Photoresonant plasma produced in an open discharge in a metal vapor

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A pulsed open discharge produces a photoresonant plasma outside the discharge channel. In this paper some results of the studies of photoresonant plasma in a Zn–He mixture are described. Oscillograms of radiation pulses from an open discharge zone and a neighboring medium are compared. The peaked structure is detected in radiation pulses at some lines. The important processes resulting in the population inversion and ionization of metal atoms are discussed.

Introduction

The resonance optical pumping (ROP) is one of the first methods for producing active media of gas lasers. In some practically important cases ROP results in ionization of a gas medium and appearance of the photoresonant plasma (PhRP), which, in turn, can serve as an active medium for a laser, as the gas discharge plasma itself. The PhRP attracts attention of investigators due to its unique properties and possibilities of using these properties in plasma chemistry, quantum electronics, and spectroscopy.[1,2] It is characterized by low temperature of electrons at high population of levels, as well as by the absence of instabilities, and significantly nonequilibrium distribution over the energy levels.[2]

The photoresonant plasma with relatively high density was first studied in Ref. 3. Cesium vapor was pumped by a resonant cesium lamp, that is, multiline resonance optical pump was used. At a medium and high pressure, as well as with gases or vapor possessing a high ionization potential, production of the PhRP faces some problems. (Pumping of the lead vapor with the emission from the resonance lead lines of a pulsed quartz lamp was studied in Ref. 4.) The wall of such a lamp often does not transmit radiation in the UV and VUV spectral regions. These regions include the resonance lines which can be absorbed by atoms in the ground state and thus pump the high excited levels near the ionization limit and above it. In spite of the low intensity of these lines these, in some cases, can in our opinion play an important part in the formation of the photoresonant plasma.

For radiation of the lines in the VUV region to be not absorbed by the wall of a discharge chamber, the pulsed open gas discharge (OGD) was used in formation of the PhRP.[5] The capabilities of a pulsed open gas discharge were studied as applied to the production of a photoresonant plasma in the mixtures of noble gases and a noble gas + hydrogen. In Ref. 6 the concentration of electrons in the photoresonant plasma produced using resonance optical pumping by high-power pulsed OGD was measured. It appeared to be about 10^{15} cm^{-3}. The use of a photoresonant plasma for producing active laser media was proposed in Ref. 2. Note that the OGD pumps the medium within the whole spectrum from IR to VUV. The radiation at VUV lines can excite levels near the ionization limit and above it. At high population of the low states, the absorption from them can play an important role in the energy balance of the PhRP. Disadvantages of the OGD for the resonance optical pumping of a gas and a vapor–gas medium in order to produce the photoresonant plasma and laser active media (LAM) are its hard initiation at high gas pressure and problems associated with the control over the discharge parameters.

In this paper we consider a PhRP in the mixture of zinc vapor with helium. The PhRP is produced by irradiation of a medium placed inside a chamber with the resonance radiation from an open gas discharge.

Gas-discharge and measuring instrumentation

The experimental setup used in the experiments consisted of a plasma source, power supply, and a recording unit. The plasma source is shown in Fig. 1. A quartz chamber (a tube with the inner diameter of 35 mm and length of 960 mm) was housed in a furnace (not shown in the figure). The tube ends were closed with quartz windows. The hot zone within the furnace was 320 mm long. Electrodes *1* and *2* were spaced at 420 mm distance. The electrode *1* was bent for convenient insertion of the quartz chamber into the furnace. The electrode *2* served as a cathode. The

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temperature in the furnace was measured with a chromel–alumel thermocouple. An auxiliary electrode *3* connected to cathode *2*, as in Ref. 7, was kept close against the outer wall of the tube along the line connecting the electrodes *1* and *2*. The outer auxiliary electrode produced pre-breakdown ionization, and the principal discharge current followed that way after the breakdown, not propagating laterally and not bending. Two types of the outer electrodes were tested: copper foil strip and round steel bar. Zinc in tantalum boats *4* (liquid zinc can damage quartz) was put in a quartz tube. The buffer gas (helium) pressure was chosen after preliminary experiments to be 100 Torr. The zinc vapor pressure in the chamber could be varied from 0.01 to 1 Torr and higher with the increase of temperature inside the furnace.

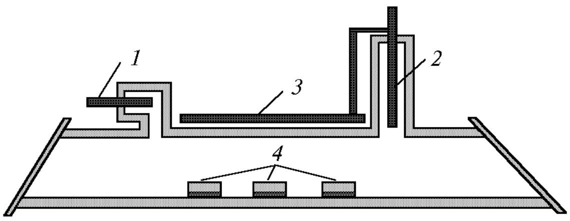

Fig. 1. Geometry of the gas-discharge tube for producing the photoresonant plasma.

The pulsed power supply of the gas-discharge tube (GDT) consisted of a high-voltage unit and a discharge circuitry with a 0.1 μF energy storage capacitor. The discharge circuit was switched with a RU–62 spark gap in the self-breakdown mode at the voltage of 11 kV. If necessary, two series-connected RU–62 spark gaps were used. In the latter case the self-breakdown voltage was 22 kV.

The oscillograms of current pulses and of the spectral lines recorded have then been studied. The GDT emission spectrum was recorded with a monochromator, PMT, and oscilloscope. The diffraction orders needed were isolated with the corresponding filters.

Experimental results and discussion

In our experiments the discharge filament of the open gas discharge was far less in diameter than the quartz tube. To produce the PhRP and an active medium based using it, the discharge filament was being localized near the wall along the line connecting the electrodes in such a way that it could irradiate the rest volume inside the tube by the resonance UV and VUV radiation capable of exciting the upper levels, including the shifted ones lying above the ionization limit. To prevent propagation of the discharge current across the volume of the quartz chamber, we used the property of a high-power discharge filament to self-contract. As was noted in Ref. 7, at the same breakdown field there exist both the current zone (filament) and currentless environment. In our setup we tried to minimize the longitudinal field at some distance from the filament. Naturally, as the voltage is applied across the electrodes prior to the main discharge, the discharge of the corona type always arises in the chamber volume and produces weak pre-ionization.

Usually, in a large-diameter tube at a medium or high pressure the discharge filament is formed, and it does not necessarily follow a straight line, but curves and shifts thus producing inhomogeneities in plasma. To eliminate these phenomena, a copper foil strip connected with the cathode was placed between the electrodes outside the discharge chamber, as it was done in Ref. 5 when dealing with inert gases. However, (maybe, because of the tube diameter was larger than that in Ref. 5) the quality of the discharge filament was insufficiently good, and its shape was irregular when observed from the end. Upon the replacement of the foil with a steel bar, we obtained the best results. In this case the discharge evenly crept over the upper wall of the tube along the straight line. The pulsed discharge filament had the cross size within 5 to 8 mm. Such a design helps, to some extent, to localize the pre-breakdown electric field and the corona discharge it induces in the area of the electrode system.

The OGD current measured with a Rogovskii belt proved to be a decaying sinusoid with the first half-period of 3 μs. Under the exposure of the chamber space beyond the discharge filament to resonance radiation from the OGD (including VUV radiation), PhRP is formed as is evidenced by the presence of zinc ion spectral lines (in particular, at 491.1 and 492.4 nm). The duration of emission at these lines in the zone of localization of the pulsed discharge is far longer (about 60 μs) than that in the zone irradiated by the OGD (about 4–5 μs). Evidently it was for the first time that the discharge in metal vapor with such properties had been realized.

The study of total radiation from all zones of the tube has shown that the shapes of the emission oscillograms at different spectral lines differ drastically. Figure 2 shows the oscillograms of the emission for spectral lines of zinc atoms and ions, all the conditions being the same. The oscillograms of zinc atomic lines have a pronounced peaked structure, whereas no such a structure is observed in the oscillograms of ion emission lines. The oscillograms of the helium lines also have no this structure. Peaks in the emission oscillograms of the zinc atomic lines correlate with the half-periods of the discharge current, however the mechanism of excitation of these lines requires further investigations. At the low zinc vapor pressure (below 0.1 Torr) in the chamber beyond the discharge filament zone, the first peak is most intense. As the zinc vapor pressure increases (above 0.1 Torr), the intensity of the first peak decreases as compared with the second and even third peak. Closer to the discharge zone, the amplitude of the first peak further decreases with respect to the second and third peaks.

The singlet lines of zinc atoms are far weaker than the triplet ones, although they can be excited optically at absorption from the ground state with a higher probability.

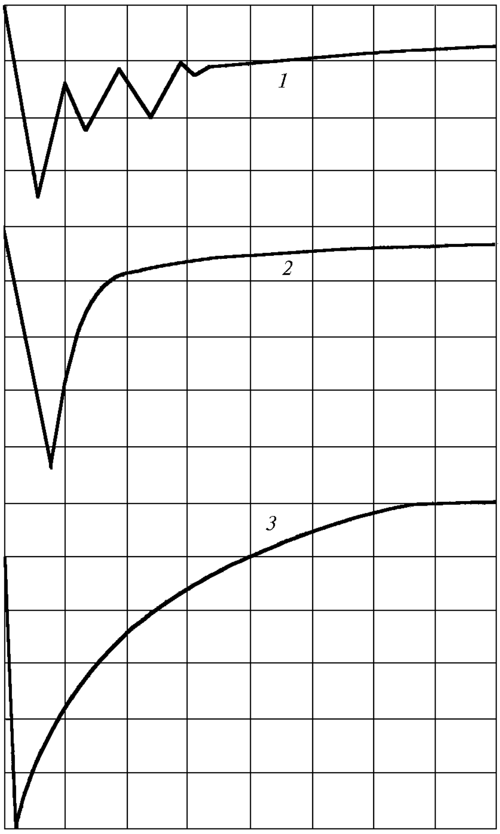

Fig. 2. Oscillograms of the radiation pulses at the following spectral lines: λ = 468.15 nm (Zn I), 2 μs/mark (total duration 25 μs) (curve *1*); λ = 491.15 nm (Zn II), 2 μs/mark (total duration 60 μs) (curve *2*); λ = 587.6 nm (He I), 10 μs/mark (curve *3*).

The oscillograms of zinc atomic lines at the same wavelength but from different chamber zones are qualitatively different. Figure 3 shows such oscillograms recorded through a 3-mm-wide slit diaphragm (the wavelength at 468 nm). The smaller value of the first peak in comparison with the rest ones is most likely caused by the prevalence of stepwise processes in the excitation of zinc atoms, although "depletion" (that is, almost complete ionization) of zinc atoms under the effect of the exciting pulse, which is strongest in the first half-period of current, also cannot be excluded.

The qualitative differences in the oscillograms of radiation pulses of zinc and helium as, in particular, the absence of peaked structure in the oscillograms of helium lines, indicates that the direct or indirect role of helium levels in populating of the levels of zinc atoms is insignificant.

In the area of the discharge filament, the long (up to 50 μs and longer) weak emission at the spectral lines of zinc ions (weak emission at the lines of the zinc atom lasts no longer than 25 μs) after an intense leading edge of the pulse is most likely caused by the processes of collisions among helium ions and zinc atoms that are followed by relaxation.

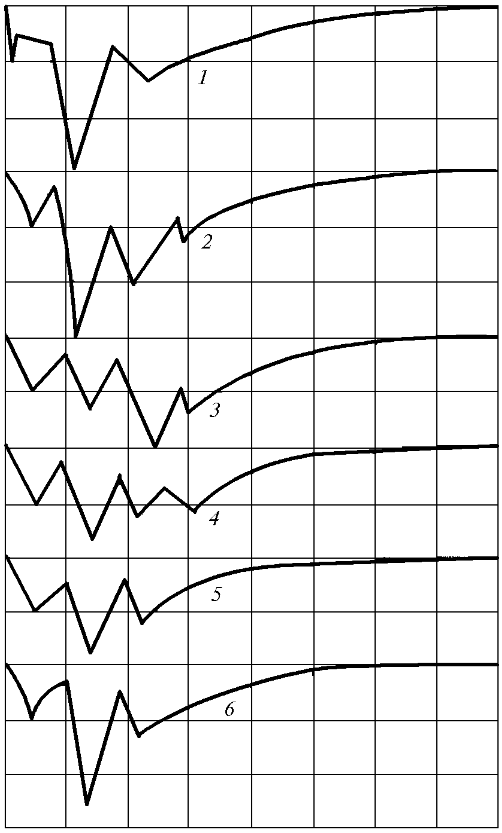

Fig. 3. Oscillograms of radiation at the line λ = 468.15 nm (Zn I) from different zones of the chamber: discharge channel (*1*); 2 (*2*), 7 (*3*), 12 (*4*), 18 (*5*), and 22 mm (*6*) far from the discharge channel.

Let us list some important processes resulting in the ionization and PhRP formation in the mixture of zinc vapor and helium, as well as excitation of the zinc atom.

I. Primary direct processes

Absorption of resonance radiation with the transition:

(a) to the low-lying states

$$\text{Zn} + h\nu \Leftrightarrow \text{Zn}^*; \tag{1}$$

(b) to the states lying below the ionization limit by the value of the electron temperature:

$$\text{Zn} + h\nu \Leftrightarrow \text{Zn}^{**}; \tag{2}$$

(c) to the shifted states lying above the ionization limit:

$$\text{Zn} + h\nu \Leftrightarrow \text{Zn}^{***}. \tag{3}$$

II. Stepwise processes

Absorption of the resonance radiation from the low-lying highly populated levels:

(a) with the transition to the states near the ionization limit

$$Zn^* + h\nu \Leftrightarrow Zn^{**}; \quad (4)$$

(b) with the transition to the shifted states above the ionization limit

$$Zn^* + h\nu \Leftrightarrow Zn^{***}. \quad (5)$$

III. Cascade processes

$$Zn^{**} \Leftrightarrow Zn^* + h\nu, \quad (6)$$

$$Zn^* \Leftrightarrow Zn + h\nu. \quad (7)$$

IV. Collisions of the second kind

$$Zn^* + e \Leftrightarrow Zn + e + \Delta W, \quad (8)$$

$$.Zn^{**} + e \Leftrightarrow Zn^* + e + \Delta W. \quad (9)$$

V. Ionization due to absorption of continuous-radiation from the levels lying lower the ionization limit by the value of the electron temperature

$$Zn^{**} + h\nu \Leftrightarrow Zn^+ + e. \quad (10)$$

VI. Autoionization from the shifted levels lying above the ionization limit of zinc atoms

$$Zn^{***} \Leftrightarrow Zn^+ + e. \quad (11)$$

VII. Collisions leading to ionization

$$Zn^{**} + e \Leftrightarrow Zn^+ + e + e, \quad (12)$$

$$Zn^* + Zn^* \Leftrightarrow Zn^+ + Zn + e, \quad (13)$$

$$He_m + Zn \Leftrightarrow Zn^{+*} + e + He, \quad (14)$$

$$He_{res} + Zn \Leftrightarrow Zn^{+*} + e + He, \quad (15)$$

$$He^+ + Zn \Leftrightarrow Zn^{+*} + He. \quad (16)$$

Here Zn, Zn^*, Zn^{**}, Zn^{***}, Zn^+, and Zn^{+*} are the zinc atom, zinc atom in the low highly populated state, zinc atom near the ionization limit, zinc atom in the shifted state, zinc ion, and zinc ion in the excited state; He, He_m, and He_{res} are the helium atom, helium atom in the metastable state, and helium atom in the resonant state; e is electron; ΔW is the kinetic energy of the electron. In the stationary and quasistationary states of the PhRP every process is balanced by the opposite one.

Conclusions

The process of collision with the excited helium atoms does not play a leading part in populating the emitting levels of zinc atoms within intense peaks of radiation pulses. This is true both for the discharge filament zone and for the rest space of the quartz chamber.

The processes of excitation of zinc atoms in the chamber (at the zinc vapor pressure higher than 0.1 Torr) are of stepwise character. Most processes (stepwise) resulting in the ionization are active in the triplet spectrum.

In the initial period of PhRP formation, direct processes (3) and (11) as well as processes with participation of zinc atomic levels lying below the ionization limit by the value of the electron temperature (to be more precise, by doubled or tripled values of the electron temperature) are of great importance. Then, as charged and excited particles are accumulated in the PhRP, the stepwise processes with the participation of the low-lying highly populated states become dominating.

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