THE M-EFFECT IN ARGON-HYDROGEN GAS MIXTURES

G. Musa*, C. L. Ciobotaru, P. Chiru, A. Baltog

Low Temperature Plasma Laboratory, National Institute for Lasers, Plasma and Radiation Physics, MG-36, 6900, Bucharest

The decrease of the emission spectra of an electronegative-electropositive gas mixture discharge to only one line in the case of Ne-H₂ gas mixture was explained in our previous published papers [1]-[14]. In the present paper, the general character of this behavior is outlined. Also, similar results for Ar-H₂ mixtures discharges are reported. In this case, at least two different argon lines were found to increase and give rise to the M-effect.

(Received January 23, 2004; accepted June 3, 2004)

Keywords: Gas discharge, Ne-H2 gas, Monochromatisation-effect

1. Introduction

In a number of previously published papers we reported that, on hydrogen addition to neon, reduction of neon emission spectra to practically one line was observed. The wavelength of this line was $\lambda_1 = 585.3$ nm and corresponds to the $2p_1$ - $1s_2$ transition [1]-[14]. Due to the fact that the spectra obtained had practically one line, this phenomenon was called the Monochromatisation-effect (Meffect).

In order to characterize the "intensity" of this effect, we introduced the M parameter defined as the relative intensity ratio of the increased single line and an arbitrary reference line. For neon, the two lines are $\lambda_1 = 585.3$ nm and $\lambda_2 = 614.3$ nm and the M parameter is:

$$M = \frac{I_{\lambda_1 = 585.3nm}}{I_{\lambda_2 = 614.3nm}} \tag{1}$$

In the case of pure neon discharges, the value of the M parameter is of the order of a few units, whereas at 40% hydrogen content in the neon-hydrogen gas mixture, a value as high as 40 was found. These results were obtained for a dielectric barrier discharge in Ne+ H_2 mixture at pressures around 10^{+2} torr.

The emission spectrum of a discharge in pure neon is presented in Fig. 1. In Fig. 2, the spectra of 70% Ne +30% H₂ mixture discharge under identical experimental conditions is shown.

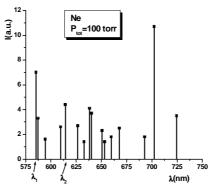


Fig. 1. Recorded spectrum of pure neon PDP discharge ($\lambda_1 = 585.3$ nm, $\lambda_2 = 614.3$ nm).

^{*} Corresponding author: musa@alpha1.infim.ro

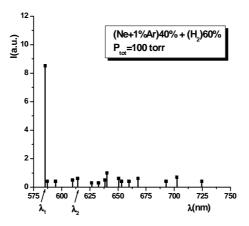


Fig. 2. Recorded spectrum of (Ne+1%Ar)40% + (H₂)60% gas mixture PDP discharge $(\lambda_1 = 585.3 \text{ nm}, \lambda_2 = 614.3 \text{ nm}).$

The M-effect can be observed in various types of discharges. Extensive studies on the M-effect were necessary for explaining it, as can be observed in references [1]-[14]. It can be concluded that two processes are responsible for the appearance of this effect:

- 1. ion-ion recombination, a process with high cross-section [15]
- 2. resonant three body collision reaction of heavy particles (with nearly zero energy defect) [16]-[35].

Being observed in various types of discharges and also in various electronegative-electropositive gas mixtures, the M-effect is considered to have a general character [18]-[35]. A number of experimental conditions are necessary in order to obtain the M-effect, namely:

- low gas temperature in order to increase the triple collision reaction cross-section
- elevated pressure of the gas mixture in order to promote triple collisions between heavy particles
- high density of the negative ions. In order to maintain high density of the negative ions, the discharge must be generated in electronegative-electropositive gas mixtures. Low electric field in the plasma and high electron densities can increase the density of negative ions.

In the present paper we report for the first time our results on the M-effect in argon-hydrogen gas mixtures. The peculiarity of the M-effect observed in this case is that there is a number of emission lines with increased intensity, not just one. In the cases of Ne mixed with H_2 or Cl_2 or O_2 , only the line at $\lambda_1 = 585.3$ nm was observed to be selectively emitted.

2. Experimental arrangement

The A.C. dielectric barrier discharge in argon-hydrogen gas mixture was produced between two thin, parallel and linear aluminium electrodes of 5 mm width and 200 mm length. Both electrodes were covered with 20 μm dielectric layer. The distance between the electrodes (i.e. discharge space) was 1 mm. The thin film conductors were obtained by deposition in vacuum through a mask on two glass plates. The electrical supply was an A.C. square wave voltage generator with a frequency of 10-50 kHz.

The PDP discharge device can be pumped down and filled with various gas mixtures. The optical emission spectra of the discharge were obtained using VARIAN TECHTRON spectrometer, a RCA photomultiplier and a recorder.

3. Experimental results and discussion

In Fig. 3, the spectrum of the most intense lines emitted by the dielectric barrier type discharge in pure argon is given. In Fig. 4, the emission spectrum of $15\%H_2+85\%$ Ar discharge taken under identical conditions are presented. Comparison of these two spectra clearly shows appearance of the M-effect which consists on selective supply of a limited number of different energy levels of argon atoms.

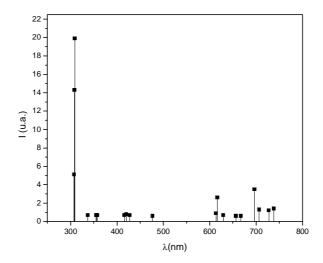


Fig. 3. The emission spectra of a dielectric barrier type discharge in pure argon (total pressure 30 torr).

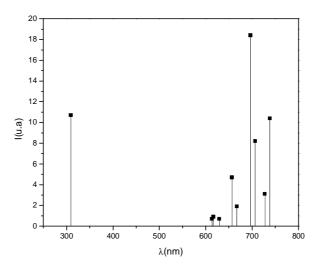


Fig. 4. The emission spectra of a dielectric barrier type discharge in $(85\% Ar + 15\% H_2)$ gas mixture (total pressure 30 torr).

In the case of $Ar + H_2$ mixtures, the M parameter was defined similarly:

$$M = \frac{I_{\lambda_n}}{I_{\lambda_3 = 614.3nm}} \tag{2}$$

where $I_{\lambda n}$ is the intensity of one of the few argon lines presenting increased intensity. In equation (2), λ_3 is a reference argon line different from the λ_n lines. In our case, we used $\lambda_3 = 616.3$ nm as argon reference line.

The M - parameter was plotted against percentage of the added H_2 to Ar, at a constant total pressure of 30 torr.

In Fig. 5, the M-effect for the argon line $\lambda_4 = 738$, 4 nm is given. An increase of the M parameter on the percentage of added H_2 to argon was observed. In this figure, a maximum at M = 21 can be observed for 15% H_2 from total pressure.

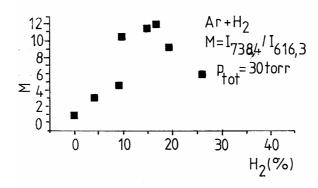


Fig. 5. The dependence of the M parameter value on the percentage of added H_2 from the total pressure of Ar+H₂ gas in a PDP type discharge ($\lambda = 738.4$ nm)

A similar dependence for the argon line at $\lambda_5 = 696.5$ nm was also observed (Fig. 6). In this case, a maximum value of 12 for M, corresponding to 17% H₂ was observed.

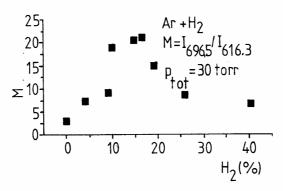


Fig. 6. Dependence of the M parameter value on the percentage of added H_2 from the total pressure of Ar+ H_2 gas filling ($\lambda = 696.5$ nm).

As shown in our previous papers, the M-effect is due to three body collision reactions of ionized and excited atoms [17]. In the case of $Ar + H_2$ mixture, the equation of the reaction can be written as:

$$Ar^{+} + H + H^{*}(n=2) \rightarrow Ar^{*} + H^{*}(n=3) + H$$
 (3)

with nearly zero energy defect.

In this equation, the excited hydrogen atoms $H^*(n = 2)$ in the left hand side behave as metastable atoms due to hydrogen resonant radiation trapping at pressures higher than 10 torr. On the right hand side of this equation, excited argon and excited hydrogen atoms appear.

4. Conclusions

Selective population of a number of energy levels of Ar in the case of discharges in $Ar+H_2$ gas mixtures at pressures of the order of $10-10^2$ torr is reported in this paper.

The M-effect proved to be an important collision process in plasmas. This effect can disturb the usual distribution of excited states. Consequently, we believe that currently published data for discharges taking place at pressures higher than 10 torr in electropositive-electronegative gas mixtures might be erroneous in the case of gas mixtures where the M-effect occurs.

References

- [1] G. Musa, A. Popescu, A. Baltog, I. Mustata, N. Niculescu, C. P. Lungu, Revue Romaine de Physique **26**, 125-134 (1981).
- [2] G. Musa, A. Popescu, A. Baltog, L. Nastase, N. Niculescu, I. Mustata, A. Cormos, C. P. Lungu, Proc. XV-th Int. Conf. on Ionized Gases- ICPIG, Minsk, 1607-1608, (1983).
- [3] G. Musa, A. Popescu, A. Baltog, I. Mustata, Journal of Physics, D Applied Physics 18, 2119 (1985).
- [4] G. Musa, I. Mustata, A. Popescu, A. Baltog, C. P. Lungu, A. M. Oancea, Proc. XVII-th Int. Conf. on Ionized Gases- ICPIG, Budapesta, (1985).
- [5] G. Musa, A. Popescu, A. Baltog, C. P. Lungu, I. Mustata, Proc. XX-th Int. Conf. on Ionized Gases- ICPIG, Pisa, (1991).
- [6] G. Musa, C. P. Lungu, A. Popescu, A. Baltog, IEICE Transaction on Electronics E **75-C**, 241-245 (1992), Japan.
- [7] G. Musa, A. Popescu, A. Baltog, C. P. Lungu, Romanian Reports in Physics **45**, 287-341 (1993) (revue paper).
- [8] G. Musa, A. Baltog, C. P. Lungu, Proc. XXIII-th Int. Conf. on Ionized Gases- ICPIG, Toulouse 94-95 (1997).
- [9] G. Musa, A. Baltog, G. Bajeu, C. P. Lungu, R Raiciu, I. Borcoman, A. Ricard, Romanian Reports in Physics **49**, 165-169 (1998).
- [10] G. Musa, A. Baltog, G. Bajeu, C. P. Lungu, R. Raiciu, I. Borcoman, A. Ricard; The European Physical Journal. Applied Physics. 4, 165-169 (1998).
- [11] G. Musa, A. Baltog, R. Raiciu, Proc. XIV-th Int. Conf. ESCAMPIG, Malahide 1998, p.86-87, 12, A. Baltog, R. Raiciu, G. Musa, Proc. XXIV-th Int. Conf. on Ionized Gases-ICPIG, Warsaw, 1999
- [12] M. A. Lieberman, A. J. Lichtenberg; Principles of plasma discharges and material processing, John Wiley & Sons, N.Y. (1994).
- [13] A. Baltog, R Raiciu, G. Musa, Contribution to Plasma Physics, 40, 537-544 (2000).
- [14] A. Baltog, G. Musa, Proc. XXV-th Int. Conf. on Ionized Gases- ICPIG, Nagoya, (2001).
- [15] A. Baltog, G. Musa, Contributions to Plasma Physics (submitted to publication).
- [16] G. Musa, L. C. Ciobotaru, A. Baltog P. Chiru, Contributions to Plasma Physics (submitted to publication).
- [17] Byoung-Kuk Min, S-H. Lee, Hun-Gun Park; J. Vac. Sci. Technol. 18, 349-355 (2000).
- [18] N. P. Curran, M. B. Hopkins, D. Vender, B. W. James, Plasma Sources Sci. Technology 9, 169-175, (2000).
- [19] J. J. Thompson, Phil. Mag. 47, 337 (1924).
- [20] Tae Hyuk Ahn, K. Nakamura, H. Sugai, Jpn. J. Appl. Phys. 34, L1405-L1408 (1995).
- [21] D. Hayashi, K. Dakota, J. Appl. Phys. 83, 697-702 (1998).
- [22] H. M. Katsch, E. Quandt; J. Phys. D. Appl. Physics **25**, 430-435 (1992).
- [23] T. Mosbach, H. M. Katsch, H. F. Dobele, Plasma Sources Science Technol. 7, 75-81 (1998).
- [24] K. N. Mellon, B. P. Coonan, M. B. Hopkins, J. Phys. D. Appl. Physics 28, 473-478 (1995).
- [25] L. Landau, Phys. Z. Sowjet. 2, 46-51 (1932).
- [26] C. Zener, Proc. Roy Soc. A 137, 696-702 (1932).
- [27] T. D. Gaily, M. F. A. Harrison; J. Phys. B Atom Molec. Phys. 3, 1098-1104 (1970).
- [28] R. D. Rundel, K. I. Aitkin, M. F. A. Harrison, J. Phys. B Atom Molec. Phys 2, 954-965 (1969).

- [29] W. H. Aberth, J. R. Peterson, Phys. Rev. 1, 158-165 (1970).
- [30] R. E. Olson, J. R. Peterson, J. Moseley, J. Chem. Phys. 53, 3391-3397 (1970).
- [31] J. R. Peterson, W. H. Aberth, J. T. Moseley, J. R. Sheridon, Phys. Rev A 3, 1651-1657 (1971).
- [32] J. Weiner, W. B. Peatman, R. S. Berny, Phys. Rev. A 4, 1824-1834 (1971).
- [33] P. A. Pogarelli, A. M. Shukhtin, Optica Spectrosc. 28, 134-137 (1971).
- [34] I. V. Komarov, P. A. Pogorelli, A. S. Tibilov; Optic. Spectrosc. 27, 198-203 (1969).
- [35] V. Mihailov, V. Ghenceva, R. Djulgerova, J. Phys D. Appl. Phys. 34, 2185-2190 (2001).