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The kinetics of monochromatization of plasma light emission

C C Surdu-Bob[1] **and G Musa**[2]

[1] National Institute for Lasers, Plasma and Radiation Physics, Str.Atomistilor 409, MG-36, 077125, Bucharest, Romania
[2] Ovidius University, Bd. Mamaia 124, 900527, Constanta, Romania

E-mail: cristina.surdubob@plasmacoatings.ro

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Abstract
This paper is concerned with the monochromatization of the visible spectrum of rare gas discharges upon addition of either oxygen, hydrogen or chlorine gas: the M-effect. This phenomenon, observed over 20 years ago at the National Institute for Lasers, Plasma and Radiation Physics (Bucharest), is clearly explained here for the first time. The explanations contain references to all most relevant previous work on this subject. Assuming that the exacerbation of a given emission line results from the collisional interaction between an electropositive gas (the rare gas) and an appropriate electronegative gas, a general method, based on calculation of the energy balance of their excited energy levels, is developed for determining the gas mixtures for which the M-effect can be obtained, along with the corresponding wavelength of the monochromatic emission. These calculations show that the M-effect is highly probable in discharges combining inert gases with oxygen or hydrogen. The experimental results are in perfect agreement with these calculations. The M-effect is an important fundamental effect in physics and has a high potential for applications requiring monochromatic light sources for both research and industry.

AQ1

1. Introduction

In the course of previous work aimed at studying dielectric barrier discharges (DBDs) in multiple gas mixtures, a new physical effect was observed at the National Institute for Lasers, Plasma and Radiation Physics (Bucharest) [1–5]. It consisted in the spectacular exacerbation of the emission intensity of a single line in the visible region of certain gas discharge mixtures. An illustration of this effect is presented in figure 1 where the emission spectrum of a DBD discharge ignited in pure Ne (*a*) and in a Ne–H_2 mixture (*b*) are presented for comparison. The discharge was sustained in both cases in a plasma display panel-type reactor with a 0.15 mm gap at 100 Torr total gas pressure using a 35 kHz square-wave driven power supply providing 1 kV. The emission spectra were acquired using an optical multichannel analyser (OMA); no optical filters were used. As can be observed from figure 1, on addition of hydrogen, the well-known optical multi-line emission spectrum of Ne has turned monochromatic.

Further extensive investigations of the M-effect resulted in determining the specific experimental conditions for its appearance in gas mixture discharges. The effect was found to depend, among other parameters, on the composition of the gas mixture, on the partial pressure of its component gases and on total pressure as well as on the type of plasma source used [7]. More specifically, the M-effect was evidenced experimentally in the following conditions: tens to hundred Torr total pressure range, electropositive–electronegative gas combinations, for cathode temperatures lower than 250 °C in dc luminescent discharges as well as in low frequency pulsed discharges, ac DBDs and RF discharges. It was also observed [7] that the addition of the electronegative gas in percentages up to 50% increased the intensity of the monochromatised line to a maximum; a further increase in the content of the electronegative gas resulted in an intensity decrease of this line.

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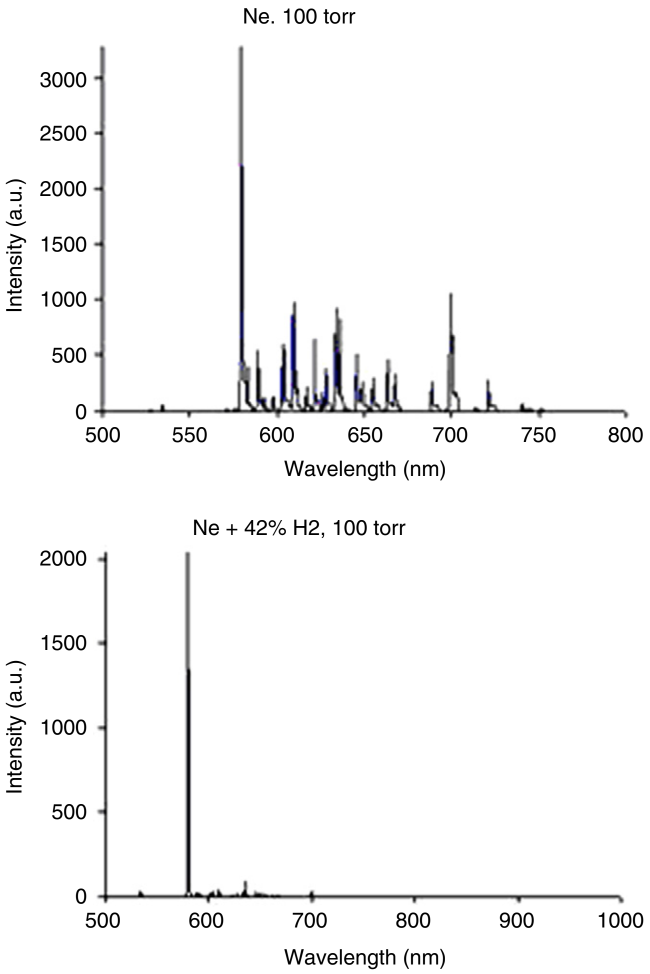

Figure 1. (*a*) Emitted spectrum from a dielectric barrier discharge ignited in pure Ne, under a 0.15 mm gap, at a 100 Torr total gas pressure, 35 kHz square-wave power supply having 1 kV peak-to-peak voltage. (*b*) Spectrum of a dielectric barrier discharge ignited in Ne + H_2 mixture, 0.15 mm narrow gap, 100 Torr total gas pressure, 35 kHz square wave power supply having 1 kV peak-to-peak voltage.

The magnitude of the M-effect can be obtained by first introducing the M parameter, namely the intensity ratio of two emission lines of the same spectrum, specifically that of the (eventual) monochromatized line and that of a reference line, $M = I(\lambda_M)/I(\lambda_{\mathrm{ref}})$, respectively; for example, $\lambda_M = 585.3$ nm, $\lambda_{\mathrm{ref}} = 614.3$ nm in neon. Then, given the ratio of the M parameter calculated for the mixture over that in the pure gas, one obtains the μ parameter: the higher the value of μ, the stronger the M-effect. The M parameter of the spectrum in figure 1(*a*) is 3.3, while that of the spectrum in figure 1(*b*) is 49.7, this yielding a μ parameter of 15. More recent studies of the M-effect sought optimized operating discharge conditions that would increase the *relative intensity of the monochromatic line* (i.e. the increase in μ) [6]. It was found, in particular, that the most significant M-effect in dc discharges is observed only in their negative glow, where the highest negative-ion density in the discharge tube is achieved [7].

A basic challenge in promoting the use of the M-effect is the possibility of readily finding the gas combinations that give a required monochromatized spectrum. Trying to determine experimentally the adequate gas combination for a given M-line would be a huge task. Solving that issue through (straightforward) calculations, as shown in the following section, opens the door to the development of an enormous range of new monochromatic light sources. Such sources may be used for scientific research as well as in industry by including them in standard reference sources, analytical tools, new plasma-based lasers, plasma displays, etc.

2. Basic principle of the *M*-effect

The extensive amount of experimental data gathered to date on the M-effect has enabled us to formulate the following physical model. It considers a three-body collision of the atoms (molecules) formed in the plasma [7]. The M-effect reaction can be written formally as

$$P^+ + N^- + N^{\mathrm{met}} \rightarrow P^* + N + N^{\mathrm{met}*}, \qquad (1)$$

where the letters P and N designate electropositive and electronegative gas atoms, respectively. P^+ refers specifically to positive ions, N^- is for negative ions while N^{met} stands for electronegative metastable atoms before the three-body interaction; then, P^* is an excited state of electropositive atoms after interaction, N is the ground state of the electronegative atoms and $N^{\mathrm{met}*}$ refers to electronegative atoms in a metastable state after interaction.

The probability of a three-body reaction, in contrast to a two-body one, is by essence small, but there are some factors intrinsic to the gas mixture discharge used that ensure a non-negligible probability to this particular reaction: (i) the simultaneous existence of negative and positive ions which provides a high probability of collisions due to the attraction of coulombian forces of opposite sign particles. The negative ions form mainly in the afterglow of pulsed discharges, where the electrons have low energies, hence a high affinity in forming negative ions with neutral atoms. The low energy collisions do not break these negative ions. In the case of hydrogen atoms (molecules), the particularly high density of negative ions stems from an electron attachment energy of 0.75 eV. Further, previous work [7] where the time variation of the pulse current was acquired simultaneously with the emission line intensity demonstrated that the effect nonetheless continues to exist for a while in the off-time (time afterglow); (ii) the total gas pressure is relatively high (tens to hundred Torr), which ensures a higher probability of the three-body interaction; (iii) metastable-state atoms are by definition long-lifetime species, a feature favouring their participation in this three-body reaction.

When the collision reaction (1) takes place (equivalent to a reaction probability equal to 1), a monochromatic light is emitted via the radiative desexcitation of the P^* atoms. This happens provided that the energy balance condition is met, i.e. the energy defect of reaction (1) is 0 eV (ideally). The energy defect (Δ) is given by the difference in the energy of the participating particles before the 3-body interaction and after it: the positively ionized and excited neutral atoms, P^+ and P^*, respectively, and the metastable-state atoms N^{met} and $N^{\mathrm{met}*}$ have, say, positive energy values while the negative ions have, then, negative energy values. This is discussed in more detail in the following section.

AQ2

Table 1. Ar + H_2.

P^+ (eV)	N^- (eV)	N^{met} (eV)	P^* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
15.76	0.75	10.2	13.98	0	12.09	−0.86	1371.9
15.76	0.75	10.2	14.07	0	12.09	−0.95	1067.4
15.76	0.75	10.2	12.91	0	12.09	0.21	965.8
15.76	0.75	10.2	13.17	0	12.09	−0.05	922.5
15.76	0.75	10.2	12.91	0	12.09	0.21	912
15.76	0.75	10.2	13.28	0	12.09	−0.16	852.1
15.76	0.75	10.2	13.09	0	12.09	0.03	842.5
15.76	0.75	10.2	13.3	0	12.09	−0.18	840
15.76	0.75	10.2	13.33	0	12.09	−0.21	826.5
15.76	0.75	10.2	13.08	0	12.09	0.04	811.5
15.76	0.75	10.2	13.15	0	12.09	−0.03	810.4
15.76	0.75	10.2	13.09	0	12.09	0.03	801.5
15.76	0.75	10.2	13.17	0	12.09	−0.05	763.5
15.76	0.75	10.2	13.48	0	12.09	−0.36	**750.4**
15.76	0.75	10.2	13.3	0	12.09	−0.18	738.4
15.76	0.75	10.2	13.33	0	12.09	−0.21	696.5
15.76	0.75	10.2	15.17	0	12.09	−2.05	621.2
15.76	0.75	10.2	15	0	12.09	−1.88	591.2
15.76	0.75	10.2	15.35	0	12.09	−2.23	568.1

Table 2. Ar + O_2.

P^+ (eV)	N^- (eV)	N^{met} (eV)	P^* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
15.76	1.46	9.146	13.98	0	10.74	−1.27	1371.9
15.76	1.46	9.146	14.07	0	10.74	−1.36	1067.4
15.76	1.46	9.146	12.91	0	10.74	−0.20	965.8
15.76	1.46	9.146	13.17	0	10.74	−0.46	922.5
15.76	1.46	9.146	12.91	0	10.74	−0.20	912.3
15.76	1.46	9.146	13.28	0	10.74	−0.57	852.1
15.76	1.46	9.146	13.09	0	10.74	−0.38	842.5
15.76	1.46	9.146	13.3	0	10.74	−0.59	840.8
15.76	1.46	9.146	13.33	0	10.74	−0.62	826.5
15.76	1.46	9.146	13.08	0	10.74	−0.37	811.5
15.76	1.46	9.146	13.15	0	10.74	−0.44	810.4
15.76	1.46	9.146	13.09	0	10.74	−0.38	801.5
15.76	1.46	9.146	13.17	0	10.74	−0.46	763.5
15.76	1.46	9.146	13.48	0	10.74	−0.77	750.4
15.76	1.46	9.146	13.3	0	10.74	−0.59	738.4
15.76	1.46	9.146	13.33	0	10.74	−0.62	696.5
15.76	1.46	9.146	15.17	0	10.74	−2.46	621.2
15.76	1.46	9.146	15	0	10.74	−2.29	591.2
15.76	1.46	9.146	15.35	0	10.74	−2.64	568.1
15.76	1.46	9.146	15.1	0	10.74	−2.39	565.1
15.76	1.46	9.146	15.12	0	10.74	−2.41	560.7
15.76	1.46	9.146	15.51	0	10.74	−2.80	559.8

Table 3. Ar + Cl_2.

P^+ (eV)	N^- (eV)	N^{met} (eV)	P^* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
15.76	3.62	8.98	13.98	0	10.27	−3.13	1371.87
15.76	3.62	8.98	14.07	0	10.27	−3.22	1067.36
15.76	3.62	8.98	12.91	0	10.27	−2.06	965.78
15.76	3.62	8.98	13.17	0	10.27	−2.32	922.45
15.76	3.62	8.98	12.91	0	10.27	−2.06	912.3
15.76	3.62	8.98	13.28	0	10.27	−2.43	852.1
15.76	3.62	8.98	13.09	0	10.27	−2.24	842.5
15.76	3.62	8.98	13.3	0	10.27	−2.45	840.8
15.76	3.62	8.98	13.33	0	10.27	−2.48	826.5
15.76	3.62	8.98	13.08	0	10.27	−2.23	811.5
15.76	3.62	8.98	13.15	0	10.27	−2.30	810.4
15.76	3.62	8.98	13.09	0	10.27	−2.24	801.5
15.76	3.62	8.98	13.17	0	10.27	−2.32	763.5
15.76	3.62	8.98	13.48	0	10.27	−2.63	750.4
15.76	3.62	8.98	13.3	0	10.27	−2.45	738.4
15.76	3.62	8.98	13.33	0	10.27	−2.48	696.5
15.76	3.62	8.98	15.17	0	10.27	−4.32	621.2
15.76	3.62	8.98	15	0	10.27	−4.15	591.2

Table 4. Ne + H_2.

P^+ (eV)	N^- (eV)	N^{met} (eV)	P^* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
21.56	0.75	10.2	18.80	0	12.09	0.12	**750.40**
21.56	0.75	10.2	19.66	0	12.09	−0.74	966.54
21.56	0.75	10.2	20.14	0	12.09	−1.22	878.38
21.56	0.75	10.2	20.05	0	12.09	−1.13	878.06
21.56	0.75	10.2	20.14	0	12.09	−1.22	865.44
21.56	0.75	10.2	18.37	0	12.09	0.55	743.89
21.56	0.75	10.2	18.38	0	12.09	0.54	724.52
21.56	0.75	10.2	18.57	0	12.09	0.35	717.39
21.56	0.75	10.2	18.38	0	12.09	0.54	703.24
21.56	0.75	10.2	18.6	0	12.09	0.32	702.4
21.56	0.75	10.2	18.63	0	12.09	0.29	692.95
21.56	0.75	10.2	18.68	0	12.09	0.24	671.7
21.56	0.75	10.2	18.69	0	12.09	0.23	667.82
21.56	0.75	10.2	18.71	0	12.09	0.21	665.21
21.56	0.75	10.2	18.71	0	12.09	0.21	659.89
21.56	0.75	10.2	18.6	0	12.09	0.32	653.29
21.56	0.75	10.2	18.55	0	12.09	0.37	640.22
21.56	0.75	10.2	18.61	0	12.09	0.31	638.3
21.56	0.75	10.2	18.57	0	12.09	0.35	633.44
21.56	0.75	10.2	18.62	0	12.09	0.30	630.48
21.56	0.75	10.2	18.69	0	12.09	0.23	626.65
21.56	0.75	10.2	18.61	0	12.09	0.31	621.73
21.56	0.75	10.2	18.72	0	12.09	0.20	616.36
21.56	0.75	10.2	18.63	0	12.09	0.29	614.3
21.56	0.75	10.2	18.69	0	12.09	0.23	609.62
21.56	0.75	10.2	18.71	0	12.09	0.21	607.43
21.56	0.75	10.2	18.72	0	12.09	0.20	603
21.56	0.75	10.2	18.69	0	12.09	0.23	594.48
21.56	0.75	10.2	18.71	0	12.09	0.21	588.19
21.56	0.75	10.2	18.96	0	12.09	−0.04	**585.25**
21.56	0.75	10.2	18.96	0	12.09	−0.04	540.06
21.56	0.75	10.2	20.7	0	12.09	−1.78	534.11
21.56	0.75	10.2	20.71	0	12.09	−1.79	533.08

3. Calculation results and discussion

Numerical calculations of the energy balance implied in reaction (1) have been achieved for few inert gases combined with either oxygen, hydrogen or chlorine. These are presented in the form of tables where the entries are published energy data on the (positive and negative) ions, the metastable and excited atoms [8–10] for the particles before and after their interaction, the output data being the energy defect. As an example, consider the first line of table 1: total energy before interaction is (in eV) $15.76 + 10.2 - 0.75 = 25.21$ while after interaction, we obtain $13.98 + 12.09 = 26.07$, hence we obtain an energy defect $\Delta = -0.86$. The M-effect is obtained only for the combinations of P^* and N^{met*} that give a very small energy defect (close to 0 eV). The gas combinations that can give the effect can thus be found in this way. The emission line corresponding to the reaction product P^* is the M-effect emission wavelength.

Table 5. Ne + O_2.

P$^+$ (eV)	N$^-$ (eV)	N^{met} (eV)	P* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
21.56	1.46	9.146	18.8	0	10.74	−0.29	750.40
21.56	1.46	9.146	18.37	0	10.74	0.14	743.89
21.56	1.46	9.146	18.38	0	10.74	0.13	724.52
21.56	1.46	9.146	18.57	0	10.74	−0.06	717.39
21.56	1.46	9.146	18.38	0	10.74	0.13	703.24
21.56	1.46	9.146	18.6	0	10.74	−0.09	702.4
21.56	1.46	9.146	18.63	0	10.74	−0.12	692.95
21.56	1.46	9.146	18.68	0	10.74	−0.17	671.7
21.56	1.46	9.146	18.69	0	10.74	−0.18	667.82
21.56	1.46	9.146	18.71	0	10.74	−0.20	665.21
21.56	1.46	9.146	18.71	0	10.74	−0.20	659.89
21.56	1.46	9.146	18.6	0	10.74	−0.09	653.29
21.56	1.46	9.146	18.55	0	10.74	−0.04	640.22
21.56	1.46	9.146	18.61	0	10.74	−0.10	638.3
21.56	1.46	9.146	18.57	0	10.74	−0.06	633.44
21.56	1.46	9.146	18.62	0	10.74	−0.11	630.48
21.56	1.46	9.146	18.69	0	10.74	−0.18	626.65
21.56	1.46	9.146	18.61	0	10.74	−0.10	621.73
21.56	1.46	9.146	18.72	0	10.74	−0.21	616.36
21.56	1.46	9.146	18.63	0	10.74	−0.12	614.3
21.56	1.46	9.146	18.69	0	10.74	−0.18	609.62
21.56	1.46	9.146	18.71	0	10.74	−0.20	607.43
21.56	1.46	9.146	18.72	0	10.74	−0.21	603
21.56	1.46	9.146	18.69	0	10.74	−0.18	594.48
21.56	1.46	9.146	18.71	0	10.74	−0.20	588.19
21.56	1.46	9.146	18.96	0	10.74	−0.45	585.25
21.56	1.46	9.146	18.96	0	10.74	−0.45	540.06
21.56	1.46	9.146	20.7	0	10.74	−2.19	534.11
21.56	1.46	9.146	20.71	0	10.74	−2.20	533.08
21.56	1.46	9.146	21.11	0	10.74	−2.60	495.7

Table 6. Ne + Cl_2.

P$^+$ (eV)	N$^-$ (eV)	N^{met} (eV)	P* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
21.56	3.62	8.98	18.8	0	10.27	−2.15	750.4
21.56	3.62	8.98	18.37	0	10.27	−1.72	743.89
21.56	3.62	8.98	18.38	0	10.27	−1.73	724.52
21.56	3.62	8.98	18.57	0	10.27	−1.92	717.39
21.56	3.62	8.98	18.38	0	10.27	−1.73	703.24
21.56	3.62	8.98	18.6	0	10.27	−1.95	702.4
21.56	3.62	8.98	18.63	0	10.27	−1.98	692.95
21.56	3.62	8.98	18.68	0	10.27	−2.03	671.7
21.56	3.62	8.98	18.69	0	10.27	−2.04	667.82
21.56	3.62	8.98	18.71	0	10.27	−2.06	665.21
21.56	3.62	8.98	18.71	0	10.27	−2.06	659.89
21.56	3.62	8.98	18.6	0	10.27	−1.95	653.29
21.56	3.62	8.98	18.55	0	10.27	−1.90	640.22
21.56	3.62	8.98	18.61	0	10.27	−1.96	638.3
21.56	3.62	8.98	18.57	0	10.27	−1.92	633.44
21.56	3.62	8.98	18.62	0	10.27	−1.97	630.48
21.56	3.62	8.98	18.69	0	10.27	−2.04	626.65
21.56	3.62	8.98	18.61	0	10.27	−1.96	621.73

This is a simple method to discard gas combinations that cannot provide the effect, irrespectively of the required experimental conditions. The results of this calculation put in evidence *all* gas combinations that possibly yield the M-effect provided, then, that the experimental conditions are suitable to ensure negative ions and metastable-state atoms (see section 1).

Table 7. Kr + H_2.

P$^+$ (eV)	N$^-$ (eV)	N^{met} (eV)	P* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
14	0.75	10.2	12.38	0	12.09	−1.02	1442.6
14	0.75	10.2	12.35	0	12.09	−0.99	1181.94
14	0.75	10.2	11.3	0	12.09	0.06	975.18
14	0.75	10.2	11.3	0	12.09	0.06	892.87
14	0.75	10.2	11.44	0	12.09	−0.08	877.67
14	0.75	10.2	12.1	0	12.09	−0.74	850.89
14	0.75	10.2	11.53	0	12.09	−0.17	829.81
14	0.75	10.2	12.14	0	12.09	−0.78	828.1
14	0.75	10.2	12.14	0	12.09	−0.78	826.32
14	0.75	10.2	11.55	0	12.09	−0.19	819
14	0.75	10.2	11.44	0	12.09	−0.08	811.29
14	0.75	10.2	11.44	0	12.09	−0.08	810.44
14	0.75	10.2	12.1	0	12.09	−0.74	805.95
14	0.75	10.2	11.55	0	12.09	−0.19	760.15
14	0.75	10.2	11.67	0	12.09	−0.31	**758.74**
14	0.75	10.2	12.82	0	12.09	−1.46	427.4

Table 8. Kr + O_2.

P$^+$ (eV)	N$^-$ (eV)	N^{met} (eV)	P* (eV)	N (eV)	N^{met*}	Δ (eV)	λ (nm)
14	1.46	9.146	11.3	0	−0.35	975.18	
14	1.46	9.146	11.4	0	−0.49	877.67	
14	1.46	9.146	11.53	0	−0.58	829.8	
14	1.46	9.146	11.55	0	−0.06	819	
14	1.46	9.146	11.44	0	−0.49	811.3	
14	1.46	9.146	11.55	0	−0.6	760.15	

Calculations are presented for Ar, Ne and Kr mixed with hydrogen, oxygen and chlorine (tables 1–8). A larger set of calculations that include the most intense lines from the IR to the UV regions of the spectrum will be given in a forthcoming paper.

The tables showing the results of the energy balance calculation of equation (1) are dependent on the accuracy of the published energy data: as a result, one cannot strictly predict the existence of the M-effect for a sharp zero energy defect. Values of Δ in the $(-1, 1)$ eV range should be considered as possibly reflecting the appearance of the M-effect. Clearly, more precise values of the published reference data would give a more accurate prediction of the emission lines that can present the M-effect.

It is interesting to note that all the monochromatized spectra found experimentally in different plasma conditions can be retrieved from the present computation procedure. Examples are given in the following tables. The wavelengths corresponding to experimental results are highlighted in bold format and are taken from [5–7]. There is though a large series of other wavelengths that have a high probability to be M-lines (predicted by computation) but they have not been found experimentally yet. This is either due to the fact that the input published reference data are not reliable, as pointed out earlier, or it is due to different experimental requirements. The tables also contain a few calculations where there is clear evidence that the M-effect does not appear, for example, where the defect Δ is far outside the $(-1, 1)$ eV range. The most probable M-lines are highlighted with a yellow-coloured background.

Finally, it is worth mentioning that the M-effect has recently been observed in mixtures of two electropositive with one electronegative gas. In this case, two M-effects were simultaneously observed in the same discharge [11].

A larger set of calculations that include the most intense lines from the IR to the UV regions of the spectrum will be given in a forthcoming paper.

4. Conclusion

The M-effect is an interesting and, in terms of applications, a possibly important physical effect that appears to be quite well understood as shown in this paper. Due to the complexity of experimental conditions that need to be simultaneously met, the M-effect had, to some extent, escaped to the scientific community, although it has certainly been observed sometimes without being identified as such. The effect was studied experimentally in the visible region of the spectrum but the calculations show that there is a high probability to appear beyond this region also. Further studies will be focused on the development of new applications based on this effect.

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