Time-resolved analysis of the electron temperature in He nanosecond pulsed discharges at atmospheric pressure

R. K. Gangwar[1], T. Verreycken[2], N. Sadeghi[3], P.J. Bruggeman[2,4,*], L. Stafford[1*]

[1] *Département de Physique, Université de Montréal, Montréal, Québec, Canada H3C 3J7*
[2] *Department of Applied Physics, Technische Universiteit Eindhoven, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*
[3] *LIPhy (UMR 5588) and LTM (UMR 5129), Université de Grenoble / CNRS, Grenoble F-38041, France*
[4] *Department of Mechanical Engineering, University of Minnesota, 111 Church Street SE, Minneapolis, Minnesota 55455, USA*

(*) luc.stafford@umontreal.ca and pbruggem@umn.edu

Temporal behaviour of the average electron energy is studied in a pin-to-pin He nanosecond pulsed discharge at atmospheric pressure using time-resolved optical emission spectroscopy measurements coupled with a collisional radiative model. T_e decreases from relatively high value of ~2.2eV early in the discharge pulse to ~0.8eV as the number density of He metastable atoms rises. Electron temperature then again increases to ~1.4eV as a result of neutral gas heating.

1. Introduction

Filamentary discharges (FD) at atmospheric pressure showed a great potential for various applications such as environmental remediation, biomedical treatment, plasma-enhanced combustion and water treatment [1]. Filaments are often randomly distributed in space and time and are characterized by high electron densities located in a few microns diameter region.

In this work, FDs are stabilized in space between two needle electrodes at a sufficiently short distance, which allows the application of diagnostics with a high resolution. The discharge is investigated in nominally pure He. The temporal behaviour of the average electron energy (or electron temperature assuming Maxwellian electron energy distribution function) in the pin-to-pin nanosecond pulsed discharge (NPD) is analysed using time-resolved optical emission spectroscopy (OES) and collisional-radiative (CR) modelling.

2. Experimental setup

The discharge was realized between two tungsten needles with an inter-electrode distance of ~2 mm. Positive high-voltage pulses were created using a DEI PVX-4110 HV pulser, fed by a DC high voltage from a Spellman DC power supply (10 kV, 15 mA). The pulse width, amplitude and frequency were 170 ns, 2.0 kV and 1.0 kHz, respectively. As shown in [1], this regime corresponds to the glow-like discharge mode. The rise time of the HV pulse was about 80 ns (from 10% to 90%). The discharge was sustained in nominally pure He in a vacuum chamber. The vacuum chamber was continuously flushed with 3.0 SLM (standard litre per minute) mass flow rate.

Current voltage (I-V) characteristics along with the electron density and He metastable (2^3S) population are shown in Fig. 1. The electron density is determined from the Stark broadening of H_β Balmer line [1]. A maximum electron density of 2.2×10^{22} m^{-3} is obtained during the current peak, with a decay of about one order of magnitude afterwards. As for the He (2^3S_1) metastable number density, it was measured by tunable diode laser absorption spectroscopy [1].

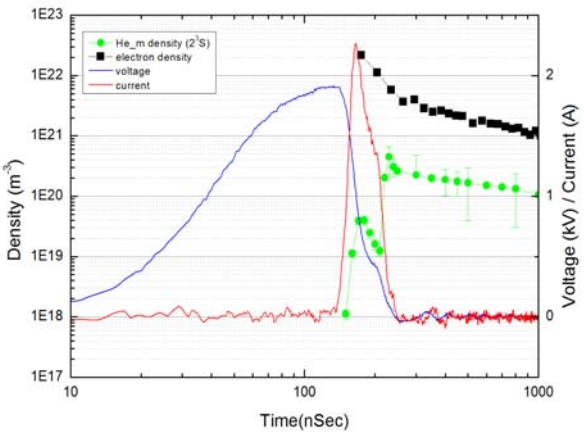

Fig 1: I-V plot from the pulsed nanosecond He discharge. Time-resolved electron and metastable 2^3S atom number densities are also shown.

3. Time-resolved OES measurements

Time-resolved OES measurements were obtained with an iCCD camera (4 Quik E SR) coupled to a 27.5cm focal length monochromator (ACTON). Emission spectra were taken between 300nm and 850nm in two frames centered at 500nm and 750nm with a 1200 grooves/mm grating yielding spectral resolution of 0.3 nm (FWHM) at 587.6 nm. Overview spectra recorded with a gate width of 10 ns at 135ns early in the discharge pulse and immediately after the current pulse at 225 ns are presented in Fig. 2. For clarity, the spectra are normalized to their maximum intensity.

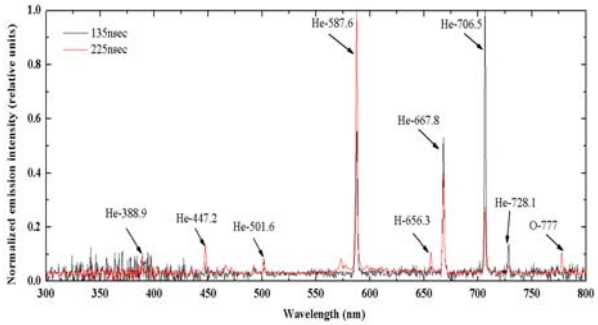

Fig. 2: Normalized OES spectra at 135 ns (early in the discharge pulse) and 225 ns (after the current pulse).

OES spectra reveal 6 emission lines at 388.9, 501.6, 587.6, 667.8, 706.5 and 728.1 nm, originating from the transitions $3^3P \rightarrow 2^3S$, $3^1P \rightarrow 2^1S$, $3^3D \rightarrow 2^3P$, $3^1D \rightarrow 2^1P$, $3^3S \rightarrow 2^3P$, and $3^1S \rightarrow 2^1P$ respectively (**L•S** coupling). While most of the observed He I lines originate from n=3 levels, emission could also be seen from higher states (4^3D) at 225 ns (after the current pulse), suggesting important modification of the electron temperature. Emission ratios between triplet and singlet state also change during the discharge pulse. This is particularly striking for the 587.6 nm-to-706.5 nm line ratio (3^3D -to-3^3S population ratio), going from about 0.6 at 135 s to about 3.3 at 225 ns. Fig. 2 further shows relatively intense emission lines from impurities, in particular H I at 656.3 nm and O I at 777 nm.

Time-resolved emission intensity from a singlet state, He-667.8nm, and a triplet state, He-587.6nm, are presented in Fig. 3 along with the discharge current. As discussed in ref. [1], the single discharge peak and the drop in voltage initiated by the power supply rather than the discharge itself is an indication that the discharge operates in a glow-like mode. Comparison of the discharge current with OES results shows that the He emission lines reach a maximum slightly before the current peak and decrease much more rapidly afterwards. A second emission peak is observed later in the discharge pulse. We will come back to this later on.

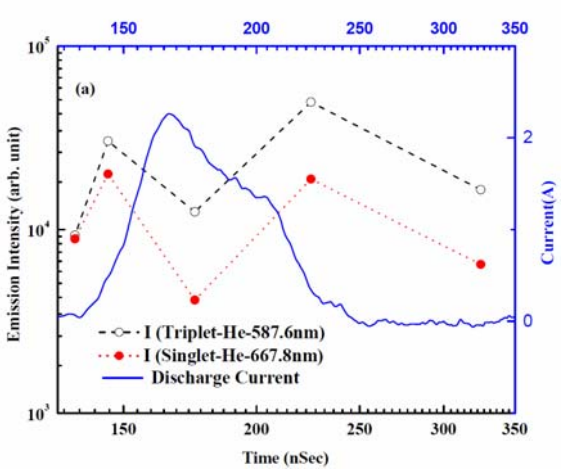

Fig. 3: Time-resolved intensities of He-667.8nm and He-587.6nm along with the discharge current.

4. Collisional-radiative model

In order to extract the information on the time-resolved behaviour of the electron temperature (assuming Maxwellian electron energy distribution function), OES measurements are coupled with a CR model developed for n=3 levels of He. These states are assumed to be excited by electron-impact processes from either ground or metastable states. The CR model also takes into account the excitation transfer processes between He n=3 levels by collisions with ground state He atoms as well as all relevant radiative decays and quenching reactions. Stepwise excitation from metastable states and excitation transfer reactions are assumed to mostly occur between triplet states and between singlet states. This assumption is consistent with the spin conservation rule (ΔS=0), which forbids population transfers between singlet and triplet levels.

At atmospheric pressure, the atoms (excited as well as in ground state) and ions are significantly converted into excimer molecules and ions. Therefore, the associative ionization (AI) reaction (He(n=3)+ He → $He_2^+ + e^-$) as well as the reverse dissociative recombination (DR) process could also influence the populations of emitting He n=3 levels. While DR processes represent additional creation terms in the particle balance of these excited states, AI reactions correspond to collisional losses. Thus, in the present CR model, the contribution of AI was implicitly included in the quenching reaction terms. On the other hand, various reports have revealed that the vibrationally excited He_2^+ states can produce

excited He atoms with n=2, n=3, n=4, etc. by collisions with electrons. However, our recent studies have revealed that these processes can be neglected for atmospheric-pressure dielectric barrier discharges (DBDs) but also for higher-density discharges such as glow NPD [2,3].

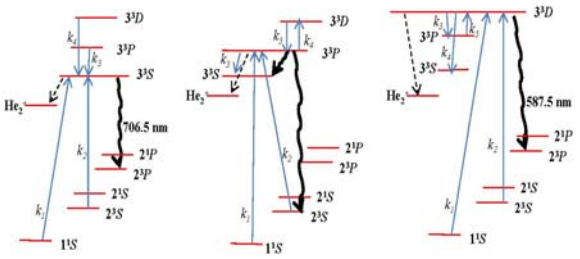

Fig. 4: Framework of the CR model used to simulate the populations of He n=3 triplet states as a function of electron temperatures and metastable-to-ground number density ratio. Similar plot can be drawn for singlet states.

Based on this framework (see Fig. 4), the coupled steady-state particle balance equations of n=3 levels can be solved to extract their population density using the electron temperature and the number density of metastable He atoms as the only adjustable parameters [2,3]. Early in the discharge pulse, a high reduced electric field (E/N) (steamer ignition) is present, such that high-energy electrons start exciting ground state He atoms by electron-impact. As a consequence, the number density of metastable He atoms (2^3S and 2^1S) is low with respect to ground state He atoms. In this regime, He n=3 levels are expected to be mostly populated through electron-impact excitation on ground state He atoms. As the discharge current rises with a corresponding increase in the electron number density, the population of He metastable atoms must increase. At the maximum current and beyond, giving the number densities reported in Fig. 1, excitation of He n=3 levels mainly occurs by electron-impact excitation on metastable states. Based on these findings, triplet-to-triplet (TT) and singlet-to-singlet (SS) line ratios in both regimes become solely dependent on the electron temperature through the reactions rates $k_j(T_e)$ displayed in Fig. 4.

5. Result and discussion

Electron temperatures, T_e, were deduced by comparing line-ratios predicted by the CR model to those obtained from time-resolved OES measurements. Fig. 5 shows TT and SS line ratios as a function of electron temperature assuming that both states are populated by stepwise excitation (assumption valid late in the discharge pulse). Dashed lines represent the experimental measurements for the various TT and SS ratios obtained immediately after the current pulse at 225 ns. For both TT and SS line ratios, a good match between the predictions of the model and the experimental data can be found at T_e=1.5 ± 0.2 eV (dashed zone in Fig. 5).

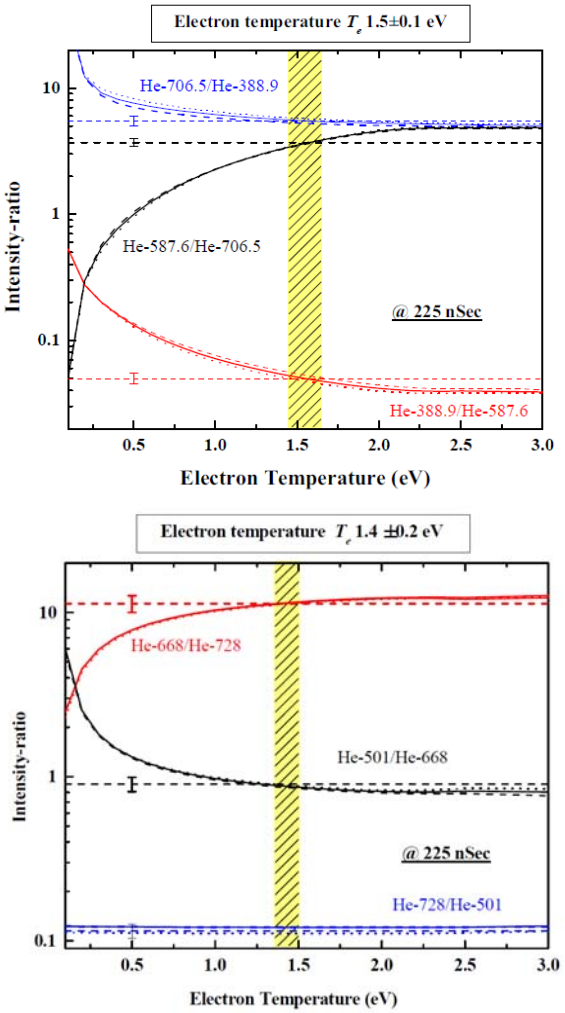

Figure 5: Influence of the T_e on the emission intensity ratios from triplet and singlet states of He n=3 levels.

A similar approach was used to determine the temporal behaviour of the electron temperature from time-resolved OES measurements (except early in the discharge pulse where the population of metastable atoms is low and excitation of He n=3 states results from electron-impact excitation on ground state He atoms). The results are shown in Fig. 6. For comparison, the discharge current and metastable He 2^3S population (obtained from tunable diode laser absorption spectroscopy, see Fig. 1) are also shown. In the first moments of the pulsed

discharge, i.e. at very low discharge currents, electron temperatures are close to 2.2 eV. T_e then rapidly decreases to 0.8 eV. This decrease of the average electron energy can probably be ascribed to a change in the ionization dynamics, going from electron-impact ionization on ground state He atoms to electron-impact ionization on metastable He atoms (stepwise ionization). A similar feature was observed in dielectric barrier discharges (DBDs) [2,3]. However, in contrast to the results obtained in DBDs, the electron temperature rises again to ~1.4eV after the current peak. Such "second rise" of T_e can probably be ascribed to neutral gas heating, leading to an increase of the reduced electric field and thus to a rise of T_e. In refs. [1,4], neutral gas temperatures close to 400 K were observed late in the pulse of low-density mode NPDs operated in He in presence of water vapours.

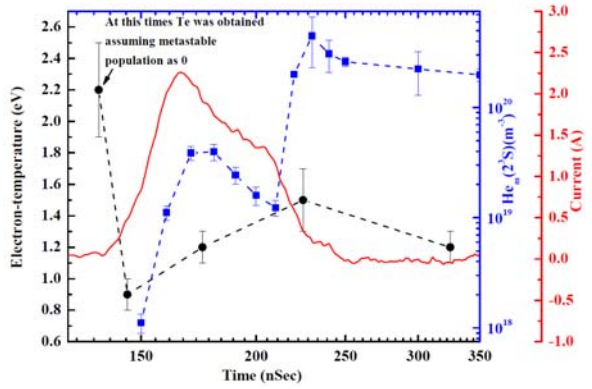

Figure 6: Time-resolved electron-temperature along with He_m (2^3S) metastable density. The first point was obtained assuming that the population of metastable He atoms is low such that excitation of He n=3 states results from electron-impact excitation on ground state He atoms.

We have also performed analysis with triplet-to-singlet (TS) line ratios (for example, 587.6nm-to-667.8nm). In this case, the results are not only dependent on the electron temperature as for TT and SS ratios but also on the 2^3S-to-2^1S metastable number density ratio. Fig. 7 presents the influence of this metastable population ratio on various TS emission line intensity ratios. The horizontal dashed lines correspond to the experimental measurements, obtained immediately after the current pulse at 225 ns. The results were plotted using the T_e value deduced from TT and SS ratios (see Fig. 5).

A good match for all TS ratios is obtained for a 2^3S-to-2^1S population ratio of 1.6. This value is much lower than the one expected from the ratio of the statistical weight of He 2^3S (triplet state; $2J$+1=5) and He 2^1S (singlet state; $2J$+1=1) in Boltzmann equilibrium. It is also lower than the one obtained assuming that these levels are mainly populated through electron-impact excitation on ground state atoms and having equal losses (2^3S-to-2^1S population ratio of 2.2 at 1.5 eV). This slight discrepancy can probably be ascribed to the different losses between both states.

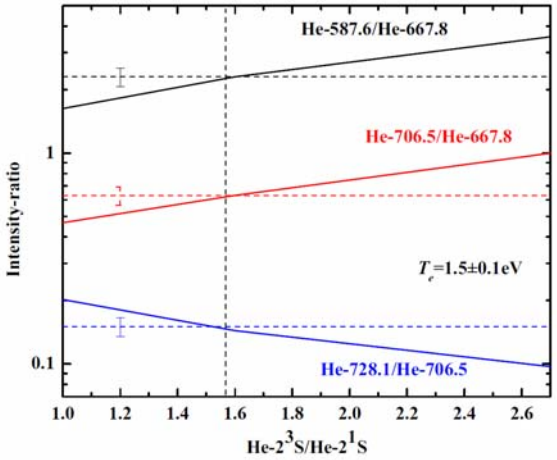

Figure 7: Influence of 2^3S-to-2^1S metastable population ratio on TS line intensity ratios.

6. Acknowledgements

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7. References

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