Absorption spectroscopy measuremente of argon metastable and resonant atomic densities in a low pressure Ar/N_2 ICP

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The influence of nitrogen concentrations ranging from 0 to 50% on the argon metastable and resonant atom density in a low pressure (up to 20 mTorr) inductively coupled plasma has been studied by means of absorption spectroscopy. The increase of resonant atom density with pressure is related to the radiation trapping at higher pressures, while the decrease of both resonant and metastable atom densities with increasing nitrogen content is due to the increasing importance of excitation transfer processes between argon excited atoms and nitrogen molecules.

Over the last few decades, several materials processing strategies using low-temperature plasmas have been developed to fullfil the increasingly needs of many industries, including microelectronics and photonics, the performance of these processes depending strongly on the plasma characteristics. In particular, in the case of a Ar/N_2 inductively coupled plasmas (ICP), the density of Ar metastable and resonant atoms play a key role in the discharge kinetics even at low pressure [1], influencing the degree of dissociation of N_2 molecules and thus the production of nitrogen atom, which play a central role in the surface chemistry for various thin film deposition and etching processes.

In our experiments, the plasma was generated in an ICP cylindrical reactor (customized Oxford Plasmalab 100 model ICP380). The chamber consists of an upper 25 cm high alumina tube in which the plasma is generated and a 15 cm high spacer made of anodized aluminum and equipped with several diagnostic ports. The plasma is generated at a frequency of 2 MHz with power levels up to 5000 W. In the present experiments, the ICP power was maintained at 1000 W. Several Ar-N_2 gas mixtures of high-purity (>99.998%) were used as plasma gas keeping the total flow rate at 20 sccm, allowing the introduction of several gas mixtures with N_2 concentrations ranging 0 – 50 %.

In order to measure the metastable and resonant density, a spectral argon lamp (Philips 91300E) was placed in front of a port facing a spectrometer located on the other side of the plasma. The lamp emission was collected by a 10 cm lens placed near the entrance of the chamber to force a parallel beam. A second, 22 cm focal length lens located on the other side of the chamber was used to refocus the lamp light on the entrance slit of a 750 mm focal length spectrometer equipped with a 3600 grooves/mm grating and an ICCD camera. When both lamp and plasma are on, the signal ($I_{Plasma+Lamp}$) collected by the ICCD is composed of the lamp signal partially absorbed by the plasma and of the plasma emission. The global optical absorption (A_L) can then be calculated as

$$A_L = 1 - \frac{I_t}{I_0} = 1 - \frac{I_{Plasma+Lamp} - I_{Plasma}}{I_{Lamp}} \tag{1}$$

where I_{Plasma} is the plasma emission with the lamp off and I_{Lamp} is the lamp emission with the plasma off. Several lines were used to measure the densities of the $^3P_2$ (696.54, 801.47 and 811.53 nm), $^3P_1$ (800.61, 810.36, 842.46 nm), $^3P_0$ (794.81 nm) and $^1P_1$ (826.45, 840.82, 852.14 nm) levels under different pressure conditions (1 to 20 mTorr) and nitrogen concentration (0 to 50%). A detailed description of the absorption method can be found, for instance, in [2].

In Figure 1, the variation of the density of the Ar metastable and resonant levels with pressure is shown for different nitrogen concentrations in the discharge. As can be seen, the metastable density is dominated by the $^3P_2$ level by about one order of magnitude. In contrast, for resonant levels, both $^3P_1$ and $^1P_1$ contribute in equal fractions. In addition, while the density of resonant levels increases with pressure, that of metastable levels weakly depends of it. The behaviour of the resonant level densities is opposite to that found in [1]. However, in the later study the pressures considered were varying from 20 to 200 mTorr, which is significantly higher than those of the present work. A similar behaviour of the resonant atoms with pressure has been found in RF discharge (13.56 MHz) in [3] in the same pressure range (5 to 50 mTorr) as a consequence of the increase of radiation trapping with pressure. Therefore, the present results demonstrate that the depedence of the resonant atom density is determined by the pressure rather than by the detailed electromagnetic field structure and frequency.

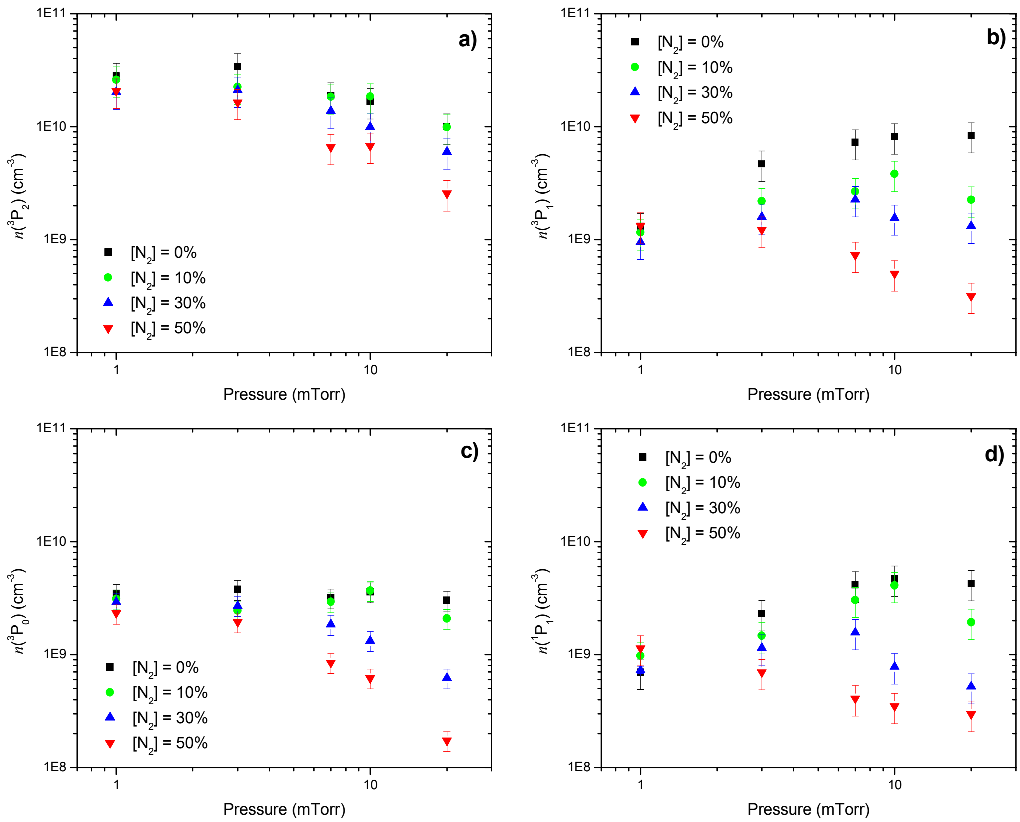

Fig. 1: Variation of the argon metastable ((a): $^3P_2$; (c): $^3P_0$) and resonant ((b): $^3P_1$; (d): $^1P_1$) densities with pressure for different nitrogen concentration.

As the nitrogen content increases, both metastable and resonant densities decrease with increasing pressure, with a more pronounced effect in the case of metastable atoms. For example, a nitrogen concentration of 50% results in a decrease of the metastable atom density decreases by a factor of ten from 1 to 20 mTorr . In the same conditions, the resonant atoms density is reduced by a factor of 3 . This decrease can certainly be related to the variations in electron density and temperature. However, it has been previously reported in [1] that the enhanced importance of excitation transfer processes (Eq. (2)) involving ground state nitrogen molecules at higher pressures play a significant role in depopulating Ar 4s levels to populate N2($C^3\Pi_u$) states.

$$N_2\left(X^1\Sigma_g^+\right) + Ar(4s) \rightarrow N_2\left(C^3\Pi_u\right) + Ar(4s) \qquad (2)$$

References

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