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PULSED PLASMA SOURCES FOR THE PRODUCTION OF INTENSE ION BEAMS BASED ON "CATALYTIC" RESONANCE IONIZATION

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Аннотация

In this paper we describe a technique to produce planar and volumetric ion sources of nearly every element. This technique is based on a generalization of the LIBORS-process (Laser Ionization Based On Resonant Saturation) which because of its similarity to chemical catalytic reactions has been called CATRION (CATalytic Resonance IONization). A vapor containing the desired atomic species is doped with a suitable element possessing resonance transitions that can be pumped to saturation with a laser. By superelastic collisions with the excited atoms and by stimulated bremsstrahlung absorption seed electrons are heated. It is the heated electron component which then by collisional processes ionizes the desired atomic species and are multiplied. This technique will work efficient if both the atom and the ion of the dopant posses resonant transitions that can be pumped by the same laser. Instead of using tunable dye lasers we propose to apply gas lasers which are more robust and appropriate for repetitive long term operation. We present a large number of coincidences between gas laser wavelength and resonant transitions in suitable atoms.

PACS numbers: 29.25.Ni, 52.50.Jm, 52.75.Pv

Импульсные плазменные источники для получения интенсивных ионных пучков методом "Каталитической" Резонансной Ионизации

Б.А. Князев, П.И. Мельников, Г. Бакш

Аннотация

В настоящей статье мы описали способ получения плазмы для плоских и объёмных источников ионов почти любого элемента. Этот способ основан на *ЛИОРИ*-эффекте (Лазерная Ионизация Основанная на Резонансной Накачке). Благодаря своей схожести с химическими каталитическими реакциями новый метод был назван КАТРИОН (КАТалитическая Резонансная ИОНизация). В облако содержащее необходимые атомные элементы вводится небольшая примесь элемента, обладающего резонансным переходом, который *может быть возбужден лазером. Затравочные электроны греются в* сверхупругих столкновениях с возбуждёнными атомами и при вынужденном бремштралунговском поглощении. Затем нагретые электроны, сталкиваясь с возбуждёнными атомами, ионизуют их и размножаются. Этот метод будет эффективным если и атомы и ионы примеси обладают резонансными переходами, которые могут быть возбуждены одним и тем же лазером. Вместо использования перестраиваемых лазеров на красителях мы предложили применять газовые лазеры, которые более мощны и способны длительное время работать в частотном режиме. Мы нашли большое число атомов с переходами резонансными длинам волн газовых лазеров.

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1 Introduction

Many industrial and scientific applications require the production of spatially localized plasma layers near to or in contact with material surfaces in vacuum [1,2]. E.g., such layers are needed in the formation of plasma electrodes for laser ion sources [1, 2] and in different types of high power accelerators [3, 4, 5]. A standard way for the production of the plasma layers is to illuminate a suitable material surface with intense non-resonant laser radiation [6]. At sufficiently high exposure levels a laser plasma" with a relatively small number of particles but with high ion-temperatures is formed. A more effective way to produce the plasma is by a two-step process [7, 8], where first a neutral gas cloud is produced in one way or another (e.g. by laser radiation) which then can be ionized by means of laser produced shock waves [7], an external UV-flashlamp [8, 9], or a near-surface discharge [10].

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2 Ionization of a dense gas by resonance radiation

If the atomic density in the vapor cloud is sufficiently high the LIBORS process (Laser Ionization Based On Resonance Saturation) [11, 12] can be applied for its ionization. In this case an intense resonant laser radiation provides a high non-Boltzmann population of the corresponding state. Then through high-frequency super-elastic collisions of seed electrons with excited atoms and stimulated bremsstrahlung absorption the radiation energy is transferred to free electrons. Eventually after a subsequent chain of events, the gas becomes ionized practically fully.

The LIBORS process is very complex and there are different points of view on its details [12, 13, 14, 15, 16, 17]. However, evidence for the super-elastic mechanism in laser-induced ionization was found in [18] and spectra of hot electrons were observed directly [19]. Most of the LIBORS-experiments were carried out under steady-state conditions in metal vapor furnaces (see, for example, [13]), and only one group [20] used this method for the ionization of a transient thin lithium-vapor-layer on the anode surface of the PBFA-II high-power ion diode. Recently, a reduction of the threshold intensity for plasma production was observed [21], when the same laser pulse was used both to evaporate a thin target layer and to resonantly excite and ionize the vapor.

The advantages of the LIBORS-method are: a more effective use of the available laser energy for ionization, and a low temperature of the resulting plasma ions. This allows, in principle the production of ion beams with low transverse velocities, i.e. small divergence. However, the necessity of a tunable flash-lamp pumped dye laser for the resonant excitation of the vapor limits its practical usefulness.

Mainly, because of their photochemical instability [22] and their thermo-optical distortion dye solutions are unsuitable for operation at high repetition rates and for long time periods. In addition, to ionize different atoms one must change from one dye solution to another and tune a dispersive element of the laser. Furthermore, the resonance transitions of many elements fall into the UV- and VUV-range where no powerful tunable lasers are available.

3 LIBORS with gas laser excitation

In contrast many intense gas lasers have adequate operating characteristics and could be used for a repetitive long term operation of a LIBORS ion source. Unfortunately there are only a few coincidences between resonant atomic transitions and wavelengths of gas lasers.

The matching between resonant transitions in TiI and AlI with the N_2 and XeCl-laser wavelength at 337 nm and 308 nm respectively had been known to us before beginning of a systematic search [23, 24]. We explored resonance transitions for atoms of all elements [25, 26, 27, 28] and found 18 transitions whose wavelengths are sufficiently close to eximer laser lines.

Fig.1 shows the positions of these atomic lines together with the experimentally observed emission spectra of ArF, KrF, XeBr, XeCl, and XeF eximer lasers and the N_2-laser as well [23, 36]. Since the fluorescence spectra of these laser transitions are quite wide the laser lines can be tuned within these limits to match the resonance lines falling into the fluorescence band [31, 32, 33, 34, 35]. It should be pointed out that active media of the eximer lasers are more stable than dye solutions. The most stable medium among eximers is XeCl [36].

More details about the relevant parameters of the above mentioned elements and their resonance lines are listed in Table I. Some of these atoms have more than one suitable transition. In order not to overload the Table with too many data we have included only 3 of 9 possible transitions in V, and 2 of the 5 in Ti^+.

Those transitions, whose lower-level energies deviate from zero have been marked with asterisks. The ionization potentials for the neutral atom and the first two ionization states are presented in the Table I too. One can see that for all selected elements the first ionization potential I_1 is always less than $2\hbar\omega_{21}$; thus, in contrast to alkali atoms, photo-ionization from a resonance level is an additional possible mechanism to generate electrons (see Fig. 2). In the other rows of Table I the statistic weights g_1 and g_2 and the value of g_2A_{21} for the resonance transitions have been listed.

At a high vapor density, atoms inside the cloud can only be excited in the saturation mode. Let us estimate the laser intensity required to achieve saturation. Under steady-state conditions a balance equation for the population of a resonance level can be derived (see, for example, [37]):

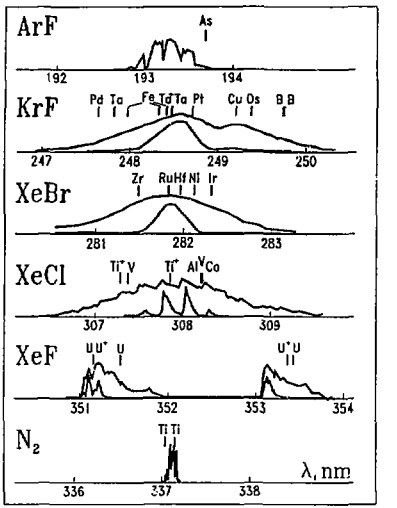

Fig. 1. Fluorescence and generation spectra of pulsed gas lasers and wavelengths of resonance transitions of atoms and ions.

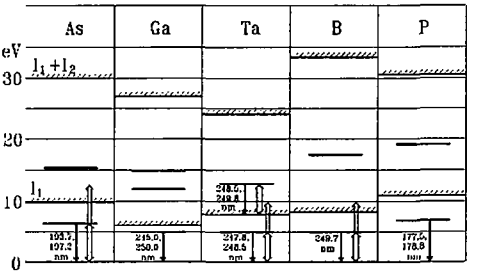

Fig. 2. Energy levels and ionization potentials of certain elements.

$$n_l \int j_\omega(\omega)\sigma_{12}(\omega)d\omega - n_2\Big[\int j_\omega(\omega)\sigma_{21}(\omega)d\omega + A_{21} + \sum_k A_{2k}\Big] = 0. \qquad (1)$$

Here A_{21} and A_{2k} are the Einstein coefficients for the resonance and for the cascade transitions respectively and j_ω is the spectral power density of the laser radiation. The cross-sections for the stimulated transitions σ_{12} and σ_{21} are equal to

$$\sigma_{21}(\omega) = \frac{\lambda^2}{4} a_{21}(\omega)A_{21}, \quad \sigma_{12} = \frac{g_2}{g_1}\sigma_{21}, \quad \int a_{21}(\omega)d\omega = 1.$$

By definition, the saturation parameter is equal to unity, when the rates of spontaneous and stimulated transitions from the upper level (the terms in the square brackets of eq. (1)) are equal. If the laser line is sufficiently broad as compared to the atomic line-width, the saturated radiation intensity j_ω^S is given by the following relation:

$$j_\omega^S = \frac{A_{21} + \sum_k A_{2k}}{\int \sigma_{21}(\omega)d\omega} = \frac{4}{\lambda^2}\left(1 + \frac{\sum_k A_{2k}}{A_{21}}\right). \qquad (2)$$

Table I. Resonance transitions and relevant atomic data

atom/ ion	λ_{21}; nm	Ionization potential I_1	I_2	I_3	g_1	g_2	$g_2 A_{21}$; $10^8 s^{-1}$
As	193.69	9.81	18.6	28.4	4	4	3.5
B	249.68	8.30	25.2	37.9	2	2	7.0
	249.77				4*	2	14.0
Fe	248.33	7.87	16.2	30.6	9	11	34.
	247.98				5*	5	15.0
	248.42				3*	3	14.0
Cu	249.21	7.72	20.3	36.8	2	4	0.31
Pd	247.64	8.33	19.4	32.9	1	3	1.1
Ta	247.82	7.88	16.2	22.3	4	4	0.85
	248.49				4	4	2.8
Ta^+	248.47	—	16.2	22.3	3	5	3.7
Os	249.38	8.7	17.0	25.	9	9	0.27
Pt	248.72	9.0	18.6	28.5	7	9	5.7
Ni	282.13	7.63	18.2	36.2	7*	7	0.72
Zr	281.49	6.84	12.9	24.1	5	5	6.5
Ru	281.84	7.36	16.6	30.3	11	9	1.5
Hf	281.97	5.5	14.9	21.	5	7	0.61
Ir	282.32	9.2	17.	27.	10	8	0.89
Al	308.22	5.98	18.8	28.4	2	4	2.7
V	308.21	6.74	14.65	29.3	10	8	1.4
	307.38				8*	6	1.4
Co	308.26	7.86	17.05	33.5	10	12	0.60
Ti^+	307.29	—	13.6	28.1	4	2	3.8
	307.86				8*	6	8.0
U	351.14	6.2	:::	:::	:::	:::	0.96
	351.46		:::	:::	:::	:::	6.2
	353.43		:::	:::	:::	:::	1.2
U^+	351.16	—	:::	:::	:::	:::	0.081
	353.36	—	:::	:::	:::	:::	0.40
	485.97	—	:::	:::	:::	:::	0.011
Ti	337.04	6.82	13.6	28.1	5	3	2.5
	337.15				9*	11	11.0

Let us design the expression in the brackets by ξ. In the case of a two-level system $\xi = 1$, and $j_\omega^S = 4/\lambda_{21}^2$ [cm^{-2}]. Multiplying j_ω^S by the photon energy

$\hbar\omega_{21}$ and transforming from frequency to wavelength, we obtain for J_λ^S:

$$J_\lambda^S = \xi \frac{16\pi^2\hbar c^2}{\lambda^5} \left(\frac{erg}{cm^2 \cdot s \cdot cm} \right) = \frac{1.4 \cdot 10^{14}\xi}{\lambda^5[nm]} \left(\frac{kW}{cm^2 nm} \right). \qquad (3)$$

The quantity J_λ^S/ξ depends only on the laser wavelength and is presented for several gas lasers in Table II. If cascade transitions from the resonance level are possible these intensity values can be much higher.

Table II. Laser wavelength (nm) and saturated intensity ($kW{=}cm^2nm$)

laser	ArF	KrF	Xe_2Br	XeCl	N_2	XeF
λ	193	248	282	308	337	351
$J_\lambda^S =$	530	150	80	50	33	27

It should be noted that, on the one hand, the specific value of saturation must be sufficiently low, to produce the most effective absorption of the laser light in the vapor cloud, and on the other hand, it should be sufficiently high to provide an adequate rate of energy transfer to the electrons. To reach a saturation parameter close to unity and to deposit the laser radiation with high efficiency, the lasing spectrum and the atomic absorption line must overlap at least partially.

From Fig. 1 one can see that for certain atoms the overlapping is not too good, and very high laser intensities may be required to saturate these transitions. However, one important point to remember is that the absorption line can be significantly broadened in the intense resonant light field [39] or due to collisions among gas particles ("pressure broadening" [38]). Overlapping may also be improved by tuning of the laser spectra with a selective resonator within the spectral ranges shown in Fig. 1. An ultimate solution to the problem of optimizing both the vapor density and the radiation intensity is beyond the scope of this work and needs experimental investigations.

4 Catalytic resonance ionization

Thus, at least for the atoms listed in Table I the quoted disadvantages of LIBORS can be overcome. Furthermore, using the coincidences found, we can come to a more universal LIBORS process if admixed atoms are acceptable in the vapor cloud. This may be tolerable in some ion sources from which a

multicomponent ion beam is extracted at first which then can be separated into its components after acceleration. In such cases one may dope the cloud of desired atoms with some impurity atoms whose resonance transitions agree with the laser spectrum.

These impurities will play a dominant role in the LIBORS process as acceptors for laser radiation and as subjects participating in super- elastic collisions and resonant bremsstrahlung absorption with seed electrons. The main ("nonresonant" atomic component will be excited and ionized by means of electron impacts and other atomic collisions. Ionization of "non-resonant" contaminations has been observed experimentally in [13], where Sr atoms were ionized in a Ba-vapor.

Thus, during the first phase of the LIBORS process we have atoms in the cloud, which drive the process without being changed themselves, like those of a catalyst in a chemical reaction. Because of this property, we call the modified LIBORS process CATalytic Resonance IONization (CATRION) [29].

Table III. Ionization potentials, (eV)

	Be	*Mg*	*Si*	*P*	*S*	*Ga*	*Ge*	*Sr*	*Ag*	*Sb*	*Ba*
I_1	9.3	7.6	8.2	11.0	10.4	6.0	7.9	5.7	7.6	8.4	5.2
I_2	18.2	15.0	16.3	19.7	23.4	20.4	15.9	12.2	21.5	18.0	10.0
I_3	153.8	78.2	33.4	30.2	35.0	30.6	34.1	43.6	34.8	24.7	37.

In Table III the ionization potentials for certain elements which are of interest for many industrial applications are presented. A vapor cloud consisting of the desired atoms and the "catalyst" atoms can be produced by laser evaporation of a suitable target mixture. Consequently, multicomponent targets (alloys, pressed powders, crystals) with the required elemental composition and suitable thermodynamical, optical and electrical characteristics must be provided.

As an example, GaAs or TaB_2 [41] can be used as targets. The Grotrian diagrams for these elements as well as for P are shown in Fig. 2. The absorption lines of As and Ta coincide well with the laser spectra of ArF and KrF, respectively. The feasibility of the ionization of B with a KrF-laser must be studied separately. During laser excitation the energy spectrum of the electrons will not be in thermodynamic equilibrium but contain two maxima near $E = \hbar\omega_{21}$ and $E = 2\hbar\omega_{21} - I_1$. The electrons will ionize both elements, but the rate of ionization is expected to be higher for the catalyst atoms because of the saturation of their resonance level and the scaling of the cross section for electron impact ionization with the 4th power of the principal quantum number n [30]. In addition the catalyst will be ionized by laser

photons. As a consequence we expect, that the catalyst atoms will be fully ionized while the other atoms will only be partially ionized. This difference in the degree of ionization can be especially dramatic for atoms with a high ionization potential, like phosphors.

The possibilities to completely ionize the main (wanted) component in the vapor have to be studied experimentally. However the problem will be mitigated if Ta or U are applied as catalysts. Both, atoms and singly-charged ions of these elements (see Table I) can be excited with the same laser (KrF or XeF, respectively). This means that the radiation will continue to be absorbed by the plasma even after full ionization of the catalyst atoms, and, as a consequence, the full ionization of the main component can be achieved. In this case a heating of the electron fluid in the plasma can also be provided. Therefore, the term "catalytic resonance ionization" is especially correct for this process.

5 Examples of CATRION ion sources

Let us consider two versions of the CATRION ion source. The first one is an ion source for an ion accelerator for technological or scientific applications (implantation, lithography etc.). A multicomponent target is installed on the anode surface (see Fig. 3). External non-resonant pulsed-laser radiation with an energy of the order of 0.01 J and pulse duration of 30–100 ns is focused to point on the target and produces a vapor cloud. The depth of evaporation is of the order of 1 μm, and the number of atoms in the cloud is of the order of 10^{16}. To exclude burning of holes in the target it can be designed for slow rotation and fast replacement. The vapor cloud from the point source expands almost spherically with a velocity of about 0.1 $cm/\mu sec$ [8], and after 10 μsec will be irradiated with resonant eximer-laser radiation with an energy of about 1 J.

When the diameter of the cloud reaches a value of 10 cm its ion density has decreased to $10^{13}\,cm^{-3}$. At this stage it is possible to extract ions from a cloud by means of a tungsten grid at a voltage of 3–10 kV. Using a wire diameter of 200 μm and an inter-wire distance of 200 μm the heating per pulse can be very high, and the grid must be cooled down before the next pulse. For this purpose the voltage should be switched off between pulses and the grid should be cooled with a neutral vapor cloud. The repetition rate of such a system can be as high as 100 Hz. Consequently, we can expect an average ion current with this laser source of the order of several tens of A/cm^2.

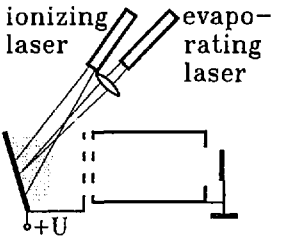

Fig. 3. Optical system for laser evaporation of anode surface in a high-power ion diode.

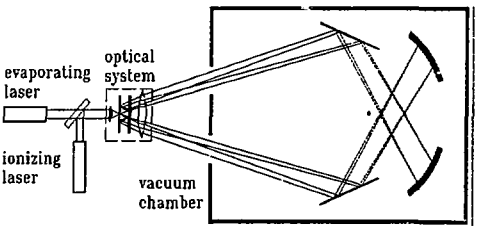

Fig. 4. Schematic of the CATRION laser source.

Another possible application of the CATRION ion source is in high power accelerators for inertial confinement nuclear fusion. A key problem in the production of intense light ion beams is the formation of a thin, uniform, and pure plasma layer on the anode surface of the high-voltage diode [4, 5]. At diode voltages between 1 and 3 MV proton and deuterion sources are suitable for beam-target interaction experiments. However, to reduce the divergence of the ion beams it is desirable to use heavier mass ions and accelerating voltages above 10 MV. Therefore, suitable sources for heavier ions like Li, Be, B are needed.

Since in these high power diodes the intense ion beams are produced space charge limited lighter ions are predominantly accelerated and the presence of hydrogen impurities is not permitted, whereas to a certain degree heavier ions may be present in the plasma.

To produce a Be or B plasma conventional techniques can probably not be used. We propose a possible system for the production of a suitable boron containing anode plasma in the diode of a high power accelerator: One may start from a LaB_6, TaB_2 or VB_2–target and evaporate and excite it by a KrF or XeCl laser. Suppose, that the anode has an active area of 300 cm^2 and that its configuration is similar to that shown in Fig. 2 of Ref. [5]. To form a plasma layer with a thickness of 1 mm and an areal density of $10^{16} cm^{-2}$ with a conventional laser we should take into account that the pulse duration is one of the most "rigid" parameters for each type of laser, and, for lasers we are interested in, is equal to 30–100 ns. To produce a uniform vapor-layer the radiation must be evenly distributed over the anode surface.

For this purpose one can use, for example, a system of lenses and prisms [8] or other optical systems. The estimates show that a laser with an energy of about 10 J is required for evaporation and that the laser to produce the catalytic resonance ionization must provide another 10–40 J to the vapor cloud. A schematic of the possible geometry of the optical system is shown in Fig. 4. For a more accurate determination of the required laser parameters, detailed experimental studies have to be carried out.

6 Summary

In summary we can say that Catalytic Resonance Ionization using eximer lasers can produce large area plasma layers near surfaces with a large variety of element compositions. Such plasmas can be used as pulsed ion sources with high repetition rate suitable for many industrial applications. In particular, it seems possible to replace the aggressive As- and P-containing

gases on which many industrial ion sources are presently based. The ion temperature of such a plasma may be less than the temperature of a typical discharge plasma and, therefore, if no other phenomena would contribute to the beam divergence very high brightness ion beams could be generated. Another important field for the application of the CATRION process is in high power light ion inertial confinement fusion drivers. Using this method gives access to a large variety of new sources of low and medium mass ions.

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