Experiments on "catalytic resonance ionization" of dense tantalum-contained ruby-laser-produced gas clouds with KrF-laser

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ABSTRACT

In this paper we describe the principle of a technique to produce planar and volumetric plasma sources of nearly every element and results of experiments on verification of this technique. 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). Characteristics of vapor clouds formed near wide variety of mono- or two-component targets in vacuum with a pulsed ruby laser were investigated. Effect of an intense KrF laser radiation on expanding tantalum-contained and, for comparision, titanium vapor clouds was studied. Photoresonance ionization in saturation mode of tantalum cloud was detected with the help of a frame camera, Langmuir probes and spectroscopic diagnostics.

1. CATALYTIC RESONANCE IONIZATION AS A TECHNIQUE FOR MULTI-COMPONENT PLASMA PRODUCTION

Many industrial and scientific applications require the production of spatially localized plasma layers near to or in contact with material surfaces in vacuum. For example, such layers are needed in the formation of plasma electrodes for laser ion sources [1,2] and in different types of high power accelerators [3-5]. 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].

An alternative way for a dense gas layer ionization is LIBORS technique (Laser Ionization Based On Resonance Saturation) [11-13]. A resonant dye-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 advantages of The LIBORS-method (in contrast to "laser plasma") is a more effective use of the available laser energy for ionization, and a low temperature of the resulting plasma ions. However, the necessity of a tunable flashlamp-pumped dye laser for the resonant excitation of the vapor limits its practical usefulness. However, due to the photochemical instability of dye lasers [14] and strong thermo-optical distortion of dye solutions they 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.

In the papers [15,16] we suggested the development of the LIBORS technique to ionize gas/vapor layers with eximer lasers. We have found about twenty elements resonance transitions of those overlap or lie close to the generation spectra of ArF, KrF, XeBr, XeCl, and XeF eximer lasers. These lasers have adequate operating characteristics and could be used for a repetitive long term operation of a LIBORS plasma source. Fig. 1 shows the positions of certain atomic lines with respect to generation band of KrF laser and kriptonium fluoride fluorescence spectrum.

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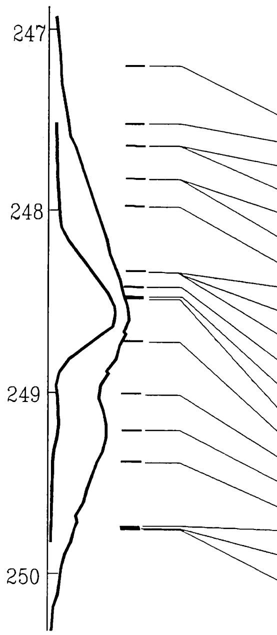
Since the last is quite wide the laser spectrum can be, in principle, tuned within these limits to match the resonance lines falling into the fluorescence band.

Thus at least for the above mentioned atoms 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 plasma 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. Such a process because of its similarity to chemical catalytic reactions has been called CATRION (CATalytic Resonance IONization) [15,16]. Schematic of plasma production by this process is shown in Fig. 2.



Atom/ion	λ,nm	gA,10^8 c^{-1}	low level, cm^{-1}	Δλ,nm
Ni	247.21	2.5	2217	-1.4
Li	247.51		0	-1.1
Pd	247.64	1.1	0	-1.0
Pb	247.64	7.8	7819	-1.0
Ta	247.82	0.85	0	-0.8
C	247.86	19	21648	-0.7
Fe	247.98	15	704	-0.6
Fe	248.33	34	0	-0.3
Rh	248.33	1.0	3473	-0.3
Sn	248.34	6.0	3428	-0.3
Fe	248.42	14	888	-0.2
Ta^+	248.47	3.7	0	-0.1
Ta	248.49	2.8	0	-0.1
Pt	248.72	5.7	0	0.1
Y	249.04	0.77	530	0.4
Cu	249.21	0.31	0	0.6
Os	249.38	0.27	0	0.8
B	249.68	7.0	0	1.1
B	249.77	14	16	1.2
Ge	249.80	2.4	0	1.2

Figure 1: Laser generation and molecular fluorescence spectra of kriptonium fluoride vs. wavelength (nm) and the table of resonance transitions of certain elements

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 [17]. In addition the excited atoms of catalyst can be ionized by laser photons directly. 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.

The feasibility to completely ionize the main (wanted) component in the vapor have to be studied experimentally. However in a case of KrF laser the problem will be mitigated if Ta is applied as catalyst. Both atoms and singly-charged ions of this element (see Fig. 1) can be excited with the laser. 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.

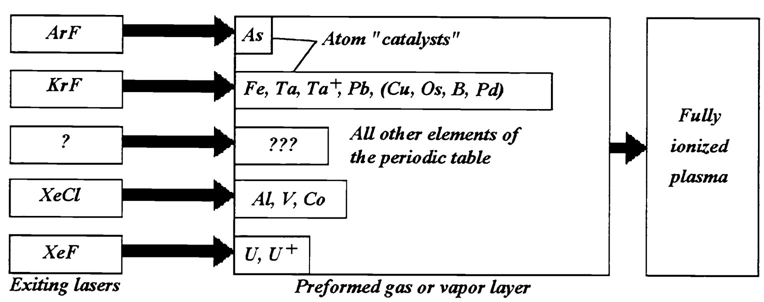


Figure 2: Schematic of plasma source based on catalytic resonance ionization: one of the eximer lasers (the left boxes) by means of excitation of an element "catalyst" (in the right boxes) ionizes all components of gas mixture.

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.

The final goal of our work is to verify experimentally realizability of the CATRION technique for ionization of multi-component gas clouds and to find for what range of gas and UV-radiation parameters the efficiency of the process is maximum. In this paper we present results of the first stage of the experiments. We studied characteristics of vapor clouds produced with a ruby laser for wide variety of targets of different elemental composition and have found optimum regimes for vapor clouds production. For the reasons given above we chose for the first experiments on catalytic resonance ionization tantalum and tantalum contained clouds.

2. EXPERIMENTAL APPARATUS

2.1 Experimental setup

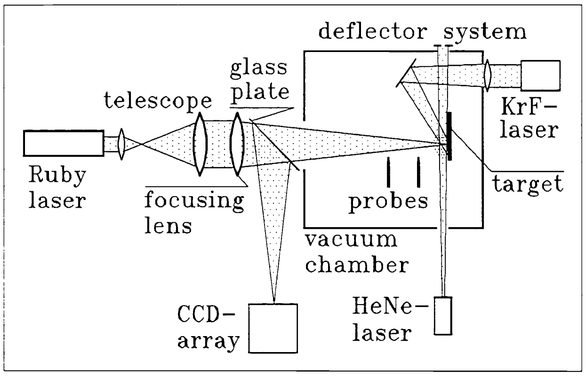


Figure 3: Schematic of experimental setup.

Experimental setup (see. Fig.3) consists of a 80 dm^3 vacuum chamber, a 0.3 J ruby laser for vapor production, a 0.1 J KrF laser for the vapor ionization. A set of diagnostic tools and a PC-based data acquisition system were used for record and processing of experimental data. Vacuum chamber is equipped with six glass and quartz windows and pumped down to $3 \cdot 10^{-6}$ Torr. A target is positioned into the chamber in the focus of output lens of ruby laser optical system and connected to ground. As a pulse length of ruby laser is about 25 ns and minimum diameter of focal spot on the target is 0.8 mm, maximum power density is equal to 2 GW/cm^2, whereas the density of 460 MW/cm^2 is sufficient to provide so called optimum evaporation of tantalum. The power density on the target was varied with two methods: by the attenuation of the laser beam with glass filters and by increasing of the beam diameter at the same beam energy. A fraction of the laser beam was splitted with a glass plate to CCD system to record distribution of power density on the target. To ionize an expanding vapor cloud a long-pulse TEA eximer laser [18] was used as a source of UV radiation. The laser beam with the cross section 3.5 cm $\times 0.7$ cm was conducted to the cloud with a system of quartz lenses and dielectric mirrors. Because of reflection of the beam from lens surfaces only 60 % (50 mJ) of radiation energy reached the target. The length of the laser pulse is equal to 150–200 ns. Generation spectrum of the laser covers the spectral range from 2481.5 to 2487.7 Å. Thus both TaI and TaII resonance lines lie close to maximum of the spectrum.

2.2 Diagnostics and data acquisition system

The CATRION setup is equipped with diagnostic tools intended for measurement of laser beams characteristics and gas cloud and plasma parameters. To measure a pulse energy, power and power density of the laser beams we used calorimeters, vacuum photodiodes, and CCD array. Spectral distribution of KrF laser radiation was taken with quartz prism spectrometer (linear dispersion is 9 Å/mm at 250 nm).

In the first stage of our experiments a dense vapor cloud have to be formed. To study the cloud evolution we used the following diagnostic instruments. As the initial temperature of vapor clouds is close to boiling point a cloud emits visible radiation. This radiation was taken with an electron-optic frame camera with MCP amplifier to observe the cloud shape during a few microseconds after laser evaporation. As recombination in expanding cloud becomes "frozen" in the last stage of expansion we used two Langmuir probes to study ion distribution in a cloud. The probes are a 20 or 120 μm diameter, 5 mm length tungsten wire. The probes were operated in ion collector regime. They were placed at the distances of 35 and 70 mm from the target.

One of the critical points in diagnostics of a laser-produced gas and plasma layers is determination of particle density near the target at the early stage of the plasma expansion. Electrical probes for many well-known reasons cannot be applied at a dense small-size laser plasma. Spectroscopic techniques is rather sophisticated and time consuming. To obtain more detailed information about cloud density near the target we used so-called "laser deflector system" [19]. For the goals of our experiment the laser deflection technique seems to be the most adequate due to its high sensitivity as well as tolerable time and space resolution. In addition, this technique permits to distinguish easily between electron and neutral induced refractive index gradients. The last advantage, on one hand, is the most important for determination of required ruby laser energy density on a target surface, and on the other hand, makes it possible to observe directly gas ionization as the result of the eximer laser irradiation of the cloud.

In Fig. 4 schematic of laser deflection system is depicted . The beam of a He-Ne laser passes through a cloud and bends to the direction of the refraction index gradient. The deflection angle is measured with a couple of photodiodes with differential amplifier. The value of the angular deflection is given by equation

$$\delta\phi = \frac{1}{\tilde{n}_0}\left|\int ds\nabla_\perp \tilde{n}\right|, \qquad (1)$$

where $\tilde{n}$ is the index of refraction, $\tilde{n}_0$ is the unperturbed index of refraction, integral is taken over the path of the laser beam in the plasma. The index of refraction of an atomic component (atoms, ions, or molecules) of plasma is given by

$$\tilde{n} = 1 + Kn \qquad (2)$$

where n is the atomic component density, and the constant K is proportional to polaribility α of the particle:

$$K = 4\pi\alpha.$$

The values of K for the most of atoms and molecules lie in the region from $0.5 \cdot 10^{-23}$ to $50 \cdot 10^{-23}$ cm^3.

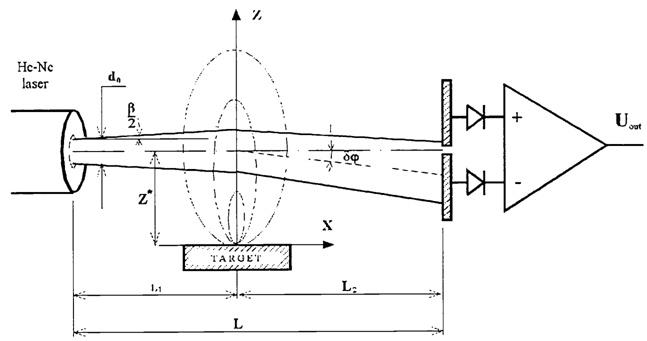


Figure 4: Helium-neon laser beam deflection system for measurement of refraction index gradient in the expanding vapor cloud.

To define density gradient of a multi-component cloud from measured beam deflection one have to know percentage as well as polaribility each of the component. Unfortunately, polaribilities of a very many atomic systems are unknown. The other problem at the deflector data interpretation is the dependence of polaribility on electron excitation of the atomic system.

The polaribility of plasma electrons is negative. It depends on the wavelength of probing beam and given by the expression

$$\alpha_e = \frac{e^2}{m\omega^2} = 7.1 \cdot 10^{-29} \cdot \lambda^2\,(nm).$$

For the wavelength of He-Ne laser this value is equal to $17.9 \cdot 10^{-23}$ cm^3. The constant K is the highest for the elements of first and second groups of periodic system and can even exceed the value of K_e. For other atoms and molecules this value is is usually one order less.

In our deflection system (which will be further referred as "deflector") we apply HeNe laser LG-208A with the power about of 3 mW, the beam divergency β of $5 \cdot 10^{-4}$ rad, and the diameter d_0 of 0.5 mm. As $L_1 = 49$ cm and $L_2 = 30.5$ cm, the distance L from the laser to the photodiode couple is equal to 79.5 cm, that is close to the optimum value of 100 cm. As photodiode couple we used silicon quadrant photodiode FD-K-142 with the sensitive area diameter of 13.7 mm. The space between quadrants is equal to 120 μm. The high-frequency limit is equal to 80 MHz. The sensitivity of deflector is 28 μ rad / V. When we were studying vapor clouds expansion the level of electromagnetic noise in the deflector system in our experiments was about 10 mV. This gives the sensitivity limit of the system the order of 0.3 μrad.

Distribution of electrons and ions in the cloud can be calculated using the results of measurements at several distances from a target. If the plasma/vapor density distribution can be approximated with Gaussian distribution, the main cloud parameters can be inffered even from one-chord measurements [19]. In a case of particles adhesion to the target the distribution has to be less sharp and results obtained at the above mentioned assumption can be accepted as a lower estimation.

To record spectral lines emitted by gas and plasma we applied two spectroscopic systems. Time dependence of a spectral line in visible spectral region could be recorded with a 0.25 m grating monochromator with photomultiplier. A 0.5 m grating monochromator with an electron-optic dissector was applied to record time evolution of radiation in different intervals of UV and visible ranges. A system of quartz lenses and fibers was used to conduct the light from cloud to the monochromators.

All signals were recorded with data acquisition system which consists of a personal computer 486DX-33 and CAMAC crates.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Targets

Catalytic resonance ionization is a rather intricate process. The efficiency of ionization of a preformed vapor cloud with the eximer laser radiation is dependent on many parameters of the cloud (total number of particles, component ratio, temperature, spatial density distribution, initial degree of ionization, wavelength of the resonance line of a catalyst) and the radiation (power, spectral power density). For these reasons we are planning to study in our experiments several types of targets.

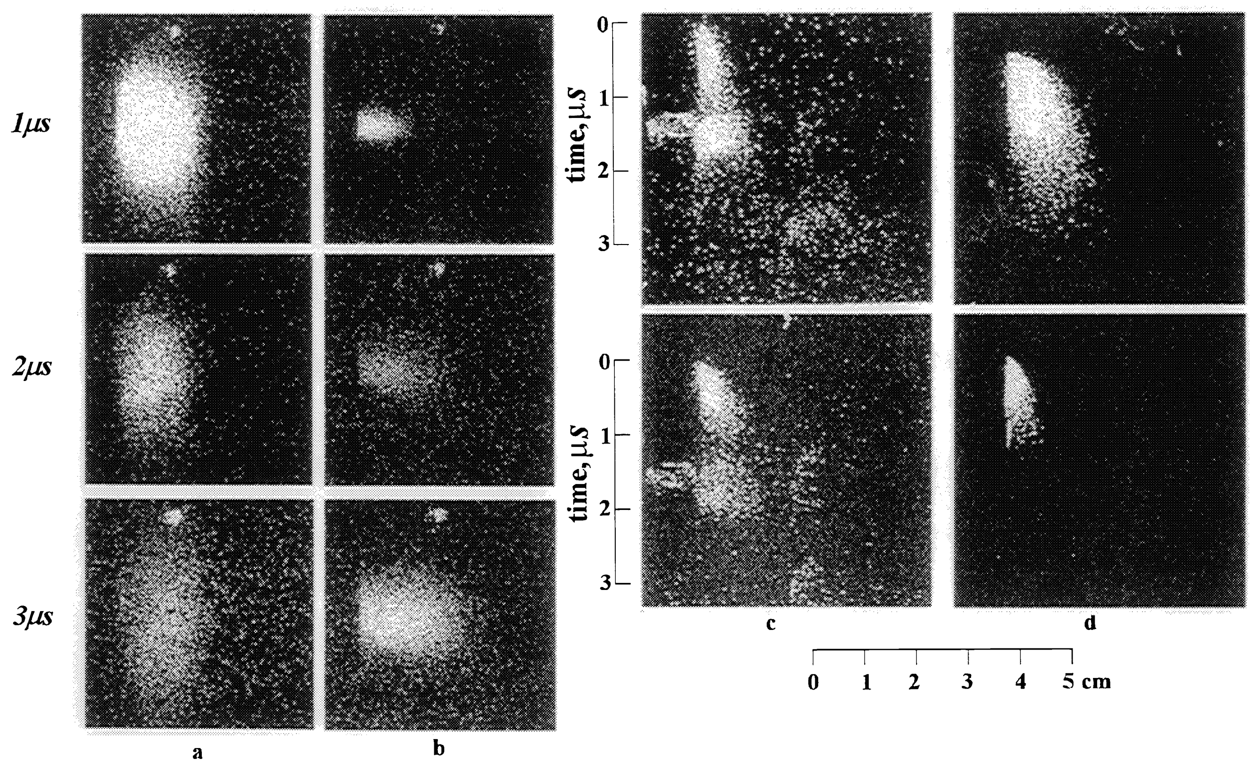


Figure 5: Self-luminosity of Fe_2 B (a) and Ta (b) clouds at different time after ruby laser pulse taken with the frame camera. Luminosity of tantalum (c) and titanium (d) clouds irradiated with KrF laser (frames with vertical sweeping).

Monocomponent tantalum and iron targets consist of the atoms which resonance transitions overlap with KrF laser generation spectrum (see Fig. 1). These atoms can be directly excited by the laser radiation. The next set of mono-component targets includes copper, lead, an boron targets which wavelength lie into the limits of fluorescence spectrum of KrF mixture. They can be excited, in principle, either with a powerful eximer flashlamp or because of probable resonance line broadening in the intense KrF-laser field ("dressed atom"). The rest monocomponent targets (nickel, titanium, molybdenum, and tungsten) with similar thermodynamic characteristics but without resonant transitions near to the laser line were studied for comparison. Each of two-component targets contains both an element catalyst and an element which resonance transitions are far from KrF laser line. We have fabricated Ta-Ti, Ta-B, and Fe-B targets with different ratio of elements.

3.2 Vapor clouds forming with a ruby laser

We have investigated characteristics of vapor clouds as a function of energy density E_s (or power density F) of ruby laser radiation on the target. Their maximum values reach 66 J/cm^2 and 3.5 GW/cm^2, correspondingly.

Expansion of laser produced clouds during a few microseconds after ruby laser pulse was studied with the frame camera and the deflector system. Exposition time of the camera was one microsecond. Self-luminosity in visible spectral range of ruby-laser produced vapor clouds near Fe, Fe_2B, Ta and Ti targets was taken at the moments one, two, and three microseconds after laser pulse. It was found that shape of a cloud depends dramatically on elemental composition of the target. In particular, the cloud expands approximately semispherically with $v_{\|} = v_{\perp}$ for Fe_2B target (Fig. 5a). For the other targets transverse velocity is less than longitudinal velocity. The ratio $v_{\|}/v_{\perp}$ is maximum (≈ 3) for tantalum target (Fig. 5b), and ≈ 2 for the other targets. Scans of the pictures made with laser microdensitometer show that in 1–2 μs after evaporation light intensity near target surface decreases, probably, both due to adhesion of particles to the surface and the gas cooling.

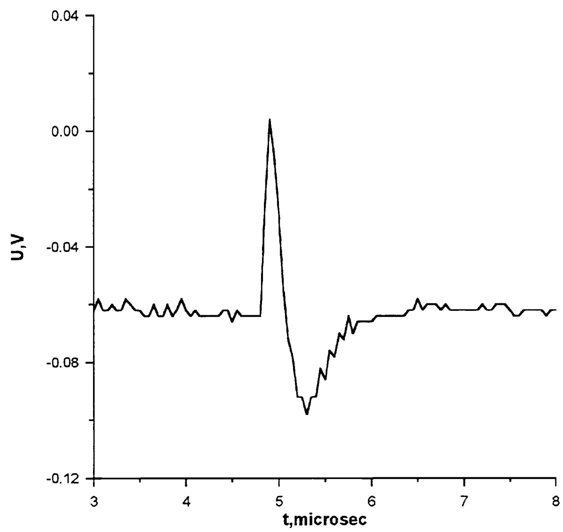


Figure 6: Signal of the deflector system after passing of HeNe laser beam through a gas cloud produced near tantalum target.

The deflector system enables to obtain information about ionization degree of a cloud, as direction of refractivity index gradient is opposite for neutral particles and for electrons. Overview of all the experimental data shows that it is easy to produce a cloud practically without electrons in a case of targets with low evaporation temperature. Almost neutral cloud for targets with high evaporation temperature can be obtained in very narrow range of laser power densities near threshold of evaporation. In this case one can see in oscillogram (Fig. 6) after positive ("electron") peak appearance of negative one. As polaribility of tantalum is unknown it is impossible to find absolute value of the vapor density from this oscillogram. In other cases (for example, for lead and titanium) we were able to calculate absolute density of neutral particle in a cloud.

The data obtained with the frame camera and the deflector system as well as data of spectroscopic measurements and Langmuir probes have shown that low-ionized vapor clouds of different elemental composition with a density of 10^{15}–10^{17} cm^{-3} at the size of about 0.5–1 cm can be obtained.

3.3 Interaction of KrF laser radiation with expanding gas clouds

Schematic of this experiment is shown in Fig. 3. Radiation of KrF laser was focused with the optical system that consists of a few quartz lenses and dielectric mirrors onto the expanding cloud under the angle of incidence of about 30°. Cross section of the laser beam near the target was rectangular in shape 1cm wide and 2 mm thick. The beam energy of 50 mJ corresponds to spectral power density of about 20 MW/cm^2nm. This value exceeds a value of 150ξ kW/cm^2nm, where $\xi = 1 + \sum_k A_2k/A21 > 1$, needed to saturate a resonance transition for $\lambda = 248$ nm [15].

To verify realizability of catalytic resonance ionization we studied the effect of KrF laser radiation on tantalum, TaTi and titanium clouds. Expanding clouds were irradiated with a pulse of KrF laser at 1–10 μs after ruby laser pulse. The cloud pictures were taken by electron-optic image converter. It was operating in the regime of frame camera with open shutter and simultaneous vertical sweeping of the frame. To cut the light of the lasers a glass filter with transparency from 330 to 640 nm was placed before the image converter photocathode. Luminosity evolution of the clouds expanding out of TaTi and Ti targets is shown in Fig. 5c,d. One can see that in a case of tantalum-contained target if the cloud is irradiated with the resonant laser in early stage of expansion (1–2 μs) the second bright flash of the cloud self-luminosity is detected. There are no second flash in a case of "non-resonant" titanium target.

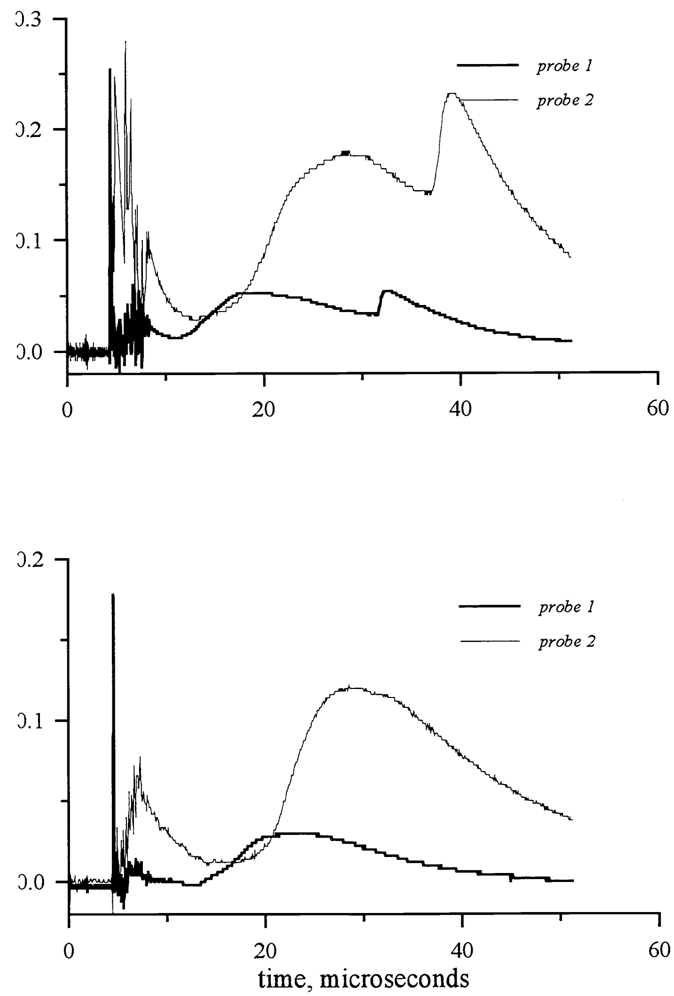


Figure 7: Langmuir probe signals for TaTi (top) and Ti targets when the clouds were irradiated with KrF laser radiation at 1.5 μs after ruby laser pulse.

Both spectral line monitor and dissector show appearence of continuous radiation and many emission lines after resonance irradiation of a dense tantalum-contained cloud. These phenomena testify that the laser energy is transferred to thermal energy of the cloud. The signals of Langmuir probes (Fig. 7) placed at the distances of 3.5 and 7 cm from the target show appearance of additional ion peak in a case of tantalum-contained cloud. This means that there is a mechanism of the cloud ionization which is a consequence of saturation of the resonance level. We have to take in our account two mechanisms. The first one is ionization by CATRION (LIBORS) process. The second process is simple photoionization from the resonance level. To distinguish what the mechanism is actually more important for the cloud ionization, it is needed to carry out more detailed experiments which now are in progress.

One can suppose that to grow up the efficiency of vapor ionization we have to decrease KrF laser power density to saturation parameter close to unity. In this case photoionization will be important to produce the seed electrons and the bulk gas will be ionized by LIBORS process.

4. SUMMARY

At the CATRION setup a number of diagnostics to study laser evaporation and photo-resonance excitation were developed and tested experimentally. Investigation of laser evaporation of mono- and two-component targets has been performed. It has been shown that for all the targets it is possible to obtain neutral vapor practically without electron component. Experiments on photo-resonance ionization of mono- and multi-component targets are started. The effect of KrF laser radiation on the "resonant" cloud has been detected. The next stage of the experiments has to be directed to more detail study above mentioned processes and determination of an optimum conditions for catalytic resonance ionization. A wide variety of two-component targets with different elemental composition and a few additional diagnostics are planned to be studied in these experiments.

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