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Advanced Laser-Photoionization Scheme of Separation of Heavy Isotopes in the Gases Separator Devices

An effective approach to determining the parameters of the optimal schemes of the method of laser selective photoionization of atoms (elements and isotopes) with finite ionization due to collisions, ionization by a pulsed electric field, ionization through high (Rydberg) states and narrow autoionization resonances for the separation of heavy isotopes has been proposed. in gas separator devices. On the basis of the theory of optimal control and previously developed quantum models for calculating the characteristics of elementary atomic processes, optimization models of isotope separation are numerically implemented in the scheme of selective laser photoionization with ionization due to collisions in gas mixtures, ionization by a pulsed electric field, autoionization, etc. etc. The data obtained quantitatively confirm the promise of the method of laser photoionization with finite ionization due to collisions, ionization by a pulsed electric field, ionization through high-lying (Rydberg) states and narrow autoionization resonances and give a set of parameters for the desired optimal schemes, in particular, the laser pulse optimal shape for rubidium and uranium isotopes.

Keywords: *gas separator, isotope separation, laser photoionization method, optimal control theory*

Introduction.   Studying of physical and chemical processes with participamce of atomic systems (gases) in the istopes and gases separator devices is of a great interest for many gas-dynamical topics, relating to physics of atoms (moleculesm gases0 interactions in presenvce of external electromagnetic fields. The intensive thepretical and experimental investigatians are carried out in a field of studying and constructiong the optimal laser—photoioniation schemes of separation of different atomic elements (isotopes) in the vapour state in the gases separator devices (e.g. [1-16]).

In this paper, we present an effective theoretical approach to the calculation of optimal schemes of the atomic systems laser-photoionzation method (based on selective resonant excitation of atoms by laser radiation into quantum states near the ionization limit and subsequent autoionization decay due to so-called gas-separators, or under the action of an external electric field, etc.) on the basis of methods of an optimal control theory as well as the theory of corresponding quantum models of elementary atomic processes [17-30]). The desired methods have previously been used in solving various problems of optimal laser effects (c.g., [11, 12]). The element of novelty is the implementation of the optimization model of selective photoionization of atoms with finite ionization by means of electric field (or autoionization or collisional mionization) (e.g.[9-17, 30-36]). The generalized

Krasnov-Shaparev-Shkedov optimization model for classical two-stage selective photoionization is used as the starting one, summarizing it in the case of electric field ionization, autoionization, and collision ionization. Using the calculated data obtained for these ionization scenarios (the last step of the scheme), one could numerically calculate the optimal schemes of the method of laser photoionization of atoms in a vapour state. The solution of the optimal control problem for multi-stage selective photoionization can be based on the model of balance relations arising from the equations for the density matrix [12-17]. The problem is formulated as a way to find the optimal shape of the laser pulse of resonant radiation, which provides the maximum of ionized particles in the scheme of selective photoionization with pulsed electric field ionization and through autoionization resonances, as well as through the collision ionization mechanism.

Theoretical quantum model of optimal control. In general, the desired task of optimal control, taking into account spontaneous relaxation can be written in the form [11, 12]:

$$J = - \int_0^{\tau_f} R(\tau) x_2 \, d\tau \to \min; \tag{1}$$

$$dx_1 / d\tau = x_2 - \overline{u}(x_1 - x_2), x_1(0) = 1; \tag{2}$$

$$dx_2 / d\tau = -[R(\tau) + 1] x_2 + \overline{u}(x_1 - x_2), x_2(0) = 0; \tag{3}$$

$$dx_1 / d\tau = \overline{u}, x_3(0) = 0, x_3(\tau_f) = E_f; \tag{4}$$

$$0 \le \tau \le \tau_f, \overline{u}(\tau) \ge 0; \tag{5}$$

where x_1, x_2 – normalized populations of the ground and excited states of the atom; $\overline{u} = u / \gamma -$ dimensionless speed of induced processes of emission and absorption of resonant radiation; $\gamma -$ the probability of spontaneous decay per unit time; $u(t) = \sigma_{12} I_1(t) / \hbar \varpi_{21} -$ rate of induced transitions (transition 1-2); $\varpi_{21} -$ radiation frequency corresponding to the transition 1-2; σ_{12} – absorption cross section at the transition 1-2; $R = R' / \gamma -$ dimensionless ionization rate from the excited state; $R'(t) = \sigma_{ph} I_2(t) / \hbar \varpi_{ph} -$ photoionization rate; $\varpi_{ph} -$ radiation frequency (~ photoionization); $\sigma_{ph} -$ cross section of photoionization; $\tau = t_\gamma -$ dimensionless time; I_1, I_2 – intensity of laser pulses, respectively, for excitation from the ground state and ionization from the excited level; $E_f, \tau_f -$ the energy of the pulse of resonant radiation and its duration. According to the standard approach of the theory of optimal control, the transition to the derivative problem is carried out by relations [11, 12]:

$$s_1 = x_1 + x_{2,}.$$

$$s_2 = (x_1 - x_2) \exp(2x_3). \tag{6}$$

The new function is the governing function:

$$(\tau) = \exp(-2x_3). \tag{7}$$

The task of optimal control takes the following form:

$$J = s_1(\tau_f) \to \min, \tag{8}$$

$$ds_1 / d\tau = -R / 2(s_1 - s_2 w), s_1(0) = 1, \tag{9}$$

$$ds_2 / d\tau = 1 / w[R / 2 + 1](s_1 - s_2 w), s_2(0) = 1. \tag{10}$$

The control entered satisfies the following obvious conditions:

$$w_0 \le w \le 1, w_0 = \exp(-2E_f). \tag{11}$$

The Hamiltonian and the equations for the related variables λ_1, λ_2 are written in the form:

$$H = (s_1 - s_2 w)[-R(\tau) / 2 \cdot \lambda_1 + \{R(\tau) / 2 + 1\}\lambda_2 / w], \tag{12}$$

$$d\lambda_1 / d\tau = R(\tau)\lambda_1 - (R(\tau) / 2 + 1)\lambda_2 / w, \lambda_1(\tau_f) = -1, \tag{13}$$

$$d\lambda_2 / d\tau = -w[R(\tau)\lambda_1 - (R(\tau) / 2 + 1)\lambda_2 / w], \lambda_2(\tau_f) = 0, \tag{14}$$

Then the problem can be reduced to the corresponding nonlinear two-point boundary of the maximum principle. Optimal modes of qualitatively different types can be separated on the basis of the KSSH condition. Condition:

$$\arg\max_{w_0 \le w \le 1} H = w_0 \tag{15}$$

is the solution of the derivative problem at a given time interval. The condition for the existence of optimal laser exposure in the form of a single pulse has the form:

$$p = \exp(-2E_f)\{1 + 2 / R_0 + \exp[-(R_0 + 1)\tau_f]\} /$$
$$(1 + 2 / R_0)\{1 - \exp[-(R_0 + 1)\tau_f]\}, p \ge 1 \tag{16}$$

A formal expression for optimal control in the general case:

$$\tilde{u}(\tau) = \begin{pmatrix} E_1\delta(\tau) + \tilde{u}'(\tau), \tau \in [0, \tau_1] \mid p < 1 \\ 0, \dots\dots\dots / . \tau \in [\tau_1, \tau_f] \mid p < 1 \\ E_f\delta(\tau), \dots\dots\dots\tau \in [0, \tau_f] \mid p \ge 1 \end{pmatrix}. \tag{17}$$

The process of isotope separation is described by the following system of equations (see [12, 17]):

$$d\rho_0 / dt = -W_1(\rho_0 - \rho_1) + \rho_1 T_1 + K_1\rho_0'\rho_1,$$
$$d\rho_1 / dt = -W_1(\rho_0 - \rho_1) - \rho_1 T_1 - R\rho_1 - K_1(\rho_0'\rho_1 - \rho_1'\rho_0)$$
$$d\rho_0' / dt = -W_1(\rho_0' - \rho_1') + \rho_1' T_1 + K_1\rho_0\rho_1'$$
$$d\rho_1' / dt = -W_1(\rho_0' - \rho_1') - \rho_1' T_1 - R\rho_1' - K_1(\rho_0\rho_1' - \rho_1\rho_0'), \tag{17}$$
$$dn / dt = R\rho_1 - n_1(K_2^{(o)}\rho_0' - K_2^{(1)}\rho_1') + n'(K_2^{(0)}\rho_0 - K_2^{(1)}\rho_1) - n / t,$$
$$dn' / dt = R\rho_1' - n_1'(K_2^{(o)}\rho_0 - K_2^{(1)}\rho_1) + n(K_2^{(0)}\rho_0' - K_2^{(1)}\rho_1') - n / t,$$

де $\tau = \gamma \cdot t$ − dimensionless time; γ − the probability of spontaneous decay per unit time; ρ_0, ρ_0' − the concentration of atoms of matter and impurities in the ground state; ρ_1, ρ_1' − the concentration of atoms of matter and impurities in an excited state; n, n' − the concentration of their ions; coefficients K_1, K_2 − accordingly determine the speed of the process of resonant transfer of excitation energy: $K_1 = \sigma_w v$; σ_w − Weiskopf cross section; v − the speed of atoms; K_2 − the speed of the recharging process: $K_2 = \sigma_{ch}(v_i) v_i$; $\sigma_{ch}(v_i)$ − cross section of resonant recharging; v_i − the relative

velocity of ions relative to neutral atoms; W_1, W_2 – probabilities of radiation transitions. Probabilities of radiation transitions W_1, W_2 are determined by expressions:

$$W_1 = I_1 \sigma_1(\omega) / \hbar\omega_1 = I_1 (16\pi / cT_2)(\mu_{01} / 2\hbar)^2 \cdot [(1 / T_2)^2 + (\omega - \omega_{01})^2]^{-1},$$

$$W'_1 = I_1 \sigma'_1(\omega) / \hbar\omega_1 = I_1 (16\pi / cT_2)(\mu_{01} / 2\hbar)^2 \cdot (1 / T_2)^2 + (\omega - \omega'_{01})^2]^{-1}. \qquad (18)$$

where I_1− the intensity of the resonant radiation field; T_2 − transverse relaxation time; other designations are standard.

The above optimization model was implemented by us to determine the optimal scheme of selective ionization of radioactive isotopes by laser radiation with ionization by a pulsed electric field and through autoionization resonances, as well as ionization due to collisions. Note that in the future in the case of the implementation of the scheme of selective ionization by laser radiation with ionization by a pulsed electric field (and ionization through autoionization resonances) as R substituted values: $R \to W/\gamma$, where W − the ionization rate of excited atoms by a pulsed electric field. In the case of the implementation of the scheme of selective ionization by laser radiation with the mechanism of ionization due to collisions under R means the value: $R \to S/\gamma$, where S − the ionization rate of excited atoms due to the collision mechanism (e.g. [12, 17]).

Modelling results and conclussions. In the practical implementation of selective photoionization schemes by laser radiation, as a rule, a mixture of buffer and fissile gases (isotopes) moves across the zone of electric discharge, and the discharge zone is irradiated by an electromagnetic field resonant with one of the selected isotopes. (see Fig. 1, and [9, 10, 12-16]).

A buffer gas determines the characteristics of the discharge (temperature T, particle concentration n). The ionization time corresponds to the time of flight of the atom through the zone of electromagnetic fields: $t_f = L/v$, where L − the size of the irradiated
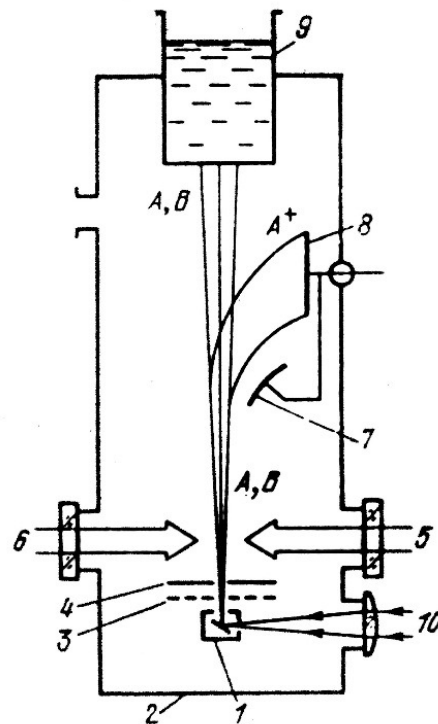

Fig. 1. Possible scheme for separation of isotopes in gas separator (atoms A in a beam in mixture with other atoms B): 1 – source of atomic beam; 2 − vacuum box; 3 – collector of non-selective ions; 4 – diaphragm; 5 – laser ray for the first step excitation; 6 – laser ray for second-step excitation to Rydberg states and further autoionization in a laser pulse or ionization by external electric field or the collisional ionization; 7 − deflecting electrodes; 8 – sublayer; 9 – cold sublayers for freezing atoms; 10 – laser ray for vaporising the substance

area; v − particle flow rate. If the radiation saturates the resonant transition and the ionization mechanism is realized due to collisions of excited atoms, then the ionization condition of the resonant component has the form: $1/nS'(T) > 1/v = t_f$, where S' − the ionization coefficient of excited atoms in the collision. These parameters satisfy the following value: $L = 1$ cm, $v = 10^4 \text{cm/s}$, $n = (10^{11} - 10^{13}) \text{cm}^{-3}$. Then it is convenient to make replacements: $R \rightarrow S'n/\gamma$, $\tau \rightarrow \gamma z/v$, $E_f = \sigma_{12}\overline{w}_0 / vd\hbar\varpi_{21}$, where z − spatial coordinate along the flow, d − transverse dimensions of the irradiated area. Typical parameter values: $S'n = 10^4 \text{c}^{-1}$, $\gamma = 10^4 \text{c}^{-1}$, $t_f = 4 \cdot 10^{-4} \text{c}$, $E_f = 2.5$. Consider the scheme for Rb. Stage 1 uses laser radiation with a wavelength of 7950A (excited to the state $5p\,^2P_{1/2}$); laser pulse ionization (quantum energy 2.62 eV). For Rb vapors at 100ºC (pressure 10^{-4} torr) Doppler absorption width $\Delta\omega_D = 4 \cdot 10^9 \text{c}^{-1}$, cross-section of excitation $\sigma_1 = 10^{-11} \text{cm}^2$, cross section of photoionization from the excited state $\sigma_2 = 10^{-18} \text{cm}^2$. In the case of the photoionization scheme with excitation of Rydberg S and D states с $n = 12$–18 and ionization by an electric field (~30kV/cm) the calculation (c.g.[17,33]) gives transition $5\,^2P_{1/2} - 16\,^2D_{3/2}$ cross-section of excitation − $\sigma_2 = 0.88 \cdot 10^{-14} \text{cm}^2$. This is 10 (+5) more than the cross section of ionization from the ground state and 10 (+4) – from the low excited state. For the classical scheme of photoionization of Rb atoms, the energy density for saturation of resonant absorption: $\Phi_{sat}^{(1)}\hbar\varpi_1 = 1,2 \cdot 10^{-8} \text{J/cm}^2$ and to saturate the photoionization transition: $\Phi_{sat}^{(2)}\hbar\varpi_2 = 0,42 \text{J/cm}^2$. Parameter (16) depends in a complex way on all physical parameters of the optimal control problem: relaxation rates, photoexcitation and photoionization, energy and duration of the laser pulse, electric field pulse (e.g. [11-17, 32-36]). With moderate requirements for the parameters of the laser pulse by appropriate selection of pulse durations and quantum transitions, it is possible to achieve a maximum of up to 100% ionization yield. This implies a fairly short exposure time $\exp[-(R+1)\tau_f] \sim 1$ in formula (16) at a sufficiently economical value of the energy of the laser radiation E_f. In the laser photoionization scheme with different final ionization scenarios, the optimal scheme will be if the atom is excited by laser radiation to a state that has a probability of decay in an electric field (autoionization decay) greater than the probability of radiation decay. Fig. 2 illustates the results of modeling the optimal shape of a laser pulse in the problems of selective photoionization of the Rb isotopes (rubidium vapuor by laser radiation with ionization by a pulsed electric field (autoionization resonances), as well as typical behavior of populations of for the ground (curve 1) and excited states (curve 2).

Analysis shows that, as in the problem of classical two-stage photoionization, depending on the physical parameter p (16), two qualitatively different modes are realized from the point of view of the theory of optimal control: p>1,p<1. For Rydberg levels, the cross section of photoionization (due to electric field-induced autoionization resonances) increased sharply in comparison with ionization from the

low-excited state. In this case, the δ-pulse provides the maximum possible level of excitation of the upper state and then parasitic processes such as spontaneous relaxation and the corresponding collision processes in a short time can not significantly change the degree of excitation achieved. If the pulse of the electric field is turned on after the end of the laser pulse, it provides a high degree of ionization (100% ionization occurs only from the last highly excited state). In the case of using a continuous electric field that devastates the final state of the atom during laser pulses, the scheme will not be optimal due to the strong Stark shift of the highly excited levels. Although the advantage, as already mentioned, is that, acting on the atom by an external electric field, it is possible to control the structure of the levels of highly excited states and the optical properties of atoms. The strong Stark effect allows you to adjust the absorption spectra at the last stage of excitation to the generation frequency of a narrowband unconstructed laser (gas discharge type). As a result, the efficiency of the whole process of photoionization of atoms increases if the parameter $p<1$ (formula (15)), i.e. one should talk about large values of a given energy of the pulse of resonant radiation $E_f >>1$ and not very low energy and pulse duration ($R\tau_f \geq 1, \tau_f \sim 1$) ionizing radiation. The optimal mode of laser exposure will include at the end of the so-called passive control area (see formula (16), for the first time it is provided for the scheme of classical photoionization in the KSSH model [11]. In our case, its appearance is associated with the final ionization rate and the subsequent inefficiency of the energy input into the resonant channel of the optimization process. The dispersion of part of the energy in the time interval of finite length will reduce the negative role of the reverse forced spontaneous radiation processes and reduce the equalization of populations of all levels used, which is especially important for the circuit with ionization by electric field. In our case, its appearance is associated with the final ionization rate and the subsequent inefficiency of the energy input into the resonant channel of the optimization process. The dispersion of part of the energy in the time interval of finite length will reduce the negative role of the reverse forced spontaneous radiation processes and reduce the equalization of populations of all levels used (e.g. [12-17]).

Further let us consider a laser photoionization scheme of the uranium isotopes. Iot is worth to remind that a detailed account of the experiments on laser fission of uranium isotopes in the framework of the "Exxon Nuclear" and "Avco Everett" programs is given in [6,7,12,31] (and refs therein). According to Ref. [17], the possibnle scheme of selective photoionization of uranium isotopes could include an excitation of atoms [235]U from the main ($5f^3 6d7s^2$-$^5L_6^o$) and low-lying metastable state ($5f^3 6d7s^2$-$^5K_5^o$ with energy 620,32 cm^{-1}) by a laser radiation in the first stage, the further transition to a narrow autoionization state with a double-excited outer shell and then ionization by an electromagnetic field. To remove atoms [235]U from the ground and low-lying metastable state (with energy 620 cm^{-1}) dual-frequency radiation is used in the first place. The required uranium vapor pressure (about 1 torr) is created by heating liquid uranium to 2500°C using an electron beam. Uranium vapors emitted by a hot source pass through parallel plates, between which the atoms are irradiated with 4-frequency laser radiation, photoions formed [235]U gather on the collector
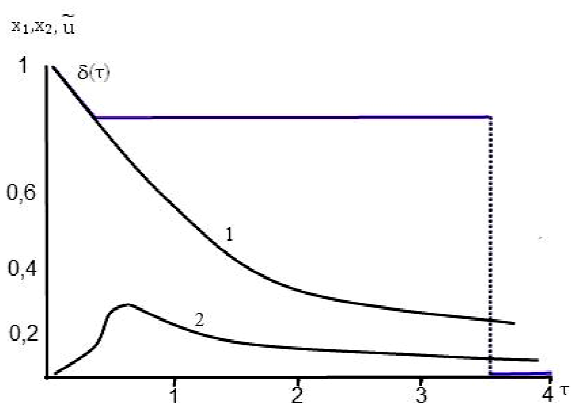
Fig. 2. The results of numerical simulation of the problem of photoionization of Rb atoms by laser radiation with ionization by a pulsed electric field: δ+ dotted line − the optimal shape of the laser pulse, curves 1 and 2 − the corresponding behavior of the populations of the ground and excited states
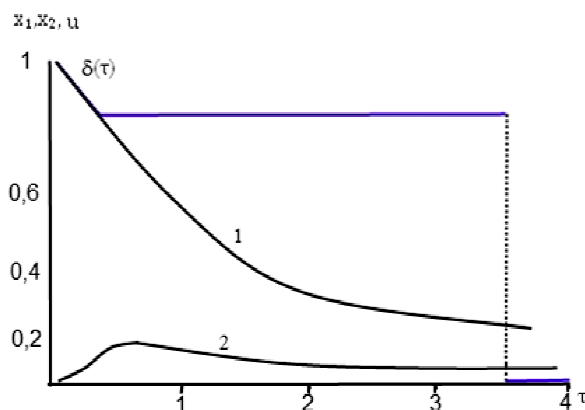
Fig. 3. The results of numerical modeling of the optimal shape of the laser pulse in the problem of photoionization separation of uranium isotopes: δ+ dotted line − optimal shape of the laser pulse, curves 1 and 2 − the corresponding behavior of the population of the ground and excited states

plates, and neutral atoms ^{238}U pass by. The collision of ions with U atoms significantly limits the selectivity of separation [12]. The ionization time corresponds to the time of flight of the atom through the zone of electromagnetic fields: $\tau = t_f, = L/v$ (L − the size of the irradiated area; v − particle flow rate). If the radiation saturates the resonant transition and the scheme of highly excited atoms is realized by pulsed laser radiation, then the ionization condition of the resonant component has the form: $1/nS'(T) > 1/v = t_f$, where S' − the ionization coefficient of excited atoms. The stage of optimization of the laser separation model is to find the optimal shape of the electromagnetic pulse of resonant radiation, which provides max of ionized atoms in the gas isotope separation scheme (one of the possible formulations). Fig. 3 shows the results of simulation of the optimal shape of the laser pulse in a problem of the uranium isotopes separation.

The δ-pulse could provide the maximum possible level of excitation of the upper state and then parasitic processes such as spontaneous relaxation, resonant excitation transmission and resonant recharging in a short time, which can not significantly change the degree of excitation. As a result, the efficiency and optimality of the entire separation scheme could increase. If the above conditions are not met, the optimal mode of laser exposure will contain at the end of the so-called passive control area, first provided for the scheme of classical photoionization (e.g. [11, 12]). Its appearance is associated with the final ionization rate and the subsequent inefficient input of energy into the resonant channel at the end of the process. The redistribution of radiation energy eliminates the harmful role of reverse forced and spontaneous radiation transitions for the process of photoionization separation. The main result of

the calculations is that the described method of modeling optimal schemes of laser photoionization separation of isotopes allows to choose optimized values of key physical parameters, the most optimal variant of the scheme as a whole, which can ensure the efficiency of isotope and nuclear isomer separation technology.

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Оптимальна лазерно-фотоіонізаційна схема розділення важких ізотопів в газових сепараторних приладах

АНОТАЦІЯ

Запропоновано ефективний підхід до визначення параметрів оптимальних схем методу лазерної селективної фотоіонізації атомів (елементів і ізотопів) з кінцевою іонізацією за рахунок зіткнень, іонізацією імпульсним електричним полем, іонізацією через високо лежачі (рідбергівські) стани і вузькі автоіонізаційні резонанси для поділення важких ізотопів в газових сепараторних пристроях. На основі теорії оптимального управління і розроблених раніше квантових моделей обчислення характеристик елементарних атомних процесів чисельно реалізовані оптимізаційні моделі поділення ізотопів в схемі селективної лазерної фотоіонізації з іонізацією за рахунок зіткнень в газових сумішах, іонізацією імпульсним електричним полем, автоіонізацією і т.і. Отримані дані кількісно підтверджують перспективність методу лазерної фотоіонізації з кінцевої іонізацією за рахунок зіткнень, іонізацією імпульсним електричним полем, іонізацією через рідбергівські стани та вузькі автоіонізаційні резонанси і дають набір параметрів шуканих оптимальних схем, зокрема, оптимальної форми лазерного імпульсу для ізотопів рубідію та урану.

Ключові слова: газовий сепаратор, поділення ізотопів, метод лазерної фотоіонізації, теорія оптимального управління

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Оптимальная лазерно-фотоионизационные схема разделения тяжелых элементов в газовых сепараторных устройствах

АННОТАЦИЯ

Предложен эффективный подход к определению параметров оптимальных схем метода лазерной селективной фотоионизации атомов (элементов и изотопов) с конечной ионизацией за счет столкновений, ионизацией импульсным электрическим полем, ионизацией через высоко лежащие (ридберговских) состояния и узкие автоионизацийные резонансы для разделения тяжелых изотопов в газовых сепараторных устройствах. На основе теории оптимального управления и разработанных ранее квантовых моделей вычисления характеристик элементарных атомных процессов численно реализованы оптимизационные модели разделения изотопов в схеме селективной лазерной фотоионизации с ионизацией за счет столкновений в газовых смесях, ионизацией импульсным электрическим полем, автоионизацией и т.д. Полученные данные количественно подтверждают перспективность метода лазерной фотоионизации с конечной ионизацией за счет столкновений, ионизацией импульсным электрическим полем, ионизацией через высоко лежащие (ридберговских) состояния и узкие автоионизацийные резонансы и дают набор параметров искомых оптимальных схем, в частности, оптимальной формы лазерного импульса для изотопов рубидия и урана.

Ключевые слова: газовый сепаратор, разделения изотопов, метод лазерной фотоионизации, оптимальное управление.