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NATURE OF IONIZATION IN A PHOTORESONANT CESIUM PLASMA

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As a continuation of earlier work,[1] the nature of ionization in a photo-resonant cesium plasma is further studied: 1) The optical pumping spectral region active for ionization is elucidated. 2) The properties of the plasma electron gas are determined. 3) Molecular as well as atomic cesium ions are detected in the plasma by the mass-spectrometer technique[6] and the ratio of the ions under various experimental conditions is measured. 4) The dynamics of the development and decay of plasma is measured separately for molecular and atomic ions. Some conclusions are drawn concerning the nature of processes in such a plasma.

IN our preceding communication[1] we described a method of obtaining a cesium plasma of a new "photo-resonant" type. This method consisted of optical pumping, i.e., exposing neutral cesium vapor predominantly to resonant radiation from an external arc-discharge plasma also in cesium vapor. The investigated plasma was produced in a glass detector filled with neutral cesium vapor and equipped with suitable probes. The detector was placed inside the free cavity of a glass Dewar-like discharge device. It was assumed that the mechanism of the ionization and formation of the plasma, observed in the detector in this case, corresponds to the following two-step associative scheme with use of resonant emission of cesium[2,3]: 1) $Cs + h\nu_d \rightarrow Cs^*$, and then 2) $2Cs^* \rightarrow Cs_2^* \rightarrow Cs_2^+ \rightarrow e$. Thus, such a plasma should consist of molecular cesium ions. However, as noted in[1], this conclusion calls for an exhaustive, primarily direct mass-spectrometric, verification.

The present paper is devoted mainly to experimental investigations aimed at determining the nature of the ionization processes in such a plasma. In parallel, we investigated further certain physical properties and determined certain plasma parameters; the results of these investigations are reported elsewhere[4].

We begin with the elucidation of a very important question, namely which region of the spectrum of the external discharge cesium plasma is decisive. To answer this question, a light filter was placed between the discharge plasma and the detector with the cesium vapor; the spectral characteristic of the filter is shown in Fig. 1. The filter has a very sharp transmission boundary at $\lambda = \lambda_0 \approx 5800$ Å, i.e., $h\nu_0 = 2.1$ eV, and when $\lambda < \lambda_0$ its transparency is D = 0, while when $\lambda > \lambda_0$ we have $D \approx 80\% \approx$ const. When a powerful discharge-current pulse is turned on in the radiation source, current is produced in the detector; oscillogram I in Fig. 2 is the ion component of this pulse as measured by the double probe in the detector. When this light filter is replaced by an ordinary transparent glass plate (of the same dimension and in the same position), we obtain oscillogram II, which is perfectly analogous to the preceding one. When the light filter is subsequently replaced by an opaque screen, the result is oscillogram III, which shows no trace of the probe current.

Thus, only the cesium-spectrum region with $\lambda > 5800$ Å is active in our case. When this radiation enters the neutral cesium vapor, the only possible doublet transition is $6S_{1/2} - 6P_{3/2,1/2}$ and is connected with absorption of the resonant doublet with $\lambda = 8521$ and

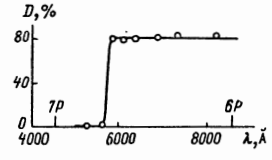

Fig. 1

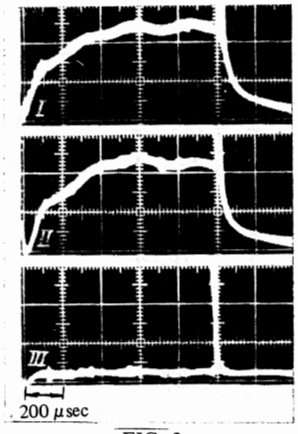

FIG. 2

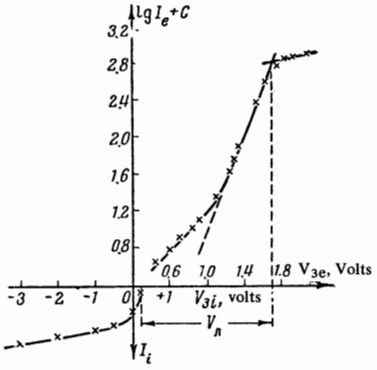

FIG. 3

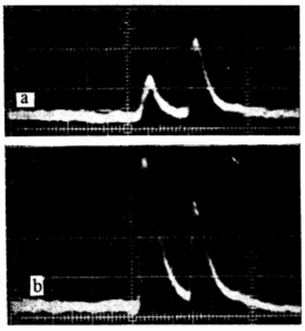

FIG. 4

8944 Å. The excited Cs* atoms obtained in such a way can cause subsequent ionization only by the aforementioned associative method, with formation of molecular cesium ions; we know of no other ionization method under these conditions. One can therefore assume that our ideas concerning the primary mechanism of photoresonant ionization under these conditions are valid.

We now turn to a more detailed clarification of what the plasma produced in the detector actually consists of. A definite answer to this question is provided by the typical probe characteristic shown in Fig. 3. It was obtained with the aid of a very small and thin (20 μ diameter) single cylindrical probe placed on the detector axis and equipped, to close the circuit, with two very large counter probes located at the ends of the detector. It follows from this characteristic that our plasma is non-isothermal and that it contains two groups of electrons.

The first is a large group of slow electrons with a Maxwellian distribution and a temperature $T_e \approx 1800°K$. It is contained in the "trap" produced here by the negatively charged glass detector with potential $V_{tr} \approx -1.4$ V. The strange appearance of such a group of electrons is apparently connected[1] with the numerous acts of energy transfer in collisions of the second kind between the photoexcited cesium atoms and the free electrons produced by the associative ionization ($Cs^* + e \rightarrow Cs + e + \mathscr{E}_e$) and their subsequent "Maxwellization"[5] as a result of Coulomb electron-electron interaction. The number of such collisions is $N_{II} = q_{II} u_e n_e n_a^*$ and is quite large under our conditions; for example, in the typical case of electron density $n_e = 10^{12}$ cm^{-3} and excited-atom density $n_a^* = 10^{13}$ cm^{-3}, it is larger by approximately four orders of magnitude than the number of acts of direct associative ionization, if the cross section of the latter is $q_{ai} = 7 \times 10^{-18}$ cm^2 and $q_{II} = 5 \times 10^{-15}$ cm^2. Simultaneously, the time of "Maxwellization" of these electrons $\tau_c = a\mathscr{E}^{3/2}/n_e \ln\Lambda \approx 10^{-7}$ sec, turns out to be, as expected, smaller than the time of energy transfer to these electrons $\tau_{II} = (q_{II} u_e n_a^*)^{-1} \approx 10^{-6}$ sec.

The second group of faster electrons, the number of which is approximately 1/40 of that in the first, cannot have simultaneously a Maxwellian distribution. It is probably simply a group of electrons with random direction of motion to the cylindrical probe, which acquire in similar collisions of the second kind an excitation energy $\mathscr{E}_e = eV_d = 1.4$ eV, but have not yet time to become completely Maxwellized.

Thus, the resultant plasma turns out to be quite close to the corresponding non-isothermal plasma[5] with very small electric field, produced in a short arc discharge with a cathode potential jump $V_c = V_d$, where probe characteristics of a similar kind can be obtained. On the other hand, the energy acquired by the electrons from its sources after Maxwellization, flows in both cases subsequently through identical and well known channels.

Finally, in accordance with the indication made in[1], we proceed to a direct mass spectrometric analysis of the composition of the ions in the investigated plasma. We used a completely sealed glass time-of-flight laboratory mass spectrometer, developed and successfully used by us in the past[6] for a discharge cesium plasma, with an electron multiplier at the output. The investigated detector, with appropriate probes, constituted a single completely sealed system together with the mass spectrometer. It was located inside the source of the pumped radiation. The latter, as in[1], was a powerful pulsed arc discharge with an incandescent cathode in cesium vapor and in a Dewar-like housing. This discharge pulse had a sharply rectangular form of duration $t_p \approx 1200$ μsec (when we expected to obtain a quasi-stationary state) and a repetition frequency ≈ 1 sec^{-1}. The shutter passing the investigated ions from the detector, where they were produced, into the drift space of the mass spectrometer, could be connected, in adjustable fashion, at different instants of time both during the time of the pulse and after its termination. This made it possible to trace additionally the composition of

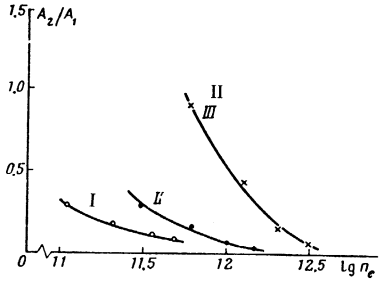

FIG. 5

the ions in the plasma both during the process of its development and during its subsequent decay.

The plasma was investigated at the following cesium-vapor pressures in the detector: p = 0.01 (I), 0.03 (II), and 0.08 (III) mm Hg, and at different electron concentrations in the plasma: $n_e = 1 \times 10^{11} - 3 \times 10^{12}$ cm^{-3}. n_e was varied by changing the discharge current in the external source, i.e., the radiation power.

A typical example of the mass spectrograms obtained in this manner on the screen of the S1-17 oscilloscope is shown in Fig. 4 for a photo-resonant plasma at p = 0.08 mm Hg and $n_e = 7 \times 10^{11}$ cm^{-3} for two instants of time, 200 μsec (a) and 1200 μsec (b) after turning on the external discharge. We see the presence of two signals, indicating the presence in our plasma of ions of two types, the left-hand signal A_1 corresponding to atomic cesium ions, and the right-hand A_2 to molecular ones; sometimes even $A_1 > A_2$. The ratio of the drift times (μsec) of these ions is $48/35 = \sqrt{2}$, and the ratio of their signals $A_2/A_1 = \gamma_2 n_2 u_2 / \gamma_1 n_1 u_1$, where γ is the coefficient of secondary ion-electron emission and n and u are the concentration and velocity of the corresponding ions. From the known published connection between the values of γ_1 and γ_2 it follows that $\gamma_2/\gamma_1 \gtrsim 1$, and at $u_2/u_1 = 0.7$ we obtain $A_2/A_1 \approx n_2/n_1$, i.e., the ratio A_2/A_1 is proportional to the ratio of the concentrations of these ions n_2/n_1.

The mass spectrometric analysis of the composition of the ions in the cesium plasma has led to the following unexpected results:

A. In the case of a discharge cesium plasma, where impact ionization of a stepwise character should predominate ($Cs + e \rightarrow Cs^* + e$, followed by $Cs^* + e \rightarrow Cs^+ + 2e$),[7] there was observed, instead of the expected atomic composition of the ions under the corresponding conditions, the presence of a rather noticeable amount of molecular cesium ions[6]. We attribute their appearance to the secondary process of ion conversion in accordance with the scheme $Cs^+ + 2Cs \rightarrow Cs_2^+ + Cs$, similar to the well known scheme in the case of a plasma in inert gases[8].

B. In the case of a photoresonant cesium plasma, where associative ionization should prevail (see above) we observed instead of the expected purely molecular composition of the ions under the corresponding conditions, the presence of a rather noticeable amount of atomic cesium ions; we shall now proceed to consider the peculiarities and the possible nature of this strange phenomenon.

Figure 5 shows the result of the determination of the ratios of the signals at the output of the instrument $A_2/A_1 \approx n_2/n_1$, where $n_1 + n_2 = n_e$, obtained at the end of a pulse of duration 1200 μsec. This ratio was determined as a function of n_e (probe measurements) at the indicated values of p (curve I—III, respectively). The course of this dependence $A_2/A_1 = f(n_e, p)$ is characterized by a sharp decrease of A_2/A_1 with increasing n_e, and by a sharp rise with increasing p. Outwardly this dependence is quite similar to that obtained by us for the case of a discharge cesium plasma[6], but in fact the two have nothing in common.

Of very great importance for the understanding of the nature of the phenomena in a photoresonant plasma are the results of separate investigations, in terms of the masses A_1 and A_2, of the evolution of the plasma after an abrupt turning on of the discharge-current pulse in the external irradiation source, and the process of plasma decay after an abrupt turning off of this current pulse. The results of such measurements are shown in Fig. 6 for the case p = 0.01 and 0.08 mm Hg. Here curves 1 and 2 pertain to the case of the concentration, respectively, of the atomic and molecular ions of cesium in the evolution of the plasma, and curves 1′ and 2′ correspond to the same ions in the decay of the plasma. The data of these interesting figures lead us to the following conclusions:

1. During the course of evolution of the plasma, the concentration of the molecular ions is established relatively rapidly, and subsequently remains unchanged; the establishment time τ_2 ($\sim$ 300—600 μsec) increases with increasing cesium-vapor pressure. Since the resonant radiation entering the detector from the outside is strongly absorbed in its near-wall region, producing there a noticeable associative ionization[4], it can be assumed that the time τ_2 is connected with subsequent diffusion of both this radiation and of the charges obtained here in the interior of our detector.

2. Unlike the preceding case, the concentration of the atomic cesium ions, experiencing at the very start of the pulse a certain still inexplicable irregularity, increases subsequently much more slowly (the establishment time is $\tau_1 > \tau_2$), especially at increased pressures, and frequently reaches a value even larger than the concentration of the molecular ions. We see also that the growth of curve 1 can occur against the background of a constant curve 2, and the initial irregularity on the former is not reflected in any way in the latter; one can therefore probably assume that the nature of formation of the atomic ions has a character independent of the molecular ions, although this conclusion is, naturally, not yet final.

3. The rates of decay of the plasma relative to the atomic and molecular ions separately (β_1 and β_2) are qualitatively approximately the same; for example, the half life for both curves at p = 0.01 mm Hg is approximateyl 200 μsec, and at p = 0.08 mm Hg it is approximately 600 μsec, i.e., it increases with increasing pressure. This circumstance is obviously connected with the fact that the plasma decay observed in the region adjacent to the entrance into the drift space of the mass spectrometer is determined mainly not by the volume recombination, but by the diffusion of the ions to all the walls, the surface of which is quite large here. In this case, unlike the recombination coefficients of these ions, which differ by approximately two orders of magnitude[9], their diffusion coefficients D, or, accord-

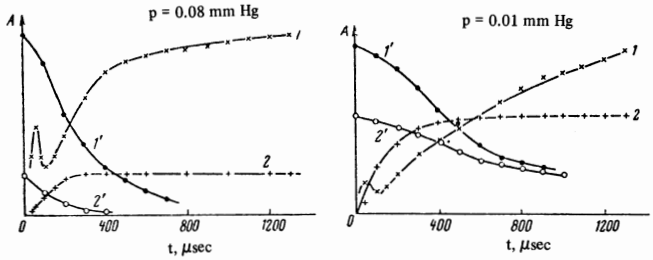

FIG. 6

ingly the mobilities μ, differ much less—only by 2–4 times[10]; moreover, the relations $D \infty 1/p$ and $\mu_2 > \mu_1$ are confirmed here. The conclusion drawn in[1] that a predominant role is played by volume recombination of the molecular ions, is connected with the different experimental conditions, namely much larger p = 0.3 mm Hg, t_p = 300 μsec, and the presence of only a small probe in the volume of the instrument; all this should contribute to the obtained tendency.

4. The time evolution of the ionic component of the probe current of the plasma $I_{pr}(t)$, has shown by measurements during the course of evolution and decay of the plasma, reflects well the sum of each pair of curves $A_1(t)$ and $A_2(t)$ in Fig. 6. However, the similar relations usually determined in the corresponding plasma investigations contain relatively little information, when it is assumed that the plasma contains ions of several types; in this case the use of the mass-spectrometric analysis becomes vital.

Thus, the data presented here point to a large number of interesting features of such a currentless and magnetless cesium plasma with a very noticeable charge density, $\gtrsim 10^{12}$ cm^{-3}. At the same time, whereas the question of the nature of formation of molecular ions in a photoresonant plasma is more or less clear, if one bears in mind associative ionization, the same cannot be said as yet concerning atomic ions. In this connection, one can indicate tentatively, for example, the following two extreme possibilities whereby the latter can be produced: 1) dissociation of molecular ions by electron impact: $Cs_2^+ + e \rightarrow Cs^* + Cs + e$ and 2) ionization of photoexcited cesium atoms likewise by electron impact $Cs^* + e \rightarrow Cs^+ + 2e$. However, arguing against the first possibility are, as it were, the considerations advanced above; experiments lead, purely qualitatively, also to some preliminary evidence favoring the second possibility. For a conclusive solution of this problem we are still lacking many data, primarily the values of the effective cross sections of the indicated processes; this will be the subject of future work.

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