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PHYSICS OF THE LASER-PHOTOIONIZATION ATOMIC PROCESSES IN THE ISOTOPES AND GASES SEPARATOR DEVICES: NEW OPTIMAL SCHEMES

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Abstract. We present review of the models for key laser photoionization processes and propose the corresponding new optimal schemes of the separating heavy isotopes and nuclear isomers, substances cleaning at atomic level, which are based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionizationThe operator relativistic perturbation theory method, optimal laser action model and density matrices formalism are used for numeric calculation of the optimal scheme parameters for the U and other isotopes.

Key words: laser photoionization method, isotopes and gases separation, new optimal schemes

ФІЗИКА ЛАЗЕРНО-ФОТОІОНІЗАЦІЙНИХ АТОМНИХ ПРОЦЕСІВ В УСТАНОВКАХ ДЛЯ ПОДІЛЕННЯ ІЗОТОПІВ ТА ГАЗІВ: НОВІ ОПТИМАЛЬНІ СХЕМИ

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Анотація. Представлений огляд моделей основних лазерно-фотоіонізаційних процесів та запропоновані відповідні нові оптимальні схеми лазерного поділення важких ізотопів, ядерних ізомерів, газів, очищення речовини на атомному рівні, які базуються на лазерному збудженні атомів ізотопів у рідбергові стани та подальшій іонізації зовнішнім сталим електричним полем. Удосконалені метод релятивістської теорії збурень, модель оптимальної лазерної дії та формалізм матриці густини використані для чисельного моделювання параметрів оптимальної схеми поділення детектування важких ізотопів урану та інших елементів.

Ключові слова: лазерний фотоіонізаційний метод, поділення ізотопів та газів, нові оптимальні схеми

ФИЗИКА ЛАЗЕРНО-ФОТОИОНИЗАЦИОННЫХ АТОМНЫХ ПРОЦЕССОВ В УСТАНОВКАХ ДЛЯ РАЗДЕЛЕНИЯ ИЗОТОПОВ И ГАЗОВ: НОВЫЕ ОПТИМАЛЬНЫЕ СХЕМЫ

А. В. Глушков, Я. И. Лепих, Г. П. Препелица, С. В. Амбросов, О. В. Бакунина, А. А. Свинаренко, А. В. Лобода

Аннотация. Представлен обзор моделей ключевых лазерно-фотоионизационных процессов и предложены соответствующие новые схемы лазерного разделения изотопов, ядерных изомеров, газов, очистки вещества на атомном уровне, базирующиеся на лазерном возбуждении атомов изотопов в ридберговы состояния и дальнейшей ионизации внешним постоянным электрическим полем. Усовершенствованные метод релятивистской теории возмущений, модель оптимального лазерного воздействия и формализм матрицы плотности использованы для численного моделирования параметров оптимальной схемы детектирования и разделения изотопов урана и других элементов.

Ключевые слова: лазерный фотоионизационный метод, разделение изотопов и газов, новые оптимальные схемы

1. Introduction

For the last 30 years, the investigations and developments aimed at creation of new highly efficient, alternative methods for separating isotopes of chemical elements were actively performed in some countries and attempts were made for their industrial employment [1–5]. Among those methods, isotope separation with the help of laser radiation is of particular interest. The methods of atomic (AVLIS process–photoionization method) and molecular (MLIS process) isotope separation are well known and developed. Since the early 1980s, the further development of AVLIS process was additionally stimulated and brought up to a qualitatively new level due to achievements in laser spectroscopy, and plasma physics and technique. The acceleration in this period is mainly explained by the fact that in the USA the photoionization method of isotope separation was chosen after thorough study and comparison of the abovementioned methods from the point of view of the highest accessible efficiency for uranium isotope separation. Throughout 1980s, Lawrence Livermore National Laboratory (USA) in cooperation with some large companies extensively developed, updated, and demonstrated the elementary basis of AVLIS. Up to the late 1980s, the noticeable success in all directions of the program was achieved. In 1989, it became possible to start with specifying the date of the development of AVLIS technology for full-scale industrial production.

Now it is well known that the laser photoionization method is one of the most perspective methods for the sensing single atomic particles, separating isotopes, nuclear isomers and nuclear reactions products (see Refs. [1–20]). The standard laser ionization sensor scheme may be 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 photoionization of the excited states by additional laser radiation, has been at first proposed and realized by Letokhov et al (see Refs. [1,2]). 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 excitement σ_{exc} and photo-ionization σ_{ion} ($[\sigma_{exc}/\sigma_{ion}]>10^4\div10^8$). It requires the using very intensive laser radiation for the excited atom ionization. The situation is more simplified for autoionization resonances in the atomic spectra, but detailed data about characteristics of these levels are often absent. Main problems here 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 is not possible hitherto (see Refs. [16–24]).

In a number of papers (see Refs. [1,10,11,19,25]) 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) moves into continuum and other levels become 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 excitement by laser field (it is trivial task) and probability of ionization of the atoms in the highly excited states (autoionization 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 [8–26]. As a rule, non-relativistic approximation has been used [1]. More consistent approach to solution of such a problem must be based on the relativistic models [11,19,23–30], as the most interesting elements for laser isotope separation are heavy ones and a role of relativistic corrections is often very dramatic. Below we use a new consistent relativistic quantum mechanical approach for numeral calculation of the autoionization resonances decay in the external DC electric field [11,23,24].

This paper goes our research on carrying out optimal schemes for the laser photoionization sensors of separating heavy isotopes and nuclear isomers. We present review of the models for key laser photoionization processes and propose the corresponding new optimal schemes of the separating heavy isotopes and nuclear isomers, substances cleaning at atomic level. It is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further DC electric field ionization and laser pulse autoionization mechanisms. The matter is in a fact that the velocity of ionization is significantly higher than one in the usual regime (see Refs. [1,5,11,22]). A result, the effectiveness of the laser photoionization sensor scheme is significantly increased. The prйcised data for the autoionization resonance characteristics for Cs, Ga, U, Tm atoms in an electric field are presented. Two autoionization resonances decay channels (traditional Beutler-Fano channel and new Letokhov-Ivanov channel) are examined. An appreciable dependence of the reorientation decay velocity of the autoionization resonances at an electric field is analyzed. An effect of the giant autoionization resonance width broadening in the relatively weak DC electric field is found more drastic than it is discovered in paper [5]. To carry out modelling the optimal scheme of the *U* and other isotopes (nuclei) sensing, we use the advanced optimal laser action models and density matrices formalism [11–16,24–26].

2. Laser photoionization isotope separation scheme. Calculation model of the laser and DC electric field ionization

Let us consider a process of the uranium *U* and thulium *Tm* isotopes separation (c.f. [1,26]). The first ionization step: excitation of the uranium and thulium atoms into the rydberg states: $5f^3 7s^2 np$, $5f^3 6d 7sns$ (*U*: electron external shell configuration) and $4f^{13} 6sns$, $4f^{13} 6snp$ (*Tm*). Principal quantum number n may be equal 10–50. The further step is ionization by external DC electric field. 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: $\sigma_2 \sim \cdot 10^{-15\div 12}$ 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 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). Further to carry out a modelling the optimal scheme of the *U, Tm* isotope (nuclei) sensing we need data about probabilities of ionization by external DC electric field. To get these data, we use a new consistent relativistic quantum mechanical approach for numeral calculation of the autoionization resonance decay in the external DC electric field [11,23,24]. Let us describe it in brief.

Probability of the ionization (autoionization width) for highly excited atoms by electric field is given by the full flow of probability through the plane, which is perpendicular to z-axe. Calculation of the probability requires a solution of the axially symmetrical problem, when a potential barrier separates two classically allowed regions. The examples

of the application of such problem are an atom in a uniform electric field, the two-centre problem etc. Let us find the wave function in the below-barrier region. In this case, the wave function is localized in the vicinity of the most probable tunnelling way, that is the potential symmetry axis z. For the bi-spinor

$$\psi = \begin{pmatrix} \xi \\ \eta \end{pmatrix}, \qquad (1)$$

the Dirac equation has the following form:

$$\begin{aligned} c\vec{\sigma}\rho\xi &= (E - V + c^2)\eta, \\ c\vec{\sigma}\rho\eta &= (E - V - c^2)\xi, \end{aligned} \qquad (2)$$

where ρ and σ, as usually, denote the momentum and Pauli operators.

Inserting first equation of the system (2) into second one and using substitution

$$\begin{aligned} \xi &= (W^+)^{1/2}\Phi, \\ W^{\pm} &= E - V \pm c^2, \end{aligned} \qquad (3)$$

we obtain the second-order equation

$$\Delta\Phi + K^2\Phi = 0,$$

$$K^2 = \frac{1}{\hbar^2 c^2}\left[(E-V)^2 - c^4\right] - \frac{\Delta V}{2W^+} - \\ - \frac{3}{4}\left(\frac{\vec{\nabla}V}{W^+}\right) + \frac{i}{W^+}\vec{\sigma}\left[\vec{\nabla}V, \vec{\nabla}\right]. \qquad (4)$$

Since the potential is axially symmetrical, we seek a solution to' this equation in the cylinder system of co-ordinates (z, p) as follows:

$$\Phi = \begin{pmatrix} F_1(z,p)\exp\left[i(m-1/2)\phi\right] \\ F_2(z,p)\exp\left[i(m+1/2)\phi\right] \end{pmatrix}. \qquad (5)$$

Substituting (5) into (4) gives the matrix equation:

$$(\Delta + \partial)F = (\hbar^{-2}q^2 + \gamma)F, \qquad F = \begin{pmatrix} F_1 \\ F_2 \end{pmatrix}, \qquad (6)$$

$$q = \frac{1}{c}\left[c^4 - (E-V)^2\right]^{1/2},$$

$$\partial = \frac{1}{W^+}\left(\frac{\partial V}{\partial p}\frac{\partial}{\partial z} - \frac{\partial V}{\partial z}\frac{\partial}{\partial p}\right)\begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix},$$

$$\gamma = \begin{pmatrix} a_{m-1/2} & b_{m+1/2} \\ b_{m-1/2} & a_{-m-1/2} \end{pmatrix},$$

$$a_\mu(z,p) = \frac{\mu^2}{p^2} + \frac{1}{W^+}\left[\frac{\mu}{\rho}\frac{\partial V}{\partial p} + \frac{\Delta V}{2} + \frac{3}{4}\frac{\left(\vec{\nabla}V\right)^2}{W^+}\right],$$

$$b_\mu(z,p) = -\frac{\mu}{pW^+}\frac{\partial V}{\partial z}.$$

A solution for system (6) is found numerically on the basis of the finite differences numerical algorithm [27]. In a case of atomic ionization by the pulsed field, probability of process is defined by the following expression:

$$W(nlm) = \sum_{n2}(a_{n1n2}^{nlm})^2 W(n_1 n_2 m). \qquad (7)$$

Here, as usually *nlm* are the quantum number of electron state; $W(n_1 n_2 m)$ 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. 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 [7–9,28]. The diagonalization of the complex energy matrix leads to complex energy correction: ReE -iΓ/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 standard energy secular 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:

$$\text{Im } M_{ik} = \sum_{ij} C^*_{mi} M_{ij} C_{jk}.$$

The other details of calculation procedure are given in refs. [10,11,21,25–30].

3. Results of numerical calculating the ionization characteristics for heavy isotope atoms and modelling the parameters for optimal scheme of the laser photoionization sensor

Here we present some key results of numerical calculating the ionization characteristics for heavy isotope of the uranium and thulium and modelling the optimal scheme parameters of the laser photoionization sensor. In fig.1 we present our calculation results on the critical electric field strength in dependence upon effective quantum number n^* for atom of U (dots- experimental data; line 1 is theoretical estimate from classical relation E~ $1/16n^4$ without account of the Stark shift and electron tunnelling effect [1]; line 2 is calculation result on the basis of the H-like non-relativistic model [2];

dashed line is corresponding to our relativistic results). It is stressed that the hydrogen-like approximation gives an inaccuracy 15–20 % [2].

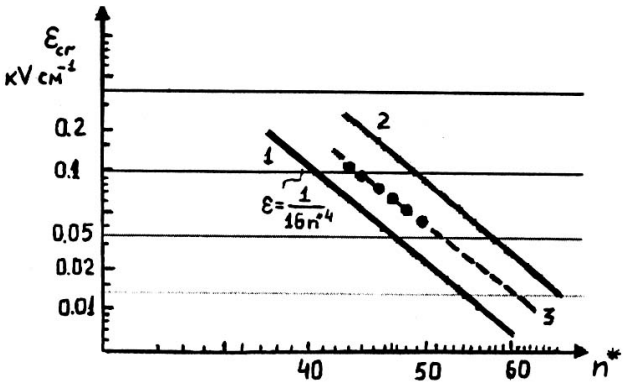

Fig. 1. Critical electric field strength in dependence upon effective quantum number n^* for the uranium atom

At the same time consistent relativistic calculation has given the results in an excellent agreement with experiment. The same situation takes a place with the thulium isotope. The most interesting feature of the ionization in an external electric field is connected with an effect of electric field on the autoionization resonances in the heavy isotopes (the last step of the laser photoionization scheme).

Let us consider the thulium isotope. It should be noted that studying the autoionization in an external DC electric field represents an undoubted interest for experimental laser spectroscopy, in controlling the population and decay kinetics of excited states or the selective ionization under laser radiation action [1]. Study of such processes is very important for development of the multistage ionization method schemes. One can mention a great role of the autoionization resonances in many processes in plasma and gases. Especially interesting effects occur in the complex heavy atom when its broad AR mix with much narrower resonances of opposite parity by means the external electric field. Authors of paper [5] have predicted new effects, connected with behaviour of the Tm autoionization resonances in an external electric field and discovered a drastic broadening of the reorientation type autoionization resonances already in a weak electric field. We have carried out the same calculation, but with the use of the relativistic approach. Note that the availability of two pairs of near-lying ionization limits (with vacancy states $4f7^1/2$ and $4f5^1/2$) in the thulium atom provides two main types of the autoionization resonance decay:

(BFD) $4f^1_{5/2}6s1/2(J12)nl-4f^1_{7/2}6s1/2[J12']Tm^+ +$ $+ leje$, $n>7$, $J12=2; 3, J12'=3; 4$,

(ROD) $4f^1j6s1/2(J12)nl$-$4f^1j6s1/2[J12']Tm^+ +$ $+leje$, $n>25$, $J12=3$, $J12'=2;4$ $j=5/2,7/2$.

Here the ROD means the reorientation autoionization resonance decay, and the BFD means the traditional Beutler-Fano autoionization resonance one. The states $4f^1_{5/2}6s_{1/2}(J12=3)nl$ undergo simultaneously both BFD and ROD. Remember that contrary to the BFD, the ROD is a low energy process preserving all the single electron quantum numbers of atomic residue: $4f^1j$ and $6s_{1/2}$. The ROD can be of the monopole or quadruple character. We mean here the multi-polarity of the inter-quasiparticle interaction causing the autoionization resonance decay. The states with $J12=2;4$ do not undergo the ROD. Nevertheless, their admixing with states undergoing the ROD can significantly amplify the monopole ROD. For Rydberg series, the only possible autoionization resonance decay is the reorientation one. Any two states of different parity can be mixed by the external electric field. The mixing leads to redistribution of the autoionization widths. In the case of degenerate or near-degenerate resonances this effect becomes observable even at a moderately weak field. In the case of the thulium we deal with reorientationally decaying *ns* and *np* series, converging to the same ionization limit, i.e. they are nearly degenerate states of different parity. Among them one can find some pairs of *ns* and *np* states with widths Γ, differing by several orders.

We consider the $f^1_{7/2}$ $6s(3)25s[5/2]$ state, decaying due to the quadruple interaction and $f^1_{7/2}$ $6s(3)25p_{1/2}[5/2]$ undergoing to the monopole ROD. In table 1 we present the calculated values of the energy E, autoionization width Γ for the $4f7^{-1}/26s(3)ns,np$ Tm states ($n=25$) for different values of the DC electric field strength ε.

A strong change of the autoionization resonance width occurs at a moderately weak electric field and this effect is found more drastic (~10 %) than it is discovered in paper [5]. The same effect is at first discovered for the uranium isotopes. We have calculated the energies and widths for higher members of the uranium Rydberg series (members accessed from 32.899,79 cm^{-1} level). Excitation sequence: 6056,81+6098,10+(5880–5890) E. For $5f^37s^2np$ $(n=40–44)$ levels the following results are obtained: i). For ε=0 V·cm^{-1}, configuration $5f^37s^242p$, E=49877,49 (experimental value [7]: 49877,5); $\Gamma(5f^37s^242p)$=1,794D-01; $\Gamma(5f^37s^242s)=2$,702D-05;

ii). For ε = 100 $V{\cdot}cm^{-1}$; $I(5f^37s^242p)$=1,675D-01; $I(5f^37s^242s)$=5,913D-04. So, we have here a strong change of the autoionization resonance width at a moderately weak electric field too.

Table 1

Calculated values of the energy E (cm^{-1}) and autoionization width Γ (cm^{-1}) for the $4f7^{-1}/26s(3)ns,np$ Tm states (n=25) for different values of the DC electric field strength ε. ($V{\cdot}cm^{-1}$)

State	$4f^{13}{}_{7/2}6s1/2$ (3) 25s [5/2] Ref. [5]	$4f^{13}{}_{7/2}6s1/2$ (3) 25s [5/2] Present paper	$4f^{13}{}_{7/2}6s1/2$ (3) 25$p_{1/2}$ [5/2] Present paper
E, ε = 0	49854,7 cm^{-1}	49854,7 cm^{-1}	49865,3 cm^{-1}
Γ, ε = 0	1,80D — 05	1,861D — 05	1,196D — 01
Γ, ε = 50	1,16D — 04	1,847D — 04	1,194D — 01
Γ, ε = 100	4,27D — 04	7,130D — 04	1,189D — 01
Γ, ε = 150	9,340D –04	1,330D –03	1,182D — 01

Detailed spectroscopic information about autoionization resonances is needed to optimize the excitation and ionization of the atom. An optimal scheme presumes a compromise between high excitation probability and high decay rate that determines the lower and upper boundaries for the autoionization resonances decay rate. The use of the ROD channel essentially increases the possibilities of such a compromise. It is very important for carrying out the optimized scheme of the selective photoionization sensor.

Further we present the results of numerical modelling the optimal scheme parameters of the laser photoionization sensor. As usually [16–21], 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 the laser separation and sensing the uranium and thulium isotopes. All necessary excitation and ionization constants for solution of the density matrice equations system are calculated within above described model and methods [16–21,23–30].

The laser photoionization scheme for U isotopes included the following steps: i). Laser excitement of the ^{235}U isotopes from the ground $5f^36d7s^2$–$^5L_6^o$ state and low lying metastable $5f^36d7s^2$–$^5K_5^o$ state with energy 620,32 $см^{-1}$; ii). Transition into the autoionization state with doubly excited external shell and then ionization by the DC electric field.

In figure 2 we present the results of numerical modelling the optimal form of laser pulse in the laser photoionization sensor scheme for the uranium isotopes. The following notations are used: δ pulse + dotted line is the optimal form of the laser pulse; curves (1) x_1 and (2) x_2 are the populations of the ground and excited (n=42) states. In the qualitative relation obtained data are similar to data for other *Na, Rb* and *Tm* isotopes [16]. At the first step of the laser photoionization scheme the 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 recharging etc. [1]) begin to destroy and change an achieved excitement level of atoms.

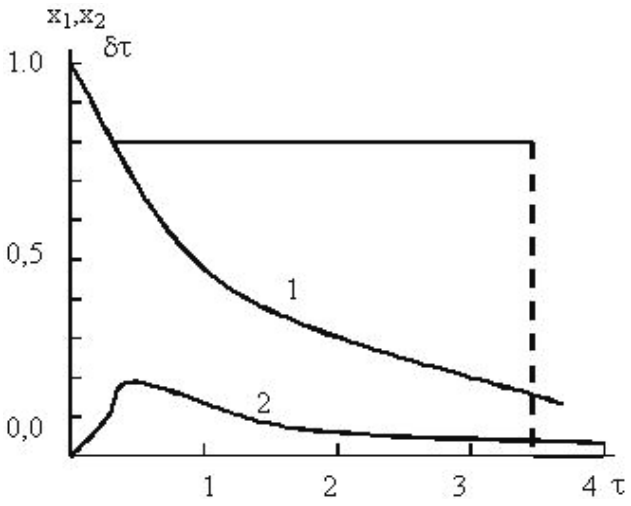

Fig. 2. Results of numerical modelling for the optimal form of laser pulse in the laser photoionization sensor scheme for the uranium isotopes: δ pulse + dotted line is the optimal form of the laser pulse; curves (1) x_1 and (2) x_2 are the populations of the ground (6s) and excited (np) states

Using the DC electric filed ionization mechanism significantly increases the output of charged particles, improve in whole the energetics of the

laser photoionization sensor scheme and its optimality. It is possible to accept the special measures to provide very high ionization output (approaching to 100 %) that requires using specially separated autoionization levels. General analysis shows that creation of the laser photoionization scheme on the basis of considered scheme is more perspective in comparison to traditional two- and three-step laser photoionization schemes with ionization by laser pulse at the final step [1,2].

4. Laser-photoionization method for preparing the films of pure composition at atomic level

The laser-photoionization method could be very effective for creation of new optimal schemes for preparing the films of pure composition at atomic level. The first possible schemes for preparing the films of pure composition by means of the two-stepped selective ionization of atoms has been proposed by Letokhov [2]. As easily understood from above text, such scheme was not experimentally checked, however it is obvious that the two-stepped laser ionization scheme is not optimal one. The main feature and innovation of our scheme is connected with using the electric field on the last ionization step and using decay of autoionization resonances in a laser pulse (i.e. autoionization at the last step). Such an optimal scheme can be used as basis for devices for preparing the films of super pure composition during sedimentation of ions of the A^+ kind, which are obtained by optimal method of selective photo-ionization of the A kind atoms in the beam in mixture with other atoms. In fig. 3 we propose the possible construction of scheme for preparing the films of super pure composition during sedimentation of ions. In fact our scheme generalizes the known Letokhov and Glushkov-Lepikh-Ambrosov scheme [2,21], but it looks more effective.

The wide spread method for getting complex hetero-structures of super lattices type is a method of molecular epytaxy. At the same time one can note that it allows creating only 1D super lattices (the known example is layers $Ga_{1-x}Al_x$as of the width 10E and GaAs of the width 60 E with full number of layers around 100. The 3D hetero-structure super lattices can be created with using the photo-ionic beams of Ga^+, Al^+, As^+ with help of the electromagnetic focusing and deflecting systems.

As it had been at first underlined in ref.[2], this opportunity of spatial control of the sedimentation of ions is of a great importance for semiconductor atomic technology of the materials in future. But the key topic is creation of the effective technology (profitable from commercial point of view).

Let us consider further laser photo-ionization method of obtaining the Ga^+ ions and optimal scheme for preparing the films of pure composition. We consider the ionization scheme of obtaining the Ga^+ ions, following to ref.[12]. The transition scheme is as follows; $4p^2P_{3/2} \rightarrow (\lambda_1=417{,}2\ nm) \rightarrow 5s^2S_{1/2} \rightarrow (\lambda_1=420–440\ nm) \rightarrow np^2P_{1/2}$.(n=14–70). Modelling the optimal parameters for last process, i.e. ionization of the Rydberg states by electric field, can be carried out on the basis of methodise [9–12]. In figure 4 the results of the numerical modelling the Ga atoms separation process from the mixture on the basis of the laser photoionization method are presented.

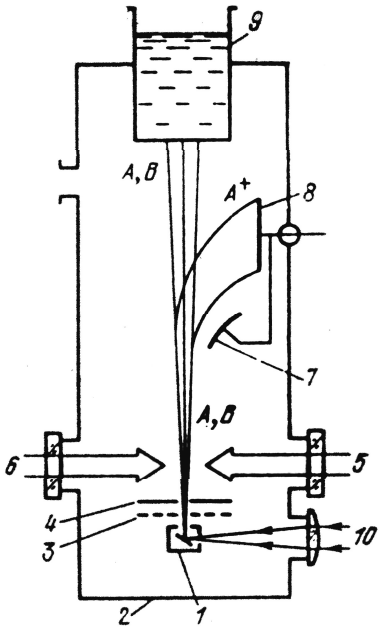

Fig. 3. Possible scheme for preparing the films of super pure composition during sedimentation of ions of A^+ kind, which are obtained by optimal method of selective photo-ionization of the A atoms in a beam in mixture with other atoms: 1 — source of atomic beam; 2- vacuum box; 3 — collector of non-selective ions; 4 — diaphragm; 5 — laser ray for the first step excitation; 6 — laser ray for second-step excitation to Rydberg states and further autoionization in a laser pulse and ionization by external electric field; 7 — deflecting electrodes; 8 — sublayer; 9 – cold sublayers for freezing atoms; 10 — laser ray for vaporising the substance

The following definitions are used: δ+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 Ga. The δ -pulse

provides maximum possible level of excitation (the excitation degree is about ~0,25; in experiment [2] with rectangular pulse this degree was ~ 0,1). In further the parasite processes such as spontaneous relaxation, resonant re- exchange can't change the achieved excitation level during a little time. The last step of the process is an ionization of excited atoms by the electric field pulse [9] (the field strength is 8,8 kV/cm). To get a high level of the optimality an electric field has to be switched on during the time, which is less than the excited state radiative decay time.

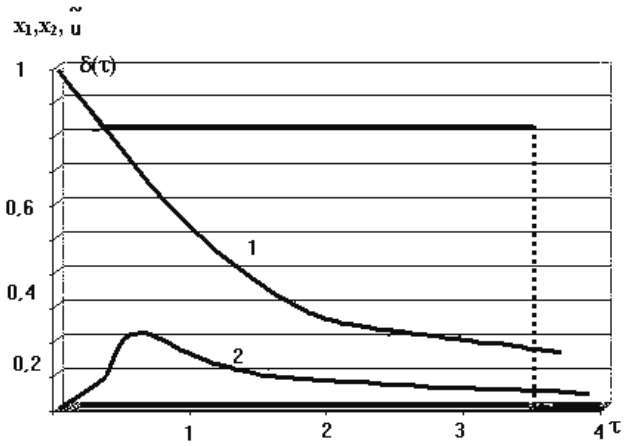

Fig. 4. Results of modelling Ga separation process from Ga-X mixture by the photo-ionization method (δ+dashed — laser pulse optimal form; curves x_1, x_2 are corresponding to populations of the ground and excited states)

The decay of Ga atoms and ions in the high-excited state demonstrates the properties of the H-like systems at the qualitative level. But, there is quite significant quantitative difference. We have found that the ionization velocity for states with n>14 is more than the radiative decay velocity in electric field with strength E less than 15 kV/cm. Our estimate for the Ga atom ionization cross section is $1{,}5\cdot10^{-13}$cm^2 that is higher than the corresponding cross section of ionization process by laser pulse in the two- stepped photo ionization [2] scheme (~10^{-17}cm^2). Using δ-pulse provides a quick ionization, but the ionization yield will be less than 100 % because of the sticking on intermediate levels. Experimentally obtained dependence of the critical ionization field strength E upon the effective quantum number n* is usually approximated by simple theoretical dependence $E_{cr}=(2n^*)^{-4}$. Our calculation results show that this is very approximate estimate and only consistent quantum calculation (c.f.[21,26,29] provides an acceptable agreement with experiment. In any case, the laser photo ionization scheme with ionization by electric field (with optimal set of energetic and radiative parameters: pulse form, duration, energetic for laser and electric field pulses etc.) could provide significantly more high yield and effectiveness of the whole process than the other known schemes.

5. Conclusions

So, we presented a brief review of the models for key laser photoionization processes and propose the corresponding new optimal schemes of the separating heavy isotopes and nuclear isomers, which are based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionization or autoionization (decay of autoionization resonances in a laser pulse). The operator relativistic perturbation theory method, optimal laser action model and density matrices formalism are used for numeric calculation of the optimal scheme parameters for the U and other isotopes. Besides, we considered the advances optimal scheme for preparing the films of super pure composition during sedimentation of ions of A^+ kind, which are obtained by optimal method of selective photo-ionization of the A atoms in a beam in mixture with other atoms. The key innovation is linked with using the new physical (effects) mechanisms of the ionization (autoionization resonances in a laser or electric pulses) at the last step of the whole scheme.

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