Correlating Metastable-atom Density, Reduced Electric Field, and Electron Energy Distribution in a 1-mbar Argon Plasma Discharge

J.B. Franek\textsuperscript{1}, S.H. Nogami\textsuperscript{1}, M.E. Koepke\textsuperscript{1,2}, V.I. Demidov\textsuperscript{1,3}, E.V. Barnat\textsuperscript{4}

\textsuperscript{1} West Virginia University, Morgantown, WV, 26506 USA
\textsuperscript{2} University of Strathclyde, Glasgow, G40NG United Kingdom
\textsuperscript{3} St. Petersburg State University, St. Petersburg, 198504 Russia
\textsuperscript{4} Sandia National Labs, Albuquerque, NM, 87123 USA

Introduction

Finding correlations between plasma parameters and alternate means to measure them may be of great utility to the plasma scientist. By employing diagnostics that rely on observing plasma emission, rather than probes or laser techniques, advantage is taken of being less invasive; the only drawback to these techniques being the difficulties associated with understanding the atomic processes that lead to these emissions.

Collisional-radiative models have been used to model plasmas with impressive accuracy in various discharges and pressures. While these models have been tested by experimental observations, their great complexity inspires one to hope that simplifications are still possible without sacrificing accuracy in the process. Efforts have been made to simplify these collisional radiative models for extracting diagnostic signatures, however, these simplifications still require algebraic overhead. These methods also often utilize the 2px→1sy transitions which are known to suffer from significant radiation-trapping effects \cite{1}. While the authors take care to account for these effects, eliminating the need to account for these effects would be desirable. In this paper we demonstrate the correlation between the electron energy distribution function, (EEDF) metastable-atom density, and reduced electric field suggested by Adams et al. \cite{2} and use this correlation to temporally predict metastable-atom density in the late stage of a pulsed positive column of argon plasma.

THE CORONA MODEL AND THE INCLUSION OF METASTABLE-ATOMS

Before we may gain insight regarding an observed line-ratio measurement, we must have some understanding of the raw emission processes. In corona equilibrium, \cite{3} the only source for a given excited-state’s electron population is electron-impact excitation from the ground state into the given excited state and the only sink (relaxation or decay process) is electron transition via photon emission. Whereas using the corona-equilibrium model is
convenient for low-pressure plasma, describing argon at higher pressure requires an extended corona-equilibrium model similar to that of Boffard et al. [4]. The numerical model in Ref [2] shows that the stepwise excitation rates for these excited states are much greater than direct excitation rates; thus, for a first-order correction, the longer lived 1s5 metastable state (in Paschen’s notation) is included as a source in the corona equilibrium equation. The photon flux $\Phi_{ij}$ (number of photons per second) is proportional to observed intensity $I_{ij}$, (watts per unit area) given by Ref [2] as

$$I_{ij} = K n_o R_{ij} \left[ n_o k^0_{ij} + n_m k^m_{ij} + \cdots \right].$$

(2)

The transitions associated with relevant photon emission are now written in terms of the two greatest atomic argon populations - the ground state and the first metastable state.

The reaction rate constants $k^l_{ij}$ calculated from the numerical model [2] are functions of optical emission cross sections and assumed EEDFs; therefore, the effects of radiative cascades into the 3px states, the resultant branching ratios, metastable-atom density, electron density, and reduced electric field are all accounted for in this step. These calculations are performed in Ref. [2] to produce an array of excitation rates $k^l_{ij}$ in various limits and values of metastable-atom density and electron density as functions of reduced electric field that can be used to predict the emission line intensity ratio. Note that these reaction rate constants are actually functions of the reduced electric field ($E/n_o$) that evolves in time and will be referred to as “excitation rates” henceforth. Equation (2) is greatly simplified if stepwise excitation of an upper state $i$ is negligible compared to direct excitation of the state – as is the case for the 3p5 state, which serves as the upper state of the 419.8nm emission. The line emission ratio $\rho$ can then be written as

$$\rho = 1 + \frac{n_m k^m_{420(\varepsilon)}}{n_o k^0_{419(\varepsilon)}},$$

(3)

because the ratio of excitation rates from the ground state is near unity for the emission lines considered here. [2] This expression for observed line ratio now implicitly depends on metastable-atom density, EEDF, and reduced electric field via the excitation rates.

**Experimental set-up**

The positive column studied in this work is produced by a 40µs-duration, 2.0kV-height square-wave voltage pulse at a repetition rate of 500Hz. Multiple diagnostic measurements were sampled synchronously at positions on the cylindrical axis of a 300mm Pyrex tube having a 2.54cm inner diameter. Diameter-averaged OES and diode-laser-absorption measurements were made perpendicular to the cylindrical axis in the positive-column region
of the discharge to probe the plasma by measuring the line emission ratio and metastable-atom density, respectively. Electron density measurements were made using a microwave-resonant cavity. Plasma current, measured by a LeCroy Waverunner digital storage oscilloscope, and the results of Pack et al. [5] are combined to infer the local value of reduced electric field \((E/n_o)\) via electron mobility. The behavior of the emission ratio throughout the discharge suggests the voltage pulse duration should be divided into three different stages. Most interestingly, after 20\(\mu\)s, the emission ratio remained around 3 and the electron density overtakes the metastable-atom density. The model predicts line-emission ratios for given values of electron density in the small metastable-atom density limit, the results of which are given in figure (1).

**The Predictive Nature of the Model**

The value of reduced electric field, \((E/n_o)\) electron density, and \((n_m)\) metastable-atom density, \((n_m)\) are combined with the numerical model [2] to infer the values of the excitation rates at the various time-steps throughout the pulse. Equation (3) predicts the observed line ratio \(\varrho\) once these rates have been inferred. Distinguishing where the various models agree with the experimental data may offer some insight into the experimental conditions.

The largest contrast between metastable-atom density and electron density is observed during the post-transient stage, 20\(\mu\)s < \(t\) < 40\(\mu\)s, of the discharge. As the electron density begins to overtake the metastable-atom density, we expect the excitation rates calculated in the low metastable-atom density limit (figure 3 of Ref. [2]) to become valid in predicting the line ratio. Upon calculating the predicted ratio in various cases of electron density, the experimental data is bounded by the traces for relative electron density \((n_e/n_o) = 10^{-6}\) (squares) and \((n_e/n_o) = 10^{-5}\) (triangles) as shown in figure (1). These excitation rates for each datum are then averaged to produce a fit approximating the case \((n_e/n_o) \approx 5 \times 10^{-6}\) (diamonds). The limiting cases of relative electron density, set by the model, agree with observation within the experimental determination of relative electron density of \((n_e/n_o) \approx 4 \times 10^{-6}\). Therefore, if one
is willing to assume the effect of metastable-atoms on the EEDF is negligible, the electron density can be written as a function of the observed emission ratio, $\varrho$, metastable-atom density, $n_m$, and reduced electric field ($E/n_0$), suggesting this model may be able to estimate electron density. Assuming the electron density, reduced electric field and observed emission ratio are known, this model may be used to determine metastable-atom density in the following manner.

As long as the metastable-atom density is sufficiently low, the ratio of excitation rates are no longer functions of metastable-atom density and equation (3) can be used to determine the normalized metastable-atom density as a function of reduced electric field and electron density. In that case, the observed line emission ratios, $\varrho$, can be combined with the excitation rates given in Ref [2] for a given electron density and reduced electric field to predict the metastable-atom density. Note that all three of these factors have an implicit time dependence. After 20$\mu$s, the electron density sufficiently exceeds the metastable-atom density and can be considered the dominant species in determining photon production routes in the plasma studied here within 40% agreement with of the diode laser absorption measurements. The average deviation from the diode laser absorption measurements, however, is less than 10% and the observed metastable-atom density via laser absorption agrees, within uncertainty, with the metastable-atom density predicted by the model.