Optimized laser photoionization scheme of separation of complex isotopes in the separator devices

An effective approach to determining parameters of optimal schemes of the method of laser selective photoionization of heavy isotopes, in particular, lanthanides (Gd atoms as the example) with ionization at the final stage by a pulsed electric field, autoionization through the narrow autoionization resonances and due to collisions is developed for separation of heavy isotopes in the separator devices. On the basis of the theory of optimal control and previously developed theoretical quantum approaches (the energy formalism and the relativistic many-body perturbation theory with the Dirac-Kohn-Sham-Sturm zeroth approximation for computing the parameters of elementary atomic processes such as excitation, ionization amplitudes, cross-sections, energies, widths of autoionization resonances etc.) an optimized scheme for the separation of Gd isotopes by the method of 3-stage laser photoionization with ionization at the final stage by a pulsed electric field, ionization through narrow autoionization resonances is presented. For the first time, there are theoretically calculated parameters of the narrow autoionization resonances for Gd in a sufficiently weak electric field, which are in physically reasonable agreement with data of the known experiment by Letokhov et al. Narrow-width autoionization resonances in Gd (in general in spectra of any lanthanide & actinide atom) have a relatively long lifetime, correspondingly, their excitation, ionization cross sections have the same order as the excitation one at the initial stage of separation scheme. Therefore, the use of these long-lived states can ensure the optimal implementation of a laser scheme for the separation of heavy isotopes. The obtained results for Gd with using models of optimal governing confirm the perspective of construction of the optimized schemes of the laser photoionization method with ionization at the final stage by a pulsed electric field, autoionization and allow determining the optimal parameters of the separation scheme, including diagram of atomic transitions, shape of laser pulses, etc. It is hoped that this work will also provide new opportunities for further development of laser chemistry of lanthanides, as well as actinides, taking into account new physics of autoionization resonances in their spectra.

Keywords: separation of heavy isotopes, method of selective laser photoionization of atoms, theoretical quantum models of elementary atomic processes of excitation and ionization, narrow autoionization resonances, models of optimal governing, gadolinium isotopes

Introduction. Studying of physical and chemical processes with participation of atomic systems (gases) in the isotopes and gases separator devices is of a great interest for many applied topics, relating to physics of atoms (molecules), laser physics and quantum electronics, plasma and gases physics. The intensive theoretical and experimental investigations are carried out in a field of studying and constructing the optimal laser photoionization schemes of separation of different atomic elements (isotopes) in the vapour state in the gas separator devices (e.g. [1-19]). The problem of developing, improving, and finding new optimal implementations of methods of nonlinear selective photoionization of atoms and molecules by laser radiation is one of
the most urgent, complex, and far from definitive solutions. Their great importance is due to the unique and high efficiency of their potential application in solving many problems of quantum electronics, laser, nuclear physics, chemistry, and related technologies.

Laser methods are characterized by much lower energy consumption than traditional methods, in addition, they have exceptional selectivity and provide the possibility of non-contact control and manipulation atoms using electromagnetic fields (e.g. [1-5]). This is what determines their extremely promising use in solving the problems of distribution of isotopes, isomers, processing of radioactive waste of nuclear energy and nuclear technologies. Although the first successful experiments on the laboratory implementation of the laser method (classical two-stage schemes of selective photoionization of atoms, photodissociation of molecules) of isotope separation (V. Letokhov et al., B. Moore et al) were carried out relatively long ago (e.g. [1-4, 10]), and, moreover, their basic feasibility and prospects for further industrial implementation were successfully demonstrated on a laboratory scale, but the sought selective photoionization schemes do not have sufficient efficiency and optimality. In this light, schemes of selective photoionization of atoms by a laser field with ionization by a pulsed electric field (including through Rydberg states), autoionization (through narrow autoionization resonances), ionization due to collisions (associative ionization, etc.) can be considered more promising (e.g. [20-26]). Until now, their consideration has been carried out mostly only at the qualitative level. There is practically no information about main characteristics of the mentioned schemes, the possibilities of their optimal implementation. There is also no adequate understanding of the role of collisional ionization processes. Though a great progress has been reached in the last two decades, however the principal problems remain, and it has a critical character for heavy isotopes such as the lanthanides and actinides elements etc.

In this work we go on our search and study of the effective schemes of the laser-photoionization separation of the complex isotopes, based on selective resonant excitation of complex isotopes (neutral atoms) by laser radiation into quantum states near the ionization limit and subsequent autoionization decay under the action of an external electric field, autoionization or collision. The methods of an optimal control theory (e.g.[5-10]) and previously developed theoretical quantum approaches (the energy formalism and the relativistic many-body perturbation theory with the Dirac-Kohn-Sham-Sturm zeroth approximation) [26-33] are used. The element of novelty is connected with discovery of the optimized laser photoionization of atoms due to the autoionization, or ionization by pulsed electric field. Besides, the optimal control models could provide an effective way to determine the optimal shape of the laser pulse to get the maximum of ionized particles in the scheme of selective atomic photoionization and whole laser separation device. Gadolinium has seven naturally occurring isotopes $^{152}\text{Gd}$ (0.20%), $^{154}\text{Gd}$ (2.18%), $^{155}\text{Gd}$ (14.80%), $^{156}\text{Gd}$ (20.47%), $^{157}\text{Gd}$ (15.65%), $^{158}\text{Gd}$ (24.84%) and $^{160}\text{Gd}$ (21.86%). Among these isotopic components $^{157}\text{Gd}$ has the highest thermal neutron cross-section (254x10$^3$ b). An atomic vapour laser photoionization allows to have the process to get the highly enriched product with a content greater than 90% in $^{157}\text{Gd}$ from natural gadolinium, and many improvements will be achieved (e.g. [14,15]). The possible optimal laser separation
scheme for the Gd isotopes is experimentally earlier studied (e.g., [1, 14, 15]), however, the principal problems remain, and have a critical character.

**Theoretical model of optimal control of the atomic laser photoionization.** As the description of the models has been presented earlier, here we will focus only on the key points. The optimal control problem is formulated as [16] (e.g. [5, 6]):

\[
J = -\int_0^{\tau_f} R(\tau)x_2d\tau \to \min; \quad dx_1/d\tau = x_2 - \bar{u}(x_1 - x_2), \quad x_1(0) = 1
\]

\[
dx_2/d\tau = -[R(\tau) + 1]x_2 + \bar{u}(x_1 - x_2), \quad x_2(0) = 0,
\]

where \(x_1\) is the normalized population of the ground state of an atom; \(\bar{u} = u/\gamma\) is a dimensionless rate of induced emission and absorption processes of the resonant radiation; \(\gamma\) is the probability of spontaneous decay per unit time; \(\tau = t\gamma\) is a dimensionless time; \(R = R'/\gamma\) is a dimensionless ionization rate from the excited state; \(u(t) = \sigma_{12}I_1(t)/\hbar\sigma_{21}\) is the rate of induced transitions (1→2 transition); \(\sigma_{21}\) is the absorption cross section for the 1→2 transition; \(R'(t) = \sigma_{ph}I_2(t)/\hbar\sigma_{ph}\) is the photoionization rate; \(\sigma_{ph}\) is the photoionization cross section; \(I_1, I_2\) are the intensities of the laser pulses for the excitation from the ground state and for the ionization from the excited level respectively; \(E_f\) and \(\tau_f\) are the energy and the duration of the pulse of resonant radiation, respectively. The transition to the derivative problem is carried out using the formulas:

\[
s_1 = x_1 + x_2, \quad s_2 = (x_1 - x_2)e^{2x_3} (\text{new control governing function } w(\tau) = e^{-2x_3}).
\]

Hamiltonian and equations for conjugate variables \(\lambda_1, \lambda_2:\)

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

\[
d\lambda_1 / d\tau = R(\tau)\lambda_1 - (R(\tau)/2 + 1)\lambda_2 / w, \quad \lambda_1(\tau_f) = -1
\]

\[
d\lambda_2 / d\tau = -w[R(\tau)\lambda_1 - (R(\tau)/2 + 1)\lambda_2 / w], \quad \lambda_2(\tau_f) = 0
\]

The condition for existence of optimal laser single pulse has the form [9, 24]:

\[
p = \exp(-2E_f)[1 + 2/R_0 + \exp[-(R_0 + 1)\tau_f]] / (1 + 2/R_0)[1 - \exp[-(R_0 + 1)\tau_f]], p \geq 1
\]

where the parameter \(p\) has an expression similar to the analogous parameter in the Krasnov-Shaparev-Shkedov scheme [5]. The optimal resonant laser pulse is given by:

\[
\tilde{u}(\tau) = \begin{cases} 
E_i\delta(\tau) + \tilde{u}'(\tau), & \tau \in [0, \tau_{in}] | p < 1 \\
0, & \tau \in [\tau_{in}, \tau_f] | p < 1 \\
E_f\delta(\tau), & \tau \in [0, \tau_f] | p \geq 1 
\end{cases}
\]

where \(\delta(\tau)\) is the Dirac delta-function, \(E_i (E_i < E_f)\) is the amplitude of the \(\delta\)-pulse; \(\tau_{in}\) is the time parameter, which is matched numerically (\(\tau_{in} \approx \tau_f\)); \(R_0\) is the ionization rate from the ground state of atom. More details of the model can be found in [16-19].
From theoretical viewpoint, computing spectra, radiative transition and autoionization parameters of the rare-earth elements is very complicated task because of the necessity of the correct accounting for the exchange-correlation (including polarization and screening effects, a continuum pressure etc) and relativistic corrections (and also radiative and nuclear effects in a case of the super heavy atomic systems). The necessary data about of the characteristics of the elementary atomic processes (excitation, ionization amplitudes and cross-sections, autoionization states energies, widths, constants etc) as well as the corresponding atomic spectra have been obtained on the basis of computing within the combined approach of the relativistic energy approach and gauge-invariant relativistic many-body perturbation theory (RMBPT) formalism with the optimized ab initio density-functional zeroth approximation with correct gauge-invariance principle fulfilling and precise accounting for the complex multibody exchange-correlation effects (both for energy spectra, as well as amplitudes, excitation and ionization probabilities and cross sections), as the RMBPT second and higher orders ones, including exchange-polarization and shielding interquasiparticle interaction, effects of rapid “smearing” of the initial state over a non-sharp set of configurations and significantly non-Coulomb grouping of levels in Rydberg spectra, “pressure” of the Rydberg and continuum states within the effective relativistic Dirac-Sturm expansion method (e.g. [26-33]). The optimal RMBPT zeroth approximation is based on an accurate treating the lowest order multielectron effects, in particular, the gauge dependent contribution into the atomic levels radiation widths for the certain class of the photon propagator (i.e., the Coulomb, Feynman, Babushkin) gauge [26,29]. The dynamics of the autoionization resonance in DC & AC electric field is studied within the operator perturbation theory (e.g. [25,29]).

As the detailed description of the method is listed earlier, here we only give the key definitions. A width $G=\Gamma$ of an autoionizing resonance is determined by a link with continuum states and is given as follows [29, 32]:

$$\Gamma(n_1^0, j_1, n_2^0, j_2, J) = \frac{2\pi e^2}{K_0} \sum_{\beta_1, \beta_2} \sum_{\beta_1, \beta_2} C^J(\beta_1, \beta_2) C^J(\beta_2, \beta_1) \sum_{\beta \rho_1, \beta \rho_2} V_{\beta \rho_1, \beta \rho_2} V_{\beta \rho_2, \beta \rho_1}, \quad (5)$$

$$C^J(\beta_1, \beta_2) = C^J(n_1, j_1; n_2, j_2) A(j_1, j_2; J, m_1, m_2, JM), \quad (6)$$

$$A(j_1, j_2; J, m_1, m_2, JM) = (-1)^{j_1-j_2+M} \left( \frac{j_1 \cdot j_2}{m_1 \cdot m_2 - M} \right) \sqrt{2J+1}, \quad (7)$$

$$C^J(n_1, j_1; n_2, j_2) = N(n_1^0, j_1^0; n_2^0, j_2^0) \delta(n_1^0, j_1^0; n_1, j_1) \delta(n_2^0, j_2^0; n_2, j_2) + \delta(n_1^0, j_1^0, n_2, j_2) \delta(n_2^0, j_2^0, n_1, j_1) \quad (8a)$$

$$N(n_1^0, j_1^0; n_2^0, j_2^0) = \begin{cases} 1, & n_1^0 j_1^0 = n_2^0 j_2^0 \\ \frac{1}{\sqrt{2}}, & n_1^0 j_1^0 \neq n_2^0 j_2^0 \end{cases} \quad (8b)$$

The matrix elements of the relativistic interelectron interaction potential $V(r_i, r_j) = \exp(i\omega, r_i \cdot (1-\alpha, r_j) / r_j$ are determined as follows (e.g. [27, 29]):

$$V_{\beta \rho_1, \beta \rho_2} = (2j_1 + 1)(2j_2 + 1)(2j_3 + 1)(2j_4 + 1)(-1)^{j_1+j_2+j_3+j_4+m_1+m_2} \times$$

$$\times \sum_{\alpha \mu} (-1)^{\mu} \left( \begin{array}{ccc} j_1 & j_3 & a \\ m_1 - m_3 & \mu & \end{array} \right) \left( \begin{array}{ccc} j_2 & j_4 & a \\ m_2 - m_4 & \mu & \end{array} \right) \times Q_{a} (n_1 l_1, j_1; n_2 l_2, j_2; n_3 l_3, j_3) \quad (9)$$
where \( Q_a = Q_a^{\text{Qul}} + Q_a^{\text{Br}} \); \( Q_a^{\text{Qul}} \), \( Q_a^{\text{Br}} \) are corresponding to the Coulomb and Breit parts of the potential \( V \). In Refs. [20, 24, 25] it has been discovered a principally new spectroscopic effect of a giant broadening autoionization resonances of the complex heavy atoms (Gd, Tm, Yb etc) in sufficiently weak electric (laser) field. This new effect is of a great importance for problem of laser separation of heavy isotopes and nuclear isomers, spectroscopy and photochemistry, green chemistry [1, 6, 16-18, 29].

**Modelling results and conclusions.** Here we consider a laser photoionization scheme of the Gd isotopes. Some useful results on laser separation of the Gd isotopes are listed in Ref. [1, 14, 15] with using the magnetic field effect. The rich physics of the autoionization resonances in spectra of the lanthanides atoms opens new perspectives in different applications [20, 23-25, 32]. These perspectives seem undoubtedly promising from the point of view of considering new possibilities for optimizing the laser ionization scheme. We mean the lanthanides isotope separation schemes with excitation by a laser radiation in the first stage, the further transition to a narrow autoionization states near the ionization threshold and final autoionization or ionization by an electric (laser) pulse field. The cited effect was firstly discovered for Gd in experiment [20]. In the known experiment by Letokhov et al it had been realized the three-stage scheme of the Gd excitation and was firstly observed very interesting autoionization state. In Fig. 1 the diagram of the Gd energy levels involved in the 3-step excitation and ionization is shown, as well as the quantum transitions used in the experiment [20]. The dye laser at 1\(^{\text{st}}\) excitation stage (5618Å) transferred the Gd atoms from the ground state \( 4f^75d6s^26^2D_2^0 \) to the state \( 4f^75d6s6p^9D_4^0 \). At the 2\(^{\text{nd}}\) stage laser pulse (6351.7Å) transferred the atoms to the state \( 4f^75d6s7s^9D_4^0 \). To ensure excitation of Rydberg states lying above the ionization limit by 300 cm\(^{-1}\), the wavelength of the 3\(^{\text{rd}}\) laser was tuned in the range 6300-6100Å. In Fig. 2 there is listed the dependence of ion current for Gd upon laser wavelength at the 3\(^{\text{rd}}\) stage in the region of 6110-6240Å and the same dependence in the vicinity of autoionization resonance at \( \lambda_3 = 6133.5 \) cm\(^{-1}\) (laser spectrum width \( \Delta \nu_3 = 0.03 \) cm\(^{-1}\)): \( G_1 \) is the resonance width (without field; Letokhov experiment); \( G_3 \) -resonance width in the field 100V/cm from the Letokhov et al experiment; \( G_2 \) is our RMBPT calculation result for the width. Detailed study of this dependence in the wavelength range 6220-6225Å made it possible to find the ionization potential \( E_i = (49588 \pm 5) \) cm\(^{-1}\), which is in good agreement with the NBS experimental data (e.g., [1]). According to the data [20], the autoionization resonance width (without a field) \( G_1 = 0.07 \) cm\(^{-1}\). Our theoretical value (scheme of computing is above) for this width in a field 100V/cm: \( G_2 = 0.31 \) cm\(^{-1}\) is in the physically reasonable agreement with the experimental value [20]: \( G_3 = 0.34 \) cm\(^{-1}\). The narrow autoionization resonances have a relatively long lifetime, and their excitation cross-sections are comparable to the excitation ones of low-lying states. So, long-lived autoionization states can be effectively used in the method of laser photoionization of atoms due to the radical increase in the photoionization cross-section at the final step. The aim of optimization of the laser photoionization separation model is to find the optimal shape of the laser pulse of resonant radiation, which provides max of
ionized atoms in the isotope separation device. Above presented optimal control model has been applied to the Gd isotopes separation. Fig. 3 shows the results of simulation of the optimal shape of the laser pulse for the Gd isotopes separation.

The physical analysis shows that the initially δ-pulse could provide the maximum possible level of excitation of the upper state, and the elementary parasitic processes (first of all, spontaneous relaxation, resonant excitation transmission and resonant recharging in a short time) can not significantly change the degree of excitation. As a result, the efficiency and optimality of the entire scheme increases. The optimal mode of laser exposure will include at the end the so-called passive control section, first predicted for the classical photoionization scheme in the models [5,16]. Its appearance is connected with the final rate of ionization and the consequent efficient input of energy into the resonant channel at the end of the process. The redistribution of the energy makes it possible to eliminate the harmful role of parasitic processes.

Conclusions. The main result of the work is that the presented approach with elements of the optimal governing and control allows to choose the optimized

---

**Fig.1.** The diagram of the Gd energy levels involved in the 3-step excitation and ionization is shown, as well as the quantum transitions used in the experiment [20].

**Fig.2.** Dependence of ion current for Gd upon laser wavelength at 3rd excitation stage in the region of 6110-6240 Å; The block in a circle presents the same dependence in the vicinity of resonance at λ₃ = 6133.5 cm⁻¹ (laser spectrum width Δν₃ = 0.03 cm⁻¹): G₁ is the resonance width (without field; Letokhov et al experiment [20]); G₃ is the resonance width in the field from experiment [20]; G₂ is our RMBPT calculation result of the autoionization resonance width; (as the resonances with widths G₂ and G₃ merge in the figure, so they are depicted with a slight spacing)
parameters of the laser separation scheme with ionization by a pulsed electric field, autoionization etc and ensure the efficiency of the isotopes separation process and technology. It should be noted that the presented approach can be also effectively used in searching for optimal schemes of a gamma-laser on rapidly decaying nuclear isomers and other similar problems [34-37].

**References:**


**Fig. 3.** The results of modeling of the optimal laser photoionization separation scheme for the Gd: δ+ dotted line – optimal shape of the laser pulse, curves 1 and 2 – the corresponding behavior of population of the ground and excited states (see text)


О.В. Глушков, О.Н. Софронков, Ю.В. Дубровська, Т.О. Флорко

Оптимальна лазерно-фотоіонізаційна схема розділення складних ізотопів в сепараторних приладах

АНОТАЦІЯ

Запропоновано ефективний підхід до визначення параметрів оптимальних схем лазерної/photoіонізації важких ізотопів (як приклад розглянуто атоми Gd) з іонізацією на останній стадії імпульсним електричним полем, автоіонізацією через вузькі автоіонізаційні резонанси, або за рахунок зіткнень для поділення ізотопів в сепараторних пристроях. На основі теорії оптимального управління і розроблених раніше теоретичних моделей підходів (енергетичний формалізм, релятивістська багаточастинкова теорія з близьким наближенням Дірака-Кона-Шема-Штурму) для обчислення характеристик елементарних атомних процесів (амплітуд, перерізів збудження, іонізації, енергій, ширина автоіонізаційних резонансів, тощо) пропонується оптимізоване схема поділення ізотопів Gd методом з-ступенчатої лазерної/photoіонізації з іонізацією на фінальній стадії імпульсним електричним полем, іонізацією через вузькі автоіонізаційні резонанси. Вперше теоретично обчислені параметри вузьких автоіонізаційних резонансів для Gd в достатньо слабкому електричному полі у відмінності від аналогічних відомих експериментам. Вузькі автоіонізаційні резонанси в Gd (очевидно, вагалі для будь-якого атому лантанідів або актинідів) мають порівняно великі часи життя, відповідно їх перерізи збудження, іонізації чисельно мають такий ж самий порядок як і перерізи збудження на початковій стадії схеми. Тому використання цих станів може забезпечити оптимальну реалізацію лазерної схеми поділення важких ізотопів. Отримані результати для Gd підтверджують перспективність побудови з використанням моделей оптимального керування ефективних схем методу лазерної фотоіонізації з іонізацією на фінальній стадії імпульсним електричним полем, автоіонізацією та дозволяють використати оптимальні параметри схеми, у т.ч., діаграми атомних переходів, форму лазерних імпульсів тощо. Є сподівання, що дана робота забезпечить також нові можливості подальшого розвитку лазерної хімії лантанідів, а також актинідів з урахуванням нової фізики автоіонізаційних резонансів в їх спектрах.

Ключові слова: поділення важких ізотопів, метод селективної лазерної/photoіонізації атомів, квантові моделі елементарних атомних процесів збудження та іонізації, вузькі автоіонізаційні резонанси, моделі оптимального керування, ізотопи гадолінію.