \varepsilon) / 2\tilde{n}^2 \quad (1)$$

$$\Gamma^{(n_1, n_2, m)} = \gamma_{cl}(\tilde{n}^4 \varepsilon) p / \tilde{n}^3 \quad (2)$$

and in the under-threshold one ( $E < 0$ ) as follows:

$$E_r^{(n_1, n_2, m)} = (1 / 2\tilde{n}^2) [\varepsilon_{cl}(\tilde{n}^4 \varepsilon) + \eta \{ (\tilde{n} n^*)^2 \varepsilon - (\tilde{n} / n^*)^2 \eta (n^4 \varepsilon) \}] \quad (3)$$

where  $n^* = n - \delta$  — is the parabolic analog of the effective main quantum number for Rydberg states in the spherical basis,  $\delta = \delta(n_1, n_2, m)$ ,  $p = 2n_2 + m + 1$ ,  $\tilde{n} = n_1 + (m + 1) / 2 - \delta$ ,  $\eta(F) = [-\varepsilon_{cl}(F)]^{3/2}$ .

The equations (1–3) do in fact provide the scaling relations for the Stark resonances. In The value  $\varepsilon_{cl}$  is dependent upon the strength  $\tilde{F} = \tilde{n}^4 \varepsilon$  as follows:

$$\varepsilon_{cl}(\tilde{n}^4 \varepsilon) = 2.14 \cdot (\tilde{n}^4 \varepsilon) - 0.9067 \text{ (at. units)}. \quad (4)$$

Figure 1 demonstrates the scaling effect for the Stark resonances in the alkali atoms and hydrogen. In figure the reduced field strength  $\tilde{F} = \tilde{n}^4 \varepsilon$  ( $F^* = 0.383$ ) is listed at the abscissa axe and reduced resonance width  $\tilde{\gamma}_{cl}$  — the ordinate axe. The following notations are used: states O, •, Δ- states  $|n100\rangle$ ,  $|n101\rangle$ ,  $|n110\rangle$  in the hydrogen in the field with strength  $\varepsilon = 6.5, 8.0$  kV/cm [6]; ▲ — states  $|n1n20\rangle$  with  $n_2 = 0, 1, 1$  ( $p = 1, 3, 5$ ) in Na with  $\varepsilon = 3.59, 6.5$  kV/cm; \* — states  $|n1n20\rangle$  with  $n_2 = 0, 1, 1$  in K with  $\varepsilon = 3.59$  kv/cm; + — states  $|n1n20\rangle$  with  $n_2 = 0, 1, 1$  in Cs with  $\varepsilon = 2.25$  kV/cm and  $\varepsilon = 8.25$  kV/cm; ◆ — states  $|n1n20\rangle$  with  $n_2 = 0, 1, 1$  in Rb with  $\varepsilon = 2.189, 6.416$  kV/cm.

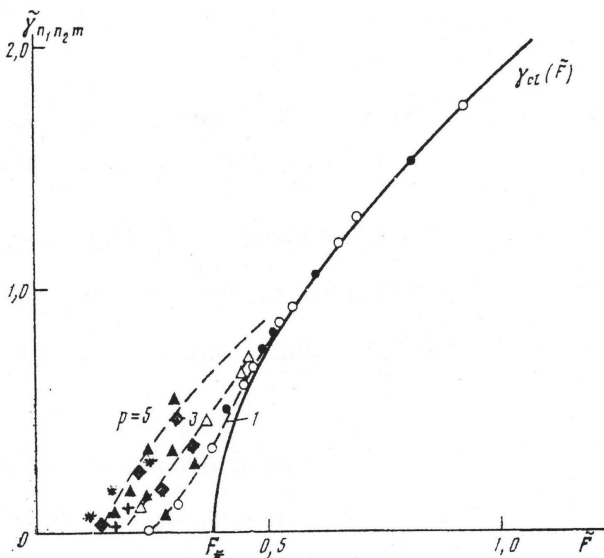


Fig. 1. Scaling for the Stark resonances on spectra of alkali elements (see text)

In whole an analysis shows that the scaling effect is observed for the Stark resonances in spectra of H, Na, K, Rb in the limits of ~3 %, but it is realized

worse for the Cs isotope. The latter is probably explained by increasing the relativistic effects role for this atom. In any case the corresponding data about the Stark resonances energies and widths (lifetimes etc) are needed for the next modeling the optimal schemes of the isotope separation. In this essence the scaling equations (1–3) provide quickly the important data about characteristics of the Stark resonances in the DC electric field and corresponding radiation parameters. In figure 2 we present the numerical modeling results of the excited and ground states populations in the photoionization scheme of the  $^{133,137}\text{Cs}$  isotopes separation process with auto- and electric field ionization by solving the corresponding differential equations system [6, 14].

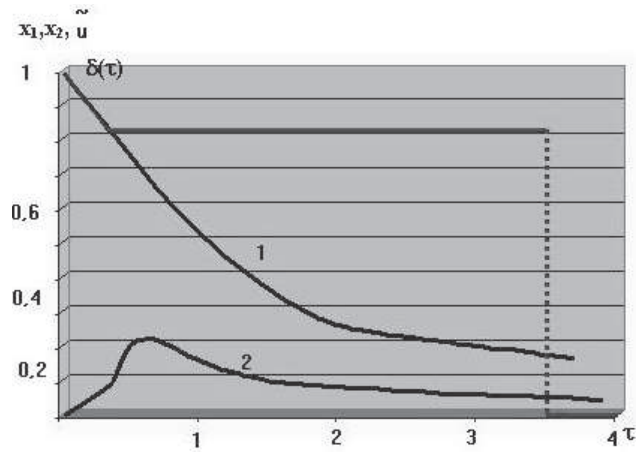


Fig. 2. Results of modelling  $^{133,137}\text{Cs}$  isotopes separation process by the laser photo-ionization method ( $\delta$ +dashed — laser pulse optimal form; see text)

The following definitions are used:  $\delta$ +dashed line is corresponding to optimal form of laser pulse, curves 1 and 2 are corresponding to populations of the ground and excited states of Cs. The  $\delta$ -pulse provides maximum possible level of excitation (the excitation degree is about ~0,25; in experiment (Letokhov, 1983) with rectangular pulse this degree was ~ 0,1). Let us note that the presented results are analogous to the similar modeling results for other elements such as Rb, Ga etc [4, 12]. Indeed, the relative populations for indicated atoms in the highly excited states are very closed to each other, however the absolute values of the radiation parameters for different isotopes naturally differ. Let us remember data regarding the Cs excitation and the ionization cross sections: the excitation cross section at the first step of the scheme is  $\sim 10^{-11} \text{cm}^2$ ; the ionization cross-section from excited  $7^2P_2$  state:  $\sigma_2 = 10^{-16} \text{cm}^2$ , from ground state  $\sigma_2 = 10^{-18} \text{cm}^2$  [2].

One can see that the relation of these cross sections is  $10^5$  and  $10^7$  correspondingly. This fact provides the obvious non-efficiency of standard photoionization scheme. Our estimate for Cs atom ionization cross section for the  $35^2S_{1/2}$  is  $3,7 \cdot 10^{-13} \text{ cm}^2$  that is higher than the corresponding cross section of ionization process by laser pulse in the two- stepped photo ionization [2] scheme ( $\sim 10^{-17} \text{ cm}^2$ ). Using  $\delta$ -pulse indeed provides a quick ionization, but the ionization yield will be less than 100 % because of the sticking on intermediate levels. So, from energetic point of view, this type of ionization can be very perspective alternative to earlier proposed classical two-step and more complicated photoionization schemes (Letokhov, 1983) [1]. The similar situation and analogous conclusions are obtained for the Sr and I isotope separation with the corresponding difference in the energetic and radiative characteristics data. So, one can say here about sufficiently optimal scheme of the separating highly radioactive isotopes and products of atomics energetics such as Cs and others. The key features of the corresponding scheme (technology) are based on the selective laser excitation of the isotopes atoms to the excited Rydberg states and further autoionization (or DC electric pulse ionization). One could remember here that a step of laser isotope separation has to be very important one in solving the modern actual problems of the transmutation of radioactive elements and decreasing the energy loses in the modern atomic energetics cycles [20,21]. One should also note that the considered scheme can be easily implemented to the possible advanced scheme of the  $\gamma$  — laser on quickly decayed nuclear isomers with using laser photoionization sorting excited nuclei  $M^*_{k+1}$  with autoionization mechanism through the Rydberg states [11,22].

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