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Coherent kinetics of a multistage λ scheme for laser isotope separation

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Analytical solutions of the Schr?dinger equations for coherent excitation of an atom in the scheme of two- and three-stage photoionization are obtained as applied to the problem of laser isotope separation. A feature of the study is the introduction of laser excitation from an additional initial state to increase the fraction of the atoms involved in the separation process. The optimal from the points of view of photoion ratios of radiation intensities at individual photoionization stages are found.

Key words: laser photoionization, laser isotope separation.

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Introduction

One of the most important parameters of laser isotope separation is the degree of the target isotope extraction using photoionization. The essence of the method is that the flow of atoms, formed by the evaporator in vacuum, passes through the working volume — the region filled with laser radiation, in which selective photoionization of the target isotope occurs. The photoions of the target isotope are pulled out of the flow by the electric field to the product collector, while the atoms of other isotopes, remaining neutral, continue their way to the waste collector. The components of the degree of the target isotope extraction are the efficiency of photoionization, the probability of irradiating an atom with pulsed laser radiation, and the probability of extraction and retention of a photoion on the collector. In this context, the photoionization efficiency is understood as the probability of photoionization of an atom, that has been subjected to laser irradiation. This probability depends, in addition to the parameters of transitions and laser radiation, on the fraction of irradiated atoms, which are generally involved in the photoionization process. Most atoms have either a ground state or split, or have nearby metastable states. Thus, the population of the starting level, which is used in the photoionization scheme, turns out to be far from 1. For example, upon photoionization of the ^{150}Nd isotope, the population of the ground state $^5I_4$ at the neodymium evaporation temperature is 0.58, while there is a metastable state $^5I_5$ with energy 1128 cm^{-1} and population 0.26 [1]. When using in the photoionization scheme the first transition to an excited level with the total electronic moment of the atom $J = 4$ or $J = 5$, it is possible to use an additional laser, that provides a transition from the metastable state $^5I_5$ to the same level, allowing the second and third power lasers to photoionize these additional atoms. This will increase the fraction of atoms involved in the photoionization process by 45%. A similar situation exists in actual cases of photoionization separation of ^{63}Ni, ^{176}Lu, ^{177}Lu, and ^{177}Lu isotopes.

In the case of lutetium, the population of the ground state at an evaporation temperature of 1700°C is ~ 0.7, but the ground state, like other levels, is split into multiplets due to hyperfine interaction. The sublevels in the multiplet are determined by the total atomic moment $F = J + I$, $J + I - 1, \ldots, |J - I|$, where I – is the nuclear spin. The splitting, as a rule, exceeds the spectral width of the laser radiation, and at a certain frequency setting of the lasers of the first, second, and third stages, photoionization occurs taking into account the selection rules ($\Delta F = 0, \pm 1$) along a certain chain of sublevels F — along the photoionization channel. For example, for ^{177}Lu the sublevels of the ground state $5d6s^2\ ^2D_{3/2}$ ($I = 7/2$) with quantum numbers $F = 2, 3, 4, 5$ are populated in accordance with their statistical weights $2F + 1$, and accordingly, when using the photoionization channel 5-6-5-4 [2], $0.7.11/32 = 0.24$ are involved in the photoionization process from all irradiated atoms. At the same time, the simultaneous use of channels 2-3-4-3 and 4-3-4-3 is possible, in this case the fraction of involved atoms $0.7 \cdot (9 + 7)/32 = 0.35$ turns out to be by 46% larger. The use of two initial states with excitation to the same sublevel leads to the formation of the so-called λ photoionization scheme. However, under certain conditions [3], the presence of a side stage can, on the contrary, suppress the atoms excitation due to „dark states“ (population trapping „dark“ states), which arise under the condition of an exact two-photon resonance between two initial levels.

In the laser separation of isotopes of rare earth elements, one has to often deal with small (about 1 GHz) frequency differences during excitation of various isotopes, that requires the use of narrow-band single-mode dye lasers with a spectral width of 120−150 MHz. When using such lasers, the experimenter often encounters the effects of splitting atomic transitions and a decrease in the photoion current

with increasing laser radiation intensity, that is explained in terms of the coherent interaction of laser radiation with an atom.

The theoretical consideration of multistage coherent photoionization of classical schemes (not λ-schemes) is widely presented in the literature [4–7]. The most universal condition for effective photoionization in such schemes is the equality of the Rabi frequencies f_i at each transition. In particular, in the case of a three-level system, when the laser frequency is tuned to resonance with transitions, the time-averaged population of the upper excited state W_3 in the absence of relaxation is proportional to the factor $\frac{f_1^2 f_2^2}{(f_1^2+f_2^2)}$ [4], which maximum corresponds to the condition $f_1 = f_2$, where f_1 and f_2 are the Rabi frequencies at the first and second transitions respectively. Unfortunately, this condition cannot be directly extended to the case of a λ-scheme. This paper is an attempt to find a condition for efficient multistage photoionization with two initial states excited by laser radiation to a common intermediate level.

Analytic assessments

Modern laser technology has in its arsenal a well-developed apparatus for measuring and stabilizing laser frequencies, so the theoretical consideration of the case of exact resonance is of great practical importance. In addition, in the case of exact resonance, the theoretical expressions are significantly simplified and it becomes possible to obtain simple analytical results, which llow to understand the common patterns. In the case of three-stage photoionization, two options of using the third stage are possible (Fig. 1). The first option is realized during photoionization into a continuum (Fig. $1,a$), the second option corresponds to the tuning of the third-stage laser radiation wavelength to resonance with the autoionization state (Fig. $1,b$). In both cases, we consider an idealized system of atom levels, which participate in photoionization by monochromatic (single-frequency) laser radiation. It is assumed, that the levels are not degenerate, the laser radiation frequencies ω_0, ω_1, ω_2 coincide with the Bohr transition frequencies. Laser pulses have a rectangular shape and their duration is much less, than the lifetime of excited levels —2¿ and —3¿. We assume, that in the first case the transitions —0¿→—2¿, —1¿→—2¿ and —2¿→—3¿ are coherent, and the transition from level —3¿ to the continuous spectrum (ionization) — is incoherent and is not considered. In the second case all transitions are coherent.

In a more general form, the N-level coherent excitation dynamics was studied in papers [4]. The difference is that in our case the excitation to the first excited level occurs from two lower levels. The presence of the second lower state —1¿ is based on the need to populate the preionization level to the maximum and obtain, accordingly, the maximum photocurrent. The level —1¿ lies close to the level —0¿ and has a comparable population at the initial moment of time. We assume, however, that the levels —0¿ and —1¿ are sufficiently separated in energy, so that there are no forced transitions from level —2¿ to level —1¿ under the radiation with a frequency of ω_0 and vice versa, there are no transitions from level —2¿ to level —0¿ induced by radiation with frequency ω_1. In contrast to the incoherent case with monotonic behavior of populations, coherent dynamics in the absence of strong relaxation is characterized by level population oscillations, and therefore the problem is to determine such ratios of radiation intensities with frequencies ω_i, which provide the maximum population of levels —3¿ or —4¿, averaged by pulse duration. For a classical three-level system, when there is one level below [4,7], this relation consists in the equality of the Rabi frequencies at the transitions —0¿→—2¿ and —2¿→—3¿. However, we did not find any papers for the case of two excited lower levels, where the solution would be obtained analytically.

The interaction of an atom with a laser field will be considered as a perturbation, and the total wave function of the atom $\Psi(t)$, which depends on time, is sought as a superposition of the wave eigenfunctions of the stationary states of the unperturbed atom. In the case of two-stage excitation, these are the wave functions Ψ_0, Ψ_1, Ψ_2, Ψ_3, so that

$$\Psi(t) = a_0(t)\Psi_0 + a_1(t)\Psi_1 + a_2(t)\Psi_2 + a_3(t)\Psi_3. \quad (1)$$

For three-stage excitation, we use five unperturbed wave functions (Ψ_0, Ψ_1, Ψ_2, Ψ_3, Ψ_4), and the total wave function is

$$\Psi(t) = a_0(t)\Psi_0 + a_1(t)\Psi_1 + a_2(t)\Psi_2 + a_3(t)\Psi_3 + a_4(t)\Psi_4. \quad (2)$$

The squares $|a_i|^2$ — are the probabilities of finding the atom in the stationary states —0¿, —1¿, —2¿, —3¿ and —4¿ at time t. At the initial moment of time the coefficients a_2, a_3 and a_4 are equal to zero. Assuming, that the electric field of laser radiation has the form of $E_j(t) = E_j(\exp(i\omega_j t) + \exp(-i\omega_j t))$ and the dipole approximation for the interaction with an atom is valid, the Schr?dinger equation for $\Psi(t)$ in the RWA (rotating wave approximation) reduces to a system of equations for two-stage photoexcitation [8]:

$$\frac{da_3}{dt} = i f_2 a_2 \quad (3)$$

$$\frac{da_2}{dt} = i f_2^* a_3 + i f_0 a_0 + i f_1 a_1 \quad (4)$$

$$\frac{da_1}{dt} = i f_1^* a_2, \quad (5)$$

$$\frac{da_0}{dt} = i f_0^* a_2. \quad (6)$$

In the left side of the equations are the time derivatives of the coefficients $a_i(t)$, and in the right side the multipliers f_i are Rabi resonance frequencies:

$$f_0 = \frac{d_{02}E_0}{\hbar}, \quad f_1 = \frac{d_{12}E_1}{\hbar}, \quad f_2 = \frac{d_{23}E_2}{\hbar}. \quad (7)$$

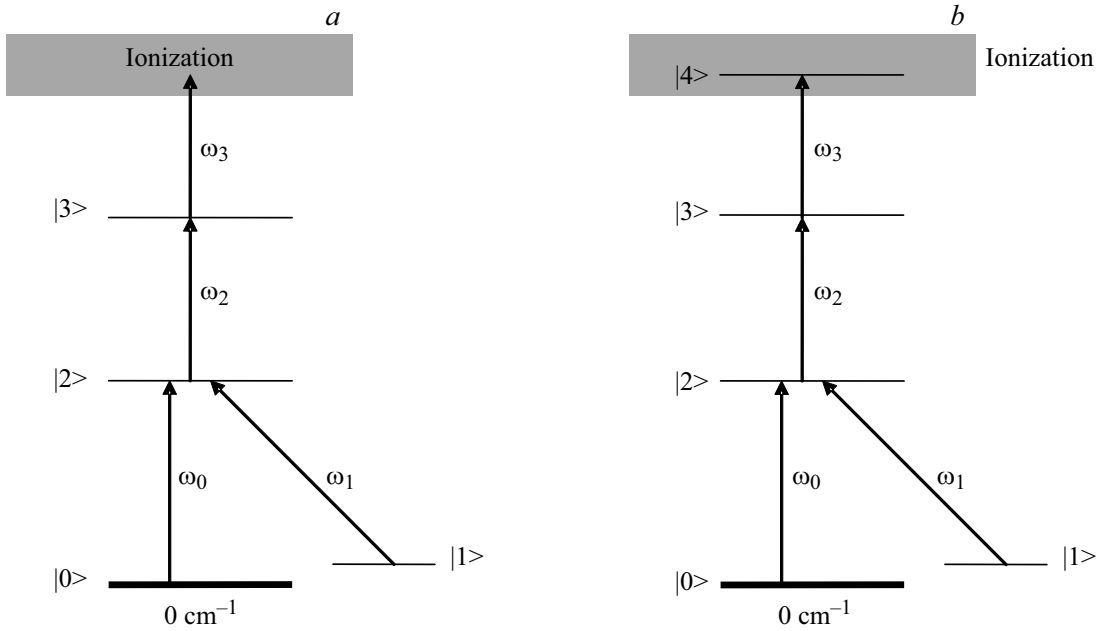

Figure 1. λ — photoionization schemes: a — two-stage scheme, b — three-stage scheme.

Here d_{ji} — the dipole matrix elements of the corresponding transitions.

If the initial states are not prepared coherently, then the phases for $a_0(0)$ and $a_1(0)$ can be chosen equal to zero, and then the coefficients a_i at the time $t = 0$ are real values and satisfy the following initial conditions:

$$a_3(t=0) = 0, \tag{8}$$

$$a_2(t=0) = 0. \tag{9}$$

$$a_1(t=0) = \sqrt{M}, \tag{10}$$

$$a_0(t=0) = \sqrt{G}. \tag{11}$$

Here G and M the initial populations of the states —0¿ and —1¿, respectively. The values G and M depend on the nature of the lower states — these can be the components of the hyperfine structure of the ground state or, for example, a combination of the ground state and a nearby metastable one. The task of the calculation is to determine the conditions, under which the population of the upper state ($W_3 = |a_3|^2$ for two-stage excitation and $W_4 = |a_4|^2$ for three-stage one) will be maximum. The solution of system (3)−(6) with initial conditions (8)−(11) for the preionization state $|a_3|^2$ can be written as

$$W_3 = |a_3|^2 = 4\left(\frac{f_0 f_2\sqrt{G} + f_1 f_2\sqrt{M}}{\Omega^2}\right)^2\left(\sin\frac{\Omega}{2}t\right)^4, \tag{12}$$

where

$$\Omega = \sqrt{|f_0|^2 + |f_1|^2 + |f_3|^2}. \tag{13}$$

Averaging the value of W_3 over oscillations with a frequency of Ω, we obtain the average value of the probability W_3

$$\bar{W}_3 = \frac{3}{2}\left(\frac{f_0 f_2\sqrt{G} + f_1 f_2\sqrt{M}}{f_0^2 + f_1^2 + f_2^2}\right)^2. \tag{14}$$

With equal Rabi frequencies $f_0 = f_1 = f_2$ and $G = M = 0.5$

$$\bar{W}_3 = \frac{1}{3}. \tag{15}$$

Considering $\bar{W}_3$ as a function of two variables f_0 and f_1 for constants f_2, G and M, it is easy to see, that the maximum of $\bar{W}_3$ is realized at

$$\begin{cases} f_0 = f_2\sqrt{G} \\ f_1 = f_2\sqrt{M}. \end{cases} \tag{16}$$

Substituting condition (16) into (14), we obtain ($G + M = 1$)

$$\bar{W}_3 = \frac{3}{8}. \tag{17}$$

Thus, in the case of two-stage photoexcitation using the λ-scheme at the first transition, the optimal distribution of intensities are the intensities proportional to the initial populations of the lower states.

For three-stage photoexcitation the following system of equations should be considered:

$$\frac{da_4}{dt} = i f_3 a_3, \tag{18}$$

$$\frac{da_3}{dt} = i f_3^* a_4 + i f_2 a_2, \tag{19}$$

$$\frac{da_2}{dt} = i f_2^* a_3 + i f_0 a_0 + i f_1 a_1, \tag{20}$$

$$\frac{da_1}{dt} = i f_1^* a_2, \tag{21}$$

$$\frac{da_0}{dt} = i f_0^* a_2, \tag{22}$$

and the solution for the top level becomes much more complicated

$$W_4 = |a_4|^2 = \left(\frac{f_2 f_3 (f_0\sqrt{G} + f_2\sqrt{W})}{\Omega_+^2 - \Omega_-^2}\right)^2 \times \left(\frac{\sin\Omega_+ t}{\Omega_+}\Omega_+ - \frac{\sin\Omega_- t}{\Omega_-}\right)^2. \quad (23)$$

Two characteristic frequencies Ω_+ and Ω_- appear:

$$\Omega_+ = \sqrt{\frac{(f_0^2+f_1^2+f_2^2+f_3^2)+\sqrt{(f_0^2+f_1^2+f_2^2+f_3^2)^2-4(f_3^2f_0^2+f_3^2f_1^2)}}{2}}, \quad (24)$$

$$\Omega_- = \sqrt{\frac{(f_0^2+f_1^2+f_2^2+f_3^2)-\sqrt{(f_0^2+f_1^2+f_2^2+f_3^2)^2-4(f_3^2f_0^2+f_3^2f_1^2)}}{2}}, \quad (25)$$

The average value of W_4 over oscillations with frequencies Ω_+ and Ω_- is equal to

$$\bar{W}_4 = \frac{f_2^2(f_0\sqrt{G} + f_1\sqrt{M})^2(f_0^2 + f_1^2 + f_2^2 + f_3^2)}{2[(f_0^2 + f_1^2 + f_2^2 - f_3^2)^2 + 4f_2^2f_3^2](f_0^2 + f_1^2)}. \quad (26)$$

If the Rabi frequencies are equal and $G = M = 0.5$, from (26) follows $\bar{W}_4 = 0.25$. If we extend the optimal condition (16) to the case of three-stage photoexcitation:

$$\frac{f_0}{\sqrt{G}} = \frac{f_1}{\sqrt{M}} = f_2 = f_3, \quad G + M = 1, \quad (27)$$

then the averaged value $\bar{W}_4 = 0.3$, which is 20% higher, than in the case of equal Rabi frequencies. Note, that the expressions for the average values $\bar{W}_3$ of the system (3)–(6) and $\bar{W}_4$ of the system (18)–(22) coincide with similar expressions of the average values from the paper [4] if we set $f_1 = 0$, $M = 0$, $G = 1$ in our formulas (14) and (26) respectively.

Fig. 2 shows the dependence of $\bar{W}_4$ on f_1 under the condition $f_0/\sqrt{G} = f_3 = 100$ MHz, $G = 0.6$, $M = 0.4$ for different values of f_2. As can be seen from the figure, a maximum is observed under condition (27), and as f_2 decreases, the maximum becomes sharper, while its amplitude changes insignificantly. This is apparently the result of the interference of oscillations of the level populations of the photoionization scheme under idealized conditions of the relaxation absence. As f_2 ($f_2 \ll f_3$) decreases, it is easy to see, that at the maximum under conditions

$$\frac{f_0}{\sqrt{G}} = \frac{f_1}{\sqrt{M}} = f_3, \quad G + M = 1 \quad (28)$$

the frequencies

$$\Omega_+ \approx f_3 + f_2/2, \quad (29)$$

$$\Omega_- \approx f_3 - f_2/2, \quad (30)$$

turn out to be very close to each other, and the sum of sines in (23) can be represented as the square of the cosine

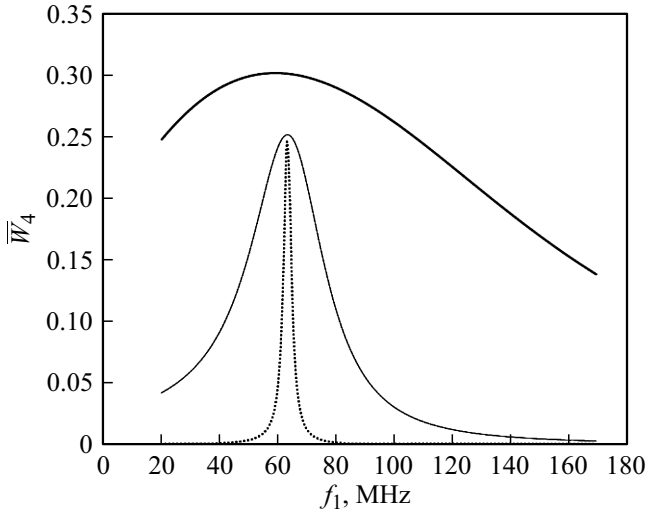

Figure 2. Dependence of the population $\bar{W}_4$ on the frequency f_1 for different values of f_2 and $f_0/\sqrt{G} = f_3 = 100$ MHz for $G = 0.6$, $M = 0.4$. $f_2 = 1$ (dotted line), 10 (thin line), 100 MHz (thick line).

f_3 modulated by the slower square of the sine $f_2/2$. The rise time of W_4 to its maximum value increases significantly and, accordingly, the requirement for the accuracy of compliance with conditions (28) increases, which leads to peak sharpening. For $f_2 \ll f_3$ the average population of $\bar{W}_4$ as a function of f_1 near the maximum can be represented with good accuracy in the form of a resonance-like curve:

$$\bar{W}_4 \approx \frac{f_2^2 f_3^2}{[(f_1^2 - Mf_3^2)^2 + 4f_2^2f_3^2]}, \quad (31)$$

where the maximum width is proportional to $f_2(f_2f_3)$. In the coordinates of laser radiation intensities (proportional to the square of the Rabi frequency), the maximum (31) has a Lorentzian shape with a width proportional to the root of the product of the second and third steps intensities.

Conclusion

The objective of this work was to obtain analytical dependences in the description of coherent excitation in a scheme of multistage laser photoionization. These results can be used to predict the behavior of the population of the system preionization state, to which it is required to transfer the maximum number of atoms from the system initial levels. In the standard formulation of the problem, effective excitation of the preionization state requires the equality of the Rabi frequencies at intermediate transitions, and this equality is realized due to different intensities to compensate for different dipole matrix elements of the transitions. In the problem, when there are two initial levels, this condition changes. To obtain the maximum ionization output, the laser radiation intensities in the first stages should be in a certain ratio with the intensities of the upper stages, and this ratio

depends on the initial population of the first levels. Such an additional correlation of intensities is a direct result of the coherent nature of the interaction of radiation with an atom. The presented results show, that in the case of coherent excitation, the process is very sensitive to the choice and maintenance of preset values of the laser radiation intensity at all stages of the photoionization circuit, and failure to comply with these conditions leads in some cases to a sharp, resonant-like drop in efficiency.

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Conflict of interest

The authors declare that they have no conflict of interest.

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