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Mode transitions and hysteresis in inductively coupled plasmas

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Optical emission spectroscopy as a noninvasive plasma diagnostic was employed to study mode transitions and hysteresis in an inductively coupled plasma in Ar and Ar/N_2 mixtures. Using selected Ar lines, basic plasma parameters, relevant to the analysis of the mode transitions, were evaluated. Small changes of the electron energy distribution function in the vicinity of the mode transition were detected. The role of metastable Ar atoms in mode transitions and in a hysteresis was clarified. Enhanced production of metastables in the hysteresis region as well as faster transitions in plasmas with higher influence of metastables were observed. © *2007 American Institute of Physics*.
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I. INTRODUCTION

High-density inductively coupled plasma (ICP) reactors have found numerous applications in various etching or deposition processes.[1–5] Detailed knowledge of plasma formation mechanisms and ability to identify and control essential plasma parameters are required for a proper design and operation of these reactors. Two plasma modes are known to exist in ICP reactors.[6–17] At low power, a low density plasma is observed (*E* mode). As power rises, a fast transition occurs to the *H* mode characterized by much higher density and brightness of the plasma. The *E-H* transition can be triggered by changing either the applied power or some other parameters such as the gas pressure or gas composition. Under certain conditions, the back *H-E* transition can occur at lower powers so that a hysteresis is established.

Different mechanisms responsible for *E-H* transitions are discussed in a number of theoretical and experimental studies,[8–11] carried out mostly in Ar plasmas. However, many issues such as the transition dynamics, the hysteresis origin, and the possible role of Ar metastable atoms are still not well understood and need further investigations. This is important, in particular, for pulsed ICP sources, since the time scale for the transitions can be comparable with the time scale of the power supply.[10,12,15]

The approach discussed by Kortshagen *et al.*[8] and then by Cunge *et al.*[10] considers the transition mechanism based on a balance between absorbed and dissipated power. As the applied power rises in the *E*-mode plasma (when the capacitive coupling dominates), the induced electric field becomes strong enough for ionization. Then, the ionization rate exceeds the loss rate, power balance no longer holds, and the electron density n_e starts to rise. For low densities the inductive coupling efficiency grows linearly with n_e. Thus, more inductive power is absorbed resulting in more ionization and so on. This nonlinear mechanism resulting in fast n_e rise is operative until a skin effect becomes significant. As the plasma resistance and absorbed power now scale as $n_e^{-0.5}$, inductive power is absorbed more slowly, and a power balance is established in the *H* mode.

Another possible source of plasma nonlinearity is two-step ionization through metastable states.[9–11] At high enough density of metastables, the two-step process (with the rate proportional to n_e^2) can become the main source of ionization in the plasma.[13] The study of transition dynamics in a pulsed Ar discharge found characteristic times of tens of microseconds, comparable with those predicted by a simple kinetic model of two-step ionization involving Ar metastables.[11]

However, it is important to note that experimental verification of the contribution of various mechanisms to the mode transitions and hysteresis is rather complicated and results are frequently ambiguous. Most studies employ Langmuir probes for the analyses,[7,9–12] which are known to interfere with the plasma thus altering the transition conditions. Moreover, the transition dynamics is studied usually for pulsed discharges with a peak power much higher than that necessary for transitions under constant power conditions.[10–12,15] Evidently, this can change considerably the relative contribution of different mechanisms to the transition. Also, most studies of the mode transitions were performed without Faraday shield,[6,7,10–12,15–17] which could affect the results due to parasitic capacitive coupling. On the other hand, optical emission spectroscopy (OES) is a noninvasive diagnostic that is increasingly used for quantitative plasma analyses due to recent advances in development of multichannel detectors and continuous efforts in determination of more accurate cross sections for atomic processes in plasmas.[6,14,18,19] In particular, by using optical emission from several rare gases added in trace quantities to the plasma,

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Donnelly and co-workers[14,18] were able to determine main discharge parameters in electronegative technological plasmas with improved accuracy.

The objective of the present work was to apply OES as a diagnostic suitable for a noninvasive analysis of ICP mode transitions and hysteresis. Using selected argon line emissions, the effective electron temperatures and the metastable fraction (the ratio of the metastable Ar density to the ground state Ar density) were calculated for different plasma conditions. The following sections discuss the experimental procedure and model employed to calculate the plasma parameters and the measurement results.

II. EXPERIMENTAL PROCEDURE AND INTERPRETATION OF SPECTROSCOPY DATA

In order to evaluate the main plasma parameters various Ar line ratios can be utilized. The 425.9 nm/750.4 nm Ar line ratio is used to monitor changes in the effective temperature $T_{e,\mathrm{eff}}$ for high-energy electrons[19] (for a Maxwellian distribution, $T_{e,\mathrm{eff}}$ is equal to the electron temperature). The 425.9 and 750.4 nm emissions originate from the $3p_1$ and $2p_1$ levels, respectively, with a considerable energy separation of 1.26 eV. Both emissions are practically not influenced by radiation trapping or metastable states, so that the variations in their relative intensities are mainly due to variations in the electron temperature.[19] On the other hand, the 811.5 nm Ar emission is strongly influenced by the $1s_5$ metastable state.[19,20] This line was used here for the diagnostic of metastables. As shown by Czerwiec and Graves[6] the metastable concentration can be calculated measuring the intensities I of the 750.4 and 811.5 nm Ar lines,

$$\frac{I_{811}}{I_{750}} = C\left(\frac{k_{811}^{\mathrm{dir}} n_{\mathrm{Ar}} + k_{811}^{m} n_{\mathrm{Ar}}^{m}}{k_{750}^{\mathrm{dir}} n_{\mathrm{Ar}} + k_{750}^{m} n_{\mathrm{Ar}}^{m}}\right), \tag{1}$$

where n_{Ar} is the ground state Ar density, n_{Ar}^m the $1s_5$ metastable density, k^{dir} and k^m are the rate coefficients for excitation from the ground and metastable states, respectively, and C is the ratio of the spectrometer spectral sensitivity for the line wavelengths. The C factor was measured here using a calibrated tungsten band lamp. The k values were calculated using the data of Boffard *et al.*[19] and assuming a Maxwellian electron distribution that is mostly valid for the H mode with its higher electron density.[8] Actually, when Eq. (1) is solved it gives the metastable fraction $(n_{\mathrm{Ar}}^m/n_{\mathrm{Ar}})$.

Note that the 811.5 nm emission can be affected by radiation trapping.[20] However, similar results were obtained using a weaker 800.6 nm line (not shown), which is known to be much less influenced by trapping.

Finally, the electron density variation can be analyzed using the Ar 750.4 nm emission. If the effective electron temperature does not suffer strong changes, the ratio I_{750}/n_{Ar} is roughly proportional to the electron density[21] and can be used to monitor n_e relative changes. Further, n_{Ar} is proportional to the Ar partial pressure (p_{Ar}) only if the gas temperature is constant. The gas heating is known to be relatively small for E mode, but in H mode the heating can be considerable. Therefore, when the ratio I_{750}/p_{Ar} is used to monitor n_e, the method should give somewhat underestimated values for H mode. Thus the increase of electron density during the E-H transition is likely to be stronger than that of the I_{750}/p_{Ar} ratio, while the opposite is valid for the H-E transition.

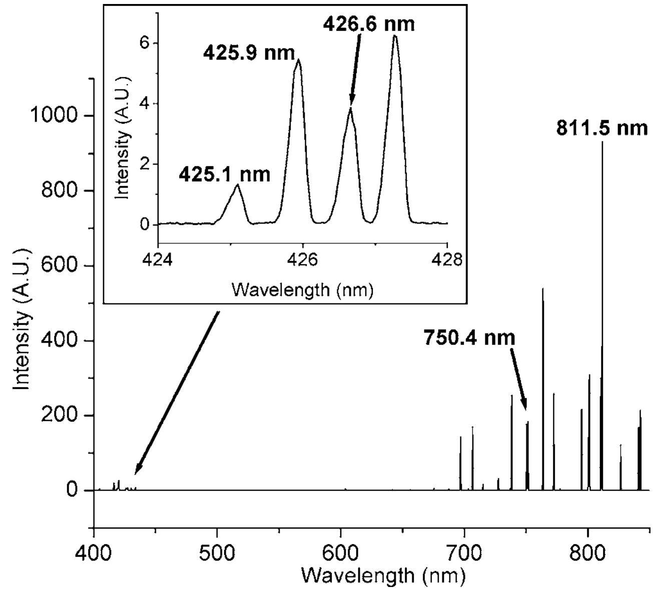

FIG. 1. Typical spectra for Ar plasma (100 mTorr, 100 W).

The measurements were performed in an rf (13.56 MHz) driven ICP discharge using Ar/N_2 gases, in a 30 cm diameter, 40 cm long vacuum chamber. A Faraday shield, consisting of radial copper stripes between a two-turn planar coil and a Pyrex window, was used to reduce capacitive coupling with the plasma. In separate experiments, a Langmuir probe was used to measure the electron density and temperature in the H mode.

A typical measured spectrum (using a Jobin-Yvon Sofie spectrometer, 35 cm focal length and a 1800 groove/mm grating) can be observed in Fig. 1. The Ar 811.5 nm emission, selected to monitor the metastable density, is the strongest of the spectrum. The Ar 750.4 nm emission is also one of the strongest in the spectrum. A number of weaker lines, originated from the $3p_x$ levels, can be observed in the 400–450 nm region, including the Ar 425.9 nm emission (used here to monitor $T_{e,\mathrm{eff}}$). However, even being much weaker than the other selected Ar emissions (mainly at higher pressures), the Ar 425.9 nm line can be recorded with a good accuracy, high signal to noise ratio, and no blending with close lines such as 425.1 and 426.6 nm (which can be influenced by radiation trapping).

III. RESULTS

Some experimental results are shown in Figs. 2–4.

In Fig. 2 it is seen that all the E-H transitions in Ar plasmas occur at approximately the same power of about 52 W. For the lowest measured pressure of 10 mTorr the transition occurs at power of 70 W (not shown) Wider hysteresis are observed for higher pressures (100 mTorr) and decreases for intermediate pressures (40 mTorr). For low pressures (≤20 mTorr), no hysteresis is observed.

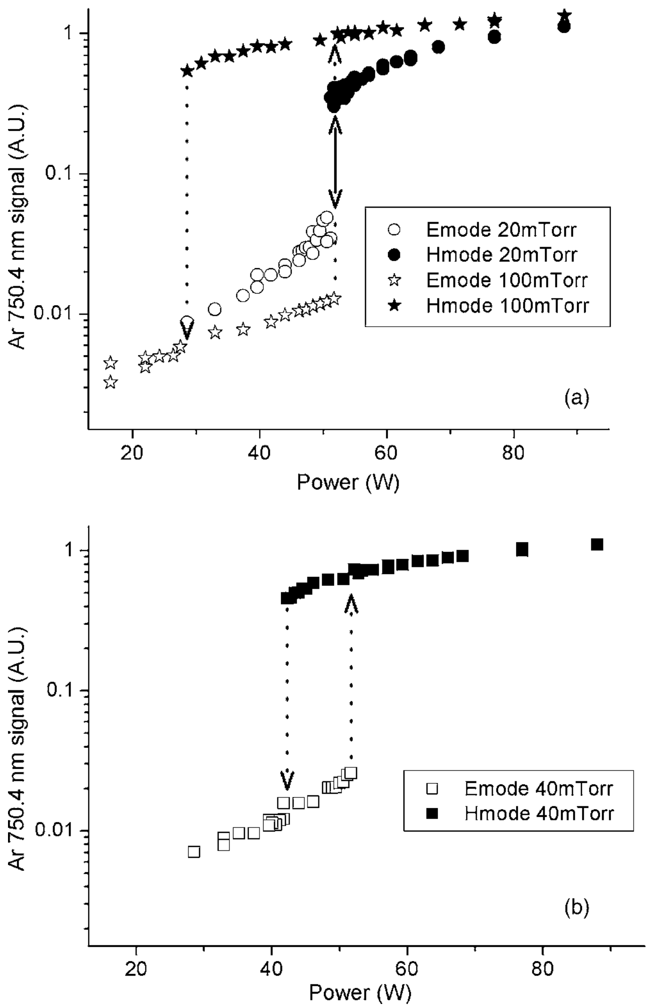

FIG. 2. Ar 750 nm line intensity in Ar plasma for (a) 20 and 100 mTorr and (b) 40 mTorr. Hollow and filled symbols correspond to E and H mode, respectively. The arrows indicate the transitions.

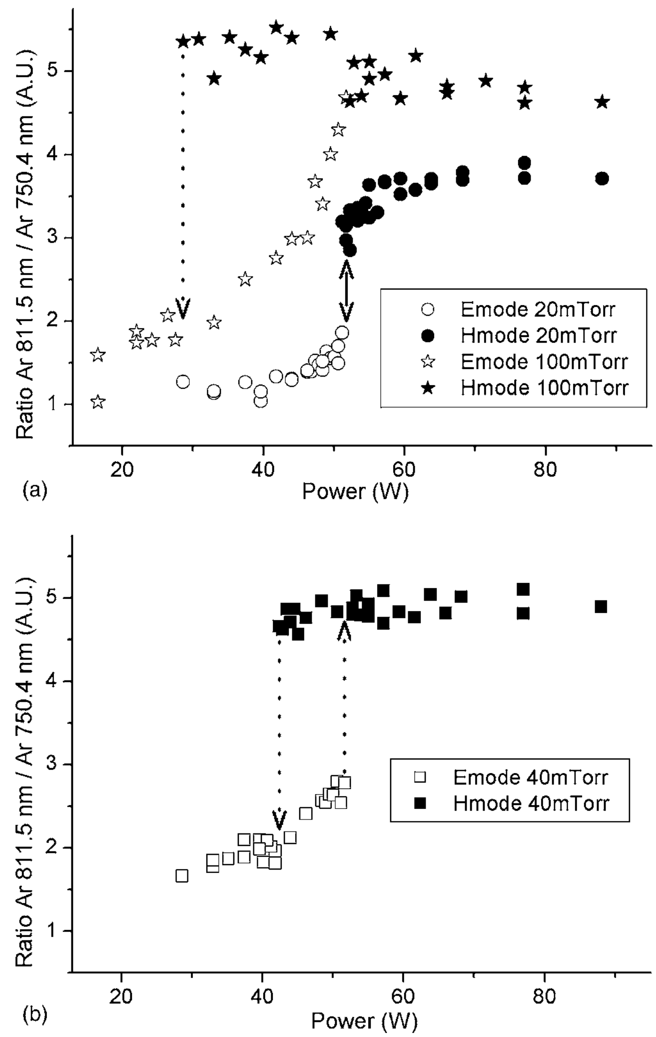

FIG. 3. 811.5 nm/750.4 nm Ar line ratios in Ar plasma for (a) 20 and 100 mTorr and (b) 40 mTorr. Hollow and filled symbols correspond to E and H mode, respectively.

In Fig. 3, increased generation of metastables (higher 811 nm/750 nm ratios) is evident for the H mode and higher pressures. Note that fast nonlinear rise of the ratio is observed just before the E-H transition for high pressures. For example, at 100 mTorr the ratio grows with power (P) as P^2 before the transition. On the other hand, for this pressure the Ar 750 nm line intensity (and hence the density of high-energy electrons) grows linearly with power before the transition [Fig. 2(a)]. Metastables are produced in collisions with high-energy electrons, and in the E mode the main losses are due to diffusion to chamber walls. Therefore, the metastable population is also expected to grow linearly with power in this mode.[6] The Ar 811 nm/750 nm ratio does not depend directly on total electron density n_e, see Eq. (1), but is sensitive to the changes in electron energy distribution function (EEDF) (through the rate coefficients k). Thus, the faster rise of the ratio just before the transition can be attributed to the growth of a low-energy fraction of plasma electrons, which can efficiently excite Ar metastables as the threshold is as low as 1.5 eV.[20] These electrons contribute to the increase of the Ar 811.5 nm line intensity but do not influence the Ar 750.4 nm line. In other words, this result indicates that the change of the EEDF shape from Druyvesteyn-like (flatter in the low-energy region) to Maxwellian-like starts *before* the mode transition, likely due to increasing ionization of metastables by low and medium energy electrons. This kind of EEDF transformation during the transition was observed by Cunge *et al.* using a Langmuir probe.[10] Therefore, this means that the 811 nm/750 nm ratio, under certain conditions, can also be used as a sensitive indicator of small changes in the EEDF, as a complementary diagnostic to Langmuir probes, being especially useful in situations where the probe data are not accurate in the low-energy part. In contrast to a "soft" change of the ratio for E-H transitions at high pressures (with higher contribution of metastables), abrupt changes are characteristic of H-E transitions independent of the pressure. This means that the change of EEDF from Maxwellian-like to Druyvesteyn-like happens abruptly during the H-E transition in Ar plasma.

In Fig. 4(a), the I_{750}/p_{Ar} ratio is plotted for different $\mathrm{Ar/N_2}$ mixture compositions. Estimations of $T_{e,\mathrm{eff}}$ were performed using both the line ratios and Langmuir probe data. The results show a weak $T_{e,\mathrm{eff}}$ dependence with power (as shown in Fig. 5), even in the transition region. The effective electron temperature was measured to be increasing with ni-

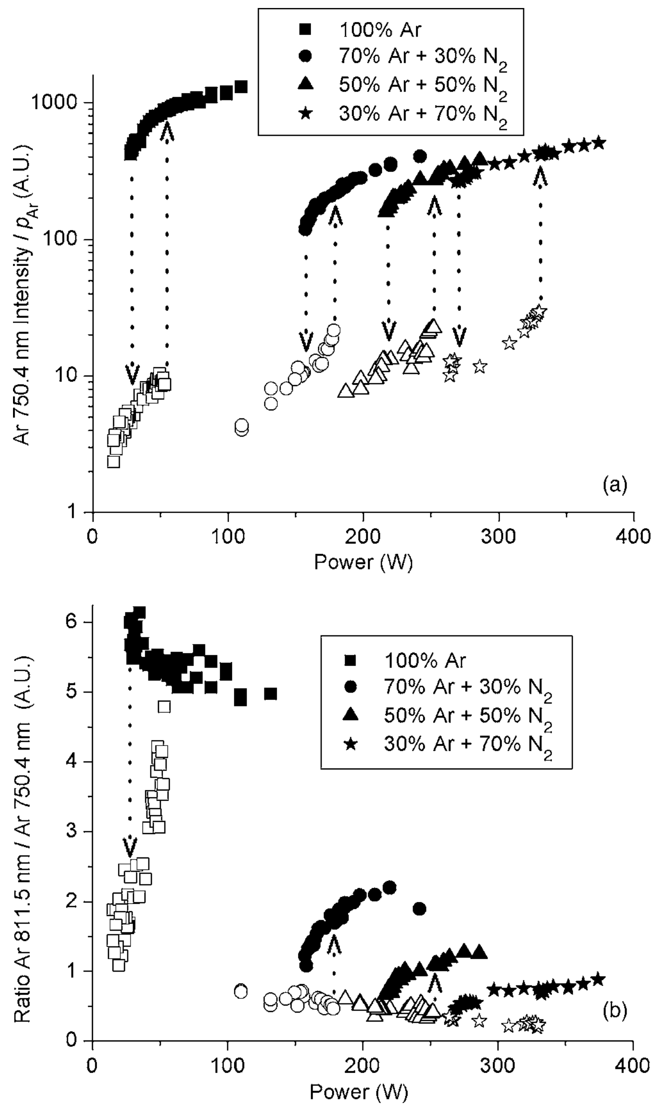

FIG. 4. (a) Ar 750.4 nm line intensities normalized by Ar partial pressure and (b) 811.5 nm/750.4 nm Ar line ratios for different Ar/N_2 mixtures (100 mTorr). Hollow and filled symbols correspond to E and H mode, respectively.

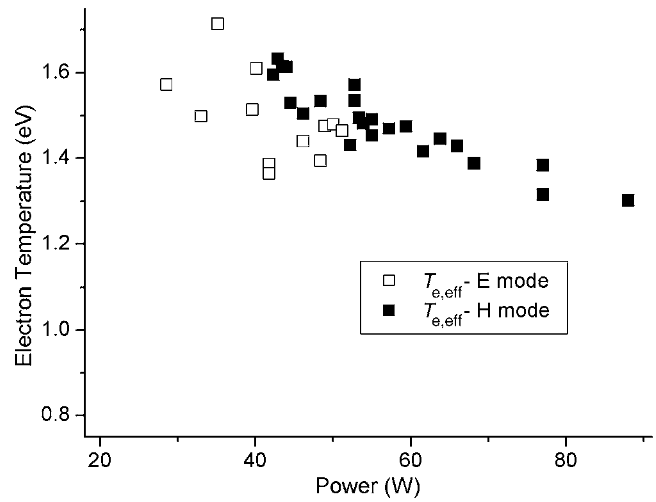

FIG. 5. Calculated effective electron temperatures (Ar: 40 mTorr).

trogen addition. However, according to both diagnostics, this $T_{e,\mathrm{eff}}$ variation does not exceed 10% as the nitrogen concentration changes from 30% to 70%. That means that for the Ar/N_2 mixtures shown in Fig. 4(a), the I_{750}/p_{Ar} ratio should be roughly proportional to n_e (as discussed in Sec. II). Therefore, while n_e jumps up to two orders of magnitude during E-H transitions in pure Ar, it rises only ~10 times in Ar/N_2 mixtures of the same pressure. The hysteresis width, determined as the ratio $(P_{\uparrow}-P_{\downarrow})/P_{\downarrow}$ where $P_{\uparrow}$ and $P_{\downarrow}$ are powers at up and down transitions, falls with N_2 addition from 0.5 to 0.15–0.2. It is also interesting to note that the I_{750}/p_{Ar} ratio (and thus the electron density) at the starting point of the E-H transition is practically the same for the three Ar/N_2 mixtures (and about twice for a pure Ar plasma). This fact strongly indicates the existence of a critical electron density necessary for the transition,[10] which depends only weakly on the gas composition.

In Fig. 4(b), lower I_{811}/I_{750} ratios were observed for Ar/N_2 mixtures indicating much lower densities of Ar metastables as compared with pure Ar plasmas. Reduction of n^m_{Ar} with nitrogen addition is expected, as N_2 is known to quench Ar metastables efficiently. Interestingly, the 811 nm/750 nm ratio behavior near the transition points for Ar/N_2 is opposite to that for Ar, with E-H transitions being abrupt and H-E ones more gradual. Again, this is consistent with the fact that Ar metastables has strong influence on the EEDF shape and are extremely important for the mode transitions in Ar plasma, while they are less abundant and thus have much less importance for the Ar/N_2 mixtures.

In order to estimate the metastable fractions $(n^m_{\mathrm{Ar}}/n_{\mathrm{Ar}})$ the OES data were used. The effective electron temperatures $T_{e,\mathrm{eff}}$ were determined from the ratios of 425.9 and 750.4 nm lines.[19] An example is shown in Fig. 5 for 40 mTorr. As discussed before, $T_{e,\mathrm{eff}}$ tends to decrease gradually with power but does not change notably during the mode transition. This result (the gradual decrease of electron temperature with power) was confirmed here by Langmuir probe measurements in special experiments, carried out in the H mode (see also Ref. 22). In contrast, abrupt changes in the electron temperature with the transition were reported in some experiments,[16,17] where Langmuir probes were employed for plasma characterization. However, in those experiments, no Faraday shield was used, and this might be responsible for the observed difference. Moreover, it should be noted that different parts of EEDF are characterized using Langmuir probes (Refs. 16 and 17) and emission spectroscopy (the present study). Next, Ar metastable fractions were estimated using Eq. (1) and the measured I_{811}/I_{750} ratios (Fig. 6). It should be noted that the accuracy of n^m_{Ar} estimates is better for the high-density mode, where EEDF is close to Maxwellian. For the E mode, the n^m_{Ar} values may be underestimated, as the contribution of low-energy electrons to excitation of the 811 nm line is smaller for the Druyvesteyn-like distribution as compared with the Maxwellian one at a given $T_{e,\mathrm{eff}}$. The important result is that for the H mode the metastable fraction grows considerably from $\sim 3\times 10^{-6}$ up to $\sim 2\times 10^{-5}$ as power decreases from 88 to 42 W. Just in the hysteresis region (52–42 W), this rise can be estimated to be about twofold. This is in contrast with predictions of a

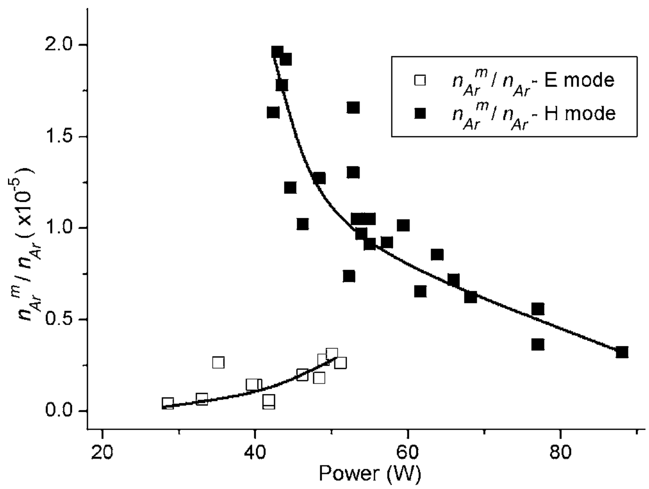

FIG. 6. Estimated Ar metastable fractions (Ar: 40 mTorr).

simple model based on the balance between metastable production and losses (in the H mode, due to quenching by electrons), resulting in $n_{\rm Ar}^m$ independent on power.[6]

This $n_{\rm Ar}^m$ rise in the hysteresis region, in part due to increasing $T_{e,\rm eff}$, may explain the origin of hysteresis. As power and n_e decrease, less metastable quenching occurs, and contribution of the two-step ionization becomes increasingly important thus maintaining the ionization balance sufficient to sustain the H mode. Our estimates (made using cross sections from Ref. 6 and references therein) indicate that for the case shown in Fig. 6 the H-E transition occurs when the relative contribution of two-step ionization falls below ~15%.

Similar results were obtained for measurements at other pressures, with a significant metastable fraction growth while approaching the H-E transition. As the pressure increases, a lower metastable fraction was estimated, as well as a lower $T_{e,\rm eff}$. This metastable behavior with the pressure was already observed by Kiehlbauch and Graves.[23] However, due to the smaller value of $T_{e,\rm eff}$ at higher pressures, the relative contribution of two-step ionization in these cases is higher. As the result, a higher contribution of metastable ionization as well a wider hysteresis was detected at 100 mTorr when compared with 40 mTorr. At a lower pressure of 20 mTorr, the relative contribution of two-step ionization in the H mode is lower than 10% and no hysteresis is observed.

This behavior of the metastable fraction with power and pressure was confirmed by laser induced fluorescence (LIF) in similar experiments carried out in a GEC reference cell adapted for an ICP discharge.[24] In these experiments, the $1s_5$ metastable state density was measured by LIF. The results show that the metastable density increases in the E mode and decreases in the H mode with power in the same manner as deduced from OES measurements (Fig. 6). In LIF measurements the highest metastable concentration was also observed in the hysteresis region, close to the H-E transition.

Dynamics of the mode transitions can give important complementary information on the role of metastables in the discharge and the underlying mechanisms. Characteristic times of the Ar line emission growth during E-H transitions ($\tau_{E\text{-}H}$) were measured to fall with Ar pressure, from 130 μs at 40 mTorr to 60 μs at 150 mTorr. Addition of N_2 to Ar had a relatively weak effect on $\tau_{E\text{-}H}$ at pressures smaller than 30 mTorr, although the threshold for transition rose considerably (Fig. 4). However, much longer transition times were detected for high pressures in Ar/N_2, being as long as 400 μs at 100 mTorr. These data support the assumption that Ar metastables contribute to the transition (accelerate the transition), as their contribution to total ionization increases with gas pressure and decreases with N_2 addition. Furthermore, the measured $\tau_{E\text{-}H}$ values are consistent with results of a model that includes two-step ionization, using an approach developed by Demidov *et al.*[11] This model predicts that a fast nonlinear rise of n_e (and hence $n_{\rm Ar}^m$) begins when critical electron and metastable densities are achieved in the E mode. In the early stage of this process, additional ionization due to metastables is no longer compensated by diffusion, which is a slow process. Further, when the critical plasma density for efficient absorption of inductive power is reached,[8,10] the mode transition process accelerates and then terminates with the establishment of a power balance. It should be emphasized that at a lower initial density of metastables or in their complete absence (e.g., when metastable quenchers like N_2 are added), the mode transition is also possible but at considerably higher powers. In this case, the mechanism of transition is that described in Refs. 8 and 10.

IV. CONCLUSIONS

OES has been shown to be a powerful noninvasive plasma diagnostic particularly suitable for studying mode transitions in ICP plasmas. Using selected Ar lines and their ratios, basic plasma parameters relevant to the analysis of the mode transitions in the plasma can be evaluated. The method proves to be sensitive for small changes in the EEDF observed in the vicinity of the E-H mode transition. A critical electron density necessary to trigger the mode transitions was identified. Finally, the role of metastables in triggering of mode transitions and establishing hysteresis has been clarified, with a maximum metastable fraction reached in the hysteresis region, and faster transitions are observed as the contribution of metastables to total ionization increases.

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