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Decay of metastable atoms in the afterglow of single-electrode breakdown

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Abstract. This paper deals with experimental studies of ionization waves igniting the breakdown in long discharge tubes in neon at low pressures. Ionization waves were produced by rectangular voltage pulses applied to the high-voltage electrode of the discharge tube. Our research focused on concentration measurements of neon excited atoms at the metastable $1s_5(^3P_2)$ level after the ionization wave passage and the breakdown termination. We also found the characteristic decay times of these states and the most significant elementary processes responsible for metastable atom deactivation.

Keywords: gas discharge, gas breakdown, ionization wave, afterglow, neon plasma, low pressure, metastable atoms, elementary processes

Introduction

Ionization wave (IW) is an electric field solitary wave formed by volume and surface charges and propagated along a gas discharge tube in the form of an ionization front (Lagarkov, Rutkevich 1994; Vasilyak et al. 1994). Due to high field strength at the IW front, high-energy electrons produce intensive ionization and excitation of the gas atoms in the time interval of 10^{-9} – 10^{-6}s, which eliminates heating. That is why IWs are used in longitudinally pumped lasers and fast gas discharge switches (Ashurbekov et al. 2000).

Atoms in metastable states play a significant role in the gas discharge. Some papers (Ashurbekov et al. 2000; 2015; Shakhsinov, Ramazanov 2013) analyze their influence on the dynamics of high-speed IWs ($v > 10^9$ *cm/s*) in long capillary tubes. The movement of the IW front is accompanied by inelastic collisions of high energy electrons with atoms, which leads to the population of metastable levels in addition to ionization. High metastable atom concentration leads to the production of a significant number of high-energy electrons due to second-kind collisions (Raizer 1991). This process affects the radial profile of the IW radiation and the distribution of ionized particles at the front.

The characteristic decay time of the metastable state is much lower than the diffusion one according to the measurements that have been made immediately after the IW passage through the discharge gap in (Ashurbekov et al. 2000; 2015; Shakhsinov, Ramazanov 2013). This fact indicates the predominance of other channels of their deexcitation. Inelastic collisions with electrons whose concentration is high after the IW passage could be one of them.

Usually, the second electrode of the discharge tube is grounded, which leads to glow discharge ignition after the breakdown. Thus, it becomes impossible to observe plasma decay after the propagation of the pre-breakdown IW only. This motivates us to work with a single-electrode discharge (Shishpanov et al. 2020). It consists of only repetitive passages of IWs with a frequency of that of high-voltage pulses applied to the tube electrode. A glow discharge does not occur in this case, and all excited atoms form at the breakdown stage only. Such discharge mode provides new information about the IW due to enhancement of its observability. A single-electrode breakdown creates a short current pulse, the duration of which is equal to the movement and IW disintegration time (< 10 μs). The beginning of the breakdown afterglow was determined by the IW registration at the opposite tube end using oscillograms of optical signals.

These features of the single-electrode mode motivated us to measure neon metastables' density in the breakdown afterglow, averaged over the tube cross section. We created breakdowns by a positive polarity voltage and chose the pressure range such that the IW front propagates most uniformly across the tube cross section (Starikovskaya 2000). The observation point was taken at a 20-cm distance from the initiating electrode in order to reduce the influence of the possible ring structure of the IW front near the high-voltage anode on the measurements, as was previously observed, for example, in helium at pressures over 10 Torr (Asinovskii et al. 1984). In this research we measured the metastables' densities at different breakdown voltages and several gas pressures after IW propagation.

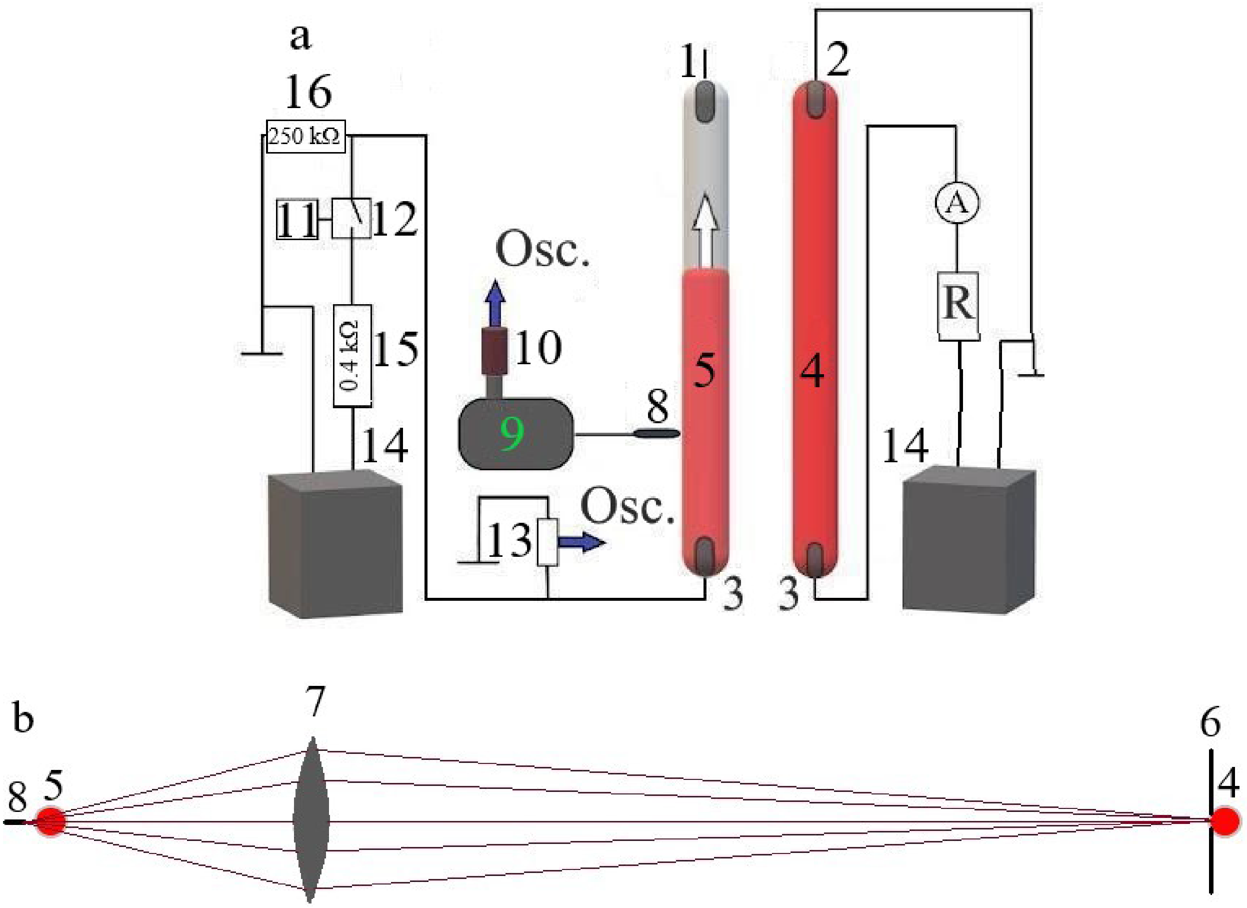

Fig. 1. a — schematic diagram of the experimental setup; b — optical scheme. 1 — unconnected electrode, 2 — grounded electrode, 3 — high voltage electrodes, 4 — radiating tube, 5 — absorbing tube, 6 — diaphragm, 7 — lens, 8 — optical fiber, 9 — monochromator, 10 — PMT, 11 — pulse generator, 12 — switch, 13 — compensated high voltage divider, 14 — high voltage power supply, 15 — resistor (0.4 kΩ), 16 — resistor (0.25 MΩ)

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Experiment

Fig. 1a shows a schematic diagram of the setup for measuring radiation absorption using the 'two tubes' method (Huddlestone, Leonard 1965). The aim of the diagnostics is to obtain data on the concentration of atoms in metastable states after the IW propagation through the discharge tube. Two parallel discharge tubes with neon, spaced by 40 cm, were used. The inner diameter of each tube was 1.5 cm, and the interelectrode distance was 60–80 cm. The tube electrodes (1–3) were made of aluminum and had the shape of a hollow cylinder, whose edges were covered with ceramic rings to prevent metal sputtering. One of the tube contained DC discharge in neon (p = 0.6 Torr, i = 3–3.5 mA) without stratification and was used as a radiation source (4). The decaying plasma in the afterglow of a single-electrode breakdown in the parallel tube (5) with neon at pressures from 0.6 to 10 Torr was used as the absorbing medium. Light from the source passed through the 0.5 cm diaphragm (6) and was then focused by the lens (7) onto a tube with an absorbing medium according to the diagram in Fig. 1b. Then, the light was collected by an optical fiber (8), aligned perpendicular to the tube axes and transmitted to the monochromator (9) with a photomultiplier (10).

To obtain reliable data, optical signals from different breakdown pulses were accumulated. It was necessary to generate identical IWs to use such recording scheme. We ensured the same initial conditions in every discharge pulse, the main of which was the exciting voltage. The IW and subsequent breakdown were initiated by a rectangular voltage pulse of positive polarity applied to the tube electrode. The pulse rise time (≈50 ns) was shorter than the average breakdown delay time. Thus, the IW appeared after the moment when the pulse voltage reached a constant U_0 value which could be varied in the range of 0.6–4.5 kV. 10 ms pulses were produced at frequencies of 1–5 Hz by the circuit consisting of a generator (Tektronix AFG3022C control oscillator (11)) and a fast switch (12). A compensated high-voltage divider (13) was connected to the oscilloscope showing the voltage behavior at the high-voltage electrode during the breakdown. When a triggering pulse from the generator was applied, the switch connected the high DC voltage power supply (14) with the electrode through a ballast resistor (15). The resistor (16) 0.25 MΩ connected in parallel to the tube provided the appropriate trailing edge of the high voltage pulse (pulse decay time ≈ 200 ns) due to the tube charge removal to the ground after switch disconnection.

The concentration of neon excited atoms at the metastable $1s_5(^3P_2)$ level has been measured by the spectrum line absorption method. For this purpose, we used neon spectral lines λ = 640.2 nm and λ = 614.3 nm. The light source was the plasma of the positive column of the neon glow discharge at low pressure, and the absorber was the afterglow plasma. The emission and absorption line profiles have been chosen Doppler with the gas temperature of T_a = 300 K. Collisional (van der Waals) broadening $\Delta\nu_c$ was estimated as follows. According to (Ochkin 2009),

$$\Delta\nu_c = 1.30 \times N{C_6}^{2/5}v^{3/5}$$

where N — concentration of unexcited atoms, v — average particle velocity and C_6 — van der Waals interaction constant, for which estimates and analysis of experimental data give the value in the range of $10^{-30} - 10^{-32}\ \frac{cm^6}{s}$. Then, the upper estimate for the $\Delta\nu_c$ value taken for the largest pressure of 10 Torr turned out to be $\Delta\nu_c \approx 3 \times 10^8\ s^{-1}$, whereas the Doppler width is $\Delta\nu_D \approx 1.3 \times 10^9\ s^{-1}$. In the experiment, the absorbed proportion of radiation was ≈ 0.8–0.95, which indicates a low absorption coefficient and hence, a small possible role of Lorentzian collisional wings. From these considerations, we considered the line profile as the Gaussian (Doppler) one.

For calculations the radial concentration distribution of metastable atoms was considered to be: $M(r) = M(0) \times J_0\left(\frac{r}{R}\mu\right)$, where J_0 is the first kind Bessel function of zero order, R is the tube radius and $\mu \approx 2.405$ is the first root of the Bessel function. It is worth noting that this assumption did not significantly affect the result which was the M value averaged over the tube cross section.

Experimental results

The peculiarity of the single-electrode breakdown mode is that the current pulse duration (< 10 μs) associated with the IW passage is much shorter than that of the discharge afterglow. Thus, the decay of metastable states was observed with no sources of their population, except for the IW. Fig. 2(a) shows the time dependence of concentrations at fixed pressure for different breakdown voltages. By fitting them to the exponential function, the metastable densities at the moment of breakdown M_0 and their characteristic decay times τ have been measured. The relative error for τ was ≲ 10% and for M_0 ≲ 15%. The main result of this experiment is the fact that an increase in the breakdown voltage leads to an increase in the number of metastable atoms remaining after the IW passage. It should be noted that the curves can be approximated by a linear dependence in a logarithmic scale, confirming that the concentration decreases exponentially with the same τ for all voltages. The curves obtained at a fixed breakdown voltage but various pressures are shown in Fig. 2 (b), while the results of their processing are in Table 1. One can see that τ depends on pressure non-monotonically with the maximum at p = 3 Torr. The concentration M_0 grows with increasing pressure, which agrees with the results of other works (Ashurbekov et al. 2000; 2015; Shakhsinov, Ramazanov 2013).

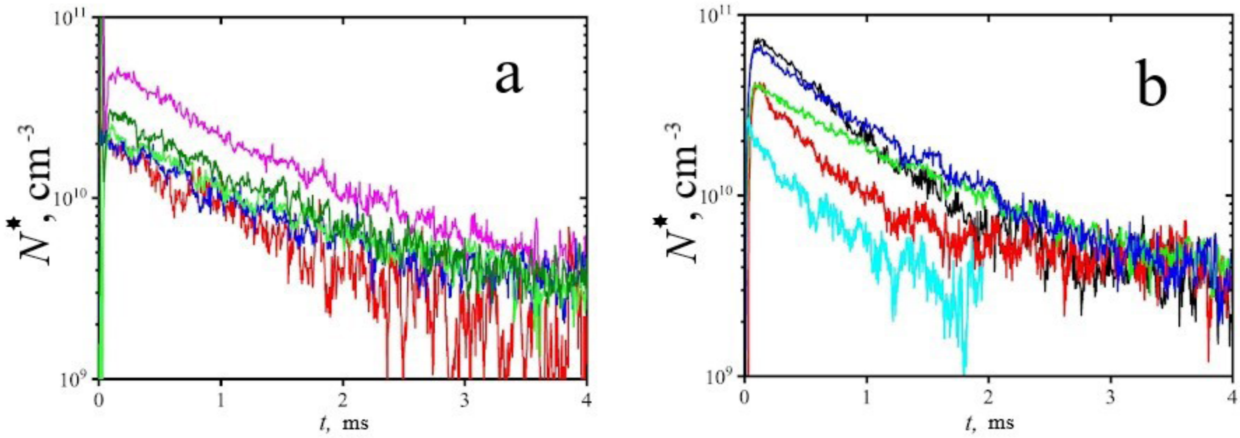

Fig. 2. Time dependences of concentrations of metastable ($1s_5$) neon atoms
(a) at different U_0. p = 3 Torr; pulse frequency is 1 Hz; voltage pulse duration is 10 ms
– 2.5 kV, – 3 kV, – 3.5 kV, – 4 kV, – 4.5 kV.
(b) at different p. U_0 = 3 kV, for 1 Hz and 10 ms.
– 10 Torr, – 5 Torr, – 3 Torr, – 1 Torr, – 0.6 Torr

Table 1. Results of approximation of the experimental time dependence of the metastable atom concentration and results of theoretical calculations

p, Torr	τ_T, ms	τ_{exp}, ms	M_0, $cm^{-3} \times 10^{10}$
0.6	0.30	0.33 ± 0.03	2.0 ± 0.3
1	0.46	0.45 ± 0.05	4.2 ± 0.5
3	0.87	1.0 ± 0.1	4.5 ± 0.6
5	0.74	0.74 ± 0.07	7.0 ± 1.0
10	0.54	0.65 ± 0.05	9.0 ± 1.5

Results and conclusions

We examined the possible channels of the metastable atom decay under the studied conditions. One of them is the diffusion to the discharge tube wall with subsequent deexcitation on it. This process is the most significant at low pressures. The excited atom diffusion lifetime can be expressed in the following way (Raizer 1991): $\frac{\Lambda^2}{D}$, where $\Lambda = R/2.405$ is the effective diffusion length for a cylindrical tube, and D is the diffusion coefficient. Another channel of the excited atoms' decay is the mixing of excited states by atom collisions:

$$Ne(1s_5) + Ne \rightarrow Ne(1s_4) + Ne$$

Since the $1s_4$ level is resonance, its concentration is substantially less than that of metastable atoms, so the reverse process was not factored into the calculations.

The dimer formation by collisions and ionization by collisions with other metastable atoms (chemo-ionization) were also taken into account.

$$Ne(1s_5) + Ne + Ne \rightarrow Ne_2^* + Ne \rightarrow Ne + Ne + Ne + h\nu$$

$$Ne(1s_5) + Ne(1s_5) \rightarrow Ne^+ + Ne + e$$
$$\rightarrow Ne_2^+ + e$$

The analysis of the obtained data as part of the proposed model was limited only to the above-mentioned processes. Therefore, the balance equation for the metastable atom concentration can be written as follows:

$$\frac{dM}{dt} = -(\frac{(DN)}{\Lambda^2 N}M + K_1 NM + K_2 N^2 M + K_3 M^2),$$

where M is the concentration of atoms at the metastable $1s_5$ level averaged over the tube cross section

N — concentration of unexcited atoms,

$DN = 6.2 \times 10^{18} \frac{1}{cm \cdot s}$ (these and subsequent data are taken from (Dyatko et al. 2006)),

$K_1 = 4.9 \times 10^{-15} \frac{cm^3}{s}$ is the rate constant of excited states' mixing,

$K_2 = 5 \times 10^{-34} \frac{cm^6}{s}$ is the rate constant of three body quenching,

$K_3 = 3.8 \times 10^{-10} \frac{cm^3}{s}$ is the rate constant of chemo-ionization.

Since the contribution of chemo-ionization appears to be negligible, the solution can be represented in the exponential form:

$$M = M_0 \, exp \, [-\frac{t}{\tau}] = M_0 \, exp \, [-t(\frac{(DN)}{\Lambda^2 N} + K_1 N + K_2 N^2 + K_3 \, M_0)], \tag{1}$$

where M_0 is the concentration of metastable atoms at the moment of time just after the IW passage.

It is important to note that one more process of the metastable $1s_5$ level quenching, *viz.* collisions with thermal electrons

$$Ne(\,1s_5) + e \rightarrow Ne(\,1s_4) + e$$

should be considered. The rate constant for it is $K_4 \approx 1 \times 10^{\wedge}7 \times$ cm^3/s (Dyatko et al. 2006). Unfortunately, we have no information on the electron number density n_e in the afterglow. However, it was shown under similar conditions for the IW in argon (Dyatko et al. 2021) that the n_e value varies strongly with the voltage pulse amplitude (two-fold increase in the voltage results in an order of magnitude growth in n_e). So, this reaction should lead to dependence of the decay curves on the pulse voltage, which is not the case (Fig. 2a). Hence, this process can be considered unnoticeable.

The decay times for different pressures were calculated according to (1), and their comparison with experimental data is given in Table 1. Note that the τ dependence on pressure is non-monotonic. For low pressures, the experimental and calculated τ values are close to each other. In these conditions, diffusion to the tube walls is the predominant decay channel. Despite the discrepancies in the measured and estimated values for the higher pressures, the τ(p) dependence is qualitatively the same in the theoretical calculation and in the experiment. This confirms the correctness of the proposed model. It also follows from the above estimations that the diffusion and mixing of excited states by atom collisions in the investigated pressure range are the dominant metastable atom decay processes.

Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

Authors contributions

All the authors discussed the final work and took an equal part in writing the article.

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