Resonance radiation plasma (photoresonance plasma)

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A plasma formed by the action on a gas of monochromatic radiation whose frequency corresponds to the energy of a resonance transition in the atom is studied. The elementary methods of creating and studying a plasma of this type are analyzed. The kinetics of formation of a photoresonance plasma is studied, including collision processes with participation of excited atoms leading to formation of molecular ions and highly excited atoms, processes of stepwise ionization and triple recombination, and radiative processes. A photoresonance plasma is characterized by a high electron density with a relatively low electron temperature; for this reason the condition of ideality is more easily violated in a plasma of this type. Some ways of utilizing a photoresonance plasma are presented.

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1. INTRODUCTION

One of the methods of creating a plasma involves the action of optical resonance radiation on a gas. This method was first realized by Mohler and Boeckner,[1] who observed the formation of ions upon irradiating cesium vapor with resonance radiation. Thus they established the possible occurrence in the gas of the process of associative ionization, in which an electron and a molecular ion are formed by collision of excited and unexcited atoms, so that the energy needed for ionizing the atom is released through formation of a molecular ion. Studies of photoresonance plasmas (PRPs) began with the study of Morgulis, Korchevoĭ, and Przhonskiĭ[2] in 1967. By illuminating cesium vapor with resonance radiation to obtain a gas with a high concentration of excited atoms, they found as a result that a plasma is formed with a rather high concentration of charged particles. Since the ionization energy of the cesium atom (3.89 eV) exceeds by more than twofold the energy of a resonance photon (1.39 or 1.45 eV), this result indicated a complex, multistep character of the kinetics of the ionization of cesium atoms under the conditions studied. The subsequent detailed studies of this kinetics[3–5] have permitted obtaining rich information on the mechanisms and rates of processes involving excited atoms.

The formation of a photoresonance plasma is accompanied by various phenomena that occur in gases. Thus, the ionization of a gas under the action of resonance optical radiation is one of the fundamental mechanisms of formation of an ionization wave in the gas, which propagates upon applying an external electric field.[6] This same mechanism plays the decisive role in the phenomenon of ionization of a gas ahead of the front of a strong shock wave in the gas.[7] Irradiation of a gas with optical resonance radiation is used as one of the methods of preliminary ionization of the active medium of high-pressure molecular lasers.[3] This enables one to create a plasma homogeneous throughout the volume, while avoiding the factors that favor the development of instabilities and spatial inhomogeneities of the active medium.[3] The stated method of creating a high-density plasma homogeneous throughout the volume has attracted the attention of investigators also in connection with the problem of heating thermonuclear targets with beams of light ions.[8] In this case the ionization of the gas under the action of resonance radiation enables one to create for a short time an extended plasma channel, which serves for transport of the ion beam to the target, while hindering electrostatic repulsion of the ions.[9]

The potentialities of study of photoresonance plasmas, as well as the set of their applications, have been expanded by the invention of frequency-tunable lasers. On the one hand, this has enabled considerable increase in the fluxes of resonance radiation transmitted through the gas, and on the other hand, study of the processes that occur upon optical excitation of various states of the atom. The photoresonance plasma formed by using tunable lasers is used as a nonlinear element in frequency transformation of laser radiation,[10] as a source of ions of a given type,[11–13] etc.

The set of phenomena that occur in a photoresonance plasma is closely bordered by the optogalvanic effect, which

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consists in a change in the electrical properties of a gas-discharge plasma or flame (e.g., the volt-ampere characteristics) when acted on by optical resonance radiation.[14] The optogalvanic effect is used for determining trace impurities of elements in a gas, in studying the mechanisms of elementary processes in a gas-discharge plasma, in controlling the parameters of a gas-discharge plasma for transmission and processing of information, and as a detection method in laser spectroscopy with superhigh sensitivity.

The process of multistep ionization of atoms opens up broad possibilities. The use for this purpose simultaneously of several frequency-tunable lasers enables one to transfer an appreciable number of atoms of a certain type to a given highly excited state. This technique, usually based on using atomic beams, is applied for detecting individual atoms, for detecting submillimeter radiation, for generating coherent radiation in the UHF range (maser), and in experiments on laser isotope separation.[15] The identification of highly excited atoms is performed by ionizing them in an external electric field. Here one uses the sharp dependence of the ionization probability in a field of given intensity on the effective value n^* of the principal quantum number of a highly excited atom. The ions formed by ionization are extracted from the system by applied fields.

The properties and specifics of a photoresonance plasma are associated with the processes that occur in them. Thus a photoresonance plasma whose properties are determined by elementary collision-radiation processes, is naturally distinguished from a laser plasma, in which the transformation of the energy of the laser radiation into the energy of plasma particles results from the excitation of collective motions in the plasma.

At the same time it seems natural to classify as a photoresonance plasma one formed by the action on a gas of radiation having a frequency that does not necessarily correspond to a resonance transition, but also to transitions between ground and highly excited states, or transitions between two excited states. In all objects of this type, the fundamental ionization mechanism is collisional processes involving excited atoms (more rarely—molecules).

This review will analyze the current status of studies of photoresonance plasmas, information will be presented on the properties and parameters of this object, and problems will be discussed involving the application of photoresonance plasmas in the technique of physical experimentation and in applied fields.

2. TYPES OF PHOTORESONANCE PLASMAS AND METHODS OF PREPARING THEM

2.1. Photoresonance non-laser plasmas

The most convenient method of obtaining a photoresonance plasma not involving the use of laser radiation consists in irradiating a gaseous substance with a gas-discharge lamp filled with the same substance. Here the parameters of the photoresonance plasma are determined by the intensity of the resonance radiation emitted by the lamp. The most interesting results have been obtained in cases in which the lamp is characterized by a high coefficient of transformation of electrical energy into energy of resonance radiation. Thus, in the pioneer study,[1] a quasistationary plasma with an electron density $N_e \sim 10^{12}$ cm^{-3} and an electron temperature $T_e \sim 10^3$ K was formed upon irradiating Cs vapor at a pressure of 10^{-2}–10^{-1} Torr with a cesium gas-discharge lamp. The radiation of the lamp corresponded to the spectral range $\lambda \geqslant 600$ nm. The bulk of the energy of the radiation of the lamp was contained in the lines at $\lambda = 894.3$ and 852.1 nm, corresponding to the 6^2S–$6\,^2P$ resonance transitions. A detailed mass-spectrometric analysis showed[1] that the main type of ions in this plasma is the atomic ion Cs^+. This indicates a complex character of the kinetics of ionization in the photoresonance cesium plasma; in particular, this implies that the process of associative ionization in the collision of two resonance-excited Cs atoms (6^2P) is not the fundamental ionization channel.[1] As is implied by the results of detailed experimental studies of recent years,[3–5] the complex kinetics of ionization of atoms in the cesium photoresonance plasma includes processes of collision of two resonance-excited atoms,

$$2\mathrm{Cs}\,(6\,^2\mathrm{P}) \rightarrow \mathrm{Cs}\,(6\,^2\mathrm{S}) + \mathrm{Cs}\,(8\,^2\mathrm{P}), \qquad (2.1)$$

processes of quenching of the excited Cs atoms (6^2P, 8^2P) by electron impact, processes of ionization of excited atoms in collisions with fast electrons formed as a result of quenching, processes of associative ionization, etc.

As the source of optical radiation for creating the photoresonance cesium plasma, not only cesium lamps, but also helium gas-discharge lamps have been successfully employed. This possibility arises from the coincidence of the wavelength of one of the effective transitions in the spectrum of He ($\lambda = 388.8$ nm) with the wavelength of the 6^2S–8^2P transition of the cesium atom. We note that this coincidence is the basis of one of the first schemes for excitation of a gas laser with optical pumping, which was proposed by F. A. Butaeva and V. A. Fabrikant[16] and realized experimentally in Ref. 17. Using this scheme, a photoresonance cesium plasma was obtained with the parameters $N_{8^2P} \approx 10^7$ cm^{-3}, $N_e \sim 10^8$–10^9 cm^{-3}, $T_e \sim 0.3$ eV, $P_{Cs} \sim 10^{-3}$–10^{-2} Torr.[18] In a plasma of this type, processes of stepwise excitation of highly excited levels from the 8^2P level by electron impact play an essential role. Figure 1 shows the dependences of the parameters of this plasma on the density of the Cs vapor with fixed intensity of resonance irradiation.

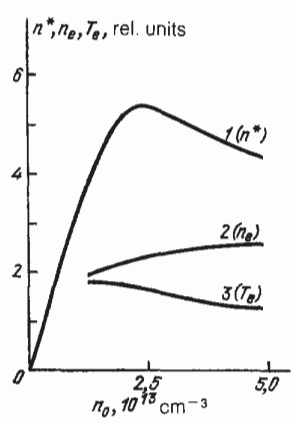

FIG. 1. Dependence of the parameters of a cesium plasma on the density of Cs vapor at fixed intensity of resonance irradiation.[18] *1*—density n^* of excited atoms (Cs, 8^2P); *2*—density n_e of electrons; *3*—temperature T_e of electrons.

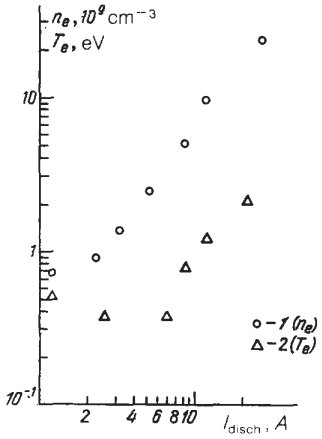

FIG. 2. Dependence of the concentration (*1*) and temperature (*2*) of the electrons of a photoresonance plasma in Hg vapor on the value of the discharge current in the optical-excitation source.[20]

Mercury-vapor lamps are also an intense source of resonance radiation, and have been successfully used to create a photoresonance plasma in mercury vapor.[19,20] Investigations in this direction were stimulated by practical problems of optical separation of mercury isotopes.[21] Upon irradiating a gas-discharge mercury lamp with the resonance line corresponding to the transition $Hg(6^3P_1^0 \to 6^1S_0)$ ($\lambda = 253.7$ nm), an increased concentration and a reduced temperature of the electrons was observed. This was associated with an increase in the efficiency of ionization under the action of the resonance radiation. The mercury lamp used as the source of resonance radiation had the form of a jacket arranged coaxially around the cylindrical cell being irradiated.[20] With a pressure of mercury vapor in the cell of ~ 0.05 Torr, the concentration of Hg atoms in the $6^3P_1^0$ state reached 10^{11} cm^{-3}. The electrical characteristics of the photoplasma that was formed are shown in Fig. 2. Owing to the coaxial excitation geometry used in this experiment, a high degree of homogeneity of the photoresonance plasma was attained.

An interesting variety of photoresonance plasma was realized in Ref. 22, where a mixture of Hg and Cs vapors was irradiated with the resonance radiation of a mercury lamp ($\lambda = 253.7$ nm). The ions were formed by the Penning reaction

$$Hg\,(6\,^3P_1) + Cs \to Hg + Cs^+ + e. \qquad (2.2)$$

According to the measurements performed using probe and UHF diagnostics, at a concentration of $Cs \sim 3\times10^{15}$ cm^{-3}, $Hg \sim 3\times10^{13}$ cm^{-3}, and a pressure of buffer gas (Ar) ~ 100 Torr, the photoresonance plasma was characterized by a density $N_e \sim 10^{12}$ cm^{-3} and a temperature $T_e \approx 2000$ K. The role of the buffer gas consists in reducing the effectiveness of the diffusion losses of charged particles, and hence, in maintaining the density of the electrons of the photoresonance plasma at a sufficiently high level. An analogous scheme for creating a photoresonance plasma was realized in Ref. 23, where a mixture of Cd and Cs vapors was irradiated with the resonance light of a cadmium lamp.

2.2. Photoresonance laser plasmas

The invention and wide spread of frequency-tunable narrow-band lasers based on dyes has stimulated to a considerable degree the study of the properties and possible applications of photoresonance plasmas. The set of studied objects has considerably expanded to encompass all the alkali metals, and also a number of metals of the second and third groups of the periodic table. The object of the studies was the mechanisms of ionization and recombination of particles of a plasma, the elucidation of the role of the buffer gas, the possibility of more complete extraction of ions in a photoresonance plasma and identifying them, etc.

Among the large number of experimental studies (see the review of the early studies[96]) on the creation and study of photoresonance laser plasmas, primary attention is owed to a series of publications reporting the practically 100% ionization of metal vapors irradiated with the resonance radiation of a pulsed laser of relatively low power. Figure 3 shows a diagram of the first of these experiments, which was performed by Lucatorto and McIlrath.[24] The radiation of a dye laser pumped with a flash lamp was tuned to a line at $\lambda = 589.6$ nm, which corresponds to the $3^2S_{1/2}$–$3^2P_{1/2}$ transition of the Na atom, and was focused on a 10-cm column of Na vapor with addition of He to a total pressure about 1 Torr. The pulses of laser radiation of duration 500 ns had an energy of 300 mJ, which corresponds to a pulse power of 0.6 MW. The degree of ionization of the vapor was determined with a vacuum-ultraviolet spectrograph, which enabled measuring the absorption coefficient in the region $\lambda = 15$–42 nm. Figure 4 shows typical densitograms of the spectrum obtained without (a) and with (b) laser irradiation. As is shown by comparison of the absorption coefficients in the region of $\lambda \approx 32.21$ nm corresponding to transitions of the Na^+ ion, the degree of ionization of Na during the laser pulse reaches 100%. The practically complete ionization of the Na vapor is also indicated by the sharp (by a factor of

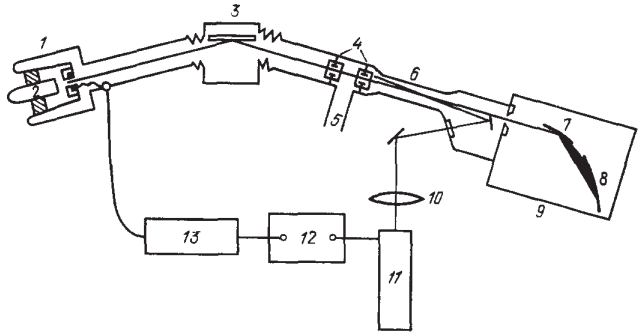

FIG. 3. Diagram of an experiment to produce and study a photoresonance sodium plasma with a high degree of ionization. *1*—radiation source with a continuous spectrum; *2*—anode; *3*—toroidal mirror; *4*—capillary rings; *5*—vacuum pump; *6*—furnace; *7*—three-meter reflecting spectrograph; *8*—diffraction grating; *9*—photoplate; *10*—cylindrical lens; *11*—laser; *12*—delay generator; *13*—pulse shaper.

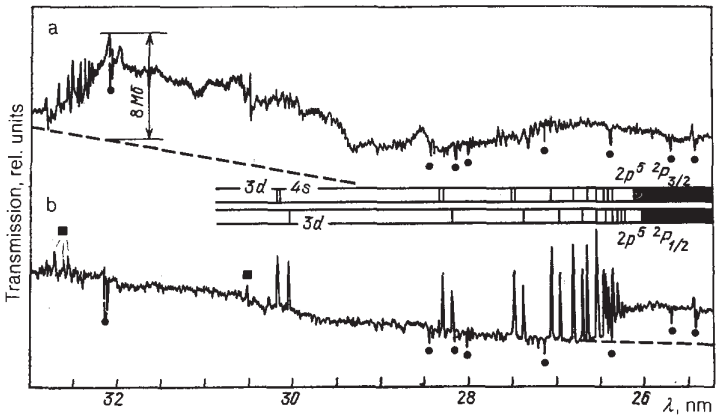

FIG. 4. Densitograms of the absorption spectrum of sodium.[24] a—Absorption of sodium vapor without irradiation; the dots indicate the emission lines of the vacuum arc, and the dotted line the absorption in He. b—Absorption of Na vapor irradiated with resonance laser radiation. The solid squares indicate the absorption lines of neutral Na.

10^6) decline in the absorption coefficient of the resonance laser radiation owing to the formation of the photoresonance plasma. Even the first rough estimates of the authors[24] indicated a complex, multistep mechanism of ionization of the vapor in the described experiments. Neither three-photon ionization nor radiation collision

$$2\mathrm{Na}^*(3p) + \hbar\omega \rightarrow \mathrm{Na}^+(2p^6) + \mathrm{Na}(3s) + e, \qquad (2.3a)$$

nor multistep collisional excitation

$$2\mathrm{Na}(3p) \rightarrow \mathrm{Na}(5s) + \mathrm{Na}(3s) \qquad (2.3b)$$

with subsequent photoionization of the excited Na (5s) atoms possess values of the rate constants high enough to explain the observed 100% ionization of the atoms.

Analogous results were obtained in Ref. 25, where a cell filled with a mixture of Na vapor and Ar at a pressure of several Torr was irradiated with pulses of radiation of a dye laser based on rhodamine 6G pumped with a nitrogen laser. The radiation pulses of 100 kW power had a duration of 10 ns and a line width ~0.1 nm. The ionization of the gas was measured by the magnitude of the photocurrent, for which a typical dependence on the Na vapor density is shown in Fig. 5. Figure 6 shows the dependence of the electron temperature on the time elapsed since the end of the laser pulse, as obtained by the Langmuir double-probe method. The results of the probe measurements of the electron density performed at a point separated by 2 mm from the focus of the laser beam are shown in Table I.

The effect described above of intense ionization of vapors under the action of resonance laser radiation has been observed in subsequent experiments with vapors of Li,[26] Cs,[27] Ca,[28], Sr,[29] Ba,[28–34] Na,[35] and Mg.[36] The properties of the PRP formed thereby have been studied in greatest detail in Ref. 36, where Mg vapor was irradiated with radiation corresponding to the resonance transition $3^1S_0 \rightarrow 3^1P_1$. ($\lambda = 285.2$ nm) of the singlet system of levels. A pulsed liquid dye laser with frequency doubling pumped with the second harmonic of a neodymium laser with a pulse repetition frequency of 3 Hz was used as the radiation source. The pulses of ultraviolet radiation had a line width ~0.1 cm^{-1}, and duration ~10 ns at a power ~1 kW. Another liquid laser ($\lambda = 280.3$ nm) tuned to the transition $3^2P_{1/2}$–$3^2S_{1/2}$ of the Mg^+ ion was used to measure the concentration of Mg^+ ions. In addition, the experiment measured the luminescence of the Mg vapor and the time-dependence of the photocurrent. The pulse of the probe radiation had a delay with respect to the pump pulse variable in the range up to 100 μs. Figure 7 shows a diagram of the experiment.[36]

The experimental chamber, which was made of stainless steel and fitted with quartz windows and internal plane electrodes to measure the photocurrent, was filled with an inert gas at a pressure ~1 Torr. The pressure of metal vapor (Mg) was varied in the range 0.1–1.0 Torr. Upon tuning the pump radiation to the frequency of the resonance transition

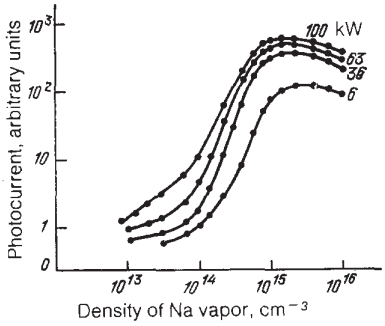

FIG. 5. Dependences of the photocurrent on the density of Na vapor obtained at different levels of laser power.[25]

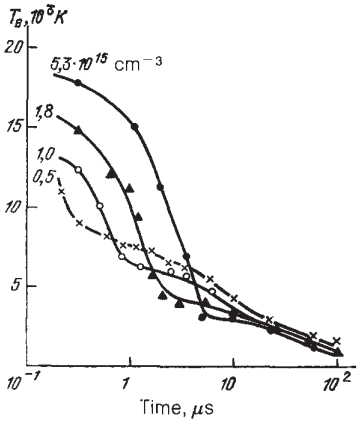

FIG. 6. Dependences of the electron temperature on the time elapsed after cessation of the laser radiation pulse.[25] The curves are marked with the different values of the concentration of Na vapor.

TABLE I. Values of the electron density N_e in a photoresonance plasma measured at different values of the density of Na vapor.[25]

N_{Na}, cm^{-3}	$5{,}9\cdot10^{14}$	$1{,}0\cdot10^{15}$	$1{,}8\cdot10^{15}$	$5{,}3\cdot10^{15}$
N_e, cm^{-3}	$6{,}6\cdot10^{12}$	$9{,}5\cdot10^{12}$	$1{,}0\cdot10^{13}$	$7{,}0\cdot10^{12}$

of Mg, an emission from the Mg vapor arose in the region of the laser-beam focus at frequencies corresponding to transitions $n\,^1D_2$–$3\,^1P_1$ ($n = 4$–10) of the singlet system and $n\,^3S$–$3\,^3P$ ($n = 4, 5$) of the triplet system of Mg levels. Here the absence was noted of radiation at the strong line $5\,^1S_0$–$3\,^1P_1$ ($\lambda = 571.1$ nm) and several other strong lines. As was shown by measurements of the density of Mg ions performed with probe radiation, the maximum concentration of ions ($\sim 2\times10^{14}$ cm^{-3}) was observed about 30 ns after the end of the pulse of pump radiation, while the total time of existence of the PRP amounted to $\sim 10\,\mu$s. The maximum degree of ionization of the plasma reached 5%. The absence of saturation in the dependence of the ion density on the intensity of pump radiation allows one to expect an increase in the PRP density upon using more intense resonance pump radiation.

Another detailed study worthy of attention on the character of formation and physical properties of a PRP with a rather high degree of ionization was performed by the authors of Ref. 35, where a dye laser (rhodamine C) was used as the source of resonance radiation. It was pumped with the radiation of the second harmonic of a solid-state pulsed laser based on yttrium aluminum garnet of type LTIP4-5 with a pulse-repetition frequency of 12.5 Hz. To narrow and tune smoothly the emission line of the dye laser, a diffraction grating (1200 lines/mm) was used and was set up in a glancing-incidence system. The width of the emission line of this laser amounted to ~ 1 nm, and the emission power was ≈ 40 kW at a pulse duration of $\sim 10^{-8}$ s. The laser was tuned either to the resonance transition of Na ($\lambda = 589.0$ nm) or to the wavelength 578.7 nm corresponding to two-photon absorption to the excited state of Na ($4d\,^2D_{5/2}$).

The Na vapor diluted with inert gases filled the discharge tube, which was made of Pyrex and had niobium tubular electrodes. The pressure of the vapor in the tube was maintained by using a heating element. Under the conditions

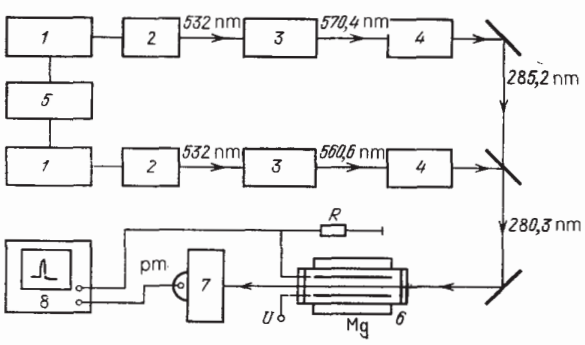

FIG. 7. Diagram of an experimental arrangement to form a quasiresonance laser plasma in Mg vapor.[36] *1*—neodymium garnet laser; *2, 4*—frequency doublers based on KDP; *3*—dye laser; *5*—delay circuit; *6*—cell with Mg vapor; *7*—monochromator with photomultiplier (PM); *8*—oscillograph.

of the experiment one pulse of laser radiation contained about 5×10^{14} photons. About the same number of Na atoms was contained in the irradiated volume. Here conditions were selected such that, on the one hand, practically complete absorption of the laser radiation was attained, and on the other hand, the intensity of absorption varied weakly along the laser beam. This was made possible by detuning the frequency of the laser beam from the center of the absorption line by four widths of the Doppler absorption contour (equal to 0.0024 nm). Whereas at the center of the absorption line the optical density of the medium amounted to $\sim 10^6$, at the detuned frequency it was close to unity.

The formation of a PRP was measured from the change in electrical conductivity of the irradiated volume. To do this, a voltage was applied to the electrodes of the discharge tube smaller than the excitation voltage of the discharge, and the current was measured that arose in the electrical circuit under the action of the laser illumination. Figure 8 shows an oscillogram of this current obtained at a voltage on the electrodes of 100 V and a vapor pressure of Na of 3×10^{-3} Torr. The pressure of the buffer gas amounted to 1 Torr. As the pressure of Na vapor was increased from 3×10^{-4} Torr to 0.2 Torr, the magnitude of the signal and its duration increased by more than an order of magnitude. Upon detuning from the resonance at $\lambda = 589$ nm, no plasma formation was observed.

Plasma formation was also observed in two-photon laser excitation of the level $4p^2D_{5/2}$. The oscillograms of the current obtained here at a pressure of Na vapor $\sim 5\times10^{-3}$ Torr are shown in Fig. 9. The rapidly growing advance front of the current pulse was associated[35] with the phenomenon of three-photon ionization of atoms via a two-photon resonance, and the subsequent, smoother increase in electrical conductivity—with a supplementary mechanism of formation of free electrons (heating of electrons by superelastic processes and subsequent ionization of atoms by electron impact).

As an analysis of the experiments to create and study a laser PRP shows, a plasma of rather high density is formed using very-low-power laser radiation. This arises from the high absorption power of gases for resonance radiation, and also the high efficiency of conversion of the energy of resonance-excited atoms into ionization energy.

2.3. Quasiresonance plasmas

As has been established in a number of experiments of recent years,[13,29,37] to form a photoresonance plasma one need not use radiation whose frequency corresponds to a resonance transition between the ground and excited states of the atom. Efficient ionization of the atoms of a metal vapor has also been observed using radiation corresponding to a transition between two excited states of the atom. Here the intensity of the laser radiation was not so great that one

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