UDC 535.42, 539.184

S. V. Ambrosov, A. P. Fedchuk, A. V. Glushkov, Ya. I. Lepikh

I. I. Mechnikov Odessa National University, 2, Dvoryanskaya str., Odessa, Ukraine
Odessa National Polytechnical University, 1, Shevchenko av., Odessa Ukraine
Odessa State Environmental University, 15, Lvovskaya str., Odessa, Ukraine
e-mail: serambr@mail.ru

LASER-PHOTOIONIZATION METHOD OF SEPARATION OF THE ISOTOPES: RADIATIVE PARAMETERS FOR ALKALI ELEMENTS

It is proposed an optimal scheme of the separating isotopes alkali elements, which is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further DC electric field ionization. Some radiative parameters data are evaluated for the alkali elements.

The AVLIS (atomic vapour laser isotope separation) and MLIS (molecular laser isotope separation) methods, based on the atomic photoionization and molecular photoionization and photodissociation processes, are well known now and actively developing [1-10]. The laser photoionization method is one of the most perspective methods for the separating isotopes, nuclear isomers and nuclear reactions products [1-20]. The standard laser ionization sensor scheme is usually realized with using a scheme of the multi-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 (look refs. [1,2]). Naturally, this scheme represents a great interest for laser separation of isotopes and nuclear isomers. However, a significant disadvantage of the two-step selective ionization of atoms by laser radiation method is a great difference between cross-sections of resonant excitation σ_{exc} and photo-ionization σ_{ion} ($[\sigma_{exc}/\sigma_{ion}]>10^4\div10^8$). It requires a using very intensive laser radiation for the excited atom ionization. The situation is more simplified for autoionization resonances in the atomic spectra because of the advanced energy parameters, however, the detailed data about characteristics of these atomic states are often absent. Here the main problems are connected with difficulties of theoretical studying and calculating the autoionization resonance characteristics. An account of complex relativistic and correlation effects (continuum states, self-energy diagrams contributions etc.) by means of the traditional quantum-mechanical methods became obligatorily.

In a number of papers (look refs. [10,11,17-21]) a possibility of the selective ionization of atoms, based on the selective resonance excitation of atoms by laser radiation into states near ionization boundary and further ionization decay of excited atoms by external electric field, has been considered. Electric field changes the electron spectra so that the part of discrete spectra levels (near the ionization boundary) part moves into continuum and other levels become by the autoionization ones. The probability of their autoionization decay quickly increases with growth of the main quantum number. The most optimal situation is when atom is excited to state, which has the autoionization probability more than the radiation decay one. To receive an accurate information about optimal laser photoionization sensor scheme, it is necessary to carry the accurate calculation of the process of sequent atomic excitation by laser field (it is the standard task) and probability of ionization of the atoms in the highly excited states (au-

toionization levels) by electric field. Now the accurate calculations of elementary atomic processes in different calculation schemes are intensively carried out, including calculation of characteristics of decay of the autoionization resonances [8-21]. As a rule, non-relativistic approximation has been used [1]. More consistent approach to this problem must be based on the relativistic models [11,17-21], as the most interesting elements for laser isotope separation are heavy ones and a role of relativistic corrections is often very dramatic.

We present a new optimal scheme of the separating isotopes, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionization. Some radiative parameters data are obtained for alkali elements. It should be noted that the excitation and ionization cross-sections of ground and low excited state for these atoms by laser pulse are as follows: the excitation cross-section $\sigma_{exc} = \sigma_1 \sim 10^{-13}$-$10^{-11}$ cm^2, ionization cross-section from excited state: $\sigma_{ion} = \sigma_2 \sim 10^{-18}$-$10^{-17}$cm^2, from ground state $\sigma_2 \sim 10^{-19}$cm^2 [1]. For selective photoionization scheme with excitation to Rydberg *ns, np* states with n = 10-50 and further ionization by the DC electric field (see below) the calculated cross-section values are as follows: $\sigma_2 \sim \times 10^{-15} \div ^{12}$ cm^2. It means that the selective photoionization scheme with using the Rydberg states (autoionization resonances) and ionization by external electric field is quite effective for studied isotopes of alkali atoms from the energetic point of view. But it is arisen a problem with the ionization output (here it may be less than 100%, so it is necessary to search the optimal levels). The corresponding theory of determination of the excitation and ionization cross-sections and radiative probabilities in a laser field is in details given in ref. [5, 11,17-20].

The important aspect of theoretical studying laser photoionization isotope separation process is linked with calculating probabilities of the autoionization resonance decay in the external DC electric field [11,17]. In a case of atomic ionization by the pulsed field, a probability of this process is determined by the following expression:

$$W(nlm) = \sum_{n2} (a^{nlm}_{nln2})^2 W(n_1 n_2 m) \qquad (1)$$

Here $W(n_1 n_2 m)$ is the state decay probability; a are the coefficients of expansion of the $\psi(nlm)$ functions on the parabolic functions $\psi(n_1 n_2 m)$. In real multi-electron atom it is necessary to account for the influence of the electron shells, which results in the changing the potential barrier and wave functions. Usually in order to take into account an influence of the electron shells one should use the corresponding model potentials. The detailed description of the «best» model potentials and corresponding schemes can be found, for example, in Refs.[8,10,22].

To define the wave functions and electron state energies in an electric field, one needs to carry out the diagonalization of energy matrice, calculated between states with the same n [11]. The diagonalization of the complex energy matrix leads to complex energy correction:

$$\mathrm{Re}E - i\Gamma/2 , \qquad (2)$$

where Re E is the level shift and Γ is the level width, including the radiation and autoionization widths simultaneously. If the effects of the autoionization resonance decay are included in the matrix M, then Γ presents only the autoionization width of the state. Only Re M is diagonalized. The imaginary part is converted by means of the matrix of eigen-vectors $\{C_{mk}\}$. The eigen vectors are obtained by diagonalization of ReM:

$$\mathrm{Im}\, M_{ik} = \sum_{ij} C^*_{mi} M_{ij} C_{jk} . \qquad (3)$$

The other details of calculation procedure are given in refs. [5, 11,17- 21].

As example, we present some characteristic results of numerical calculating the ionization characteristics for the alkali elements isotope (the rubidium). In table 1 the characteristics of the quickly decayed states of the Rb atom (electric field strength: $E = 3 \times 10^4$ V/cm) are given for states with fundamental quantum number n=7-12 (our data). In fig.1 we present the calculation results on critical electric field strength E in depen-

dence on effective quantum number n^* for atoms of Rb, Na (dots- experimental data; line 1 is theoretical estimate on the basis of classical model E~ $1/16n^4$ without an account of the Stark shift and electron tunneling effect [1]; line 2 is calculation result on basis of the H-like non-relativistic model [2]; dashed line is corresponding to our relativistic data). It is stressed that the hydrogen-like approximation gives an inaccuracy ~15-20% [17]. However, consistent relativistic calculation has given the results in an excellent agreement with experiment.

Table 1. Characteristics of the quickly decayed states of the Rb atom
(E =3×10^4 V/cm; n=7-12)

n	7	8	9	10	11	12
E, cm (-1)	31405	31904	32229	32456	32614	32761
n_2^*	4,7	5,8	7,0	8,0	8,9	9,0
a_f^2	0,28	0,27	0,13	0,13	0,011	0,12
a_s^2	<10(-6)	<10(-6)	<10(-5)	<10(-5)	5 10(-5)	0,022
a_p^2	<10(-6)	8.8 10(-6)	2,5 10(-5)	0,00016	0,0024	0,15
a_d^2	0,00027	0,0006	0,0033	0,015	0,045	0,008

The next step is modelling the optimal parameters of the laser photoionization separating scheme. As usually [16-18], the optimization procedure of the laser photoionization sensor scheme is in a searching the optimal form of the laser pulse to provide a maximum of excited particles in the gases separation scheme (naturally this is one of the possible versions). The separation process is described by the density matrice equations system (c.f. [12,13]). We considered a scheme for laser separation and sensing Na, Rb isotopes. At the first step of the laser photoionization scheme δ-pulse provides a maximally possible level of excitation for the up state. At the last step an external DC electric field ionization must be realized earlier than the parasitic spontaneous relaxation processes (resonant re-charging etc.[1]) begin to destroy and change achieved excitation level of atoms. Using DC electric field ionization scheme sharply increases the output of charged particles, improve in whole energetics of the laser photoionization scheme and its optimality. It is possible to accept the special measures to provide very high ionization output (up to 95%) that requires using specially separated autoionization levels. The analysis shows that creation of laser photoionization scheme on the basis of considered scheme is more perspective in comparison to traditional two- and three-step laser photoionization schemes with an atomic ionization by laser pulse at the final step [1,2].

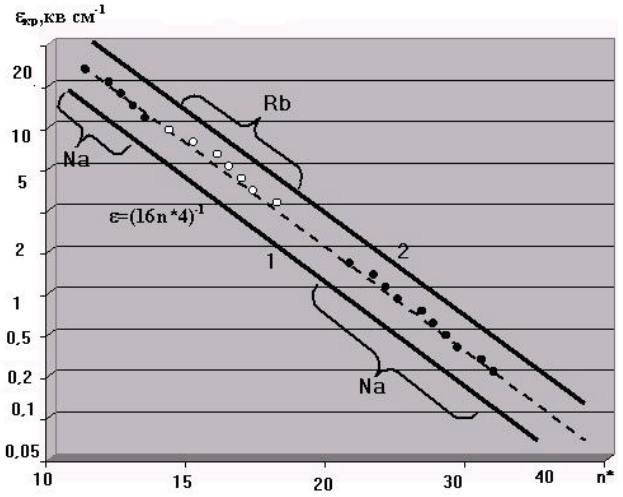

Fig.1. Critical electric field strength in dependence upon effective quantum number n^* for atoms of Na, Rb: dots – experiment (see [1]); solid line 1 – theoretical estimate on the basis of classical model; solid line 2 – calculation within the H-like approximation; dotted line –our data (eq.(1))

So, we considered the alkali elements isotopes separation scheme, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionization, and evaluated some important radiative parameters. The final presentation of the optimal parameters for the whole scheme requires more detailed search of the optimal laser and DC electric pulses characteristics and solving the task of the optimal governing [16-18].

References:

1. Basov N. G., Letokhov V. S., Optical frequency standards//Sov.Phys. — Uspekhi. — 1999. — Vol. 11. — P. 855-880.
2. Letokhov V. S., Laser isotope separation// Nature. — 1999. — № 277. — P. 605-610.
3. Bokhan P. A., Buchanov V. V., Fateev N. V., Kalugin M. M., Kazaryan M. A., Prokhorov A. M., Zakrevskў D., Laser Isotope Separation in Atomic Vapor- WILEY-VCH Verlag, 2006.
4. Bagratashvili V. N., Letokhov V. S., Makarov A. A., Ryabov E. A. Multiple Photon infrared Laser Photophysics and Photochemistry.-N-Y, Harwood Acad. Publ.1999.
5. Glushkov A. V., Ivanov L. N. DC Strong-Field Stark-Effect: consistent quantum-mechanical approach// J.Phys.B: At. Mol. Opt. Phys. — 1999. — Vol. 26. — P.L379-L386.
6. Ryabov E. A. Laser separation of isotopes on the basis of IR multi-photon dissociation of molecules//Usp.Phys. Nauk. — 2004. — Vol.174. — P. 684-688.
7. Baldwin G. G., Salem J. C., Goldanskii V. I., Approaches to the development of gamma ray lasers // Rev.Mod. Phys. — 1981.-Vol.53.-P.687-742
8. Ivanov L. N., Letokhov V. S. Spectroscopy of autoionization resonances in heavy elements atoms// Com.Mod. Phys.D.:At.Mol.Phys. — 1999. — Vol. 4. — P. 169-184.
9. Goldanskii V. I., Letokhov V. S. Effect of laser radiation on nuclear decay processes// Sov. Phys. JETP. — 1999 — Vol. 67.-P. 513-516.
10. Glushkov A. V., Atom in an electromagnetic field.-Kiev: KNT, 2005.
11. Glushkov A. V., Khetselius O. Yu., Svinarenko A. A., Pre-pelitsa G.P., Energy Approach to Atoms in a Laser Field and Quantum Dynamics with Laser Pulses of Different Shape//In: Coherence and Ultrashort Pulsed Emission, Ed. Duarte F. J. (InTech). — 2011. — P. 159-186.
12. Karlov N.,Orlov A., Petrov Y., Prokhorov A. M. Laser governing by diffusion stream of gases to vacuum through obstacle with developed surface // Izv. AS USSR.Phys-1999Vol.49-P.500-505.
13. Solatz R. W., May C. A., Carlson L. R. et al, Detection of Rydberg states in atomic uranium using time-resolved stepwise laser photoionization//Phys.Rev.A. — 1999. — Vol. 14, № 3.—P. 1129-1148.
14. Stoll W. Present Status of industrial Isotope separation by laser technology// Atomic and Molecular Pulsed Lasers.-Tomsk: SO RAN, 2001. — P. 71.
15. Buchanov V. V., Kazaryan M., Kalugin M., Prokhorov A. M. Laser separation of Silicon Isotopes by the AVLIS Technology// Atomic and Molecular Pulsed Lasers. — Tomsk: SO RAN, 2001. — P. 72.
16. Glushkov A. V., Ambrosov S. V., Shpinareva I. M. Non-linear Selective Photoprocesses in Atoms and Molecules and their Optimal Governing. Optimized Isotope Separation Schemes// Atomic and Molecular Pulsed Lasers. — Tomsk: SO RAN, 2001. — P. 70.
17. Glushkov A. V., Khetselius O. Yu., Svinarenko A. A., Prepelitsa G. P., Energy Approach to Atoms in a Laser Field and Quantum Dynamics with Laser Pulses of Different Shape//In: Coherence and Ultrashort Pulsed Emission, Ed. Duarte F. J. (Intech, Vienna). — 2011. — P. 159-186..
18. Glushkov A., Lepikh Ya., Svinarenko A., Prepelitsa G., Ambrosov S., Bakunina E., Loboda A., Physics of laser-photoionization atomic processes in isotopes and gases separator devices: New optimal schemes//Sensors Electr. and Microsyst.Techn. — 2011. — Vol. 2 (8), № 1. — P. 27-35.
19. Glushkov A. V., Prepelitsa G. P., Ambrosov S. V., Loboda A. V., Gurnitskaya E. P., Consistent QED approach to cal-

culation of electron-collision excitation cross-sections and strengths: Ne-like ions // Int. Journ.Quant.Chem. — 2005. — Vol. 104, № 4. — P. 562-569.
20. Glushkov A. V., Ambrosov S. V., Fedchuk P. O., Fedchuk O. P., Stark effect and resonances in the ionization continuum for hydrogen atom and Wannier-Mott excitons in DC electric field// Journ. Phys.Stud. — 2003. — V. 7, № 3. — P. 121-125.
21. Glushkov A. V., Lepikh Ya. I., Ambrosov S. V., Khetselius O. Yu., New optimal schemes of laser photoionization technologies for cleaning the semiconductor materials and preparing the films of pure composition at atomic level//Ukrainian Journ. Physics.-2008. — Vol. 53, № 10. — P. 1017-1022.
22. Glushkov A. V., Fedchuk P. A., Fedchuk A. P., Loboda A. V., New general approach in electron theory of catalytic processes on semiconductors and binary metallic materials// Photoelectronics.-2004. — № 13. — P. 19-22.

UDC 535.42, 539.184

S. V. Ambrosov, A. P. Fedchuk, A. V. Glushkov, Ya. I. Lepikh

LASER-PHOTOIONIZATION METHOD OF SEPARATION OF THE ISOTOPES: RADIATIVE PARAMETERS FOR ALKALI ELEMENTS

Abstract.

We present an optimal scheme of the separating alkali elements isotopes, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionization. Some radiative parameters data are obtained for alkali elements.

Key words: laser photoionization method, isotopes separation, radiative parameters

УДК 535.42, 539.184

С. В. Амбросов, А. П. Федчук, А. В. Глушков, Я. И. Лепих

ЛАЗЕРНО-ФОТОИОНИЗАЦИЙ МЕТОД РАЗДЕЛЕНИЯ ИЗОТОПОВ: РАДИАЦИОННЫЕ ПАРАМЕТРЫ ДЛЯ ЩЕЛОЧНЫХ ЭЛЕМЕНТОВ

Резюме.

Представлена оптимальная схема лазерного разделения изотопов щелочных элементов, базирующаяся на лазерном возбуждении атомов изотопов в ридберговские состояния и дальнейшей ионизации внешним постоянным электрическим полем. Определены некоторые радиационные параметры для щелочных элементов.

Ключевые слова: лазерный фотоионизационный метод, разделение изотопов, радиационные параметры

УДК 535.42, 539.184

С. В. Амбросов, О. П. Федчук, О. В. Глушков, Я. І. Лепих

ЛАЗЕРНО-ФОТОІОНІЗАЦІЙНИЙ МЕТОД ПОДІЛЕННЯ ІЗОТОПІВ: РАДІАЦІЙНІ ПАРАМЕТРИ ДЛЯ ЛУЖНИХ ЕЛЕМЕНТІВ

Резюме.

Представлена оптимальна схема лазерного поділення ізотопів лужних елементів, яка базується на лазерному збудженні атомів ізотопів у рідбергівські стани та подальшій іонізації зовнішнім сталим електричним полем. Визначені декотрі радіаційні параметри для лужних елементів.

Ключові слова: лазерний фотоіонізаційний метод, поділення ізотопів, радіаційні параметри