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LASER PHOTOIONIZATION SENSOR OF THE SEPARATING HEAVY ISOTOPES AND NUCLEAR ISOMERS: SELECTIVE IONIZATION BY DC ELECTRIC AND LASER FIELD (U, Tm)

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Abstract

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S. V. Ambrosov

It is proposed and studied a new optimal theoretical scheme for the laser photoionization sensor of the separating heavy isotopes and nuclear isomers, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further DC electric field ionization. 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 *Tm*, *U* isotopes (ions, nuclei) sensing.

Key words: laser photoionization sensor, isotopes, DC electric field ionization;

Аннотация

ЛАЗЕРНИЙ ФОТОІОНІЗАЦІЙНИЙ СЕНСОР ПОДІЛЕННЯ ВАЖКИХ ІЗОТОПІВ ТА ЯДЕРНИХ ІЗОМЕРІВ: СЕЛЕКТИВНА ІОНІЗАЦІЯ СТАЛИМ ЕЛЕКТРИЧНИМ І ЛАЗЕРНИМ ПОЛЕМ (U, Tm)

С. В. Амбросов

Запропоновано нову оптимальну схему селективного фотоіонізаційного сенсору поділення важких ізотопів й ядерних ізомерів, яка базується на лазерному збудженні атомів ізотопів у рідбергові стани та подальшій іонізації зовнішнім сталим електричним полем. Метод релятивістської теорії збурень, модель оптимальної лазерної дії та формалізм матриці густини використані для чисельного моделювання параметрів оптимальної схеми детектування важких ізотопів урана й туллія.

Ключові слова: лазерний фотоіонізаційний сенсор, ізотопи, іонізація сталим електричним полем

Анотація

ЛАЗЕРНЫЙ ФОТОИОНИЗАЦИОННЫЙ СЕНСОР РАЗДЕЛЕНИЯ ТЯЖЕЛЫХ ИЗОТОПОВ И ЯДЕРНЫХ ИЗОМЕРОВ: СЕЛЕКТИВНАЯ ИОНИЗАЦИЯ ПОСТОЯННЫМ ЭЛЕКТРИЧЕСКИМ И ЛАЗЕРНЫМ ПОЛЕМ (U, Tm)

С. В. Амбросов

Предложена новая оптимальная схема селективного фотоионизационного сенсора лазерного разделения тяжелых изотопов и ядерных изомеров, базирующаяся на лазерном возбуждении атомов изотопов в ридберговы состояния и дальнейшей ионизации внешним постоянным электрическим полем. Метод релятивистской теории возмущений, модель оптимального лазерного воздействия и формализм матрицы плотности использованы для численного моделирования параметров оптимальной схемы детектирования тяжелых изотопов урана и туллия.

Ключевые слова: лазерный фотоионизационный сенсор, изотопы, ионизация постоянным электрическим полем

1. Introduction

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 (c. f. [1-15]). The standard laser ionization sensor scheme may be realized with using a scheme of the multi-step excitement and ionization of atoms by laser pulse. The scheme of selective ionization of atoms, based on the selective resonance excitation of atoms by laser radiation into states near ionization boundary and further photo-ionization of the excited states by additional laser radiation, has been at first proposed and realized by Letokhov et al (c. f. ref. [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 (c. f. refs. [16-24]).

In a number of papers (c. f. [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) part moves into continuum and other levels become by the autoionization ones. The probability of their autoionization decay quickly increases with growth of the main quantum number. The most optimal situation is when atom is excited to state, which has the autoionization probability more than the radiation decay one. To receive an accurate information about optimal laser photoionization sensor scheme, it is necessary to carry the accurate calculation of the process of sequent atomic 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 [11,19,23,24,26], 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 opti-

mal schemes for the laser photoionization sensors of separating heavy isotopes and nuclear isomers. We propose and make modelling a new, optimized scheme for the laser photoionization sensor. It is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further DC electric field ionization mechanism. The matter is in a fact that the velocity of ionization is significantly higher than one in the usual regime (c. f. [1,5,11,22]). A result, the effectiveness of the laser photoionization sensor scheme is significantly increased. The numeral data for the autoionization resonance characteristics for U and 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 *Tm* isotopes (nuclei) sensing, we use the optimal laser action model and density matrices formalism [11-16,24-26].

2. Laser photoionization sensor 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: $5f^3 7s^2 np$, $5f^3 6d7sns$ (*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

$$\psi = \begin{pmatrix} \xi \\ \eta \end{pmatrix}, \tag{1}$$

the Dirac equation has the following form:

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

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

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

we obtain the second-order equation

$$\Delta\Phi + K2\Phi = 0,$$

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

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

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

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

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

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

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

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

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

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

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:

$$W(nlm)=\sum_{n2}(a_{n1n2}^{nlm})^2W(n_1n_2m)\,. \qquad (7)$$

Here $W(n_1n_2m)$ is the state decay probability; a are the coefficients of expansion of the $\psi(nlm)$ functions on the parabolic functions $\psi(n_1n_2m)$. 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 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. [7-9,23,24].

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 [1]; dashed line is corresponding to our relativistic results). It is stressed that the hydrogen-like approximation gives an inaccuracy 15-20% [1]. 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).

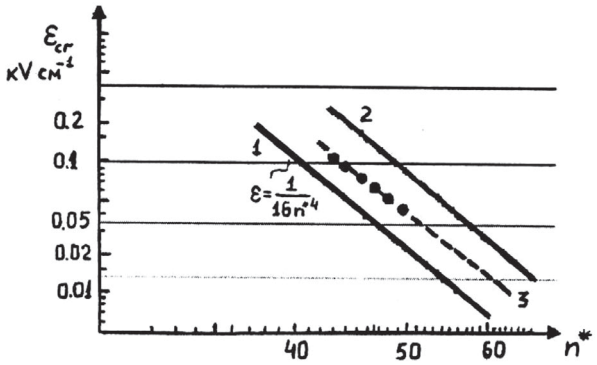

Fig. 1. Critical electric field strength in dependence upon effective quantum number n^* for the uranium atom

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 [4,5,17]:

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

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

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: $4f^{-1}j$ and $6s_{1/2}$. The ROD can be of the monopole or quadruple character. We mean here the multi-polarity of the inter-quasi-particle 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 (~10-15%) than it is discovered in paper [5].

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 e. (VЧcm^{-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	$4f^{13}{}_{7/2}\ 6s1/2\ (3)\ 25p_{1/2}$ [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

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) Å. For $5f^37s^2np\ (n=40\text{-}44)$ 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·cm^{-1}; $\Gamma(5f^37s^242p)$=1,675D-01; $\Gamma(5f^37s^242s)$=5,913D-04. So, we have here a strong change of the autoionization resonance width at a moderately weak electric field too.

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 exci-

tation 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 [10,26], 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 [7-9,23,24]. The laser photoionization scheme for *U* isotopes included the following steps: i). Laser excitement of the ^{235}U isotopes from the ground $5f^3 6d7s^2\text{-}^5L_6^o$ state and low lying metastable $5f^3 6d7s^2\text{-}^5K_5^o$ 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 [13-15]. 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. 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 sensor 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].

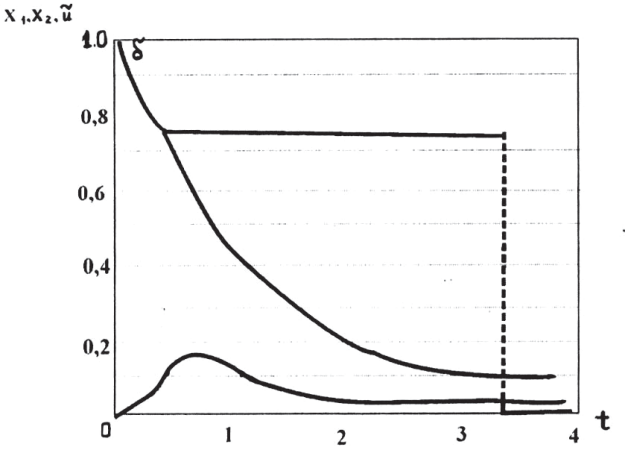

Figure 2. Results of numerical modelling for the optimal form of laser pulse in the laser photoionization sensor scheme for the uranium isotopes: δ pulse + dotted line is the optimal form of the laser pulse; curves (1) x_1 and (2) x_2 are the populations of the ground (6s) and excited (np) states.

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