Spatially-resolved modeling and measurements of metastable argon atoms in argon-helium microplasmas

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Microwave-driven plasmas operating near atmospheric pressure have been shown to be a promising technique for producing the high density of argon metastable atoms required for optically-pumped rare gas laser (OPRGL) systems. Stable microwave-driven plasmas can be generated at high pressures using microstrip-based resonator circuits. We present results from computational modeling and laser absorption measurements of argon metastable densities in such plasmas operating in argon-helium gas mixtures at pressures up to 300 Torr. The model and measurements resolve the plasma characteristics both perpendicular to the substrate surface and along the resonator length. The measurements qualitatively and in many aspects quantitatively confirm the accuracy of the model. The plasmas exhibit distinct behaviors depending on whether the operating gas is mostly argon or mostly helium. In high-argon plasmas the metastable density has a large peak value, but is confined very closely to the electrode surfaces as well as being reduced near the discharge gap itself. In contrast, metastable densities in high helium-fraction mixtures extend through most of the plasma. In all systems, increasing the power extends the region of metastable along the resonator length while the extent away from the substrate surface remains approximately constant.

I. INTRODUCTION

The optically-pumped rare gas laser (OPRGL) is a gas laser system based on the concept of using a metastable state of a rare gas atom as the lowest level of a quasi-3-level laser. Atoms in the np⁴(n+1)s¹ metastable level are excited to an upper np⁴(n+1)p level by absorption of pump laser photons. The upper p level rapidly populates a lower-energy p level via collisions with other atoms, creating a population inversion that allows for lasing on the p → s transition back to the original metastable level. Helium atoms provide efficient energy transfer collisions, eliminating the need for reactive chemical species inherent in the analogous diode-pumped alkali laser (DPAL) [1]. The use of a diode laser as the optical pump allows conversion of the power-efficient but typically divergent diode laser beam to a near-Gaussian output beam with high quantum efficiency. The OPRGL system has the potential for
scaling to high power levels while using only chemically inert gases at near-atmospheric pressures. The OPRGL was first demonstrated by Han et al. in pulsed operation [2], [3], with subsequent steady-state operation achieved by Rawlins et al. [4] using a microwave resonator discharge. The focus of much recent experimental and theoretical work has been on argon-based systems [5], [6], [7], but lasing schemes based on krypton and xenon metastable levels have also been studied [2].

Generating the rare gas metastable species required for the OPRGL requires an external process such as an electric discharge. In particular, a discharge at pressures on the order of atmospheric is required, as low-pressure discharges have not been observed to generate a sufficient density of metastables. Other effects such as faster collisional energy relaxation and pressure broadening of the pump laser absorption line also encourage operation at elevated pressures. Consequently, understanding the production and loss mechanisms of metastables in high-pressure plasmas is crucial for design studies of volume- and power-scaled OPRGL systems. In addition, the energy transfer kinetics, e.g. competition between collisional energy transfer with He and quenching by Ar, dictate operation in dilute mixtures of Ar in He [4,5], which have not been well studied in the literature. The present work continues our examination of metastable generation using microwave-driven microplasmas [8], which have shown great promise as a source of high densities of metastable atoms. We focus on the production of argon $1s_5$ metastables in argon-helium gas mixtures, as the laser kinetics of this system have received more study than other candidate mixtures.

Both the density and spatial distribution of metastable argon atoms are critical elements in the design and operation of argon-based OPRGL systems. The maximum achievable gain of the laser system is determined in part by the total number of Ar($1s_5$) atoms in the laser cavity, which in turn plays a large role in determining both the power requirements of the system as well as what laser resonator configurations can be used (i.e. stable vs. unstable). The localized density of these metastable atoms is one of the major factors in determining the laser optical gain per unit length, which in turn has a strong influence on the size requirements of the OPRGL. Finally, the shape of the region of high density determines how easily the pump laser beam can be matched to the gain region.

In this work we use a microwave resonator to generate the discharge that produces a metastable argon population. We examine the effects of total gas pressure, the fraction of argon in the argon-helium gas mixture, and
microwave power on the spatial distribution of Ar(1s$_5$) atoms in the discharge, in order to determine the degree to which this distribution can be adjusted to meet the needs of an OPRGL system.

Several previous studies have examined the spatial distribution of argon metastable atoms in discharges operating at hundreds of Torr. Belostotskiy et al. measured the metastable densities along the direction parallel to the electric field in a parallel-plate DC microdischarge [9]. Miura and Hopwood measured the metastable density transverse to the electric field through a cutout in the discharge gap of a split-ring microwave resonator discharge [10]. Both of those works examined discharges in pure argon, and found that neutral gas heating played a significant role in the metastable population. In contrast, the present work looks at discharges including significant fractions of helium, which have been shown to reduce the neutral gas temperature [8]. Crucially to the potential use of these discharges in an OPRGL system, we also measure metastable density simultaneously in the two dimensions which correspond to the spatial cross section for absorption of a pump laser beam. Niermann et al. have previously measured 2-D spatially-resolved metastable densities generated by a radio-frequency plasma jet operating in a 3%-argon, 97%-helium gas mixture [11] similar to one of the mixtures studied here. For the purposes of an OPRGL system, the Ar* densities produced by the RF jet in that work were several orders of magnitude too small, motivating our study of a microwave-driven plasma. Niermann et al. also measured and modeled metastable helium in a pure helium RF jet, noting that the metastable atoms reached peak densities near the sheath-bulk boundaries, similar to the thin layers observed here.

Numerous fluid modeling studies have examined high-pressure microplasmas in argon or helium (e.g. [12], [13]), but spatially-resolved models have not included operation in mixtures of the two gases. Gregório, Parsons, and Hopwood recently modeled a microwave resonator discharge in a nearly identical geometry to the current work at a range of pressures in pure argon [14]. More recently, interest in OPRGL systems has led to the development of several kinetic models describing energy transfer between argon excited states in an argon-helium mixture in order to examine the efficiency of the OPRGL system [6], [7]. Those models, while informative about laser performance, require assumptions about the discharge that produces the argon metastables, and do not provide spatially-resolved information that is important to real laser systems as discussed above. The present modeling effort includes two-dimensionally resolved species densities in argon-helium plasmas without assumptions about the discharge state, but tracks only a limited number of “lumped” excited states of argon and helium.
II. MODEL DESCRIPTION

A fluid model is developed to describe the spatial behavior of a microplasma. The physical model and the solution techniques are nearly identical to those described in Gregório et al. [14]. The model solves the continuity and momentum equations for each particle species, as well as the electron energy equation and Poisson’s equation. In employing Poisson’s equation, we use the electrostatic approximation since the microwave wavelength (~300 mm) is much larger than the plasma characteristic dimensions (~1-4 mm). The momentum equations are in the drift diffusion approximation for all species since the collision frequency for momentum transfer is always much larger than the excitation frequency (in [14] the momentum equation included temporal inertia effects which are neglected here). For each neutral species the momentum equation reduces to a classic diffusion equation. As in [14] we also solve the energy balance equation of the neutral background gas in order to determine the local neutral temperature everywhere in the domain. The gas density is computed with the ideal gas law. All heavy particles are assumed to be in thermal equilibrium with the background gas. No background flow is considered, and buoyancy effects are neglected.

We use the same plasma chemistry for argon-helium mixtures described in Ref. [8], itself based on reactions in Ref. [15], with a total of 30 reactions involving electrons, \( \text{Ar}^+, \text{Ar}_2^+, \text{He}^+, \text{HeAr}^+, \) and lumped levels representing the neutral argon 1\( s \) and 2\( p \) manifolds and all excited neutral helium states. We adopt the local mean energy approximation: the rate coefficients and the electron transport parameters are parametrized in time and space by the electron mean energy. These coefficients are obtained by suitable averaging over the electron energy distribution function computed using the freeware Boltzmann solver BOLSIG+ [16] with most cross-sections presented in Yanguas-Gil et al. [17] and extracted from LXCAT [18].

The model equations are solved in a two-dimensional Cartesian domain 16 mm wide and 10 mm tall, as pictured in Fig. 1 (note: not drawn to scale). The dielectric thickness (2.54 mm) and electrode gap size (100 µm) were chosen to match the experimental values. A sinusoidal voltage drop is applied between the two electrodes; the amplitude of this voltage is adjusted until the time-averaged total dissipated power matches the specified value. The model equations are solved given the geometric source dimensions, gas pressure, power absorbed by the plasma, and excitation frequency. All other quantities are computed self-consistently as a function of time and space without adjusting any parameters. The equations are discretized on a non-uniform rectilinear mesh with 200 by 200 grid points; higher grid densities resulted in nearly identical results. The mesh is refined around the gap region (where
gradients are steeper), with the finest mesh spacing being $29 \times 8 \, \mu\text{m}$ (width $\times$ height) at 100 Torr and $14 \times 4 \, \mu\text{m}$ at 300 Torr. The equations are integrated in time until a periodic steady state is reached.

![Model domain including coplanar electrodes separated by a 100 µm discharge gap, dielectric substrate, and ground electrode. Diagram is not to scale. The dashed rectangle indicates the approximate subset of the discharge region that is pictured in Fig. 4 and Fig. 6.](image)

**III. EXPERIMENTAL METHODS**

In this work we use a quarter-wave resonator to generate the plasma. The general design has been described elsewhere [19]. Briefly, the resonator consists of a microstrip transmission line grounded on one end and open-circuited on the other. A grounded electrode is placed 100 µm away from the open end, creating a discharge gap. Microwave power at the device resonant frequency (here 0.95 GHz) is supplied, resulting in a voltage drop across the discharge gap which breaks down the operating gas. Once the plasma forms, it typically expands beyond the gap. The quarter-wave resonator design can be arrayed to generate many microplasmas, as has been done in demonstrating OPRGL operation [4]. The resonator here was constructed by micro-milling a standard 2.5 mm thick dielectric substrate (Rogers TMM10) with copper plating on both sides, and is shown in Fig. 2. Power is supplied to the resonator from a standard microwave power train; see Hoskinson and Hopwood [20] for a full description of such a setup. The powers reported in this work are net powers, determined as the difference between the measured forward and reflected powers after correction for cable losses [21]. During experimental measurements, the quarter-wave resonator is placed into the same chamber as described in [8], which is continuously flushed at 500 sccm with ultra-high purity argon and helium in the desired ratio to maintain a high gas purity level. This level of gas flow results in linear gas velocities smaller than 0.1 m/s, small enough that it is not expected to affect the results beyond limiting impurities.
The densities of argon $1s_5$ metastable atoms are measured using tunable diode laser absorption spectroscopy (TDLAS). The setup and technique are similar to those described previously [10], and so are only summarized here. A schematic of the diagnostic system is shown in Fig. 3. A diode laser (ThorLabs L808P010) operating near 811.5 nm is expanded and aimed across the discharge gap of a resonator, as shown in Fig. 2. The beam passes through the plasma and a bandpass filter (passband: 810 ± 5 nm) before being imaged by a singlet lens at approximately unity magnification onto an intensified CCD camera (Princeton Instruments PI-MAX 1k). The laser wavelength is tuned through the argon $1s_5 \rightarrow 2p_9$ transition by modulating the diode drive current. The absolute laser wavelength is determined prior to the experiment using a 0.6 m spectrometer (SPEX 1269), while the relative variation in wavelength is monitored during the experiment using a Fabry-Perot interferometer (ThorLabs SA200-7A, 1.5 GHz free spectral range). The camera is gated open for a short time during this tuning sweep with a stepped delay time to obtain a series of images corresponding to different laser wavelengths. A ground-glass diffuser (ThorLabs DG10-600) is inserted in the beam path prior to the chamber to reduce the spatial coherence of the beam and thereby reduce interference fringes on the camera as the laser wavelength is modulated. While significantly mitigated by this technique, the remaining interference patterns (which shift as the laser frequency is scanned) are the largest source of noise in the experiment.

The $\text{Ar}(1s_5)$ density is estimated based on the absorption of laser light as it passes through the plasma. The optical depth $\tau$ of the plasma at each location is estimated as a function of the laser frequency as
\[ \tau(f) = \ln \left( \frac{I_0(f)}{\chi I_T(f)} \right), \]  

(1)

where \( I_0 \) is the intensity of the laser incident on the plasma and \( I_T \) is the laser intensity transmitted through the plasma. The factor \( \chi \) represents a correction for the refraction of the laser beam through the plasma: it is the fraction of transmitted light that reaches the detector where absorption is negligible (e.g. at frequencies far from the nominal absorption frequency). Such refraction results from a gradient in the gas density, and therefore the index of refraction, caused by plasma heating, and results in a shift of the baseline optical density.

The optical depth versus frequency determined using (1) is numerically fit with a Voigt profile. Due to the larger relative noise levels in the spatially-resolved data compared to single-point measurements [8] we do not attempt to estimate the gas temperature from the fit, and so do not constrain the Gaussian and Lorentzian broadening parameters to match particular mechanisms. The fitted offset parameter of the Voigt profile, which directly relates to the correction factor \( \chi \), is subtracted from the profile before integrating to obtain the density \( n_{1s} \) averaged over an effective absorption path length \( H \) [9]:

\[ \langle n_{1s} \rangle H = \frac{8\pi f_0^2}{A_{ul} c^2} \frac{g_l}{g_u} \int_{-\infty}^{+\infty} \tau(f) \, df. \]  

(2)

In this equation \( A_{ul} \) is the Einstein coefficient for spontaneous emission from the upper to lower levels, \( g_u \) and \( g_l \) are the degeneracies of those levels. To improve the robustness of the Voigt fit, we apply a modest nearest-neighbor smoothing routine to the recorded image, and bin the pixels into 2 × 2 blocks before fitting and integrating as in (2). The fitting and integration procedure is repeated for each “binned” pixel block in the recorded image. The resulting spatial resolution is approximately 35 µm × 35 µm, limited primarily by the residual laser coherence and associated thin-film diffraction in images as the wavelength is scanned. Locations where the Voigt fitting routine fails due to lack of a discernible absorption peak are treated as having zero metastable density. To estimate number densities from line-integrated densities, we take the absorption path length \( H \) to be the 1 mm resonator width, which in photographs is observed to typically restrict the plasma width.
IV. RESULTS AND DISCUSSION

In previous work [8], we noted distinct differences in plasma behavior between relatively low pressures (~100 Torr) and higher pressures. In the present work, we examine the discharge at both 100 and 300 Torr. At each total pressure level, we investigated two gas mixtures: an 80% argon / 20% helium mixture, and a 5% argon / 95% helium mixture. The former, high-argon, mixture represents the maximum argon fraction that in the experiments avoided filamentation of the plasma [22] at the tested pressures and power levels. The latter, low-argon, mixture represents a reasonable expectation for the operating condition of an argon-based OPRGL system [3], [4]. Higher helium content increases the rate of collisional \( \text{Ar}(1s_4) \rightarrow \text{Ar}(1s_5) \) de-excitation [3], allowing “recycling” of the produced \( \text{Ar}(1s_5) \) atoms in the lasing scheme. At 100 and 300 Torr, higher helium content up to approximately 98% was believed to increase the \( \text{Ar}(1s_5) \) density [8]; in the present work we demonstrate that this effect is at least partially due to differing spatial distributions of the argon metastables.

A. Model and Experimental Results at 100 Torr

Model predictions for \( \text{Ar}(1s_5) \) densities for the two gas mixtures and two power levels are shown in Fig. 4. Note that the plots display only a subset of the computational domain, chosen to match the scale of the experimental results. In the high-argon mixture, the region of high metastable density is largely confined to the area immediately adjacent to the electrodes. Over 70% of the total number of metastable atoms exist within 200 µm of the electrode surfaces in this mixture. The plasma itself, as discussed further in section I.C, extends much higher. At 0.5 W, it is evident that the metastable density peaks at the left and right sides of the plasma, far from the discharge gap where the density is lower. In contrast, in the low-argon mixture (right column of Fig. 4), the metastable atoms are spread throughout a significantly larger volume and their density peaks near the discharge gap and decreases near the left and right ends of the plasma.
Fig. 4. Modeled Ar* densities at 100 Torr for 0.10 W (top) and 0.5 W (bottom). The discharge gap is located at 
\(x = 0\). The modeled results have been corrected for the degeneracy of the 1s states.

Experimentally-measured line-averaged metastable densities at the same set of conditions are shown in Fig. 5. 
Here the resonator is on the right, the ground electrode on the left, and the discharge gap at \(x = 0\) mm. The 
measurements match the model results quite well, including the absolute values of the metastable densities, although 
the experimental resolution is not sufficient to resolve the predicted sharp density peak near the electrodes for 80% 
argon. The experiments do show that the region of high metastable density extends notably further away from the 
electrode surface in the 5% argon mixture. Particularly at 0.5 W, we can also observe that in the 80% argon mixture 
the metastable density is lower near the discharge gap than further away. These two effects largely explain the 
lower line-integrated metastable density we noted for higher argon fractions in a previous study [8], where the 
absorption path was vertically through the discharge gap. We note that for a given input power, the total numbers of 
metastable atoms are nearly identical between the two gas mixtures; it is only the spatial distribution that changes. 
This effect appears in both the experimental and model results.
Fig. 5. Line-averaged Ar (1s\textsubscript{5}) densities at 100 Torr for 0.10 W (top) and 0.50 W (bottom). The left column is for an 80\% argon / 20\% helium mixture, while the right column shows results for a 5\% argon / 95\% helium mixture. The discharge gap is located at x = 0.

**B. Model and Experimental Results at 300 Torr**

We have repeated the modeling and experiments at a total gas pressure of 300 Torr, with the results shown in Fig. 6 and Fig. 7, respectively. As the total gas pressure increases, the plasma size—including the region of highest Ar(1s\textsubscript{5}) density—is generally reduced as mean free paths fall. The localized peak Ar(1s\textsubscript{5}) densities do become larger, exceeding 10\textsuperscript{19} m\textsuperscript{-3} very near the electrode surfaces. As at 100 Torr, increasing the power largely increases the extent of the plasma horizontally along the electrode surfaces. Both at 100 and 300 Torr, the experimental-determined metastable densities exhibit significantly more spatial structure than the model. While a portion of this structure is attributable to laser interference as discussed above, the presence of convective gas currents and the associated temperature variations likely also play a role. The heavy-particle energy balance equation solved in the model includes only conductive heat transport. We note that measurements were also conducted at atmospheric pressure. The discharge and region of metastables shrink below their sizes at 300 Torr. Higher noise levels and lower spatial resolution (due to the smaller discharge size), however, make further conclusions difficult.
Fig. 6. Modeled Ar* densities at 300 Torr for 0.10 W (top) and 0.5 W (bottom). The discharge gap is located at $x = 0$. The modeled results have been corrected for the degeneracy of the 1s states.

Fig. 7. Line-averaged Ar (1s) densities at 300 Torr for 0.10 W (top) and 0.50 W (bottom). The left column is for an 80% argon / 20% helium mixture, while the right column shows results for a 5% argon / 95% helium mixture. The discharge gap is located at $x = 0$.

C. DISCUSSION

One of the more interesting differences between the high- and low-argon test mixtures is that the metastable density is confined to a thinner layer near the electrodes in the high-argon case. In contrast, the electron densities in both mixtures have relatively similar spatial distributions. These spatial distributions were not resolved in our previous 0-dimensional model [8]: the inclusion of spatial effects results in the current model predicting absolute argon metastable densities comparable to those observed experimentally, whereas the 0-D model significantly over-predicted these densities under all conditions. In both mixtures the model shows, as expected, the largest electron mean energies exist near the sheath-bulk boundary just above the electrodes and lower electron energies elsewhere. To investigate the cause of the difference in metastable distribution, we focus on a single location approximately
120 µm above the discharge gap in the 300 Torr, 0.5 W model results. At this position the metastable number density in the high-argon mixture \((n_{1s5} \approx 3 \times 10^{18} \text{ m}^{-3})\) has already fallen significantly below the value in the low-argon mixture \((n_{1s5} \approx 2 \times 10^{19} \text{ m}^{-3})\) despite having a somewhat higher mean electron energy (4.1 eV to 3.3 eV, respectively). This difference appears to be due to the differing non-equilibrium electron-energy distribution functions (EEDFs) in the two mixtures. When the neutral gas is primarily helium, the resulting EEDF has a much larger high-energy tail. In Fig. 8, we show the EEDFs calculated using BOLSIG+ (and used by the model to determine rate constants) for the modeled conditions at this location. The EEDF in the low-argon mixture, despite having a lower mean electron energy, has a much larger fraction of its electrons at energies where excitation of the metastable state is possible (see the plotted excitation cross section for comparison). As a result, more excitation occurs further from the electrodes in the low-argon mixture. While the variation in EEDF affects metastable excitation from the ground state most strongly due to that process’s large energy threshold (11.5 eV), other reactions are also affected. In particular, excitation from \(\text{Ar}(1s) \rightarrow \text{Ar}(2p)\) and ionization from the \(\text{Ar}(1s)\) state are both significant loss mechanisms for metastable argon. Because these processes have smaller threshold energies of 1.7 and 3.9 eV, respectively, they are relatively less affected by the absence of high-energy electrons in the 80% argon gas mixture.

Fig. 8. EEDFs calculated using BOLSIG+ for 5% argon and 80% argon for the modeled conditions 120 µm above the center of the discharge gap at 0.5 W and 300 Torr. Also shown is the ground state \(\rightarrow \text{Ar}(1s)\) excitation cross section for comparison.

Notably, as we increase the net microwave power the regions of high metastable density extend horizontally (along the electrodes), but remain nearly constant in vertical size (perpendicular to the substrate). The precise mechanisms for the horizontal-only expansion have not been fully characterized, but are believed to generally result from the co-planar electrode geometry employed here. Strong electric fields in the system largely exist only
between the electrodes and the bulk plasma. As the microwave power is increased, the potential on the electrodes increases, allowing plasma expansion along the electrode surface. Electric fields generally do not extend upwards as they are shielded by the plasma. The lack of vertical expansions is undesirable for an OPRGL system, as it increases the difficulty of matching the pump laser beam shape and alignment to the region of high metastable density. Alterations to the electrode geometry, such as dielectric limiters [23] or directed gas flow may be able to alter the metastable spatial distribution.

V. CONCLUSION

We have presented a series of measurement and model results for spatially-resolved argon metastable atoms produced by plasmas in argon-helium gas mixtures at total pressures of 100 and 300 Torr. The experimental results confirm the model outputs, showing quite similar absolute metastable densities and verifying that metastable atoms are largely confined near the electrode surfaces, particularly in high argon-fraction mixtures. Increasing the discharge power causes the metastables to expand along the electrode lengths, but they do not extend significantly further away from the surface. Increasing the extent of the metastable density in the direction perpendicular to the substrate surface may require changes to the electrode design and/or the introduction of directed gas flow.

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