Ne Emissions from a He Discharge Flow System: I. Relative Transition Probabilities of Ne

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A windowless microwave discharge in He was used to generate Ne emissions in a flow system. Relative transition probabilities for a number of emission arrays were determined from the observed line intensities. Agreement is generally obtained with previous experimental and theoretical work.

Introduction

There has been considerable interest in transition probabilities for Ne states since the first laser oscillation on Ne transitions was obtained in 1961 [1]. Until now laser oscillation has been observed on more than 160 lines [2]. A bibliography on atomic transition probabilities including those for Ne has been published recently [3]. Although there are numerous determinations of lifetimes of individual Ne states [3, 4], an extensive set of experimental relative transition probabilities has not been reported. On the other hand there are several calculations of transition probabilities for a number of arrays [3].

Using a particular arrangement of a He discharge flow system we have generated Ne emissions mainly originating in states below 167810 cm^{-1} above the ground state [5]. In the present work we have used the intensities of these Ne emissions to determine relative transition probabilities which are then compared with experimental and theoretical values reported in the literature. The following paper [5] reports on the excitation mechanism of the observed Ne states.

Experimental

The experimental set-up is similar to that used previously for the studies of the charge transfer reaction of N_2^+ with NO [6] and for the investigation of the emissions from the NH and ND ($b\,^1\Sigma^+ \rightarrow X\,^3\Sigma^-$)-transitions [7].

A schematic diagram of the apparatus is shown in Figure 1. The apparatus consists of a discharge system connected to a flow system and of a monochromator. The microwave discharge in He can be operated in two positions. In position A, light from the He discharge irradiates the flow tube directly; when using the alternative position B the discharge is operated in a side arm in such a way that direct illumination of the flow tube is avoided. The distance between the discharge region and the flow tube was kept the same (~ 5 cm) for both positions. Two inlets (a) and (b) were used to introduce He to the flow system. Using inlet (a) the flow was usually directed towards the flow tube. In some

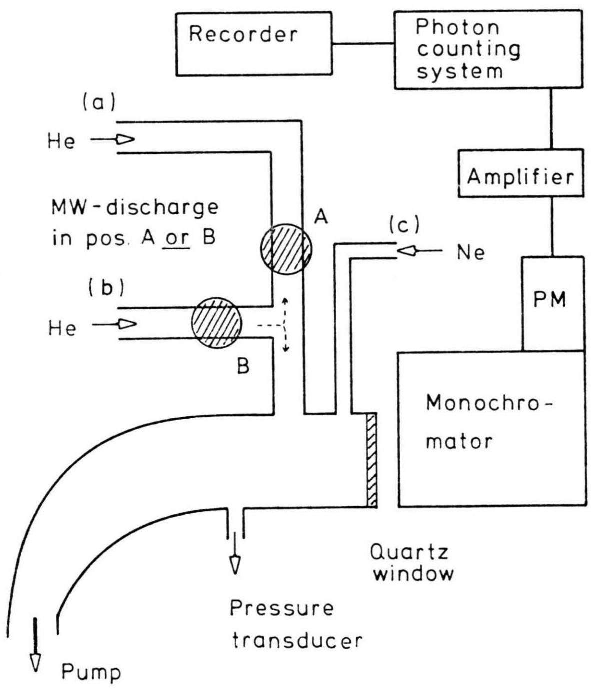

Fig. 1. Schematic diagram of the He flow apparatus and of the optical detection system.

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experiments using inlet (b) the flow was split towards the discharge in position A and the flow tube (dashed arrows in Figure 1). Through the separate inlet (c) small amounts of neon were added.

The He flow rate through the discharge was typically 0.14 cm^3 (STP) sec^{-1}. The pressure in the flow tube was measured by a capacitance manometer (MKS, Type 220). Pressures ranging from 0.5 to 2 Torr for He and ranging from 0.01 to 0.5 Torr for Ne were used. The He and Ne samples were of the following stated (Messer Griesheim) minimum purities: He, 99.996% and Ne, 99.99%. All the experiments were performed at room temperature.

The monochromator (Chromatix CT 103) was used to monitor emissions from the flow tube. Two different gratings (Bausch & Lomb, blazed at 300 and at 500 nm, 1200 grooves mm^{-1}) were combined with two different photomultipliers (EMI 9789 QB for the wavelength region from 200 to 550 nm and RCA C31034 A, cooled to -50 °C, from 400 to 890 nm). The signals from the photomultiplier tubes were registered by a chart recorder using a photon counting device (Ortec Brookdeal 5C1). Typically 0.04 nm could be resolved using the first order and a slit width of 50 μm. The relative sensitivity of this optical arrangement was determined by the air afterglow [8] for wavelengths $\lambda > 385$ nm and by a calibrated D_2-lamp (Optronic Laboratories, model UV-40) for wavelengths $\lambda < 400$ nm. The relative spectral sensitivity of the detection system is shown in Figure 2. The accuracy of this calibration is conservatively estimated to be within $\pm 20\%$.

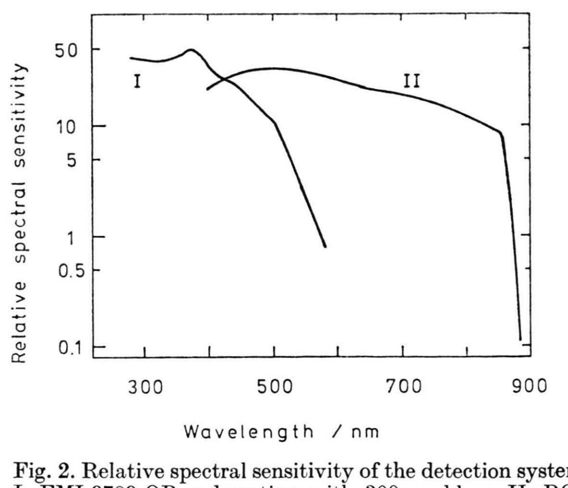

Fig. 2. Relative spectral sensitivity of the detection system. I: EMI 9789 QB and grating with 300 nm blaze. II: RCA C 31034 A and grating with 500 nm blaze.

As can be seen from Fig. 2 the sensitivity decreases gradually from shorter wavelenghts to 850 nm and drops rapidly between 850 and 890 nm by more than two orders of magnitude. In this cut-off region the calibration is estimated to be accurate within $\pm 40\%$. The reproducibility of the Ne intensities is within $\pm 10\%$.

Results

Typical Ne spectra were taken with the discharge in position A and using 0.9 torr He and 0.1 torr Ne in the flow system. As will be described in the following paper [5] the intensities of the Ne lines depended strongly on the position of the discharge (A or B), but were not influenced by the flow direction. A part of the spectrum covering the range from 580 to 650 nm is shown in Figure 3. The Ne lines were identified using the tables given by Zaidel et al. [9] and by calculating energy differences of atomic Ne-levels given by Moore [10]. Relative intensities of the observed Ne lines were determined from the peak heights of the recorded emission lines. The obtained intensities are the mean of at least five determinations and are estimated to be

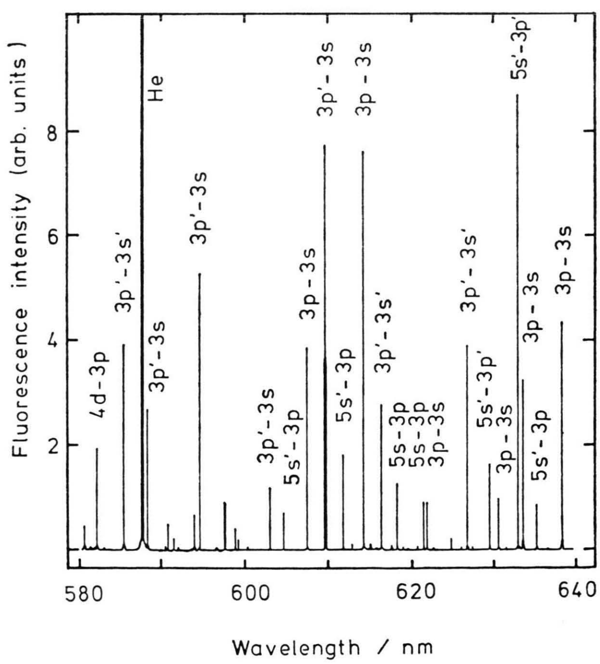

Fig. 3. A part of the neon spectrum. The resolution is 0.04 nm. The upper levels of the transitions are indicated in the spectrum.

precise within $\pm 30\%$ ($\pm 50\%$ for $\lambda \geqq 850$ nm). For all emissions resulting in deviations greater than 30% from the literature value additional spectra were taken in the wavelength region of the emission line.

Tables 1 to 4 summarize the relative intensities of almost half of the observed Ne lines. In these tables the neon transitions are arranged according to emission arrays. Only those transitions are listed which result in (almost) complete arrays. Emissions from *n*s, *n*s′ levels with $n > 5$ and from *n*p, *n*p′, *n*d and *n*d′ levels with $n > 4$ are not listed since they were either not detected ($\lambda > 890$ nm) or very weak.

Table 1. Comparison of the present relative transition probabilities with literature data for the Ne 3p and 3p′ arrays.

Level			Transition probability/10^6 s^{-1}		
upper	lower	λ air/nm	Ref. [11]	Ref. [4][b]	this work[a]
3p′(1/2) 0	3s(3/2) 1	540.06	0.9		0.79
	3s′(1/2) 1	585.25	**68.2**		**68.2**
3p′(1/2) 1	3s(3/2) 2	588.19	11.5	10.10	10.40
	3s(3/2) 1	603.00	5.61	5.15	4.97
	3s′(1/2) 0	616.36	14.6	13.54	12.01
	3s′(1/2) 1	659.90	23.2	**21.72**	**21.72**
3p′(3/2) 2	3s(3/2) 2	594.48	11.3	**10.30**	**10.30**
	3s(3/2) 1	609.62	18.1	17.07	15.82
3p′(3/2) 1	3s(3/2) 2	597.55	3.51	4.48	4.10
	3s(3/2) 1	612.85	0.67	0.677	0.73
	3s′(1/2) 0	626.65	24.9	24.27	26.21
	3s′(1/2) 1	671.70	21.7	**22.66**	**22.66**
3p(1/2) 0	3s(3/2) 1	607.43	**60.3**		**60.3**
	3s′(1/2) 1	665.21	0.29		0.38
3p(1/2) 1	3s(3/2) 2	703.24	25.3	**25.82**	**25.82**
	3s(3/2) 1	724.52	9.35	9.66	9.32
	3s′(1/2) 0	743.89	2.3	2.43	2.23
	3s′(1/2) 1	808.25	0.12	0.114	0.11
3p(3/2) 2	3s(3/2) 2	614.31	28.2	**29.11**	**29.11**
	3s(3/2) 1	630.48	4.16	4.21	4.15
	3s′(1/2) 1	692.95	17.4	19.32	17.79
3p(3/2) 1	3s(3/2) 2	621.73	6.37	6.55	7.23
	3s(3/2) 1	638.30	32.1	**36.05**	**36.05**
	3s′(1/2) 0	653.29	10.8	11.58	12.55
	3s′(1/2) 1	702.41	1.9	2.32	2.29
3p(5/2) 2	3s(3/2) 2	633.44	16.1	17.16	16.90
	3s(3/2) 1	650.65	30	**32.05**	**32.05**
	3s′(1/2) 1	717.39	2.87	3.42	3.23

[a] The present experimental values are adjusted to the bold-faced literature values.

[b] If possible, the more recent experimental values of Ref. [4] were taken for comparison.

Table 2. Comparison of the present relative transition probabilities with literature data for the Ne 3d and 3d′ arrays.

Level			Transition probability/10^6 s^{-1}		
upper	lower	λ air/nm	Ref. [12]	Ref. [13][a]	this work
3d′(3/2) 1	3p(1/2) 1	705.13	3.00	2.02	2.12
	3p(5/2) 2	792.71	0.40	0.29	0.31
	3p(3/2) 1	811.86	4.10	**3.49**	**3.49**
	3p(3/2) 2	824.87	0.34	0.30	0.24
	3p′(3/2) 1	857.14	3.18	3.04	2.73
	3p(1/2) 0	867.95	**13.65**	**13.65**	10.73
	3p′(1/2) 1	877.17	10.23	10.58	9.25
3d′(3/2) 2	3p(1/2) 1	705.91	9.35	6.75	7.38
	3p(5/2) 3	783.30	0.06	0.03	< 0.04
	3p(5/2) 2	793.70	0.96	0.78	0.90
	3p(3/2) 1	812.89	0.50	0.72	0.79
	3p(3/2) 2	825.94	2.37	**2.03**	**2.03**
	3p′(3/2) 1	858.29	0.33	1.00	1.04
	3p′(3/2) 2	864.71	4.20	3.91	3.04
	3p′(1/2) 1	878.38	30.44	31.26	27.19
3d′(5/2) 2	3p(1/2) 1	706.48	0	< 0.02	< 0.04
	3p(5/2) 3	784.00	0.01	< 0.03	< 0.04
	3p(3/2) 1	813.64	12.45	**11.42**	**11.42**
	3p(3/2) 2	826.71	0.52	0.65	0.54
	3p′(3/2) 1	859.13	30.39	30.86	30.79
	3p′(3/2) 2	865.55	4.04	5.03	4.32
	3p′(1/2) 1	879.25	0	0.15	< 0.18
3d′(5/2) 3	3p(5/2) 3	783.91	0.13	0.12	0.12
	3p(5/2) 2	794.32	4.12	**3.87**	**3.87**
	3p(3/2) 2	826.61	3.53	3.44	2.94
	3p′(3/2) 2	865.44	38.30	39.94	33.64
3d(3/2) 2	3p(1/2) 1	748.89	27.0	19.39	21.31
	3p(5/2) 3	836.58	2.26	**2.29**	**2.29**
	3p(5/2) 2	848.45	0.79	0.71	0.75
	3p(3/2) 1	870.42	1.38	1.79	1.27
	3p(3/2) 2	885.39	14.96	17.16	15.87
3d(5/2) 2	3p(1/2) 1	743.75	0		< 0.04
	3p(5/2) 3	830.15	**1.0**		**1.0**
	3p(5/2) 2	841.84	15.53		16.24
	3p(3/2) 1	863.47	21.84		18.96
	3p(3/2) 2	878.20	0.08		< 0.08
3d(5/2) 3	3p(5/2) 3	830.03	**14.27**		**14.27**
	3p(5/2) 2	841.72	1.01		1.28
	3p(3/2) 2	878.06	27.32		29.68
3d(7/2) 3	3p(5/2) 3	837.64	**2.43**		**2.43**
	3p(5/2) 2	849.54	37.95		34.55
	3p(3/2) 2	886.53	5.20		< 5.8

[a] relative determination for the 3d′(3/2) 1 array adjusted to the bold-faced value of Ref. [12]; absolute determination for all the other arrays of Ref. [13].

Also, emission arrays from 4p and 4p′ states are not listed since generally the corresponding intensities are rather weak and several emission lines are blended with other lines. Included in the tables are selected literature values.

It should be noted for some arrays that not the complete set of lines was observed because of the limited wavelength region of detection. Furthermore, some Ne lines are blended with other emission lines. If possible their intensities were estimated from the shape of the shoulders of the observed lines. Otherwise, these transitions are also omitted from the tables.

Table 3. Comparison of the present relative transition probabilities with literature data for the Ne 4d and 4d′ arrays.

Level			Transition probability/10^6 s^{-1}	
upper	lower	λ air/nm	Ref. [14]	this work
4d(1/2) 0	3p(1/2) 1	534.33	**7.84**	**7.84**
	3p(3/2) 1	593.45	0.98	0.93
	3p′(3/2) 1	617.28	0.41	0.21
	3p′(1/2) 1	627.60	1.31	0.82
4d(1/2) 1	3p(1/2) 1	534.11	**6.24**	**6.24**
	3p(5/2) 2	582.89	0.25	0.27
	3p(3/2) 1	593.18	0	< 0.03
	3p(3/2) 2	600.10	1.05	1.07
	3p′(3/2) 1	616.99	0.0	< 0.03
	3p′(3/2) 2	620.30	0.26	0.10
	3p(1/2) 0	622.57	0.66	0.41
	3p′(1/2) 1	627.30	0.98	0.66
	3p′(1/2) 0	713.85	0.36	0.17
4d(3/2) 1	3p(1/2) 1	532.64	0.43	0.43
	3p(5/2) 2	581.14	0.61	0.60
	3p(3/2) 1	591.36	**2.72**	**2.72**
	3p(3/2) 2	598.24	0.08	< 0.07
	3p′(3/2) 1	615.03	1.27	1.14
	3p(1/2) 0	620.58	1.39	1.27
	3p′(1/2) 1	625.27	0.04	< 0.04
	3p′(1/2) 0	711.23	1.09	0.48
4d(3/2) 2	3p(1/2) 1	533.08	**4.28**	**4.28**
	3p(5/2) 3	576.06	0.44	0.54
	3p(5/2) 2	581.67	0.16	0.13
	3p(3/2) 2	598.79	3.20	2.88
	3p′(3/2) 1	615.61	0.16	0.15
	3p′(3/2) 2	618.91	0.65	0.50
	3p′(1/2) 1	625.88	0.46	0.50
4d′(3/2) 1	3p(1/2) 1	511.37	0.66	0.78
	3p(5/2) 2	555.91	0.04	< 0.3
	3p(3/2) 1	565.26	0.57	0.70
	3p(3/2) 2	571.53	0.04	< 0.3
	3p′(3/2) 1	586.84	0.76	0.99
	3p′(3/2) 2	589.84	0.18	< 0.3
	3p′(1/2) 1	596.16	**1.96**	**1.96**
	3p′(1/2) 0	673.81	1.60	0.98
4d′(5/2) 3	3p(5/2) 3	511.15	0.01	< 0.16
	3p(5/2) 2	556.28	1.13	1.42
	3p(3/2) 2	571.92	1.25	1.32
	3p′(3/2) 2	590.25	**10.32**	**10.32**
4d(5/2) 3	3p(5/2) 3	574.83	2.76	2.94
	3p(3/2) 2	597.46	**5.18**	**5.18**
	3p′(3/2) 2	617.49	0.66	0.74
4d(7/2) 3	3p(5/2) 2	582.02	**7.38**	**7.38**
	3p(3/2) 2	599.17	1.15	1.09
	3p′(3/2) 2	619.31	0.16	0.19

Table 4. Comparison of the present relative transition probabilities with literature data for the Ne 5s and 5s′ arrays.

Level			Transition probability/10^6 s^{-1}		
upper	lower	λ air/nm	Ref. [15]	Ref. [16]	this work
5s′(1/2) 1	3p(1/2) 1	543.36	0.45	0.283	0.303
	3p(5/2) 2	593.93	0.22	0.200	0.224
	3p(3/2) 1	604.61	0.20	0.226	0.254
	3p(3/2) 2	611.80	0.64	0.609	0.638
	3p′(3/2) 1	629.38	0.66	0.639	0.663
	3p′(3/2) 2	632.82	3.20	**3.39**	**3.39**
	3p(1/2) 0	635.19	0.32	0.345	0.361
	3p(1/2) 1	640.11	1.30	1.39	1.463
	3p′(1/2) 0	730.48	0.31	0.255	0.262
5s′(1/2) 0	3p(1/2) 1	544.85	0.80		0.50
	3p(3/2) 1	606.45	1.7		1.68
	3p′(3/2) 1	631.37	**3.3**		**3.3**
	3p′(1/2) 1	642.17	1.84		2.01
5s(3/2) 1	3p(1/2) 1	566.26	0.51		0.51
	3p(5/2) 2	621.39	**2.70**		**2.70**
	3p(3/2) 1	633.09	1.40		1.45
	3p(3/2) 2	640.98	1.40		1.48
	3p′(3/2) 1	660.29	0.48		0.53
	3p′(3/2) 2	664.08	0.01		< 0.02
	3p(1/2) 0	666.69	0.41		0.38
	3p′(1/2) 1	672.11	0.03		0.05
	3p′(1/2) 0	772.46	0.13		0.14
5s(3/2) 2	3p(1/2) 1	568.98	1.30		1.27
	3p(5/2) 3	618.22	**3.60**		**3.60**
	3p(5/2) 2	624.67	0.78		0.81
	3p(3/2) 1	636.50	0.16		0.17
	3p(3/2) 2	644.47	1.14		1.17
	3p′(3/2) 1	664.00	0.65		0.064
	3p′(1/2) 1	675.96	0.21		0.17

Discussion

In the present study extensive Ne spectra contaminated with only a few He lines were generated using the indirect influence of the He discharge. As will be shown in the following paper [5] a major source for the observed emissions is the excitation of Ne atoms in the energy transfer reaction of Ne with metastable He(2^1S).

The emission intensities measured in the present work are taken to be proportional to the number of atoms in the upper excited state and to the corresponding transition probability. Thus, for an emission array from a common upper level, the ratios of the line intensities determine the relative probabilities for competing transitions to lower states. To facilitate comparison with literature values one of the stronger intensities of an array has been set equal to one of the corresponding literature values. Both values are bold-faced in Tables 1 to 4. If available, the most recent experimental literature values [4, 11, 13, 16] are listed in these tables, otherwise the calculated data reported by Lilly [12, 14, 15] were preferred for comparison.

Tables 1 to 4 show that generally the present experimental values agree well with those reported in the literature. However, the relative intensities of some emission lines were observed to be not in agreement with the corresponding transition probabilities reported. These discrepancies will be discussed briefly in the following section.

While there are no discrepancies observed in the 3p, 3p′, 3d, and 3d′ arrays there are several disagreeing relative intensities in the 4d and 4d′ arrays (Table 3). Particularly, in the 4d(1/2)0 array, only one relative intensity agrees with the data reported by Lilly [14]; in the 4d(1/2)1 array only half of the measured intensities agree with the previously calculated values [14]. Furthermore, in each of the 4d(3/2)1 and the 4d′(3/2)1 arrays there is one measured value not in accord with that of the calculation [14]. Unfortunately, for the 4d and 4d′ arrays, there appears to be only one set of literature data [14] available and hence a comparison with additional theoretical or experimental data is not possible.

For the 5s′(1/2)0 $\rightarrow$ 3p(1/2)1 transition (Table 4) the present intensity is smaller than that expected from the calculation by Lilly [15]. A calculation by Loginov and Gruzdev [17] confirms Lilly's calculation. Furthermore a similar disagreement between calculation [15] and experiment (Ref. [16] and the present work) is observed for the data of the 5s′(1/2)1 $\rightarrow$ 3p(1/2)1 transition.

One measured intensity of the 5s(3/2)1 array at 672.11 nm is too large. It should be noted that Loginov and Gruzdev [17] report a value of $0.038 \times 10^6\ s^{-1}$ which is almost within the error limits of the present value. In the 5s(3/2)2 array, for the intensity at 664.00 nm, there is a difference of a factor of ten between the calculation [15] and the present experiment. Loginov and Gruzdev [17], however, report a value of $0.073 \times 10^6\ s^{-1}$ in very good agreement with our experiment.

It should be noted that the intensities of all the emission lines observed to deviate from the corresponding literature values are relatively small. The intensity of the strongest of these lines is smaller than one hundredth of the intensity of the strongest Ne line observed in the present system. Finally, it should be mentioned that the rather good agreement of calculations and experiment suggests the use of Ne emissions to calibrate relative sensitivities of optical arrangements.

Acknowledgement

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