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Nonmonotonic radial distribution of excited atoms in a positive column of pulsed direct current discharges in helium

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Nonmonotonic radial distributions of excited helium atoms have been experimentally observed in a positive column of pulsed helium discharges using planar laser induced fluorescence. Computational analysis of the discharge dynamics with a fluid plasma model confirms the experimental observations over a range of pressures and currents. The observed effect is attributed to the peculiarities of electron population-depopulation of the excited states during the “dynamic discharge” conditions with strong modulations of the electric field maintaining the plasma. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4788689]

Current-modulated and pulsed DC discharges have been used to increase efficiency of gas discharge light sources¹ and control population channels of gas lasers. It is shown that in a “dynamic regime” at frequencies in the kHz range, it is possible to manipulate electron kinetics and control the electron energy distribution function (EEDF) due to strong modulation of the electric field maintaining the plasma. Low duration (~10 μs), high current intensity (~1 A), and low-repetition pulses can eliminate plasma constriction and striation, reduce gas heating, and improve discharge stability. Spatially resolved and temporally resolved laser-based diagnostics, coupled with predictive simulations of a pulsed discharge provide an effective set of tools for studying the elementary processes and electron kinetics in these regimes. In this letter, we report on experimentally observed nonmonotonic distribution of excited helium atoms in a pulsed DC positive column discharge. Coupled with a fluid plasma model, we conclude that these observed profiles result from changes in the population-depopulation kinetics as the electric fields in the pulsed positive column evolve over the duration of the current drive.

The experimental setup is depicted in Figure 1. A helium positive column was generated in a vacuum chamber that was constructed with a combination of stainless steel vacuum tubing with nominal inner diameter of ~35 mm and Kovar glass tubing also with 35 mm inner diameter. The length of the glass tube was 450 mm, the length of the biased anode was 180 mm, and the length of the grounded cathode was 200 mm for a total chamber length of 830 mm. The anode was connected to a fast switching pulsed power supply via a 1.5 kΩ current limiting resistor and a DC of ~50 mA was used to sustain the discharge at all times. Typical helium pressures range from ~1 Torr to 10 Torr and typical current pulses span 0.1 A to 2 A for a duration of 50 μs and at a rate of 20 Hz. A Pearson current probe and a microwave interferometer operating at 81 GHz were used to measure the discharge current and the line-integrated electron density. The spatial and temporal distributions of helium atoms in the 2³P state were measured using planar laser-induced fluorescence.

The output of a 20 Hz pulsed ND:YAG laser (355 nm, 10 ns pulse width) pumping an optical-parametric oscillator (OPO) was used to generate 471.32 nm radiation that excites the 2³P to 4³S excitation. The output of the laser was spatially filtered and then formed into a collimated planar sheet ~1 mm thick by a pair of cylindrical lenses. An aperture was used to limit the height of the beam to be slightly less than the inner diameter of the glass tube housing the plasma discharge. A gated, intensified CCD camera with a narrow band interference filter centered on 471 nm (<4 nm bandwidth) images the spatial distribution of the laser-induced fluorescence. Images are acquired ~20 ns after laser excitation for a duration of ~100 ns. The firing of the laser and the imaging of the fluorescence are synchronized to the pulsed current used to excite the positive column. Temporal evolution of the column can be studied by varying the delay between the current pulse and the firing of the laser.

A customized version of cfd-ace+ software was used for simulations of positive column.² Although a kinetic module for EEDF calculations was available in cfd-ace+, we used a fluid model assuming a Maxwellian EEDF. Gas heating by electrons and ions was taken into account, and a Poisson solver was used for calculations of the radial electrostatic potential. Overall, our model was similar to

![Image](https://example.com/image.jpg)

**FIG. 1.** Illustration of experimental setup used to observe transient behavior of a pulsed helium positive column.
Ref. 3. The axial electric field maintaining the plasma was found from

\[ E(t) = \frac{I(t)}{2\pi \mu_e n_e(r) r dr}, \]  

where \( I(t) \) is the discharge current, \( \mu_e \) is the electron mobility, and \( n_e \) is the electron density. In our simulations, the current is specified in the form:

\[ I(t) = I_0 + I_1(t), \]  

where

\[ I_1(t) = \begin{cases} \frac{2I}{\pi} \arctan(\omega t), & 0 < t < \tau, \\ 0, & \tau < t < T. \end{cases} \]  

Simulations were performed for a tube of radius 1 cm, for helium pressures of 1, 3, and 10 Torr, for different amplitudes of pulse currents \( I \), and inter-pulse period, \( T \). The base case is for \( \tau = 50 \mu s, \quad T = 0.5 \text{ ms}, \quad \omega = 3 \times 10^8 \text{ s}^{-1}, \quad I_0 = 0.05 \text{ A}, \quad I = 0.25 \text{ A}. \) We used a time step, \( \Delta t = 1 \times 10^{-8} \text{ s}. \) The specific heat and the thermal conductivity of helium in the code are \( c_p = 5193 \text{ J/(kgK)} \) and \( C = 0.15 \text{ W/(mK)} \). Two different chemistry models of helium plasma have been used: a simple chemistry and a detailed chemistry.

Key experimental observations made during the current pulse are presented in Figure 2 for two pressures. The upper plots correspond to averaged plasma parameters such as discharge current (\( I \)), electric field (\( E \)), electron density (\( n_e \)), and electron temperature (\( T_e \)) while the lower images show the spatial-temporal evolution of the (normalized) profiles of the \( 2^3P \) states. For both pressures, the current and electron densities reach maximum values within 20 \( \mu \text{s} \) of the current pulse. Both the electric field and electron temperatures have sharp peaks during the initial stage and subsequently decay during the pulse. Electron densities decay while the electric fields and electron temperatures recover after the current reaches its peak value of \( \sim 1.2 \text{ A} \) for the 2.3 Torr cm case. On the other hand, these parameters remain essentially constant after the current is established for the 11.2 Torr cm case. The spatial and temporal evolutions of the \( 2^3P \) are observed to depend on the pressure of the positive column. Measured and simulated radial distributions of excited atoms are plotted in Figure 3 for various pressures and for various times during the current pulse. For all three pressures, the excited state species are initially center peaked and monotonic. This period of time corresponds to the growth of the electron density in response to the current pulse. For the lowest pressure case, the distribution of the excited atoms undergoes some degree of flattening during the middle portion of the current pulse but remains monotonic. As better illustrated in Figure 2, the center-peaked nature of the measured excited atoms is recovered towards the end of the current pulse. On the other hand, for the higher pressures, nonmonotonic radial distributions of the excited state helium are generated and maintained during the remaining duration of the current pulse and center-peaked profiles are not recovered. Nonmonotonic profiles are also observed in simulations. For these predictions, the simplest chemistry model taking into account direct
ionization by electron impact, excitation of a metastable state, and stepwise ionization from this state captures qualitatively the experimentally observed behavior for radial distribution of metastable atoms and electrons for different currents and pressures. Detailed chemistry of high-pressure helium discharges gives qualitatively similar effects.

Nonmonotonic radial distributions of excitation rates in steady-state positive columns of rare gases (with exception of helium) at elevated pressures have been observed in numerical simulations and explained by non-local effects in EEDF formation. Clearly, effects described above are different from these and captured by a fluid plasma model. Nonmonotonic radial distributions of metastables have been recently observed in simulations of pulsed xenon discharges under similar conditions. In that study, the authors ascribed this distribution to the heating and rarefication of the neutral gas in the center of the column and subsequent reduction of quenching of higher lying 1s states into lower 1s states. However, no experimental observation of this effect is reported.

In attempt to understand the observed nonmonotonic distributions of excited atoms, we consider the dynamic behavior of both the electric field and the electron temperature in response to the applied pulse and how their behavior impacts the balance between production and loss of excited species. The simulation results plotted in Figure 4 help distinguish four stages in the dynamics of pulsed discharges (assuming $E_0$ is the steady-state field): (1) high-current pulse ($E > \sim E_0$), (2) afterglow ($E \ll E_0$), (3) recovery ($E < \sim E_0$), and (4) low-current discharge ($E = E_0$). During the initial part of stage 1, the electron temperature peaks and the excited state production is high. As a result, the excited state species reaches a maximum during this period of time (Fig. 2). During the latter portion of stage 1, both the electric field and the electron temperature drop to values comparable to, or less than those of the pre-pulse condition. The excited state densities are observed to obtain a nonmonotonic distribution across the column. After termination of the current pulse, the values of electric field and electron temperature drop far below their steady-state DC values (stage 2). The simulations show that the nonmonotonic profiles of metastables exist not only during the current pulse but also in the afterglow. On the other hand, the 23P state is not observed because the electron temperature becomes too low to excite the 1.14 eV 23S $\rightarrow$ 23P transition in the afterglow. Because the electron temperature drops, the electron induced reactions are switched off and the density of the excited atoms evolves slowly (become almost frozen) during the afterglow stage. Thus, during the afterglow stage, metastable density profiles are primarily governed by diffusion and chemical reactions among heavy species. The duration of the afterglow stage is on the order of the ambipolar diffusion time ($\tau_a \sim D_a/R^2$), where $D_a$ is the diffusivity and $R$ is the radius of the column. After this period of time, the electric field and electron temperatures rise (stage 3) to the nominal pre-pulse conditions (stage 4).

The formation of nonmonotonic distributions coincides with the period of the pulse when net production of excited state species at higher electron temperatures (earlier portion of stage 1) transitions to net loss of excited state species at lower electron temperatures (latter portion of stage 1). The factors that contribute to the depletion of the excited state include radiative decay (23P), excited state-exited state collisions, and electron impact (ionization). The latter two interactions are likely mechanisms for generating the nonmonotonic distribution as both excited-state densities and electron densities are initially centered peaked during the earlier portion the current pulse. The recovery of the center peaked distribution of the 23P at 2.3 Torr cm (Figure 3) likely corresponds to the recovery of electron temperatures.
prior to the termination of the current pulse. Further investigations are underway to better identify the cause on the non-monotonic profiles described here.

To summarize, we have experimentally observed nonmonotonic radial distributions of metastable atoms in positive column of pulsed DC discharges in helium over a range of gas pressures and pulse currents. The observed effects are qualitatively described by a fluid plasma model. They are attributed to peculiarities of population-depopulation of the excited states in the dynamic plasma regimes, which are characterized by strong modulation of the electric field and electron temperature. We believe that similar effects typical to pulsed plasma excitation can be observed in other types of plasma sources under dynamic regimes of plasma maintenance.

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