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TENTATIVE EXPLANATION OF SELECTIVE POPULATION OF THE $2p_1$ LEVEL OF NEON ATOMS IN THE CASE OF M - EFFECT IN *NEON+ HYDROGEN* AND *NEON+OXYGEN* GAS MIXTURES

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In electropozitive-electronegative gas mixtures discharges, the selective population of a certain level has been previously observed, this process being called the M-effect. This behaviour was explained by ion-ion recombination. However, the selective population of a certain level was not explained before. In this paper, we report on a tentative explanation of the selectivity as being due to energy resonant recombination process of a three body collision between negative and positive ions and a metastable electronegative atom. Comparisons of the spectra of $Ne+H_2$ and $Ne+O_2$ with the spectrum of pure neon are presented as an example. A single strongly enhanced line was observed in the spectra of the two gas mixtures. The energy defect of the reaction was calculated to be less than ± 0.2 eV. Various other gas mixtures containing electronegative and electropositive atoms are expected to give rise to the M –effect.

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1. Introduction

In a number of previously published papers we reported the possibility of obtaining nearly monochrome radiation spectra using discharges in multiple gas mixtures at pressures in the range 10 $–10^2$ torr. This effect of radiation monochromatisation was called the M-effect [1-4].

In order to characterize this effect we introduced the M parameter defined as the ratio of the relative intensities of the radiation of two chosen wavelengths, here λ_1 = 585.3 nm and λ_2 =614.3 nm, i.e.

$$M = I\,(\lambda_1{=}585.3\text{nm})I\,(\lambda_2{=}614.3\text{ nm}) \tag{1}$$

Using the M parameter, various recorded spectra can be compared and the value will be a measure of the M-effect "intensity".

Two comparative spectra of a narrow gap (0.15 mm) PDP type discharge at 100 torr in pure Ne and also in $(Ne+1\%Ar)50\%+(H_2)50\%$ are presented in Figs. 1 and 2, respectively. We notice the fact that the two spectra are plotted using the same arbitrary units on Y-scale. A dramatic change of spectra can be observed.

Extensive studies of the M-effect allowed identification of its parameters and determination of the nature of the gas mixtures for which the effect can be observed.

A summary of these results is presented in the following:

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- The gas mixture must contain at least one electronegative and one electropositive gas
- The M-effect increases with increasing of the percentage of the electronegative gas up to 50% in the total gas mixture.
- The M-effect increases with the total gas pressure
- The M-effect is sensitive to the plasma conditions, especially to those related to generation and maintenance of negative ions.

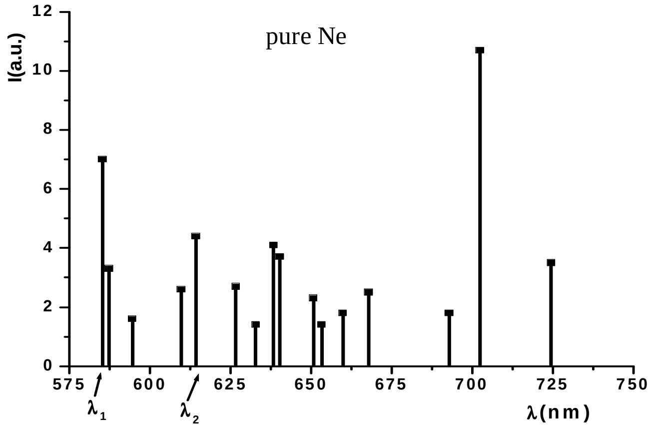

Fig. 1. The spectrum of pure neon in a PDP type discharge (λ_1=585.3nm, λ_2=614.3nm) at a total pressure of 100 torr.

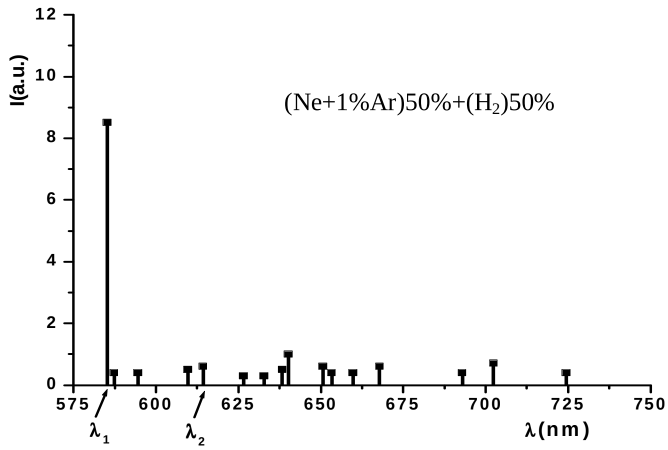

Fig. 2. The spectrum of (Ne+1%Ar)50% + (H_2)50% gas mixture in a PDP type discharge (λ_1=585.3nm, λ_2=614.3nm) at a total pressure of 100 torr.

This behaviour are closely related to the presence of the negative ions and therefore we concluded that the M-effect is due to the ion-ion recombination [5-15]. Recently was reported that this process has a large cross section [16].

The M-effect could not be explained by processes like dissociative recombination or radiative recombination of molecular neon-ions or atomic neon - ions with electrons. All the experimental data obtained for the M-effect are in good agreement with the hypothesis of ion-ion recombination. Thus an important advancement towards explaining the M effect was achieved.

However, the ion-ion recombination can not explain why after the recombination process only the neon level $2p_1$ is filled while all other levels from $2p_2$ down to 2 p_{10} remain less populated.

In the present paper we give a tentative explanation of the M-effect including selective population of the $(2p_1)$ level of neon.

Let us consider the three body reaction, which can be written as:

$$Ne^+ + H^- + P \longrightarrow Ne^*(2p_1) + \text{other products} \quad (2)$$

where P is the third reacting particle, in an excited state too.

The probability of such a reaction depends strongly on the energy balance between the right and left side of the equation (2), respectively. The probability of reaction (2) will decrease quickly with the increase of the energy defect, ΔE.

The following the three-body reaction can explain selective population of $2p_1$ level of neon:

$$Ne^+ + H^- + H^*(2p) \longrightarrow (Ne^+ H^-) + H^*(2p) \longrightarrow Ne^*(2p_1) + H^*(3s) + H(1s) \pm \Delta E \quad (3)$$

At the left hand side of this equation is written the recombination of a positive ion-negative ion the third body being atomic hydrogen excited on resonant level $2p\,^2P^0_{3/2}$.

The probability of Ne^+ collisions with H^- is high due to the coulombian attractive forces. The density of the hydrogen atoms excited on resonant level at hydrogen partial gas pressures of over a few torrs is comparable to the density of metastable atoms in low temperature plasma. Indeed, the lifetime of $H^*(2p)$ will be comparable with the lifetime of the metastable atoms due to the process of capture of the radiation $H\alpha$ at gas pressures higher than 10 Torr.

As observed in the case of M effect, the resulting particles of this reaction are the excited neon atoms $Ne^*(2p_1)$, the excited hydrogen atoms $H^*(3s)$ and atomic hydrogen. In this last equation ΔE represents the energy defect, namely the difference between the energies of the particles in the left hand side of the reaction and the energies of the particles at the right hand side of the reaction.

The excited particles resulting from the last equation emit photons:

$$Ne^*(2p_1) \longrightarrow Ne^*(1s_2) + h\nu_1 \qquad \lambda_1 = 585.3 \text{ nm} \quad (4)$$
$$\qquad\qquad Ne^*(1s_4) + h\nu_3 \qquad \lambda_3 = 540.1 \text{ nm}$$
$$H^*(3s) \longrightarrow H^*(2p) + h\nu_4 \qquad \lambda_4 = 656.3 \text{ nm} \quad (H_\alpha) \quad (5)$$

In accordance with the transition probabilities, the intensity of the neon radiation with $\lambda_3 = 540.1$ nm is 100 times lower than the one of the neon radiation with $\lambda_1 = 585.3$ nm [17], in spite of the fact that their intensities are almost equals in a certain type of discharge [17]. It is important to notice that, after the radiative dezexcitation, the excited hydrogen atom on level $3d\,^2D_{3/2}$ returns to its previous state i.e. $H^*(2p)$. Thus, the reaction (3) does not decrease the density of the metastable atoms like the excited $H^*(2p)$ atoms.

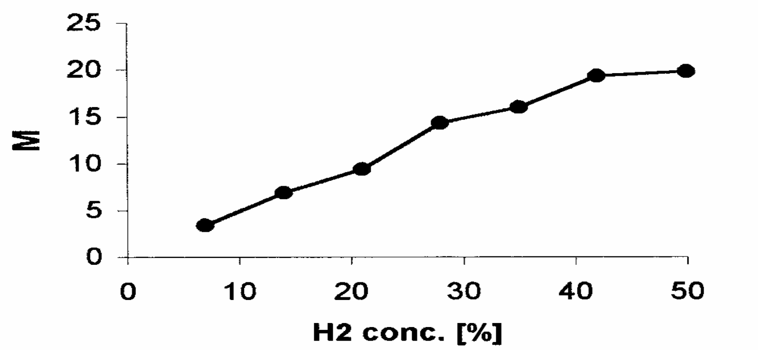

Fig. 3. The dependence of M parameter on the concentration of hydrogen at a total pressure of 30 Torr in a PDP type discharge.

Since both particles $N^*(2p_1)$ and $H^*(3s)$ resulting from the equation are equally participating in equation (3), the number of emitted photons $h\nu_1$ and $h\nu_4$ will be equal. Due to the fact that the experimental arrangement to measure the intensity of emitted radiation does not give absolute values, we can say that the experimental values of the intensities of the emitted radiation for the mentioned transitions must be proportional

In order to prove this behaviour using the recorded emission spectra we have represented in Fig. 3 the dependence of the M parameter:

$$M = \frac{I_{\lambda=585.3nm}}{I_{\lambda=614.3nm}} \tag{6}$$

and of a similar parameter for Hα radiation of hydrogen, namely:

$$M^* = \frac{I_{H\alpha}}{I_{\lambda=614.3nm}} \tag{7}$$

on the percentage of H_2 in the $(Ne+H_2)$gas mixture at a constant total pressure of 30 torr.

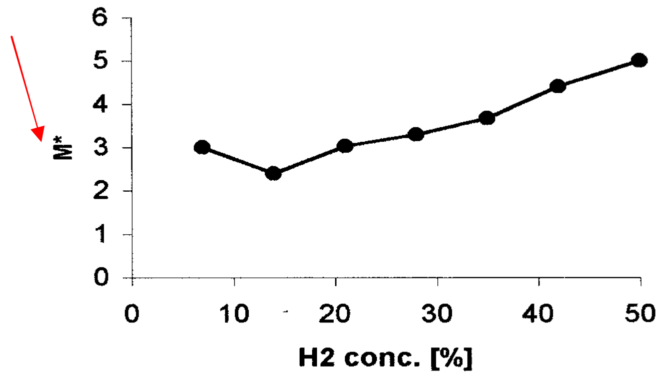

Fig. 4. The dependence of M^* parameter on the concentration of hydrogen at a total pressure
of 30 Torr in a PDP type discharge.

As it can be observed in Fig. 4, after a decrease of the Hα radiation with increasing hydrogen concentration to neon, both parameters M and M* increase similarly with hydrogen content in neon-hydrogen gas mixtures.

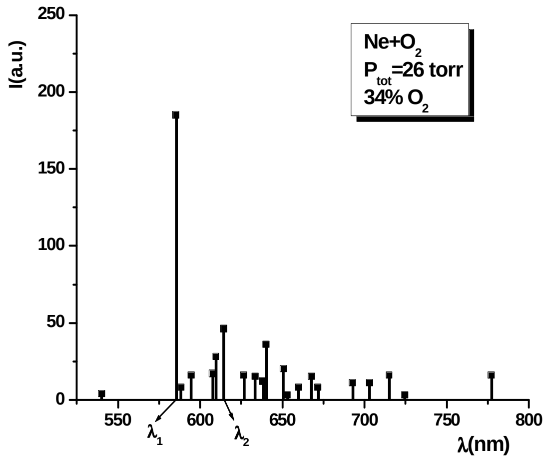

Fig. 5. The spectrum of ($Ne+34\%O_2$) gas mixture PDP type discharge at a total pressure of 26 Torr.

In this paper dedicated to a tentative explanation of the M-effect, from the large number of electronegative-electropositive gas mixtures for which the existence of the M-effect was proved, the gas mixtures containing neon-oxygen and neon- hydrogen were chosen as a case–study.

In these both cases, the dominant radiation of the emitted spectra was the neon line with the wavelength λ_1 = 585.3 nm.

In Fig. 5 is given the spectrum for (neon+oxygen) filled PDP dicharge with a content of oxygen 34% from total gas mixture pressure of 26 torr.

The spectrum is very similar to that obtained in the case of $Ne+H_2$ gas mixture (Fig. 2) with the exception of electronegative atoms radiation. Now, the $H\alpha$ radiation is replaced with the radiation of oxygen atoms, corresponding to the transition 3p $^5P_3$ – 3s $^5S_2^0$ (wavelenght λ = 777.5 nm).

In this case, the three-body reaction can be written as:

$$Ne^+ + O^- + O^*(met) \rightarrow (NeH^-) + O^*(met) \rightarrow Ne^*(2p_1) + O^*(3p\ ^5P_3) + O \pm \Delta E \qquad (8)$$

The excited oxygen atoms $O^*(3p\ ^5P_3)$ returns to the metastable state $O^*(3s\ ^5S_2^0)$ emitting the radiation with the wavelength λ = 777.5 nm.

Let us consider the energy balance for equations like equation (3) .

In Table 1 are given, in separate columns, energies of the particles involved in the reactions (3) and (8), as well as of the resulting particles.

Table 1.

Gas mixture	Electropos particle Ionisation Energy	Electroneg particle Electron Affinity	Third particle Energy	Electropos. excited particle Energy	Electroneg. particle excited Energy	Third particle Energy	ΔE
1	2	3	4	5	6	7	8
$Ne+H_2$	Ne^+ 21.57 eV	H^- 0.75 eV	H*(2p) 10.2 eV	$Ne^*(2p_1)$ 18.71 eV	H*(3s) 12.09 eV $\lambda_{H\alpha}$=656.28nm	H(1s) 0 eV	+0.2 eV

$Ne+O_2$	Ne^+ 21.57 eV	O^- 1.46 eV	$O^*(3s\ ^5S_2^0)$ 9.15 eV (met)	$Ne^*(2p_1)$ 18.71 eV	$O^*(3p\ ^5P_3)$ 10.74 eV λ=777.5nm	$O(2p^4\ ^3P_2)$ 0 eV	-0.2 eV

As observed in the table, the energy deffect was quite low. The probability of having such a reaction where all reacting particles are in energetic states above the ground state is high only due to the high cross-section of the ion-ion recombination process. This process compensates the lower concentration of the third particle wich is in a metastable state.

The three-body reaction considered is more efficient at elevated pressures. Heavy particle collisions also explains why the optimum percentages of the gas mixture is arround 50% .

2. Conclusions

This tentative explanation of the M-effect is given for neon+hydrogen and neon+oxygen gas mixtures in a PDP type discharge.

We have previously found the conditions of appearence of the M-effect from the- type of gas-point of view [15]. In this paper we try to explain the selective population of only one level of the atoms of electropositives gases.We have also found the type of reactions at an atomic and molecular scale, explaining the M-effect. Similar explanation can be found for various other gas mixtures like $Xe+Cl_2$, $Ne+Cl_2$, $Ar+H_2$, $Ar+Cl_2$, $He+Cl_2$ and $He+O_2$ for wich we proved experimentally the occurence of the M-effect. In some particular gas mixtures, binary heavy particle reactions might produce also selective population of one atomic level. Such ion-ion recombination processes are described by Landau-Zenner theory [18,19].

The fact that the M-effect does not appear in all electronegative-electropositive gas mixtures like $Ne+SF_6$ or $Xe+H_2$ can also be explained with our assumption. This behaviour is due to the fact that energetically resonant three-body or two-body reaction can not always occur.

Full knowledge of M-effect is important not only for fundamental knowledge but also for the developement of applications of this effect in the future.

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