Possibilities of UV laser oscillation on aluminium ion lines

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Abstract. An analysis of the general energy level structure of the singly ionised Al is made. It is found that in the UV spectrum of Al II there are many intensive lines starting from levels which can be populated selectively via charge transfer collisions with either helium or neon ions. The emission spectrum of aluminium in the 200-400 nm spectral range is measured in a cylindrical Al hollow cathode at discharge conditions typical for laser oscillation. An enhancement of the spontaneous emission intensity on a number of Al II lines in the UV spectral range in Ne compared to He discharges is observed. The Al atom density in the cathode for different values of current density and voltage are calculated.

1. Introduction
Hollow cathode glow discharges have been widely used as an excitation medium for a large number of metal vapour laser lines with wavelengths extending from the near infrared to the deep UV spectral range. Nowadays the hollow cathode metal vapour lasers are the only continuously oscillating direct laser source in the deep UV. Up to now laser oscillation in the spectral range bellow 300 nm is achieved on ion lines of only three metals: copper, gold and silver [1]. Cathode sputtering is used to produce the required metal atom density in the hollow cathode discharge.

In the spectrum of singly ionised Al there are more than 200 lines in the UV spectral range – from 200 nm to 400 nm. The ion spectrum in the vacuum UV range is also reach of intensive Al + lines [2]. Many of these lines originating from levels which can be populated selectively via charge transfer collisions with either helium or neon ions. Some of these levels are known to be the upper levels of already obtained Al ion laser transitions, excited in a hollow cathode discharge [3, 4].

To find whether the hollow cathode discharge used in the Al ion laser can be used as an active medium in the deep UV, we have investigated the emission spectrum of Al hollow cathode discharge at conditions typical for laser oscillation. The relative emission intensities of aluminium ion lines in the 200-400 nm spectral range are measured and compared for He and Ne as buffer gases.

The aluminium atom density in the hollow cathode discharge is produced by self-sputtering of the cathode. Aluminium compared to Cu, Au and Ag, sputters poorly, so it is important to find discharge conditions at which sufficient Al density (>10^{13} atoms/cm^{3}) could be created by sputtering. We have calculated the Al atom density in the cathode for different values of current density and voltage and discussed the possibility to obtain laser gain on a number of Al ion lines in the UV spectral range.

2. Analysis of the UV spectrum of singly ionised Al
An analysis of the general energy level structure of the singly ionised Al shows that a large number of high lying levels of Al + should be populated selectively through charge transfer collisions with He
ions. In Ne it is believed [3, 4] that the already obtained laser oscillation on Al ion transitions is a result of direct charge-transfer between Al atoms and ground state Ne ions, demonstrating the overpopulation of certain Al$^+$ levels by collisions with Ne$^+$. In Figure 1 partial energy diagrams of singly ionised Al are shown [2], regarding the levels population with He$^+$ (Figure 1-a) and with Ne$^+$ (Figure 1-b). The already obtained laser lines in Ne-Al hollow cathode discharge are also indicated.

![Energy Diagrams](image)

**Figure 1.** Partial energy diagrams of Al ion: population with He$^+$ (a) and with Ne$^+$ (b)

Several groups of transitions in the deep and vacuum UV spectral range which are likely to be excited through charge transfer with He ions, are marked by solid lines in Figure1-a. With A (Figure1-a) are specified about 20 transitions in the spectral range from 230 nm to 275 nm to the 3p$^2$S$_0$, 4$^1$P$_1$ and 3d$^3$D$_2$ lower levels, respectively. Two of these levels are the lower levels of two laser lines: 692.0 nm and 747.1 nm. There are also 17 lines in the 235-245 nm spectral range with lower levels 4p$^2$P (B in Figure1-a). About 30 lines are registered in the vacuum UV (150-200 nm) with lower levels 3p$^2$1D, 4s$^2$S, 3p$^3$P and 3d$^3$D (C in Figure1-a). Two of the lower levels are also lower laser levels and are depopulated through strong optical transitions. The same lower levels are populated also by about 20 lines in the 200-260 nm spectral range, originating from levels with energy 15.5-17.5 eV which are very likely populated by cascade transitions from higher lying levels, also excited through charge transfer with He$^+$. A similar mechanism is believed to be responsible for the oscillation of the 747.1 nm Al$^+$ line in He-Al hollow cathode discharge.

In Figure1-b the UV transitions on which enhancement due to selective excitation with by Ne$^+$ can be expected are shown. Compared to He in Ne there are only few transitions, which appear to have favourable characteristics as laser transitions in the UV spectral range.

3. Experimental

3.1. Discharge tube design

The measurements are performed using a specially designed discharge hollow cathode tube (Figure 1). The cathode is a cylinder made of high purity aluminium with 4 mm inner diameter and 28 cm length. Along the cathode length a 2 mm slit is cut. The cathode is inserted in a quartz tube with a slit above the cathode slit; hence the discharge can burn only inside the aluminium cylinder. The anode is a tungsten rod, 2 mm in diameter, placed at 3 mm distance parallel to the cathode slit. The two electrodes are mounted in a glass tube, connected to a vacuum pump.

The discharge is excited by 3 ms sinusoidal current pulses with 12.5 Hz pulse repetition rate and current amplitude up to 8 A, a value limited by discharge instabilities. The discharge voltage, depending on the type of the buffer gas, its pressure and the discharge current varies from 200 V to
550 V. We have measured the on-axis spontaneous emission intensity of aluminium, and neon or helium atom and ion lines emitted from the whole discharge volume at different discharge conditions. The voltage drop between the anode and the cathode, the cathode current, are also measured. All pulses are recorded by a digital 200 MHz Tektronix oscilloscope (TDS 420A) with a 20-MHz cut-off filter and are stored in a computer. A Digikrom spectrograph equipped with a CCD camera and a 1200 g/mm grating (blaze wavelength 300 nm) has been employed to scan the emission spectrum in the 200-400 nm spectral range.

### 3.2. Results

The discharge is excited in Ne, He or a mixture of He with 5% Ar (to promote sputtering) at buffer gas pressure from 0.2 to 1.0 kPa which is typical for laser oscillation [2, 3]. In only He as a buffer gas it is found hard to excite the aluminium ion lines in the UV range. By adding a small amount of Ar in the He the intensity of the UV emission spectrum increases but it is much weaker compared to the spectrum in Ne buffer gas. At our experimental conditions only few very weak emission lines are found in He-Ar mixture as a buffer gas in the spectral range from 200 nm to 400 nm. In Ne we have registered several Al ion lines and enhancement is achieved on only 3 lines: 247.5 nm, 263.2 nm and 358.7 nm.

![Discharge tube](image)

**Figure 2.** Discharge tube

**Figure 3.** Recorded spectra in He&Ar (upper spectrum) and in Ne (lower spectrum) buffer gas.

Figure 3 indicates spectral scans in the wavelength range from 245 nm to 286 nm (Figure 3 a) and from 350 nm to 365 nm (Figure 3 b) measured in Ne and in He&Ar buffer gas.

### 4. Calculations

To sustain laser oscillation a metal atom density higher than $10^{13}$ atoms/cm$^3$ is needed. The sputtering yield of Al is worse than that of Cu, Ag and Au, but as it is demonstrated in [3,4], it is possible to
produce sufficient for laser oscillation aluminium atom density by sputtering in hollow cathode discharge. To evaluate the average active particle densities responsible for the formation of population inversion and lasing on aluminium ion lines in the Ne-Al and He-Al discharges, we have used the discharge model of Warner et al. [5].

4.1. Model
This simple analytical model explains many of the operating characteristics of sputtered metal-ion charge-transfer excited hollow cathode lasers and has been applied earlier for evaluating the average particle densities as well as the radial dependence of particle densities in slotted and in cylindrical copper hollow cathodes [5-7].

The main simplifications and approximations, described in details elsewhere [5-7], are the following. The negative glow region inside the cathode is nearly field-free and the voltage drop is nearly equal to the discharge voltage. The plasma is homogeneous in the whole cathode cavity and there is a little or no variation of discharge parameters along the optical axis. Only the three major species in the discharge are considered: Al, Al+ and buffer gas (He+ or Ne+) ions. The buffer gas atoms are ionised mainly by beam electrons. The aluminium ions are created primarily by a charge transfer process between metal atoms and buffer gas ions. The metal atoms are produced by sputtering of the cathode: The sputtering is caused by both buffer gas ions and metal ions. The metal atoms, metal ions and buffer gas ions densities in the discharge are coupled by the charge transfer reaction. The dominant loss mechanism of particles in the discharge is the ambipolar diffusion. All other processes and species in the discharge are neglected under our experimental conditions.

The current density $J$ at the cathode surface is given by

$$ J = e \cdot r / 2 \cdot \sum (1 + \gamma_i) \cdot N_i \cdot H \cdot D_i. $$

$N_i$ is the spatially averaged particle density, $D_i$ is the average diffusion coefficient for the corresponding particle type and $H$ is a factor, determined by the boundary conditions and approximately $H = (2.405/r)^2$ for a cylinder with a radius $r$.

The spatial distributions of Al atoms ($i=1$), Al ions ($i=2$) and buffer gas ions ($i=3$) are determined by the diffusion equation for the corresponding particle density $N_i$. In a steady state and if the axial diffusion is neglected,

$$ -N_i / \tau_i + P_i = 0 $$

where $P_i$ is a source term, which includes various mechanisms of particle generation and losses.

The following mechanisms of generation and losses of metal atoms and ions in the discharge are considered. Metal atoms are generated by sputtering of the cathode by the remaining in the cathode metal ions and buffer gas ions, and are annihilated by charge transfer collisions with rare gas ions:

$$ P_i = \xi_2 \cdot N_2 / \tau_i + \xi_3 \cdot N_3 / \tau_i - K_{ct} \cdot N_i \cdot N_1 \cdot N_3 $$

with $\xi_2$ and $\xi_3$ being the sputtering coefficients of Al and buffer gas ions, respectively, and $K_{ct}$ the charge transfer rate constant.

The metal ions are produced mainly by charge transfer collisions and their loss is due only to ambipolar diffusion:

$$ P_2 = K_{ct} \cdot N_1 \cdot N_3. $$

Combining Eqs. (1)-(4), three quadratic equations for the spatially averaged particle densities in the discharge as a function of current density are obtained.
4.2. Results

To find the current dependencies of the particle densities, information about the voltage-current characteristics and the values of the parameters $K_{ct}$, $\gamma$, $A_0$, $\xi$, $D_0$, $T_g$ and $T_e$, approximated for the investigated discharge conditions, are needed.

The values of the parameters in Eqs. (2-5), shown in Table 1, are determined for the investigated neon and helium pressures and the measured voltage-current characteristics. As we could not find information for $A_i$ of Al, as well as for the $T_g$ and $T_e$, we have used data for He-Cu and Ne-Cu hollow cathode discharge at similar discharge conditions. All calculations are made for two different values for the charge transfer rate constants as calculated in [8].

| Parameter                    | in He                  | in Ne                  | Ref. |
|----------------------------|------------------------|------------------------|------|
| Charge transfer rate constant, [cm$^3$/s] | $k_{ct}$               | $45.63 \times 10^{-10}$ | $8.17 \times 10^{-10}$ | [8] |
|                             | $21.01 \times 10^{-10}$| $11.54 \times 10^{-10}$|      |
| Secondary electr. emission coeff., [electron/ion] | $\gamma_2$             | $0.675 + 0.002 \times U - 2.79583 \times 10^{-6} \times U^2 + 1.29167 \times 10^{-9} \times U^3$ | [9] |
|                            | $\gamma_3$             | $0.1$                  |      |
| Sticking coefficient        | $A_1$                  | $0.05 \times (1 + 10 \times J)$ | [7] |
|                            | $A_2$                  | $0.8$                  |      |
| Sputtering coefficient, [atoms/ion] | $\xi_2$               | $0.055 + 0.003 \times U + 1.6556 \times 10^{-6} \times U^2$ | [10] |
|                            | $\xi_3$               | $0.001$                | $0.02$ | [11, 12] |
| Diffusion coefficient, [cm$^3$/s] | $D_1$                  | $3.3 \times T_g^{3/2} / p_{He}$ | $1.4 \times T_g^{3/2} / p_{Ne}$ | [13] |
|                            | $D_2$                  | $2 \times D_3$         | $2 \times D_3$         |      |
|                            | $D_3$                  | $0.3 \times T_g^{3/2} / p_{He} (1 + T_e/T_g)$ | $0.1 \times T_g^{3/2} / p_{He} (1 + T_e/T_g)$ | [14] |
| Gas temperature, [K]        | $T_g$                  | $537 + 5441 \times J - 14548 \times J^2 + 13722 \times J^3$ | $700 + 1778 \sqrt{J} + 155 \times J$ | [15] |
| Electr. temp. [K]          | $T_e$                  | $716 + 3516 \times J$  |      | [16] |

In Figure 4 the calculated dependence of Al atoms, ions and He and Ne ions on discharge current density are shown at 270 Pa gas pressure. The solid lines are for the first set of charge transfer rate constants ($45.63 \times 10^{-10}$ in He and $8.17 \times 10^{-10}$ in Ne) and the dotted lines are for the second set ($21.01 \times 10^{-10}$ in He and $11.54 \times 10^{-10}$ in Ne) [8]. As it is seen in the figure, the influence of the charge transfer rate constants is small for Al atoms and there is practically no change for the He and Ne ions.

The Al atom density increases with current and is comparable in He and Ne discharge, presumably because the Al atoms are produced primarily by sputtering with Al ions. At low current the Al ion density grows with current, but then it saturates, probably due to the low density of gas ions ($\sim 10^{12}$ cm$^{-3}$). At the comparatively low gas pressure, typical for the Al-Ne laser [3,4], nearly all gas ions are converted to atoms as a result of the charge exchange collisions with Al atoms and the further increase of metal atom density does not contribute to raising of Al ion density. The density of Al ions in He discharge is nearly 2 orders of magnitude lower than that in Ne, which is in agreement with the observed UV spectrum in He and Ne discharges.

The results from calculations for higher gas pressure – 600 Pa, show that although the density of metal atoms decreases, due to the higher gas ion density, the Al ion density grows by almost one order of magnitude.

5. Conclusion

At the investigated discharge conditions in the conventional hollow cathode discharge it is hard to excite the UV spectrum of singly ionised Al in He as buffer gas. In Ne discharge an enhancement of the intensity of several Al ion lines in the 200-300 nm spectral range is observed, demonstrating
selective population of certain levels, excited by charge-transfer collisions with Ne ions. We suppose that if discharge conditions for more efficient excitation of these transitions are created, it is possible to obtain laser gain in the deep UV spectral range on Al ion lines.

**Figure 4.** Calculated Al atoms and ions, and He and Ne ions particle densities in He (○) and in Ne (△) discharge: $k_{ct} = 45.63 \times 10^{-10}$ in He and $k_{ct} = 8.17 \times 10^{-10}$ in Ne (solid lines), and $k_{ct} = 21.01 \times 10^{-10}$ in He and $k_{ct} = 11.54 \times 10^{-10}$ in Ne (dotted lines).

The calculation results show that Al atom density, comparable to that in He-Cu hollow cathode discharge [5] is obtained. As the ion density in the conventional hollow cathode discharge saturates with current, we suggest that more favourable conditions for excitation will be realised in the high-voltage variants of the hollow cathode discharge [17-19].

**References**

[1] Gerstenberger D C, Solanki R and Collins G J 1980 *IEEE J.Quant. Electron.* QE-16 820
[2] *NIST Atomic Spectra Database Data* http://physics.nist.gov/cgi-bin/AtData/main_asd
[3] Gerstenberger D C, Reid R D and Collins G J 1977 *Appl.Phys.Lett.* 30 466
[4] Schuebel W K 1977 *Appl.Phys.Lett.* 30 516
[5] Warner B E, Persson K B and Collins G J 1979 *J.Appl.Phys.* 50 5694
[6] Koch H and Eichler H J 1983 *J.Appl.Phys.* 54 4939
[7] Hamisch J and de la Rosa J 1987 *Appl.Phys.* B 43 189
[8] Temelkov K, Vuchkov N and Sabotinov N *Plasma Processes in Polymers*, to be published
[9] Thomas E W ed. 1985 *ORNL Atomic Data for Fusion*, v.3 Particle Interaction with Surfaces
[10] Hayward W and Walter A 1969 *J.Appl.Phys.* 40 2911
[11] Laegreid N and Wehner G 1961 *J.Appl.Phys.* 32 365
[12] Rosenberg D and Wehner G 1962 *J.Appl.Phys.* 33 365
[13] Sekido H et al. 1993 *J.Phys.D:Appl.Phys.* 26 1441
[14] McDaniel E and Mason E 1973 *The Mobility and Diffusion of Ions in Gases* (NY: Wiley)
[15] Arslanbekov R 2000 *J.Phys.D:Appl.Phys.* 33 524
[16] McNeil J R, Collins G J and DeHoog F J 1979 *J.Appl.Phys.* 50 6183
[17] Rozsa K, Janossy M, Csillag L and Bergou J 1977 *Phys.Lett.* 63A 231
[18] Grozeva M and Sabotinov N 1984 *Opt.Commun.* 51 417
[19] Peard K, Donko Z, Rozsa K, Szalai L and Tobin R *IEEE J.Quantum Electron.* QE-30 215