Note on Population Densities in an Expanding Helium-Neon Plasma Flow

Michio Nishida *

Stoßwellenlabor, Institut für Luft- und Raumfahrt Technische Hochschule, Aachen

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The axial population distributions in an expanding helium-neon mixture plasma flow have been calculated for three cases of the concentration of neon ions. The results show that the electron temperature distributions are strongly dependent on the concentration of the neon ions and that the populations of the upper levels of the helium atom depend on the electron temperature near downstream and on the flow expansion far downstream.

Hurle and Hertzberg firstly proposed the promotion of population inversion between electronically excited states by fluid mechanical technique 1, where the possibility of population inversion between the 3p- and 4s-state of the neon atoms in a rapidly expanding helium-neon plasma flow was stated. The He(2s3S) metastables transfer their energy to the near-resonant Ne(4s) levels and the Ne(4s) levels decay to the Ne(3s) levels through the Ne(3p) states. The radiative lifetime of the 3p-4s and 3s-3p transitions are 1.5×10^{-7} sec and 7×10^{-9} sec, respectively 2. Therefore, if the expansion is made so rapidly that the flow time becomes sufficiently small compared with collisional transition time, the population inversion between the 3p- and 4s-state of the neon atoms will become possible. In the present paper the behaviour of the populations of the helium and neon atoms in a source flow expansion of a He-Ne plasma is investigated as a preliminary study to pursue the possibility of a helium-neon laser by fluid mechanical technique. In a previous paper 3 the populations of the helium atoms in a source flow expansion were studied, where it was stated that the behaviour of the electron temperature was quite similar to that of the heavy particles. This phenomenon was explained as the result of the small mass of the helium atoms. In the helium-neon gas mixture the different behaviour of the electron temperature will be expected, which will likely produce the different behaviour of the helium populations. In addition the energy transfer from He(2s3S) to Ne(4s) will also affect the population distributions.

Since the excitation model of the helium atoms to be used here is the same as that used in the previous paper 3 , it is simply stated here. The excitation model of the helium atoms is shown in Table 1. For $n \le 4$ (n: the principal quantum number) the singlet and triplet levels are individually treated because of the large energy difference between them. For n > 4 both members with equal principal quantum number are treated as one level. The popula-

Table 1. Groupings of actual quantum states of the helium

| j | Singlet | Triplet |
|---|---|------------------|
| 0 | 1s ² ¹S | |
| 1 | $2s^{1}S$ $2s^{3}S$ | |
| 2 | $2p^{1}P$ $2p^{1}P$ | |
| 3 | 3s ¹ S, 3d ¹ D, 3p ¹ P | 3s3S, 3d3D, 3p3P |
| 4 | n=4 | n=4 |
| 5 | n=5 | |
| 6 | n = | 6 |
| • | : | |
| : | : | |

Table 2. Groupings of actual quantum states of the neon atoms.

| j | g | terms | radiative transition probability |
|---|-----|--------------------|--|
| 0 | - 1 | 2p ⁶ ¹S | |
| 1 | 12 | 3s | |
| 2 | 36 | 3p | $A(2,1) = 1.42 \times 10^8 \text{ sec}^{-1}$ |
| 3 | 12 | 4s . | $A(3,2) = 6.67 \times 10^6 \text{ sec}^{-1}$ |
| 4 | 96 | 3d, 4p | $A(4,1) = 8.86 \times 10^{5} \text{ sec}^{-1}$ $A(4,2) = 1.24 \times 10^{7} \text{ sec}^{-1}$ $A(4,3) = 2.16 \times 10^{6} \text{ sec}^{-1}$ |
| 5 | 12 | 5s | |
| 6 | 164 | 4d, 4f, 5p | |



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^{*} Present Address: Department of Aeronautical Engineering, Kyoto University, Kyoto, Japan.

tions of $n \ge 7$ are considered to have a relaxation time of de-excitation sufficiently smaller than the flow time so that the populations of $n \ge 7$ are calculated from consideration of steady state population. It is assumed that the levels of $n \ge 20$ are populated according to Saha equilibrium. The excitation model of the neon atoms is shown in Table 2. All levels higher than the sixth level (i = 6) of the neon atoms are taken to be in thermal equilibrium with free electrons, and the fifth level (j=5) of the neon atoms is estimated from the steady state consideration. The populations lower than the fourth level are solved from a set of population continuity equations coupled with the continuity equations of the helium ions, the neon ions and the helium populations, and the electron energy equation. As transition model we are considering the conventional collisional-radiative transition, and in addition the energy transfer from He(2s3S) to Ne(4s) is taken into account. Except for the transitions from $He(np^{1}P)$ to $He(1s^{2} {}^{1}S)$ and from Ne(3s) to Ne(2p⁶ ¹S), the plasma considered here is taken to be optically thin. The effective value of the radiative transition rates from $He(np^{1}P)$ to $He(1s^{2} {}^{1}S)$ and from Ne(3s) to Ne(2p⁶ 1S) are taken to be zero due to radiation trapping. However, as the flow expands, the population density of the ground state (which is approximately equal to the atom density) decreases so that the above consideration will produce error far downstream. Nevertheless this consideration is used to investigate the behaviour of the He-Ne plasma as a preliminary study. The plasma of interest here is partially ionized, which is justified if we are interested only in the behaviour of the populations and the charged particles. Such a plasma expands into a vacuum as a freejet expansion, that is replaced by a source flow expansion in the analysis. Using the conventional assumptions for a partially ionized plasma, we have the following governing equations 3:

Continuity of ions:

$$\frac{1}{r^2} \frac{\mathrm{d}}{\mathrm{d}r} \left[r^2 N_+(k) u \right] = N_e \sum_{j=0}^{n^*} Q_l^{(k)}(j, e^-) \left(N_j^{(k)} - N_{j, E}^{(k)} \right) (k = \text{He, Ne}) . \tag{1}$$

Continuity of population of the jth level:

$$\frac{1}{r^2} \frac{\mathrm{d}}{\mathrm{d}r} (r^2 N_j u) = \sum_{s=0}^{\infty} Q(s, j) N_e N_s - \sum_{s=0}^{\infty} Q(j, s) N_e N_j + Q_l(j, e^-) (N_{j,E} - N_j) N_e + \sum_{s=j+1}^{\infty} A(s, j) N_s - \sum_{s=0}^{j-1} A(j, s) N_j.$$
(2)

Electron energy:

$$\frac{3}{2}kN_{e}u\frac{dT_{e}}{dr} + N_{e}kT_{e}\left(\frac{du}{dr} + \frac{2u}{r}\right) = 2km_{e}N_{e}(T_{a} - T_{e})\sum_{k=He, Ne} \frac{\nu_{ei}^{(k)} + \nu_{ea}^{(k)}}{m_{a}^{(k)}} - \sum_{k=1}^{n^{\bullet}} \sum_{j=0}^{n^{\bullet}-1} (E_{k} - E_{j})\left[Q(j,k)N_{j}N_{e} - Q(k,j)N_{k}N_{e}\right] - \sum_{j=0}^{n^{\bullet}} (I - E_{j} + \frac{3}{2}kT_{e})Q_{I}(j,e^{-})(N_{j} - N_{j,E})N_{e}$$
(3)

where the conventional notations are used and r is the distance from the source; $N_+(k)$ the ion density of the k-species $k=\mathrm{He},\ \mathrm{Ne}$); N_j the population density of the jth level; $N_{j,\mathrm{E}}$ the equilibrium population density of the jth level; A(s,j) the Einstein A coefficient for the radiative transition from the sth level to the jth level; I the ionization energy; E_j the excitation energy of the jth level; $N_{\mathrm{e}}=N_+(\mathrm{He})+\mathrm{N}_+(\mathrm{Ne})$; Q(j,k) and $Q_I(j,\mathrm{e}^-)$ are the collisional rate constants for the transition from the jth level to the kth level (k>j) and the collisional ionization rate constant from the jth level, respectively, which are expressed as, when the modified

Thomson formula 4 for the encounter of the electrons and the excited atoms is used,

$$Q(j,k) = \pi e^4 \left(\frac{8}{\pi m_e}\right)^{1/2}$$
 (4)

$$\left. \cdot (kT_{\rm e})^{-3/2} \left[\frac{\exp{(-u_{j,k})}}{u_{j,k}(u_{j,k}+1)} - \frac{\exp{(-u_{j,k+1})}}{u_{j,k+1}(u_{j,k+1}+1)} \right],$$

$$Q_I(j, e^-) = \pi e^4 \left(\frac{8}{\pi m_e}\right)^{1/2} (k T_e)^{-3/2} \frac{\exp(-u_{j,\infty})}{u_{j,\infty}(u_{j,\infty} + 1)}$$
(5)

where $u_{j,k} = (E_k - E_j)/k T_e$ and $u_{j,\infty} = (I - E_j)/k T_e$. The expression of the rate constants for the

collisional transition from the singlet to triplet of the helium atoms and the inverse transition were proposed by Drawin and Emard ⁵.

$$Q^{\text{TS}}(j,j) = 7.2 \times 10^{-7} j^{6} T_{e}^{-1/2} \frac{E_{j}^{\text{S}} - E_{j}^{\text{T}}}{E_{1}^{\text{H}}}$$

$$\cdot \exp \left\{ -\frac{E_{j}^{\text{S}} - E_{j}^{\text{T}}}{k T_{e}} \right\} (\text{cm}^{3} \text{sec}^{-1}) .$$
(6)

For k > j, the following expression was proposed:

$$\begin{split} Q^{\rm TS}(j,k) = & 2.862 \times 10^{-6} \bigg(\frac{E_1^{\rm S} - E_1^{\rm T}}{E_k^{\rm S} - E_j^{\rm T}} \bigg)^2 T_{\rm e}^{-1/2} & (7) \\ & \cdot \exp \bigg\{ -\frac{E_k^{\rm S} - E_j^{\rm T}}{k T_{\rm e}} \bigg\} \, (\text{cm}^3 \, \text{sec}^{-1}) \; . \end{split}$$

In Eq. (6) $E_1^{\rm H}$ represents the ionization energy of hydrogen; the superscript TS means the transition from the triplet to the singlet; T and S denote the triplet and singlet levels, respectively. Equation (7) with the superscripts S and T interchanged holds for the collisional excitation transition from the singlet to the triplet level. The collisional de-excitation rate constant Q(k, j) (k > j) is easily estimated from the principle of detailed balance. The term of the resonant energy transfer from He(2s3S) to Ne(4s) is contained in the population continuity equations of their levels. The total velocity-averaged cross section for the encounter of He(2s3S) and $Ne(2p^{6} {}^{1}S)$ is taken to be $3.7 \times 10^{-17} cm^{2}$ (see Reference 6). The radiative transition probabilities of the helium and neon atoms, which are given in Ref. 2, are used in the present analysis. Equations (1) - (3) have been non-dimensionalized in the same way as in Ref. 3 and numerically solved by using the Runge-Kutta-Gill method, on the digital computer CD 6400 at the Computer Center of the Technische Hochschule Aachen.

The following conditions have been taken at the starting point $(r/r^*=1)$: $T_e^*=T_a^*=7500 \,\mathrm{K}$, $N_a^*(\mathrm{He})=10^{16} \,\mathrm{cm}^{-3}$, $N_+^*(\mathrm{He})=10^{14} \,\mathrm{cm}^{-3}$, where the superscript * denotes the condition at $r/r^*=1$. In this analysis r^* has been taken to be 1 cm. The calculations have been carried out for three cases of the neon concentration; Case I: A=B=0.5, Case II: A=B=0.1, Case III: A=B=0.01 where

$$A = N_a^* (\text{Ne}) / \{N_a^* (\text{He}) + N_a^* (\text{Ne})\}$$
 and $B = N_+^* (\text{Ne}) / \{N_+^* (\text{He}) + N_+^* (\text{Ne})\}$.

The initial conditions of the populations were determined from a steady state consideration and the numerical calculations started at the sonic point $(r/r^*=1)$. The ground level population density N_0 (which is equal to N_a in the present condition) and the atom temperature T_a , which appear in Eqs. (1)-(3), have been estimated from an isentropic expansion consideration ³.

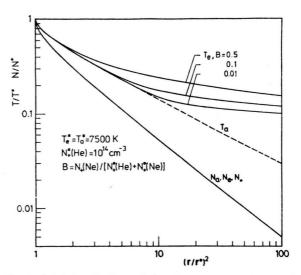


Fig. 1. Axial distributions of the electron temperature, and the electron density.

Figure 1 shows the axial distributions of the electron temperature, which reveal that the position, where the electron temperature deviates from the atom temperature, approaches the sonic point with increasing neon concentration and that after the deviation the electron temperature gradient gradually becomes small. When the plasma consists of only helium-species, the electron temperature behaves in the same way as the heavy particle temperature, that was already stated in Reference 3. Surprisingly, the effect of the neon-species on the electron temperature is considerably strong even in the case of only 1% neon (Case III). It is expected that in a two-species mixture plasma with a large difference between their masses one can control the electron temperature by altering the concentration of heavier ions. Figure 1 also illustrates the axial distributions of the atom, ion and electron densities. Calculated results of the ion and electron densities show the negligible difference among the density distributions of these three species. For the case of A = B = 0.1, the degree of ionization of helium varies only from 9×10^{-3} at the starting point (sonic point or $r/r^* = 1$) to 8.97×10^{-3} at r/r^* = 10 [or $(r/r^*)^2 = 100$ on Figure 1]. The decrease

in the degree of ionization is only 0.33%. According to Drawin and Emard 7 , the relaxation time of recombination of helium is of the order of 10^{-2} sec for the present condition at the initial point ($N_{\rm e}=10^{14}~{\rm cm}^{-3}$ and $T_{\rm e}=7500~{\rm K}$) while the characteristic time of the expansion flow is of the order of 10^{-5} sec. Therefore one may state that the behaviour of electrons (or ions) follows the flow expansion.

The axial population distributions of the helium atoms are shown in Figs. 2 to 4, which represent a

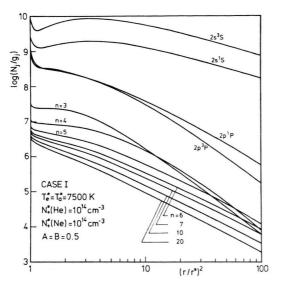


Fig. 2. Axial population distributions of the helium atoms for Case I.

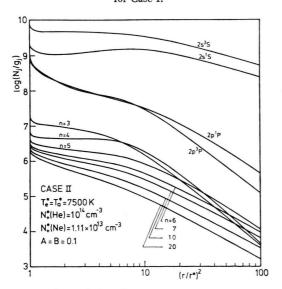


Fig. 3. Axial population distributions of the helium atoms for Case II.

similar tendency. In Fig. 4 (Case III) the bend of the distribution can be found downstream of $(r/r^*)^2$ = 10, and the similar bend is appreciable in Fig. 3 (Case II). The explanation of this bend is given as follows: In Case III the electron temperature gradient becomes small downstream of $(r/r^*)^2 = 10$. Since the exponential part in the expression of collisional transition probability is very sensitive to the electron temperature, the decrease in the electron temperature up to $(r/r^*)^2 = 10$ makes the value of the exponential part quite small, i.e. the collisional excitations become small. Consequently the collisional de-excitations relatively increase, which produces a small gradient of the populations up to $(r/r^*)^2 = 10$. After the electron temperature gradient becomes small, the decrease in the populations is dominated by the behaviour of the electrons which follow the expansion of a flow. Especially for $n \ge 5$ it may be stated that the decrease in the populations results wholly from the decrease in the electron density due to the expansion of the flow while the gradient of the populations of n=3, 4, and $2p^{1}P$ and $2p^{3}P$ is larger than that of $n \ge 5$. The latter results from the fact that the relaxation time of populations of these lower levels due to radiative transitions is still smaller than the flow time even far downstream. On the other hand the populations of both 2s3S and 2s1S have a small gradient. The radiative decay from these two levels is optically forbidden and the collisional decay is

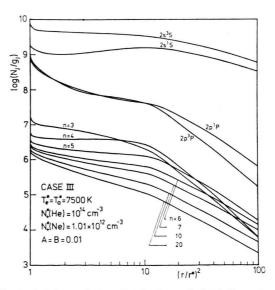


Fig. 4. Axial population distributions of the helium atoms for Case III.

quite small. The populations of these levels are therefore controlled mainly by the income from the upper levels.

Figures 5-7 show the axial distributions of populations of the neon atoms. The effect of the decreasing electron temperature on the population distributions is clearly observed. The population of j=6 is considered to be in equilibrium with free electrons so that it has been calculated from the Saha equilibrium equation. Although the electron and ion densities rapidly decrease with the atom density as shown in Fig. 1, the exponential part in the Saha equation is still predominant rather than the part of NeN₊ even far downstream.

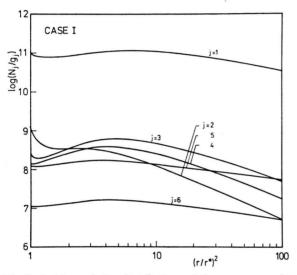


Fig. 5. Axial population distributions of the neon atoms for Case I.

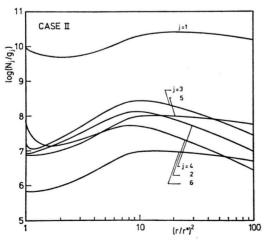


Fig. 6. Axial population distributions of the neon atoms for Case II.

The axial distributions of the population inversion between the 3p- and 4s-level of the neon atoms, which is non-dimensionalized by the population density of the 4s-level at sonic point, is shown in Figure 8. The distributions obviously depend on the neon concentration. If one lets Fig. 8 correspond with Figs. 5 to 7, one can easily see that this result comes from the electron temperature distribution. In the case of A=B=0.5, the population influx to Ne(4s) due to the energy transfer from He(2s³S) to Ne(4s) is 1.0×10^{15} cm⁻³ sec⁻¹ at $r/r^*=1$ and

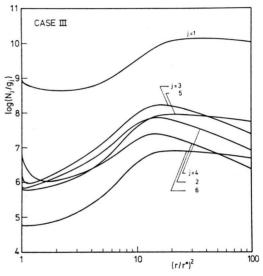


Fig. 7. Axial population distributions of the neon atoms for Case III.

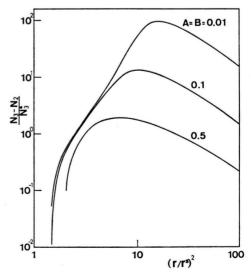


Fig. 8. Axial distributions of population inversion.

 $8.6 \times 10^{10} \, \mathrm{cm^{-3} \ sec^{-1}}$ at $r/r^* = 10$, while the population flux from the 4th level of the neon atoms to Ne(4s) varies from $8.8 \times 10^{18} \, \mathrm{cm^{-3} \ sec^{-1}}$ at $r/r^* = 1$ to $6.2 \times 10^{15} \, \mathrm{cm^{-3} \ sec^{-1}}$ at r/r = 10, and the population influx to Ne(4s) due to the recombination varies from $7.2 \times 10^{14} \, \mathrm{cm^{-3} \ sec^{-1}}$ at $r/r^* = 1$ to $9.6 \times 10^8 \, \mathrm{cm^{-3} \ sec^{-1}}$ at $r/r^* = 10$. The population influx to Ne(4s) due to the resonance energy transfer is quite small compared with that from the 4th level of the neon atoms. Under the present condition of the neon atom density, it may be stated that the

population of Ne(4s) is supplied mainly from the 4th level of the neon atoms.

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