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B.A. Knyazev

FEASIBILITY OF NON-LIOUVILLEAN ION INJECTION VIA RESONANTLY ENHANCED PHOTOIONIZATION

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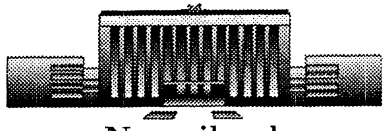

Novosibirsk
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О возможности нелиувилевской инжекции ионов с помощью резонансно-усиленной фотоионизации

Budker INP 2003-56

Ответственный за выпуск А.М. Кудрявцев
Работа поступила 5.09.2003 г.

Сдано в набор 10.09.2003 г.
Подписано в печать 11.09.2003 г.
Формат бумаги 60×90 1/16 Объем 1.5 печ.л., 1.2 уч.-изд.л.
Тираж 120 экз. Бесплатно. Заказ № 56

Обработано на IBM PC и отпечатано на
ротапринте ИЯФ им. Г.И. Будкера СО РАН
Новосибирск, 630090, пр. академика Лаврентьева, 11.

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Abstract

Charge-exchange method is now one of the main techniques for ion injection into accelerators and storage rings. Disadvantages of conventional methods, based on the atom or ion stripping at the material target, are growth emittance and energy struggling. In this paper a variant of stripping technique based on the resonantly enhanced two-photon ionization is considered. The technique allows ionization of singly charged ions of the elements from helium to bismuth. The technique can be also employed for proton injection. Numerical examples are given for several elements.

О возможности нелиувилевской инжекции ионов с помощью резонансно-усиленной фотоионизации

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Аннотация

Метод перезарядки является сейчас одним из основных методов инжекции ионов в ускорители и накопительные кольца. Недостатком обычных методов, основанных на обдирке атомов или ионов в материальной мишени является рост эмиттенса и разброса по энергиям. В данной работе рассмотрен вариант обдирочного метода, основанный на резонансно-усиленной двухфотонной ионизации. Этот метод позволяет ионизовать однократно заряженные ионы элементов от гелия до висмута. Метод можно использовать и для инжекции протонов. Приведены численные примеры для нескольких элементов.

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Introduction

Charge-exchange, or stripping, method of ion injection into accelerators and storage rings has essential advantages compared to the other conventional methods because this technique has no restrictions imposed on the storage ions on orbit by Liouville's theorem. A. J. Dempster in 1932 [1] had obtained protons with double energy of 45 keV in a tandem "ground–22.5 keV–ground" accelerator using charge-exchange of protons in a hydrogen target. Double stripping of H^- ions in a thin foil was first proposed by L. Alvarez [2] in 1951 for the tandem proton accelerators. He suggested the reconstruction of existing Van de Graaf accelerators to a tandem configuration, placing a 180°-magnet inside a high-voltage conductor. In the same paper he suggested also the application of the charge-exchange method for tracing of orbits in the proton synchrotrons. However, he did not expected application of this technique as a basic injection method because of a lack at that time of intense negative hydrogen ion sources.

Impressive development of the negative ion sources (see [3]) allowed in few years obtaining high negative hydrogen ion currents. First experiments on the charge-exchange injection into a small storage ring using a gas target with a beam of accelerated H^- ions have been carried out at the Institute of Nuclear Physics (Novosibirsk) in early 1960s [4, 5]. A detailed analysis of the charge-exchange injection for the proton accelerators was performed by G. I. Dimov [6]. Later development of this technique is described in the review [7]. Nowadays the charge-exchange injection is a preferred injection method for proton machines. As an example one can mention light ion storage ring COSY [8].

Since a hydrogen atom has only one electron, the charge exchange injection of protons can be realized practically only by means of formation and acceleration of the negative hydrogen ions. The use of the molecular ions, such as H_2^+ or H_3^+ as primary particles is possible, but this technique has some disadvantages and is not applied in practice [7]. For heavier elements there are more variants for the charge-exchange injection, and it is now em-

ployed as injection technique for many heavy ion accelerators and storage rings. Suffice it to mention Nuclotron (Dubna) [9], SIS and ESR (Darmshtadt) [10], and TSR (Heidelberg) [11].

Charge-exchange injection is a subject of special interest for the projects of heavy ion driven inertial confinement fusion (HIF) [12, 14, 15]. There are several schemes for the HIF In the project ITEP-TWAC [12, 13] mulitiple-charged ions are considered to be accelerated and directed to a D-T pellet. In American and European projects [16, 17, 18] to reduce space charge effects it is considered the acceleration of low-charged ions. In both cases, however, to provide a necessary power density on the D-T pellet the ion beams have to be additionally compressed before aiming at the pellet. This assumes multi-turn beam injection and bunch stacking, which can be performed by the charge-exchange technique.

Advantages and disadvantages of the charge exchange injection of the heavy ions have been recently analyzed in detail by Dinev [19]. There are several problems in the standard charge exchange scheme when one uses a material target on the orbit. First, the ions loose energy in the stripping foil due to atomic excitation and ionization that increases the energy straggling of the beam. Second, the Coulomb elastic scattering causes a change in the ion trajectory slope and leads to transverse emittance growth. In a case of the multi-turn injection the four-dimensional transverse phase volume increases proportionally to the number of injection turns. The other important disadvantage, which does not exist in a case of proton machines, is a charge distribution function $F(q)$ for the ions passing through the target. This leads to the successive cut by the accelerator of all ions in charge states different from the equilibrium state charge and substantial losses of the accelerated particles.

A non-Liouvillean injection technique based on ion stripping via photoionization by hard vacuum ultaviolet (VUV) radiation has been recently proposed [20]. This technique promises to overcome the above mentioned disadvantages of the standard charge-exchange schemes but requires development of an intense VUV source. In this paper I suggest an injection method based on resonantly-enhanced two-photon photoionization (RETPI). For many ions this injection technique can be implemented with commercially available excimer lasers. In the following sections of the paper we briefly review the existing projects of the photoionization stripping technique, describe principles of the novel technique and analyze its potential capabilities.

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Table 1. Kinetic energy of ions T, for which the resonance transitions coincide with Doppler shifted laser wavelength, and laser energy density providing the ion photoionization length $L_{ph\,0}$ to be equal to one meter.

		KrF laser		ArF laser	
Ion	Transition	T, Gev	F_{L0}, J/cm^2	T, Gev	F_{L0}, J/cm^2
Sn II	5p–5d			6	1.4
Sr II	5s–6p	3	1.3	0.3	0.54
Ca II	4s–5p	3.5	2.4	0.5	1.2
Mg II	3s–4p	6	7 4	2.5	6.7
He II	1s–2p	11	53	9	67
H I	1s–2p	0.3	8.9	0.1	7.5

to the transitions given in the second column of the Table. Practically for all ions we have $\sigma_{ph} \sim 10^{-18} - 10^{-19}$ cm^2, $G \sim 1 - 2$, and $\beta \sim (0.1 - 1)$. Necessary photon number per cm^2 lies within the limits

$$N_{ph} \sim 10^{18} - 5\ \ 10^{19}$$

Required laser energy density is about 1 J/cm^2 for heavy ions (quite reasonable energy, easy achieved for such lasers) and abruptly grows up to 50 J/cm^2 for the lightest ions.

Thus. an excimer laser with rather moderate parameters can ensure practically complete photoionization of the ion bunch in a straight channel of several meter length during 25-nanosecond laser pulse. The estimation above is correct if total ionization energy is much less than the laser pulse energy. The ionization of 10^{14} ions, for example, requires as little as 200 μJ. This means that excimer laser beam can be, if to use adequate reflectors and optical delays, multiply employed for ionization of many subsequent ion bunches.

Acknowledgments

Author is indebted to G.I. Dimov, I.B. Khriplovich, I.N. Meshkov, A.I. Milstein, K.A. Nasyrov, and A.M. Shalagin for helpful comments and discussions. This work is based, in part, on experimental results obtained under grant from the program "Russian Universities. Fundamental Research" on the CATRION setup (reg. No. 06-06, NSU) supported by the Ministry of Industry, Science and Technology of Russian Federation.

Ion stripping via direct VUV photoionization

Application of intense radiation for ion beam injection had been first proposed by Arnold et al. [21] in 1977 They considered storing the atomic ions I^+ by direct or stepwise photodissociating the molecular ions HI^+, having been previously accelerated, at injection into the storage ring. In the second case photodissociation of the molecular ion has to be carried out through an excited molecular state by xenon flashlamp or ruby laser radiation. The excitation energy of the molecular band is equal approximately to 3.6 eV, and to match the ruby laser photon energy(1.78 eV) to this value the laser beam has to collide with the molecular beam being accelerated to $\beta = 0.6$. Although today there exists a very wide variety of laser sources and the Arnold's scheme can be improved substantially, nevertheless, as it was mentioned above, the injection scheme with primary molecular ions does not seem to be best for practical implementation.

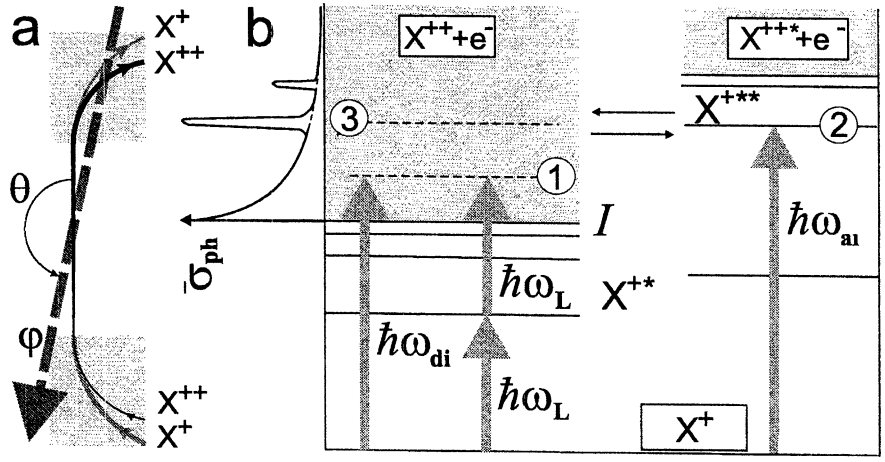

Fig. 1. Ion stripping via photoionization techniques: *a* – schematics of photoionization; *b* - Grotrian diagrams for direct photoionization (photon energy $\hbar\omega_{di}$), photoionization through an autoionization state ($\hbar\omega_{ai}$), and resonantly enhanced photoionization ($\hbar\omega_L$); *1* and *3* – states in continuum, *2* – autoionization state; σ_{ph} – typical cross-section of photoionization.

Direct photoionization of singly-charged heavy ions

$$X^+ + \hbar\omega_{di} \rightarrow X^{++} + e^-$$

had been proposed by C. Rubbia [20] for injection of ions from linac into a storage ring. I. Hofmann [16, 22] suggested application of this technique for the final stage of pulse compression in the heavy-ion fusion reactor systems. The both schemes suppose employing a VUV laser for direct photoionization of the ions and the capture of the double-charged ions produced on the equilibrium orbit. Fig. 1 (a). One of the great advantages of this method in comparison with the foil stripping is its absolute selectivity. Since the next ionization potential is more than the photon energy, the radiation interacts only with the primary ions and the ions produced are not further disturbed being circulated on the same orbit.

The ionization potentials of singly-charged ions lie between 10.0 eV (Ba^+) and 75.6 eV (Li^+). The cross sections for direct photoionization near a threshold are as low as 10^{-17}–10^{-18} cm^2 and intense flux of hard radiation is required for efficient ion stripping. To decrease required light intensity I. Hofmann considered ion stripping via the excitation of an ion autoionization state (see Fig. 1, *b*) that can be symbolically written for an ion X^+ as

$$\mathrm{X}^+ + \hbar\omega_{\mathrm{a1}} \rightleftharpoons \mathrm{X}^{+**} \rightarrow \mathrm{X}^{++} + e^- \,,$$

where X^{+**} denotes the autoionization state. An autoionization cross-section can reach a value of the order of 10^{-15} cm^2 within narrow (~ 0.1 eV) peaks [23], which lies, however, at much higher photon energies than the direct ionization threshold. For example, for Ba^+ three autoionization peaks with $\sigma = (2.4\text{–}2.8)\ \ 10^{-15}$ cm^2 lie at 21.2, 26.9, and 32.2 eV According to [16] similar cross-section can be expected for Bi^+ at 25 eV

Thus stripping the ions during time of flight of a bunch in a straight interaction region with a length of 1–10 m requires using a hard VUV light beam with low emittance, a short pulse length and high intensity. Since in this spectral range there are no conventional laser sources with required parameters, authors of the papers [16, 20, 22] have proposed using for stripping a free electron laser (FEL).

There are two main problems in the implementation of this technique. First, FELs with necessary characteristics do not still exist though some of the existing FELs already generate radiation in VUV [24] and can, in principle, reach required parameters. Second, focusing and reflecting optical elements are obviously necessary for guiding of the light beams especially in the multipass optical systems. The last may be required to ensure multiple interaction of the light beam with the ion beam or beams. Unfortunately, there is a lack of suitable mirrors for the hard VUV radiation. Thus development of an alternative low-cost photoionization technique using soft VUV radiation is very desirable.

be obtained at the atom energy of about 300 MeV and about 100 MeV for ArF laser (see. Fig. 2). Photoionization cross section for the excited atom is $\sigma_{\mathrm{ph\,H^*}} \approx 0.6\ \ 10^{-18}$ cm^2 These values look to be acceptable for proton injection (see Appendix). More detail analysis of RETPI for hydrogen is out the scope of this paper and will be done elsewhere.

Concluding remarks

There are many uses to which the resonantly enhanced photoionization can be put in the accelerator technique. First of all, it can be used for injection of accelerated singly-charged ions into storage rings by stripping them to the next stage of ionization. One of the promising application of RETPI may be stacking ion beams in some schemes of the heavy ion inertial fusion and neutron sources. The technique can be realized even with commercially available excimer lasers. It can be also used as a diagnostic method for study such characteristics as emittance, energy straggling, bunch length and density of ion beams in the accelerators and the storage rings.

One more possible application of RETPI is production of ions at local point within an accelerating structure. A gas jet coming across the structure is irradiated with one or two laser beams. The first beam populates an intermediate level and the next one photoionizes the excited atoms. Since the ionization potential for the atoms is lower than for the ions, it is possible to ionize the atoms resting in the laboratory frame. About twenty atoms (see Ref. [36]) have resonance transitions, which resonance lines are covered by generation bands of the excimer lasers. In this case there is no necessity to use the second laser because $\lambda \leq 4\pi\hbar c/I$ For several elements (iron, tin and tantalum) the resonance transitions for both the atom and the singly-charged ion coincide at the rest frame with the wavelength of KrF laser [36]. This coincidence allows obtaining a beam of singly-charged ions about half of which is excited to a definite term. This feature can be, probably, also used, for example, for diagnostics of the accelerating structure recording the fluorescence in a course of acceleration.

In conclusion, I present in the Table the laser energy density

$$F = N_{\mathrm{ph}} \ \ \hbar\omega_{\mathrm{L0}} \,,$$

which is required to obtain the rest-frame ionization length $L_{\mathrm{ph\,0}}$, see Eqs. (20) and (21), equal to one meter, for several singly charged ions and the hydrogen atom. Laser pulse length is 25 ns. Atomic parameters correspond

Proton generation

Although this paper is devoted to stripping the ions having at least two electrons, application of RETPI method for production of protons is a subject of special interest because of wide applicability of proton machines. Electron photodetachment from H^- and subsequent ionization of the resulting atom by laser excitation to Rydberg states and high electric field ionization was suggested in paper Zelensky et. al. [44] which devoted to production of polarizing relativistic proton beam. Yamane [45] suggested neutralization of H^- ions using Lorentz stripping in a magnetic field gradient. The resulting hydrogen atoms are excited by laser radiation to 3p state and then loose the last electron by stripping in a magnetic field gradient. This method is rather promising, taking in account that the Lorentz stripping of H^- is already realized in the Los Alamos proton storage ring, but emittance of the proton beam grows because of finite stripping length in the strong magnetic field.

Direct two-photon resonantly enhanced ionization of hydrogen atoms has, probably, formally less cross section, than the effective cross section in the Yamane method, but in our case there is no sources for growing the beam emittance and all laser photons participate in ionization, whereas in the bound-bound transitions only the "resonance" photons can participate. Thus, a scenario for RETPI proton generation is as follows. Negative hydrogen ions are produced by any conventional method and accelerated to required for RETPI energy. Then they are transformed into neutral atoms either by Lorentz stripping or by photodetachment:

$$H^- + \hbar\omega_1 \to H + e^-$$

Since the cross section of the detachment as a function of wavelength [43, P 473] has a wide maximum from 400 to 1200 nm (FWHM) with $\sigma_{\rm det}^{\rm max} \approx 3.5\ \ 10^{-17}$ cm^2, the radiation may be produced alternatively by flashlamps or a laser.

The neutrals produced are stripped by excimer laser radiation by means of the resonantly-enhanced two-photon ionization:

$$H + 2\hbar\omega_2 \to H^* + \hbar\omega_2 \to H^+ + e^-$$

Term system of hydrogen is very simple and 1s–2p transition ($\lambda = 121.57$ nm) can be used for the intermediate excitation. Excitation of the upper levels is not reasonable because they have spontaneous transitions to the metastable ns levels[4] Resonance saturation of the intermediate level for KrF laser can

[4]This loss mechanism obviously exists in the Yamane scheme.

Ion stripping via resonantly enhanced UV photoionization

One can suggest two variants of ion ionization by low energy photons. The first is a direct multiphoton ionization (see. *i.e.* [25]). Since a cross section of the multiphoton ionization is very low, extremely high laser intensity is required. Resonantly enhanced two-photon ionization (RETPI) [26], Fig. 1 (b),

$$X^+ + \hbar\omega_L \rightleftharpoons X^{+*},$$
$$X^{+*} + \hbar\omega_L \to X^{++} + e^-$$

allows to do it with substantially lower laser intensity[1] It retains the advantage of selectivity of the interaction only with singly-charged ions, but admits employment of more long-wave radiation in comparison with the case of the direct photoionization.

RETPI is a two-step process. On the first stage a singly charged ion is excited by intense resonance radiation to an intermediate level and then ionized by the next photon. Obviously, one or two lasers have to be employed to implement this technique. Selection of the laser depends on many factors: necessary wavelength, spectral bandwidth, total pulse energy, pulse length, beam divergence, and robustness. For distinctness, we assume further the excimer lasers as radiation sources. Commercially available ultraviolet excimer lasers have extremely high service life [28]. They generate $1 - 10$ J pulses at the repetition rate up to several kHz Characteristic laser pulse length is usually $10 - 40$ ns, but it can be extended to 100-300 ns.

The ionization potentials I of the singly charged ions lies between 75.6 eV (Li^+) and 10.0 eV (Ba^+). To be photoionized the intermediate level X^{+*} should lie not lower than at the energy $I/2$. Photon energies of widely used KrF and ArF lasers are equal to 4.99 and 6.41 eV, respectively. In a frame of the singly charged ion, laser radiation frequency ω_{L0} transforms to

$$\omega_L = \gamma(1 - \beta\cos\theta)\,\omega_{L0}\,, \qquad (1)$$

where θ is the angle in the laboratory frame (Fig. 1, *a*) between ion velocity and laser beam direction, and γ and β are well known relativistic variables.

We will assume below $\theta = \pi$ In this case

$$\omega_L = \sqrt{\frac{1+\beta}{1-\beta}}\ \omega_{L0} \qquad (2)$$

[1]This technique as an alternative method for ion stripping was first proposed on the HIF-2002 Symposium (see. [27]).

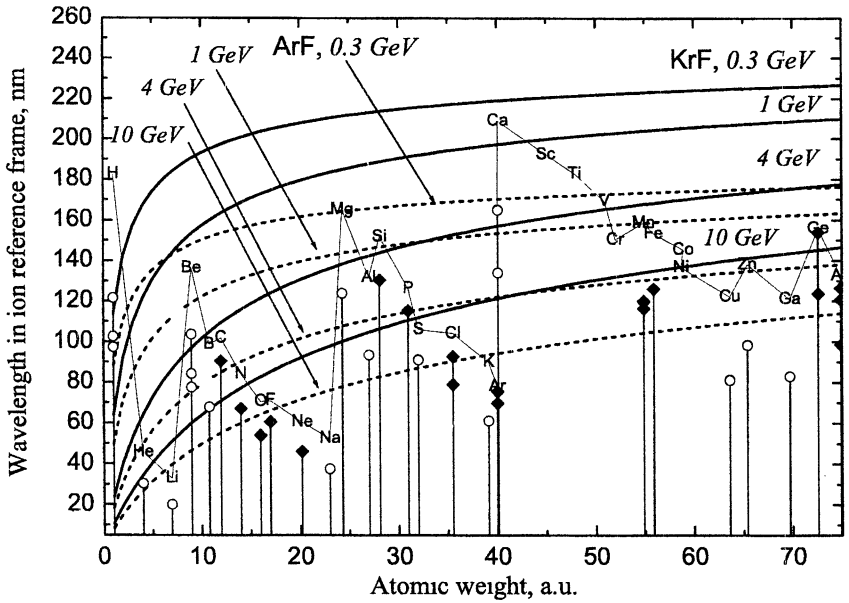

Fig. 2. Wavelengths of the excimer lasers in moving frame for the singly charged ions of the first 75 elements. The points correspond to the resonance transitions for the singly ionized ions of the elements. The open circles correspond to the transitions with a singlet ground state; the diamonds denote the transitions with a multiplet ground state (the points for hydrogen are given for a neutral atom). A thin broken line marks the wavelengths corresponding to energy $I/2$ for each ion.

Corresponding wavelengths of KrF and ArF lasers in the moving frame for the ions from He to Bi are given in Figs. 2–4 for four values of the ion energy· $T = 0.3, 1.0, 4.0$, and 10 GeV A thin broken line in the same figure is the wavelengths, corresponding to the energy $I/2 = \hbar\omega_{\rm L}$ for the singly charged ion of each element. If the ion of an element has one or several resonance transitions with the wavelengths

$$\lambda \leq \frac{4\pi\hbar c}{I} \qquad (3)$$

lying lower than this line, it can be, in principle, stripped to the next ionization state through one of these intermediate excited levels. Selected resonance transitions satisfying this requirement are also shown in the figures.

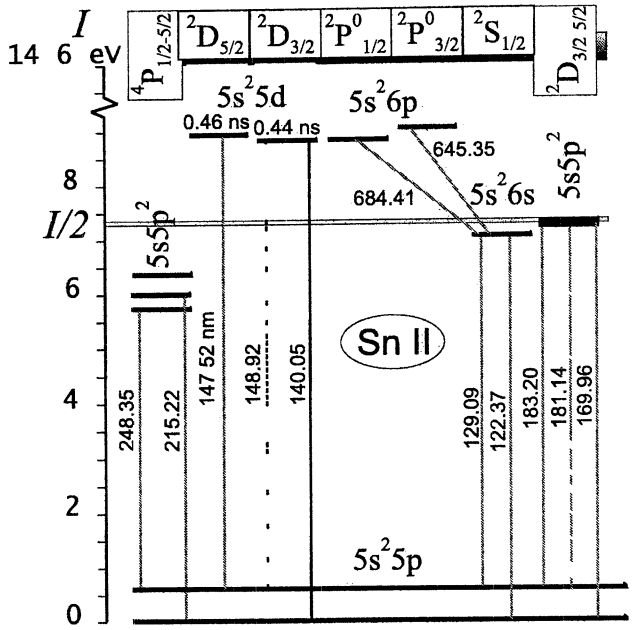

Fig. 6. Grotrian diagram for Sn II. Dark lines denote the transitions involving in the radiative processes for $\lambda_L = 140.1$ nm. Dashed lines – transitions to a long-lived sub-level that has to be "cleaned"

In specific case of Sn II ion it is better to select as the intermediate level $5s^25d\ ^2D^\circ_{3/2}$ state. It has no obvious allowed transitions to lower levels (transitions with simultaneous change of state for two electrons have low probability), besides a very fast transition to the upper level of the ground state (the last means that radiative cleaning of this level is absolutely necessary). The transition $5s^25p\ ^2P^\circ_{1/2} - 5s5p^2\ ^2D^\circ_{3/2}$ corresponds to $\lambda = 169.96$ nm. It is only 0.47 nm more than it requires for a state to be employed as the intermediate level. Although there is no data on the transition probability, by analogy with Tl II (see, e. g. [42, P 69]) it must be about twice less than for above mentioned 5p–6s transition. Using two inclined laser beams and increasing a little ion beam energy, it is easy to design a system employing this transition. These two examples show that for stripping the singly-charged ion of a specific element one has to develop an individual optical system with one or several laser beams.

Lifetimes for 5d and 5p levels are very low [38, 39] in comparison with the laser pulse length τ_L, and they cannot practically slow the ionization process down.

The most serious problem is existence of the metastable 4d levels with lifetimes $\sim$ 0.4 s [40], which are populated by 4d–5p (τ=110 ns) and 4d–6p (τ is unknown, but probably similar) transitions. Spontaneous radiation lifetimes for these transitions in practical situations must be rather long in comparison with the ionization time (19) in the ion frame, nevertheless this radiation branch may decrease efficiency of ionization. This shortcoming may be obviated by irradiation the ions on the wavelength exciting the metastable atoms up to an allowed transition. In our example it is $\lambda = 242.7$ nm. It may be a fraction of the same laser radiation propagating through the ion beam under the angle θ determined by eq. (1). The absence of data on Einstein coefficient for many transitions does not enable exact calculation of ionization time and ionization efficiency for many elements, but obviously, the ions with singlet ground state can be stripped to the next ionization state by means of RETPI technique.

Consider now the elements with a multiplet ground state. The wavelengths of resonance transitions from sub-levels of the multiplet ground state for such ions are shown in Figs. 2–4 with diamonds. For clarity sake we select for analysis (Fig. 6) a tin ion having relatively simple two-level ground state [41]. Usually the distance in the energy scale between the multiplet components is rather large and only one of the components can be excited by the laser radiation. In this case only a fraction of the ions can participate in the REPI. A value of this fraction depends on the ion pre-history (initial distribution over all the energy levels, spontaneous transition rates, time of ion travelling).

In general case, one can expect that the ions accelerated to the required velocity may be only in metastable or ground states. We already discussed how can the metastables can be cleaned off. The same method can be employed for cleaning sub-levels of the ground state. In this case required angle $\Delta\theta$ between two laser beams, obviously, is very small. Transitions between the sub-levels are forbidden in the electric dipole assumption, but slow magnetic dipole (*M1*) or electric dipole (*E2*) transitions are permissible. Probability of these transitions in comparison with *E1* transition for equal ΔE are 10^{-5} and 10^{-8}, respectively. Since $A_{km}(M1) \propto \lambda_{km}^3$ and $A_{km}(E2) \propto \lambda_{km}^5$, for small distances between sub-level components probability of the radiative decay of the upper sub-levels are very small. Degree of forbiddenness, however, decreases for heavy elements, and for some experimental configurations spontaneous cleaning may work.

The transitions and other spectroscopic data mentioning in this paper are obtained from Refs. [31, 32, 33, 34, 35][2] For example, strontium ion Sr^+, as it is seen from Fig. 3, has at least two transitions which lay lower the limit. It can be ionized with KrF laser at the ion energy 4 GeV and 10 GeV, and with ArF laser at 0.3 GeV and 3 GeV

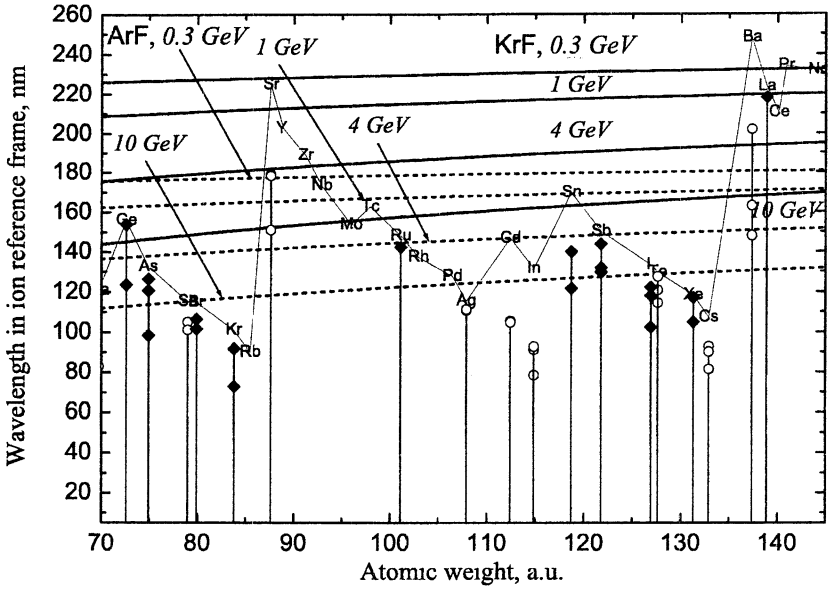

Fig. 3. Wavelengths of the excimer lasers in moving frame for the singly charged ions of the intermediate mass elements. The legend see in Fig. 2.

One can see from here that one of the restrictions of the technique is the necessity tuning ion beam energy to the value determined by required Doppler shift. This problem can be overcome in part by the use of the oblique laser beam as it follows from eq. (1). One more restriction is the absence for today of the robust hard-UV lasers that are necessary for ionization of the ions with a high ionization potential such as Ne^+, Na^+, Ar^+, Kr^+, Rb^+, Ag^+, Xe^+, Cs^+, Au^+, Hg^+, Tl^+, and Bi^+ (see Figs. 2–4). Future development of laser technology will close undoubtedly this gap. The other restrictions of the technique – characteristics of atomic term system – will be discussed further.

[2]For some elements (specially for the heavy ones) the databases do not give information on necessary UV-transitions, though such transitions with high probability exist for these elements.

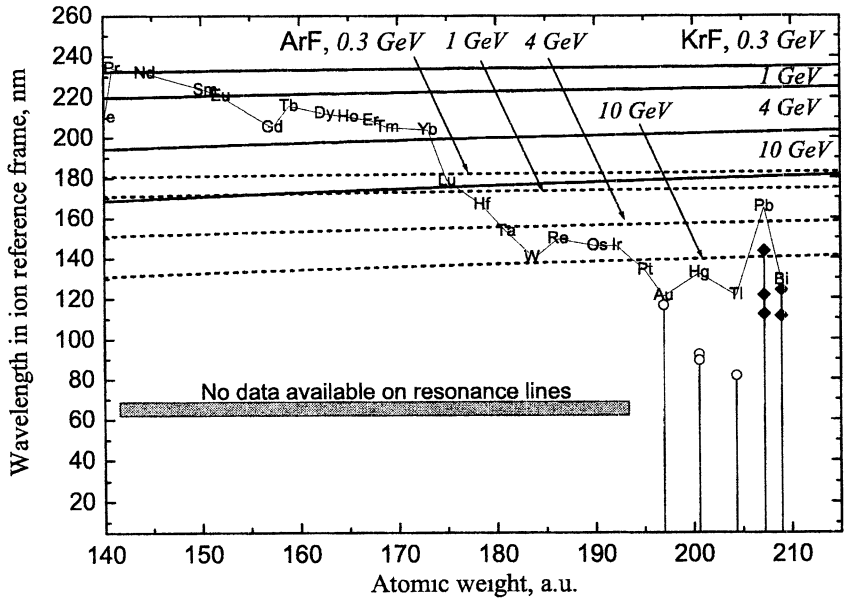

Fig. 4. Wavelengths of the excimer lasers in moving frame for the singly charged ions of the heavy elements. The legend see in Fig. 2.

Resonance saturation of intermediate level

Characteristic pulse length (*proper time* in terms of the relativistic theory) of an excimer-laser is about $\tau_{L0} = 25$ ns. In the ion reference frame it increases by γ times:

$$\tau_L = \gamma \tau_{L0} \tag{4}$$

The pulse length τ_L remains practically the same for heavy ions and converts to $\tau_L \approx 60$ ns for 10-GeV He^+ It means that each ion of the bunch, moving in the racetrack, is exposed by radiation during this time. Since the Einstein coefficient for allowed transitions (*E1*) is equal to

$$A_{km}[s^{-1}] = 6.67 \ \ 10^{13} \frac{g_k}{g_m} \frac{f_{km}}{(\lambda_{km}\,[\mathrm{nm}])^2}, \tag{5}$$

radiation lifetime of the intermediate level is the order of value 0.01 ns for $\lambda = 20$ nm and 0.5 ns for $\lambda = 180$ nm. We have assumed for the estimations

of the singly charged ions escape photoionization. Obviously, in this case Eq. (17) is not valid and occupation of the intermediate level is described by a system of kinetic equations. Analysis of the system in detail is beyond the scope of this paper and we content themselves below with simple estimations.

Initial ion distribution over the states in the ion source is usually the Boltzman one. The extracted beam may contain ions at different states: ground, short-living excited, and metastable. The short-living states decay within about 10 ns. Fraction of the metastable states strongly depends on kind of the element and for many elements can be rather little. The metastables may decay during ion storage or can be "cleaned off" by excitation to the upper levels (see below).

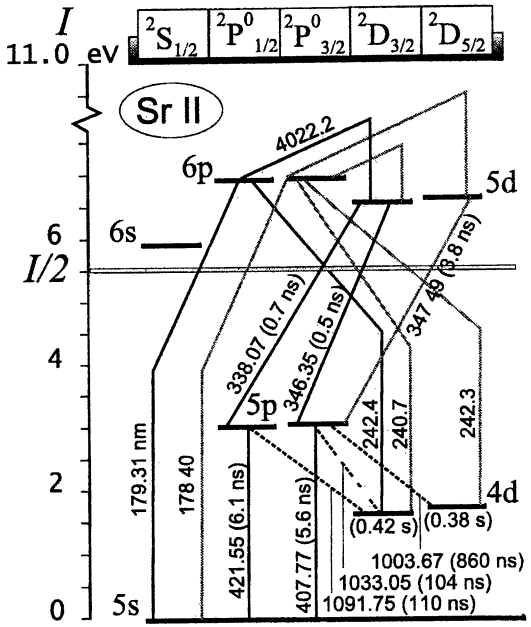

Fig. 5. Grotrian diagram for Sr II. Dark lines denote the transitions involving in the radiative processes for $\lambda_L = 179.3$ nm. Dashed lines – transitions to metastable states.

Fig. 5 shows Grotrian diagram for Sr^+ Lets all the ions are in the singlet ground state. Two 2s–2p transitions can be used for population of two possible intermediate levels. It is better to use $4p^6 6p\ ^2P^\circ_{1/2}$ intermediate level ($\lambda = 179.31$ nm), for which the cascade transition system is simpler.

can introduce the photoionization length in the laboratory system:

$$L_{\mathrm{ph}\,0} = \tau_{\mathrm{ph}\,0}\beta c = \frac{\tau_{\mathrm{L}0} G\beta c}{N_{\mathrm{ph}}\sigma_{\mathrm{ph}}} \tag{20}$$

After passing this distance 63% of the singly-charged ions are stripped to the second ionization state.

The only free parameter in the right part of the equation is the number of photons in the laser beam. Specifying acceptable photoionization length, we determine a number of photons that is necessary for photoionization:

$$N_{\mathrm{ph}} = \frac{\tau_{\mathrm{L}0} G\beta c}{L_{\mathrm{ph}\,0}\sigma_{\mathrm{ph}}} \tag{21}$$

The photoionization cross-section for an excited ion can be estimated with reasonable precision from the expression [30]

$$\sigma_{\mathrm{ph}}\,[\mathrm{cm}^2] = 7.9\ \ 10^{-18}\ \ \frac{n}{Z^2}\left(\frac{I-\hbar\omega_{\mathrm{L}}}{\hbar\omega_{\mathrm{L}}}\right)^3 , \tag{22}$$

where n is a principle quantum number of the intermediate state k and Z is a charge number of the ion core. We also have taken here in account that $\hbar\omega_{km} = \hbar\omega_{\mathrm{L}}$.

Selection of ions that can be photoionized with excimer lasers

Lets consider now the role of atomic structure details on the RETPI efficiency. In the previous sections we described a "two-level ion", which does not exist in the nature. A real atomic system has two features, which have to be considered.

First, the ground term of a real ion can be singlet or multiplet. All initial ions with singlet ground term can be excited by laser radiation. Resonance wavelengths for such ions are shown in Figs. 2–4 with open circles. The ions with multiplet ground term are distributed over several levels, further called for simplicity "sub-levels" In this case, only one of the sub-levels can be excited by laser radiation[3]

Second, for heavy ions there usually exist other levels laying below selected intermediate level. Population of these levels grow with time, and a fraction

[3]Distances between sub-levels are mostly more that the width of the generation spectrum.

$(g_k/g_m)f_{km} \sim 1$ that in fact can be much less than unity. For example, for the resonance transitions of Ba II 6s–7p (three lines near 201 nm) and 6s–8p (three lines near 162 nm) oscillator strengths are $10^{-2} - 10^{-3}$ [29] that is favorable for level population. Nevertheless, the lifetime mostly is less than the pulse length and the only way to obtain effective two-step ionization is to populate the intermediate level to saturation.

Kinetic equation for population of the upper level k in a system of two levels k and m (in our case the last denotes the ion ground state, $m = 1$), excited by resonance radiation, is

$$\frac{dn_k}{dt} = n_m \int \jmath_{\mathrm{L}}(\omega)\sigma_{mk}(\omega)d\omega - n_k \int [\jmath_{\mathrm{L}}(\omega)\sigma_{km}(\omega) + a_{km}(\omega)]d\omega\,, \tag{6}$$

where $\jmath_{\mathrm{L}}(\omega)$ is a spectral density of the photon flux, and

$$\int a_{km}(\omega)d\omega = A_{km}\,,$$

$$\sigma_{km} = \frac{a_{km}\lambda^2}{4}\,,$$

$$\sigma_{mk} = \frac{g_k}{g_m}\ \ \sigma_{km}$$

All the equations are written for the atomic system rest frame. By definition, saturation photon flux density $j^{\mathrm{s}}_{\mathrm{L}}$ corresponds to the equality

$$j^{\mathrm{s}}_{\mathrm{L}}(\omega)\sigma_{km}(\omega) = a_{km}(\omega) \tag{7}$$

Therefore, a transition is saturated if the spectral density is equal to or more than

$$j^{\mathrm{s}}_{\mathrm{L}}(\omega) = \frac{a_{km}}{\sigma_{km}} = \frac{4}{\lambda^2}\left[\frac{1}{\mathrm{cm}^2\ \ \mathrm{s}\ \ \mathrm{s}^{-1}}\right] \tag{8}$$

Since for $\theta = \pi$ the wavelength converts to $\lambda_{\mathrm{L}} = \lambda_{\mathrm{L}0}\sqrt{(1-\beta)/(1+\beta)}$, required saturation spectral density in the ion frame can be written as

$$j^{\mathrm{s}}_{\mathrm{L}}(\omega) = \frac{4}{\lambda^2_{\mathrm{L}0}}\left(\frac{1+\beta}{1-\beta}\right)\,, \tag{9}$$

were $\lambda_{\mathrm{L}0}$ is the laser wavelength in the laboratory coordinate system. The beam cross section at $\theta = \pi$ is the same in both systems. Total number of photons is a relativistic invariant

$$N^{\mathrm{s}}_{\mathrm{L}} = j^{\mathrm{s}}_{\mathrm{L}}\Delta\omega_{\mathrm{L}}\tau_{\mathrm{L}} = \mathrm{inv} \tag{10}$$

Substituting here Eqs. (2), (4) and (9), one can find relativistic transformation for photon flux spectral density:

$$j^{s}_{L0}(\omega) = \sqrt{\frac{1+\beta}{1-\beta}}\,\gamma j^{s}_{L}(\omega) \qquad (11)$$

Thus spectral density of the photon flux, required for saturation of a resonance transition of the moving ion, in the laboratory reference frame is

$$j^{s}_{L0}(\omega) = \frac{4}{\lambda^{2}_{L0}}\, f(\beta)\,, \qquad (12)$$

where

$$f(\beta) = \frac{1+\beta}{(1-\beta)^{2}}$$

Since

$$j_{\omega} d\omega = -j_{\lambda} d\lambda\,,$$

after simple evaluation one comes to a practical expression for the saturation spectral power density:

$$J^{s}_{\lambda}\left[\frac{\mathrm{kW}}{\mathrm{cm}^{2}\ \mathrm{nm}}\right] = \frac{1,42\ \ 10^{14}}{\lambda^{5}_{L0}\,[\mathrm{nm}]}\, f(\beta) \qquad (13)$$

One should emphasize that the saturation intensity does not depend on atomic parameters of the transition and is a function of the wavelength only. For the wavelengths of interest we have

$$J^{s}_{\lambda}(248.4\ \mathrm{nm}) = 150\, f(\beta)\left[\frac{\mathrm{kW}}{\mathrm{cm}^{2}\ \mathrm{nm}}\right] \quad (\mathrm{KrF\ laser})\,, \qquad (14)$$

$$J^{s}_{\lambda}(193.3\ \mathrm{nm}) = 526\, f(\beta)\left[\frac{\mathrm{kW}}{\mathrm{cm}^{2}\ \mathrm{nm}}\right] \quad (\mathrm{ArF\ laser}) \qquad (15)$$

For heavy ions relativistic factor γ is close to unity and the function $f(\beta) \approx 1$. Multiplying the required spectral power density (14) and (15) by τ_{L0}, one can find laser pulse energy necessary for ion beam saturated excitation. For KrF laser $\Delta\lambda_{L} \approx 0.4$ nm and for the laser-beam cross section of 1 cm, the laser pulse energy required for saturation of a heavy ion transition is merely 1.5 mJ. Similar calculation for ArF laser yields about 6 mJ.

For relativistic light ions $f(\beta)$ rapidly grows, and for $(1/4\gamma^{2})^{-1} \ll 1$ it tends to the limit $f(\beta) \to 8\gamma^{4}$ For the lightest (helium) ion at $T = 10$ GeV

the relativistic factor $\gamma \approx 2.5$ and $f(\beta) = 278$. This estimation shows that saturated population of the intermediate transition is easy attainable for most ions with conventional excimer lasers.

Since for the excimer lasers $\Delta\lambda_{L0}/\lambda_{L0} \equiv \Delta\lambda_{L}/\lambda_{L} \approx 2\ \ 10^{-3}$, relative Doppler width of the resonance line of the singly-charged ion has to be not more than this value. In this case only a fraction of the laser photons equal to $\Delta\lambda_{+D}/\Delta\lambda_{L}$ interacts with the ions. In the opposite case only a fraction of the ions equal to $\Delta\lambda_{L}/\Delta\lambda_{+D}$ can be excited.

Photoionization of excited singly charged ions

Lets assume bandwidth of laser radiation to be more than the Doppler width of the ion absorption line. For the beam cross section of 1 cm^{2} the spectral power density J_{λ} is about 100 MW/cm^{2}·nm that is much higher than the saturation spectral power density. Under such condition a resonance transition of a singly charged ion in the beam is populated to saturation, $n_{k}/n_{m} = g_{k}/g_{m}$, at the very beginning of the laser pulse. In contrast to the process of population of the intermediate level, all laser photons participate in the process of photoionization.

The photoionization rate can be calculated from the equation

$$-\frac{dn_{+}(t)}{dt} = n_{k}(t)\, j_{L}(\omega)\ \Delta\omega_{L}\, \sigma_{\mathrm{ph}}\,, \qquad (16)$$

where

$$n_{+} = n_{k} + n_{m}\,, \qquad (17)$$

and σ_{ph} is the photionization cross section. Since

$$n_{k}(t) = n_{+}(t)\ \frac{g_{k}}{g_{k}+g_{m}}\,,$$

density of the doubly ionized ions grows as

$$n_{+-}(t) = n_{+}(0)\ \left\{1 - \exp\left[-\left(\frac{g_{k}}{g_{k}+g_{m}}\, j_{L}(\omega)\,\Delta\omega_{L}\sigma_{\mathrm{ph}}\, t\right)\right]\right\} \qquad (18)$$

Thus, characteristic photoionization time in the moving frame, taking in account relation (10), can be written as

$$\tau_{\mathrm{ph}} = \frac{\tau_{L} G}{N_{\mathrm{ph}}\sigma_{\mathrm{ph}}}\,, \qquad (19)$$

where $G = (g_{k}+g_{m})/g_{k}$ and N_{ph} is a number of photons in the laser beam. Since τ_{ph} and τ_{L} transform to the laboratory frame in the same manner, one