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ADVANCED LASER PHOTOIONIZATION SEPARATION SCHEME AND TECHNOLOGY FOR HEAVY RADIOACTIVE ISOTOPES AND NUCLEAR ISOMERS

We present new optimal scheme of the separating highly radioactive isotopes and products of atomics energetics such as $^{210-214}\text{Fr}$, $^{133,135,137}\text{Cs}$ and others, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further autoionization or DC electric field pulse ionization. As result, requirements to energetic of the ionized pulse are decreased at several orders. And an effectiveness of a scheme increases. There are theoretically calculated values of the characteristics of heavy Rydberg atoms in an external electromagnetic field (DC Stark effect) In particular, data on the energy level, the energy widths of Stark resonances for Rydberg Cs, Fr ($n < 45$).

In series of our papers we considered the different schemes of a laser photoionization isotopes and nuclear isomers schemes. In this work, which goes on this studying, we present an advanced scheme of the separating highly radioactive isotopes and products of atomics energetics such as $^{210-214}\text{Fr}$ and $^{133,135,137}\text{Cs}$, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further autoionization or DC electric field pulse ionization. Following to [1,4], let us remind that related a search of the effective methods for isotopes and nuclear isomers separation and obtaining especially pure substances at atomic level is related to number of the very actual problem of modern nuclear technology, quantum and photoelectronics. The basis for its successful realization is, at first, carrying out the optimal multi stepped photo-ionization schemes for different elements and, at second, availability of enough effective UV and visible range lasers with high average power (Letokhov, 1977, 1979, 1983; etc) [9]. The standard laser photo-ionization scheme may be realized with using processes of the two-step excitation and ionization of atoms by laser pulse. The scheme of selective ionization of atoms, based on the selective

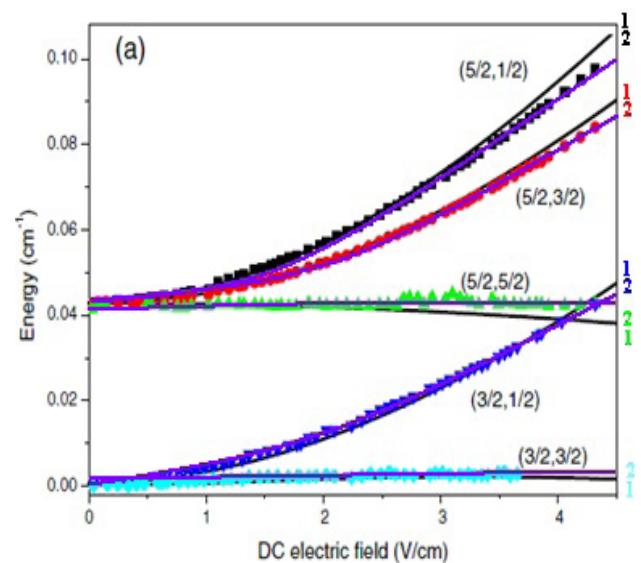
resonance excitation of atoms by laser radiation into states near ionization boundary and further photo-ionization of the excited states by additional laser radiation, has been at first proposed and realized by Letokhov et al (Letokhov, 1969, 1977) [1]. It represents a great interest for laser separation of isotopes and nuclear isomers. The known disadvantage of two-step laser photoionization scheme a great difference between cross-sections of resonant excitation s_{exc} and photo-ionization s_{ion} . It requires using very intensive laser radiation for excited atom ionization. The same is arisen in a task of sorting the excited atoms and atoms with excited nuclei in problem of creation of g-laser on quickly decayed nuclear isomers. Originally, Goldansky and Letokhov (1974) [17] have considered a possibility of creating a g-laser, based on a recoilless transition between lower nuclear levels and shown that a g-laser of this type in the 20-60 keV region is feasible. But, it is obvious that here there is a problem of significant disadvantage of the two-step selective ionization of atoms by laser radiation method. The situation is more simplified for autoionization and Stark resonance's in the atomic spectra, but detailed data about characteristics of these levels are often

absent [1-16]. Several new optimal schemes for the laser photo-ionization sensors of separating heavy isotopes and nuclear isomers are proposed [1,4]. It is based on the selective laser excitation of the isotope atoms into excited Rydberg states and further autoionization and DC electric field ionization mechanisms.

Let us remind that in a classic scheme the laser excitation of the isotopes and nuclear isomers separation is usually realized at several steps: atoms are resonantly excited by laser radiation and then it is realized photo ionization of excited atoms. In this case photo ionization process is characterized by relatively low cross section $s_{\text{ion}} = 10^{-17} - 10^{-18} \text{cm}^2$ and one could use the powerful laser radiation on the ionization step. This is not acceptable from the energetics point of view [1-8]. The alternative mechanism is a transition of atoms into Rydberg states and further ionization by electric field or electromagnetic pulse. As result, requirements to energetic of the ionized pulse are decreased at several orders. The main feature and innovation of the presented scheme is connected with using the DC electric field (laser pulse) autoionization on the last ionization step of the laser photoionization technology. There is a principal difference of the simple ionization by DC electric field. The laser pulse ionization through the auto ionized states decay channel has the advantages (more high accuracy, the better energetics, universality) especially for heavy elements and isotopes, where the DC electric field ionization from the low excited states has not to be high effective. This idea is a key one in the realization of sorting the definite excited atoms with necessary excited nuclei of the A^+ kind, obtained by optimal method of selective photo-ionization of the A kind atoms at the first steps. The suitable objects for modeling laser photoionization separation technology are the isotopes of alkali element Cs, long-lived transuranium elements etc.

We considered the isotopes of $^{210-214}\text{Fr}$ and $^{133,135,137}\text{Cs}$. For example, the resonant excitation of the Cs atoms can be realized with using dye lasers with lamp pumping (two transitions wavelengths are: $6^2S_{1/2} \text{ @ } 7^2P_{3/2}$ 4555A and $6^2S_{1/2} \text{ @ } 7^2P_{1/2}$ 4593A).

The next step is in the further excitation to the Rydberg S,P,D states with main quantum number $n=35-50$. The final step is the autoionization of the Rydberg excited atoms by a laser pulse or DC electric field pulse ionization and output of the created ions. The scheme will be optimal if an atom is excited by laser radiation to state, which has the decay probability due to the autoionization (pulse ionization) higher than the radiation decay probability. So, one could guess that the accurate data on the autoionization states energies and widths and the same parameters for the DC Stark resonances are needed. The consistent and accurate theoretical approach to calculation of these characteristics is based on the operator perturbation theory formalism [18] and corresponding advances relativistic version with model potential approximation [22, 23]. In Fig.1a we present the energy dependence (note that the level energy in the absence of field is taken as zero) of Stark components ($j, |m_j\rangle$) of the caesium state $39D$ Cs field strength: Experiment - squares, circles, triangles, diamonds [33]; Theory 1 - semi-empirical perturbation theory on the field by Zhao et al [25]; 2 - our data; In Fig.1b we list the Stark shift (in MHz) for the state $46D$ Cs in dependence on the square of the field: the experiment - squares, circles, triangles [25]; Theory - continuous.



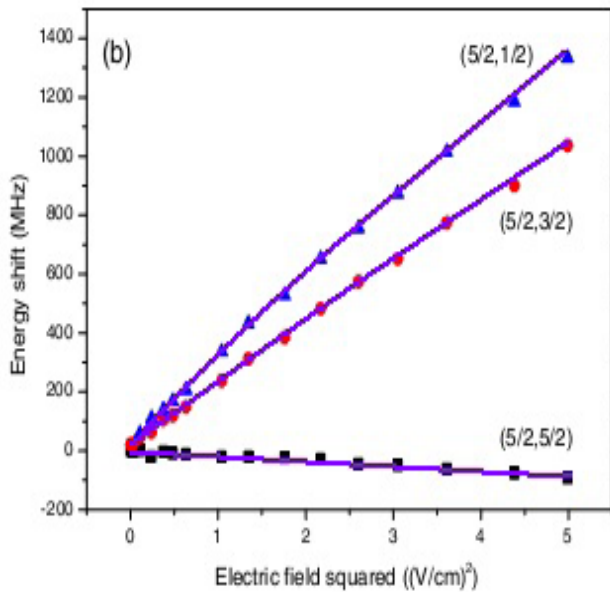


Fig.1 a – The energy dependence (note that the level energy in the absence of field is taken as zero) of Stark components ($j, |m_j\rangle$) of the caesium state 39D Cs field strength: Experiment - squares, circles, triangles, diamonds [25]; Theory 1 – semi-empirical perturbation theory on the field by Zhao et al [25]; 2 – our data; **b** – Stark shift (in MHz) for the state 46D Cs in dependence on the square of the field: the experiment - squares, circles, triangles [25]; Theory – continuous

In Fig.2 we present a dependence of the Stark component ($j, |m_j\rangle$) energies (MHz) of the state 44D for the francium atom of the square of the electric field (the first data).

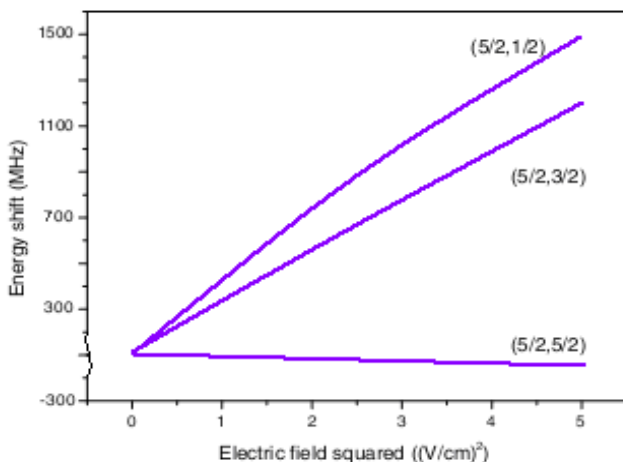


Fig.2. Dependence of the Stark component ($j, |m_j\rangle$) energies (MHz) of the state 44D for the francium atom of the square of the electric field (our data)

In figure 3 we present the numerical modeling results of the excited and ground states populations in the photoionization scheme of the $^{133,137}\text{Cs}$ isotopes separation process with auto- and electric field ionization by solving the corresponding differential equations system [4].

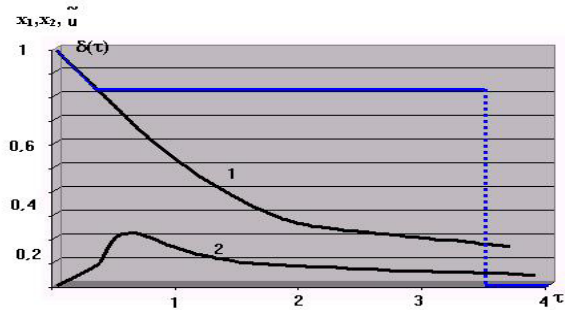


Fig.3. Results of modelling $^{133,137}\text{Cs}$ isotopes separation process by the laser photo-ionization method (d+dashed – laser pulse optimal form; see text)

The following definitions are used: d+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 Cs. The d-pulse provides maximum possible level of excitation (the excitation degree is about $\sim 0,25$; in experiment (Letokhov, 1983) with rectangular pulse this degree was $\sim 0,1$). It is worth to turn attention on some analogy between modeling results for different alkali isotopes. Indeed, the relative populations for indicated atoms in the highly excited states are very closed to each other, however the absolute values of the radiation parameters for different isotopes naturally differ. Let us remember data regarding the Cs excitation and the ionization cross sections: the excitation cross section at the first step of the scheme is $\sim 10^{-11}\text{cm}^2$; the ionization cross-section from excited $7^2P_{2/2}$ state: $s_2=10^{-16}\text{cm}^2$, from ground state $s_2=10^{-18}\text{cm}^2$ [2]. One can see that the relation of these cross sections is 10^5 and 10^7 correspondingly. This fact provides the obvious non-efficiency of standard photoionization scheme. Using d-pulse indeed provides a quick ionization, but the ionization yield will be less than 100% because of the stick-

ing on intermediate levels. So, from energetic point of view, this type of ionization can be very perspective alternative to earlier proposed classical two-step and more complicated photoionization schemes (Letokhov, 1983) [1]. The similar situation and analogous conclusions are obtained for the Sr and I isotope separation with the corresponding difference in the energetic and radiative characteristics data. So, one can say here about sufficiently optimal scheme of the separating highly radioactive isotopes and products of atomic energetics such as Cs and others. The key features of the corresponding scheme (technology) are based on the selective laser excitation of the isotopes atoms to the excited Rydberg states and further autoionization (or DC electric pulse ionization). One could remember here that a step of laser isotope separation has to be very important one in solving the modern actual problems of the transmutation of radioactive elements and decreasing the energy losses in the modern atomic energetics cycles [20,21].

References

1. Glushkov A.V., Lepikh Ya.I., Prepelitsa G.P., Ambrosov S.V., Bakunina E.V., Svinarenko A.A., Loboda A.V., Physics of the laser-photoionization atomic processes in the isotopes and gases separator devices: new optimal schemes// Sensors Electronics and Microsystems Technologies.-2011.-Vol.2(8),N1.-P.27-35.
2. Glushkov A.V., Khetselius O.Yu., Svinarenko A.A., Prepelitsa G.P., Energy Approach to Atoms in a Laser Field and Quantum Dynamics with Laser Pulses of Different Shape//In: Coherence and Ultrashort Pulsed Emission, Ed. Duarte F. J. (Intech, Vienna).-2010.-P.159-186.
3. Glushkov A.V., Lepikh Ya.I., Ambrosov S.V., Khetselius O.Yu., New optimal schemes of the laser photoionization technologies for cleaning the semiconductor materials and preparing the films of pure composition at atomic level// Ukrainian Journal of Physics.-2008.-Vol.53,N10.-P.1017-1022.
4. Glushkov A.V., Prepelitsa G.P., Svinarenko A.A., Pogosov A.Yu., Shevchuk V.G., Ignatenko A.V., Bakunina E.V., New laser photoionization isotope separation scheme with autoionization sorting of highly excited atoms for highly radioactive isotopes and products of atomic energetics//Sensors Electr. and Microsyst. Tech.-2011.-Vol.2(8),N2.-P.81-86
5. Glushkov A.V., Ambrosov S.V., Loboda A.V., Gurnitskaya E.P., Prepelitsa G.P., Consistent QED approach to calculation of electron-collision excitation cross-sections and strengths: Ne-like ions // Int. Journ. Quant. Chem.-2005.-Vol.104, N4 .-P. 562-569.
6. Glushkov A.V., Lepikh Ya.I., Fedchuk A.P., Ignatenko A.V., Khetselius O.Yu., Ambrosov S.V., Wannier-Mott excitons and atoms in a DC electric field: photoionization, Stark effect, resonances in the ionization continuum// Sensor Electr. and Microsyst. Techn. (Ukraine).-2008.-N4.-P.5-11.
7. Glushkov A.V., Ambrosov S.V., Kozlovskaya V.P., Prepelitsa G.P., Auger effect in atoms and solids: Calculation of characteristics of Auger decay in atoms, quasi-molecules and solids with application to surface composition analysis// Functional Materials.-2003.-V.10, N2.-P.206-210.
8. Khetselius O.Yu., Relativistic perturbation theory of the hyperfine structure for some heavy-element isotopes//Int. Journal of Quantum Chem.-2009.-Vol.109.-P. 3330-3335.
9. Letokhov V.S., Laser isotope separation// Nature.-1999.-N277.-P.605-610.
10. Velikhov E.P., Baranov V.S., Letokhov V.S., Rybov E.A., Starostin A.N., Pulsed CO₂-lasers and their application for isotope separation.-Moscow: Nauka, 1993.
11. Basov N.G., Letokhov V.S., Optical frequency standards//Sov. Physics Uspekhi-1999.-Vol.11.-P.855-880 (UFN.-

- 1998.-Vol.96,N12).
12. Bokhan P.A., Buchanov V.V., Fateev N.V., Kalugin M.M., Kazaryan M.A., Prokhorov A.M., Zakrevský D., *Laser Isotope Separation in Atomic Vapor*-WILEY-VCH Verlag, 2006.
 13. Duarte, F. J. , Tunable lasers for atomic vapor laser isotope separation: the Australian contribution. *Australian Physics*.-2010.-Vol.47,N2.-P.38-48.
 14. Glushkov A.V., *Atom in an electromagnetic field*.-Kiev: KNT, 2005.-450P.
 15. Baldwin G.G., Salem J.C., Goldansky V.I., Approaches to the development of gamma ray lasers // *Rev.Mod. Phys*.-1991.-Vol.53.-P.687-742
 16. Ivanov L.N., Letokhov V.S. Spectroscopy of autoionization resonances in heavy elements atoms// *Com.Mod. Phys.D.:At.Mol.Phys*.-1995.-Vol.4.-P.169-184.
 17. Goldansky V.I., Letokhov V.S. Effect of laser radiation on nuclear decay processes// *Sov. Phys. JETP*.-1994.-Vol.67.-P.513-516.
 18. Glushkov A.V., Ivanov L.N. DC Strong-Field Stark-Effect: consistent quantum-mechanical approach// *J.Phys. B: At. Mol. Opt. Phys*.-1993.-Vol.26,N16.-P.L379-386.
 19. Glushkov A.V., Ivanov L.N., Letokhov V.S., Nuclear quantum optics// *Preprint of Institute for Spectroscopy of the USSR AS, ISAN-N4, 1991*.-16P.
 20. Solatz R.W., May C.A., Carlson L.R. et al, Detection of Rydberg states in atomic uranium using time-resolved stepwise laser photoionization//*Phys. Rev.A*.-1996.-Vol.14.-P.1129-1148.
 21. Ivanov L.N., Ivanova E.P., Knight L. Energy Approach to consistent QED theory for calculation of electron-collision strengths// *Phys.Rev.A*.-1993.-Vol.48.-P.4365-4374.
 22. Ternovsky V., Svinarenko A., Ignatenko A., Nikola V., Seredenko S., Tkach T.B., Advanced relativistic model potential approach to calculation of radiation transition parameters in spectra of multicharged ions// *Journal of Physics: C Series (IOP, London, UK)*.-2014.-Vol.548.-P. 012047 (6p).
 23. Ternovsky V., Glushkov A.V., Zaichko P., Khetselius O., Florko T.A., New relativistic model potential approach to sensing radiative transitions probabilities in spectra of heavy Rydberg atomic systems/ // *Sensor Electr. and Microsyst. Techn*.-2015.-Vol.12, N4.-P.19-26.
 24. Zhi-Gang Feng, Lin-Jie Zhang, Jian-Ming Zhao, Chang-Yong Liand Suo-Tang Jia, Lifetime measurement of ultracold caesium Rydberg states/ // *J. Phys. B: At. Mol. Opt. Phys*.-2009.-Vol.42.-P.145303 (5P);
 25. J.Zhao, H.Zhang, Z. Feng, X.Zhu, L.Zhang, C.Li, S.Jia , Measurement of Polarizability of Cesium nD State in Magneto-Optical Trap/ //*J. Phys. Soc. Jpn.* 80 (2011) 034303 (7P).
 26. van Wijngaarden W.A., Xia J., Lifetimes and polarizabilities of low lying S,P, D states of francium/ *Journ. Quant. Spectr. Rad.Transfer*.-1999.-Vol.61, N4.-P.557-561.
 27. Huang S., Sun Q., Calculation of the Rydberg Energy Levels for Francium Atom// *Phys.Res.Internat*.-2010.-Vol.33.-P. 203497 (5P).

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Abstract

We present new optimal scheme of the separating highly radioactive isotopes and products of atomics energetics such as $^{210-214}\text{Fr}$, $^{133,135,137}\text{Cs}$ and others, which is based on the selective laser excitation of the isotopes atoms into excited Rydberg states and further autoionization or DC electric field pulse ionization. As result, requirements to energetic of the ionized pulse are decreased at several orders. And an effectiveness of a scheme increases. There are theoretically calculated values of the characteristics of heavy Rydberg atoms in an external electromagnetic field (DC Stark effect) In particular, data on the energy level, the energy widths of Stark resonances for Rydberg Cs, Fr ($n < 50$).

Key words: laser photoionization method, highly radioactive isotopes, new scheme

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УЛУЧШЕННАЯ ЛАЗЕРНО-ФОТОИОНИЗАЦИОННЫЕ СХЕМА РАЗДЕЛЕНИЯ ИЗОТОПОВ ДЛЯ ТЯЖЕЛЫХ РАДИОАКТИВНЫХ ИЗОТОПОВ И ЯДЕРНЫХ ИЗОМЕРОВ

Резюме

Представлена новая оптимальная схема лазерного разделения высоко радиоактивных изотопов, продуктов атомной энергетики таких как $^{210-214}\text{Fr}$, $^{133,135,137}\text{Cs}$ и других, базирующаяся на лазерном возбуждении атомов изотопов в ридберговские состояния и дальнейшей автоионизации или ионизации импульсом электрического поля. Теоретически вычислены значения характеристик тяжелых ридберговских атомов во внешнем электромагнитном поле (DC эффект Штарка), в частности, данные по энергиям уровней, энергиям, ширинам штарковских резонансов для ридберговских Cs, Fr ($n < 50$).

Ключевые слова: лазерный фотоионизационный метод, высоко радиоактивные изотопы, новая схема

ПОКРАЩЕНА ЛАЗЕРНО-ФОТОІОНІЗАЦІЙНА СХЕМА ПОДІЛЕННЯ ІЗОТОПІВ ДЛЯ ВАЖКИХ РАДІОАКТИВНИХ ІЗОТОПІВ ТА ЯДЕРНИХ ІЗОМЕРІВ

Резюме

Представлена нова оптимальна схема лазерного поділення високо радіоактивних ізотопів, продуктів атомної енергетики, зокрема, таких як $^{210-214}\text{Fr}$, $^{133,135,137}\text{Cs}$ та інші, яка базуються на лазерному збудженні атомів ізотопів у ридбергові стани та подальшій автоіонізації або іонізації імпульсом електричного поля. Теоретично обчислені значення характеристик важких ридбергових атомів у зовнішньому електромагнітному полі (ДС ефект Штарка), зокрема, дані по енергіях рівнів, енергіям, ширинам штарківських резонансів для ридбергових Cs, Fr ($n < 40$).

Ключові слова: лазерний фотоіонізаційний метод, високо радіоактивні ізотопи, нова схема