

Excitation of the 210 nm Zn II Line in a Hollow Cathode He-Ne-Zn Discharge

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Z. Naturforsch. **53a**, 955–959 (1998); received April 6, 1998

In a hollow cathode He-Zn discharge it was observed that addition of Ne stops laser oscillation at the 492.4 nm Zn II line and increases significantly the spontaneous intensity at the 210 nm Zn II line, which originates from the $4d^2D_{5/2}$ lower level of the laser transition. Based on a rate equation model, the cross-section of the Ne ion-Zn atom charge transfer collisions populating the $4d^2D_{5/2}$ level was determined. The cross-section $3.5 \times 10^{-15} \text{ cm}^2$ found by us is in acceptable agreement with the value $2.3 \times 10^{-15} \text{ cm}^2$ reported in the literature. This high cross-section together with existing lower level depopulation points to the possibility of obtaining laser oscillation at 210 nm in a Ne-Zn discharge.

Key words: Gas Lasers; Laser Excitation; Charge Transfer; Glow Discharges; Sputtering.

1. Introduction

Hollow cathode discharge [1] offers a convenient way of producing cw laser oscillation in the ultraviolet range of the spectrum. Several possibilities based on charge transfer collisions with He or Ne ions [2] exist for selective pumping of the upper level of metal vapour ionic transitions, this excitation resulting in cw ultraviolet (UV) laser oscillation. Metals for UV operation (Cu, Ag, Au) [3] should, however, be heated to temperatures above 1000 °C, which process raises several technical problems. At technically feasible currents, cathode sputtering produces metal vapour densities considerably lower than the optimum, this resulting in limiting the maximum power obtainable. However, room temperature operation and the relative ease of obtaining a stable discharge has resulted in wide application of this technique [3]. UV laser action using Cu, Ag, Au has been observed at several ionic transitions, the wavelengths being between 301.8 nm (Ag II) and 224 nm (Ag II). These laser lines have all been obtained in a hollow cathode discharge using cathode sputtering.

Collins suggested the possibility of population inversion and laser action at the 210 nm ($4d^2D_{5/2} - 4p^2P_{3/2}^0$) Zn II line in a Ne-Zn discharge, where charge transfer col-

lisions between Ne ions and Zn atoms excite the upper $4d^2D_{5/2}$ level [4]. This suggestion was based on the close coincidence between the energy of the Ne ion (21.56 eV) and that of the $4d^2D_{5/2}$ Zn II level (21.41 eV), the energy difference being 0.15 eV. Collins measured the cross-section of this Ne ion-Zn atom collision to be $2.3 \times 10^{-15} \text{ cm}^2$ [5]. However, since these measurements were carried out, to the authors knowledge no work has been done to obtain further information related to this question.

Investigations on a cathode sputtered He-Zn laser have shown that the 492.4 nm ($4f^2F_{7/2}^0 - 4d^2D_{5/2}$) Zn II laser oscillation stops, and simultaneously there is a significant increase of the intensity of the 210 nm spontaneous line, when a small amount of Ne (~0.4 mbar) is added to the discharge [6]. The lower level of the 492.4 nm Zn II transition is the upper level of the 210 nm transition, thus this observation is attributed to charge transfer excitation of the $4d^2D_{5/2}$ upper level of the 210 nm Zn II transition by Ne ions, in agreement with the suggestion in [4].

In the present paper, a rate equation model is used to describe population changes of the various Zn II transitions playing a role in populating the $4d^2D_{5/2}$ level. Applying in the model the condition that, when 492.4 nm laser oscillation stops, the upper and lower level populations at 492.4 nm are equal, the cross-section of the Ne ion-Zn atom charge transfer collisions exciting the $4d^2D_{5/2}$ level could be determined. The cross-section was found to be $3.5 \times 10^{-15} \text{ cm}^2$, which value is in acceptable agreement with that given in [5].

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2. Experimental

A description of the experiment concerned with the measurement of the intensity changes of the 492.4 nm laser oscillation and that of the spontaneous 210 nm line is given in the following. The hollow cathode was made of Zn with a 2 mm × 5 mm slot for the discharge, the Zn vapour being produced by cathode sputtering. The 492.4 nm laser was operated using four cathodes placed in series, resulting in an active length of 40 cm [6]. The discharge was excited by 5 Hz, 1 μs, 6 A current pulses. The 150 μs pulse length is long enough for the conditions to be considered being equivalent to cw operation. The 492.4 nm laser intensity and that of the spontaneous 210 nm line were measured by a Zeiss SPM2 monochromator equipped with an EMI 6256S photomultiplier. In order to detect the 210 nm line, one of the laser mirrors was removed.

It can be seen from the experimental results shown in Fig. 1 that by adding Ne to the He-Zn discharge a fast decrease of 492.4 nm laser intensity occurs and the laser oscillation stops completely at 0.4 mbar Ne partial pressure. In contrast, a steep increase in the intensity of the spontaneous 210 nm line was measured in this pressure region. Above a Ne partial pressure of 1 mbar, the 210 nm intensity increases further with a much smaller slope up to the 12 mbar Ne partial pressure used in the measurements.

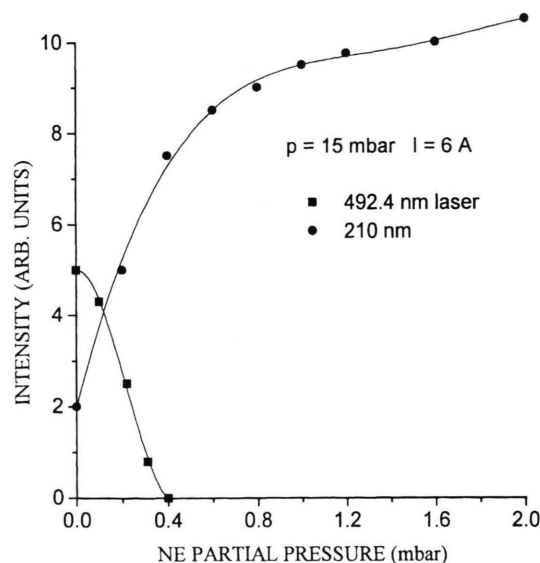
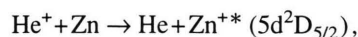


Fig. 1. Dependence of the 492.4 nm laser intensity and the 210 nm spontaneous intensity on the Ne partial pressure.

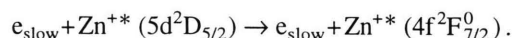
3. Determination of Ne⁺-Zn Collision Cross-section

The main processes taking part in the excitation of the 210 nm transition are considered in order to find the rate equations describing the population variations of the different Zn II levels.

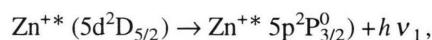
The scheme of the Zn II energy levels involved and the excitation processes taken into account can be seen in Figure 2; the energies of ground state He and Ne ions are also shown in the figure. The 5d²D_{5/2} level is excited by charge transfer collisions between the He ions and Zn atoms,



and this is followed by collisions of slow electrons carrying a part of the 5d²D_{5/2} population to the 4f²F_{7/2}⁰ upper level of the 492.4 nm laser transition,



The other significant population loss of the 5d²D_{5/2} level results from the 610.2 nm transition originating from this level,



where h is the Planck constant and ν_1 denotes the transition frequency.

The 210 nm transition is excited by two processes:

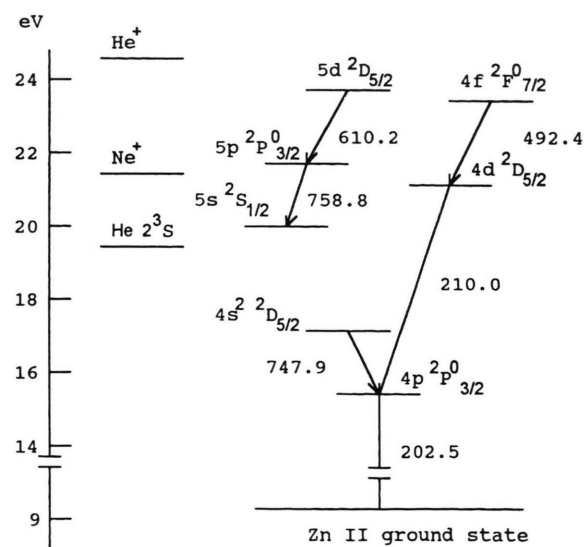
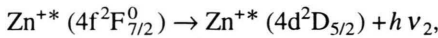


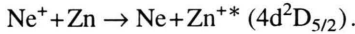
Fig. 2. Energy level diagram of Zn II. The arrows show known laser transitions. Wavelengths are in nm.

I. Cascade excitation through the 492.4 nm transition,



where ν_2 is the frequency corresponding to 492.4 nm.

II. Charge transfer excitation by collisions with Ne ions,



Taking into account these processes considered in the excitation model, the rate equations describing the population variations N of the $5d^2D_{5/2}$ level, N_2 of the $4f^2F_{7/2}^0$ upper laser level and N_1 of the $4d^2D_{5/2}$ lower laser level, respectively, are:

$$\frac{dN}{dt} = N_{\text{He}}^+ N_{\text{Zn}} \langle \sigma \nu \rangle_{\text{He}} - N (A + n_{\text{eL}} \langle \sigma_e \nu_e \rangle), \quad (1)$$

where N_{He}^+ =density of He ions, N_{Zn} =density of Zn atoms, $\langle \sigma \nu \rangle_{\text{He}}$ =charge transfer excitation rate by He ions, σ =cross-section, ν =velocity, A =transition probability from the $5d^2D_{5/2}$ level, n_{eL} =density of slow (low energy) electrons, $\langle \sigma_e \nu_e \rangle$ =destruction rate of $5d^2D_{5/2}$ level by slow electrons, σ_e =cross-section, and ν_e =velocity;

$$\frac{dN_2}{dt} = n_{\text{eL}} N \langle \sigma_e \nu_e \rangle - N_2 A_2, \quad (2)$$

where A_2 =transition probability from the $4f^2F_{7/2}^0$ level;

$$\frac{dN_1}{dt} = N_2 A_2 + N_{\text{Ne}}^+ N_{\text{Zn}} \langle \sigma \nu \rangle_{\text{Ne}} - N_1 A_1, \quad (3)$$

where N_{Ne}^+ =density of Ne ions, $\langle \sigma \nu \rangle_{\text{Ne}}$ =charge transfer excitation rate by Ne ions, and A_1 =transition probability from the $4d^2D_{5/2}$ level.

In the stationary state

$$\frac{dN}{dt} = 0, \quad \frac{dN_2}{dt} = 0, \quad \frac{dN_1}{dt} = 0. \quad (4)$$

The condition when the 492.4 nm laser oscillation stops because of the addition of 0.4 mbar Ne to the He-Zn discharge is taken to be the equality of the upper and lower level populations:

$$N_2 = N_1. \quad (5)$$

On the basis of (1), (2), (3), (4), and (5), and approximating the rates $\langle \sigma \nu \rangle$ by the product of average values $\bar{\sigma} \bar{\nu}$, the following expression is obtained for the cross-section: $\bar{\sigma}_{\text{Ne}}$:

$$\bar{\sigma}_{\text{Ne}} \approx \frac{1}{\bar{\nu}_{\text{Ne}}} \frac{N_{\text{He}}^+}{N_{\text{Ne}}^+} \frac{n_{\text{eL}} \bar{\sigma}_e \bar{\nu}_e \bar{\sigma}_{\text{He}} \bar{\nu}_{\text{He}}}{A + n_{\text{eL}} \bar{\sigma}_e \bar{\nu}_e} \frac{A_1 - A_2}{A_2}. \quad (6)$$

To obtain $\bar{\sigma}_{\text{Ne}}$, the following values were used in (6):

the ratio of the relative average velocities between He ions and Zn atoms and Ne ions and Zn atoms is $\bar{\nu}_{\text{He}}/\bar{\nu}_{\text{Ne}}=2.3$; the ratio of He and Ne ion densities is assumed to be proportional to their partial pressures: $N_{\text{He}}^+/N_{\text{Ne}}^+=37.5$.

The transition probabilities have been measured and are taken from [7]:

$$A = 3 \times 10^8 \text{ s}^{-1}, \quad A_2 = 2 \times 10^8 \text{ s}^{-1}, \quad A_1 = 2.6 \times 10^8 \text{ s}^{-1}.$$

Extrapolation of the data given in [8] give the total electron density n_{eT} . Under our experimental conditions this results in $n_{\text{eT}}=7.7 \times 10^{13}/\text{cm}^3$. The density of low energy electrons was obtained on the basis of the following consideration:

In a hollow cathode discharge the electron-energy distribution can be described by two parts [9]: the lower energy part can be approximated up to the region of the ionization potential of He by a Maxwellian distribution, whereas in the high electron energy region a constant function up to the value of the cathode fall is appropriate. To obtain the density of low energy electrons one needs only to consider the Maxwellian part of the distribution. The average electron energy kT is around 3 eV in hollow cathode discharges corresponding to our experimental conditions, low energy electrons are considered to have an energy below 0.1 eV. Using this condition, the ratio of low energy electron density and total electron density is 1/70, and on the basis of this value the low energy electron density is obtained as $n_{\text{eL}}=1.1 \times 10^{12}/\text{cm}^3$. The cross-section for the slow electron collisions $\bar{\sigma}_e$ and the average velocity $\bar{\nu}_e$ are taken from [10]: $\bar{\sigma}_e=2 \times 10^{-12} \text{ cm}^2$, $\bar{\nu}_e=1.1 \times 10^7 \text{ cm/s}$. The cross-section for charge transfer excitation of the $5d^2D_{5/2}$ level has been measured to be $\bar{\sigma}_{\text{He}}=1.8 \times 10^{-15} \text{ cm}^2$ [11]. With the above data, (6) gives the value $\bar{\sigma}_{\text{Ne}}=3.5 \times 10^{-15} \text{ cm}^2$ for the cross-section of the Ne^+ -Zn atom charge transfer collisions.

4. Discussion

The value of the cross-section $\bar{\sigma}_{\text{Ne}}$ obtained from the rate equation model based on the quenching of 492.4 nm laser operation resulting from the addition of Ne is in acceptable agreement with the value of $2.3 \times 10^{-15} \text{ cm}^2$ measured by Collins [5]. The difference between the two values is probably due to the approximations used in our model.

Laser oscillation where the population inversion is produced by charge transfer collisions between Ne ions and metal atoms occurs in several cases [3]. The small energy difference of 0.15 eV between the Ne ion and the Zn II ($4d^2D_{5/2}$) level and the corresponding high cross-section of the Ne ion-Zn atom collisions result in a high excitation rate of the upper level of the 210 nm line. The lower level of this line – which is also the lower level of the 747.9 nm laser transition – is depopulated by the strong 202.5 nm transition. In view of what has just been said, it would seem that there is the possibility of population inversion and laser action at 210 nm. On the one hand, the use of a He-Ne mixture could be advantageous since it increases the upper level population, while on the other hand the lower $4p^2P_{3/2}^0$ level is populated in a direct manner via Penning ionization collisions or by the 747.9 nm cascade line. Both excitation paths involve metastable He atoms, the density of which can be significantly reduced by resonant energy exchange collisions of the second kind with Ne atoms.

With regard to the pure Ne-Zn system, the main process for lower level population is Penning ionization by the ~ 16.6 eV Ne metastables. The ratio of the upper and lower level populations N_U and N_L can be approximated as

$$\frac{N_U}{N_L} \sim \frac{N^+}{N_M} \frac{N_{Zn}}{N_{Zn}} \frac{\bar{\sigma}_{Ne}}{\bar{\sigma}_M} \frac{\bar{v}_{Ne}}{\bar{v}_{Ne}} \frac{A_L}{A_U}, \quad (7)$$

where N^+ and N_M are the Ne ion and Ne metastable densities, A_U and A_L denote the transition probabilities from the upper and lower levels, and $\bar{\sigma}_M$ is the cross-section for Penning ionization. The ratio of the transition probabilities is 1.5 [7]. In the case of He, the charge transfer and Penning ionization collision cross-sections are not very different, the same may be supposed for Ne. Metastable densities saturate with increasing discharge current, in contrast to the continuous increase of ion densities [11]; so in this way $N^+ \gg N_M$ can be reached. This is supported by the fact that in an analogous Ne-Cu discharge no significant excitation of the Cu ionic spectrum due to Ne metastables could be observed [12]. Thus a

population inversion at 210 nm seems possible. The gain at the 210 nm transition is expected to be lower than that at other Zn II laser transitions, however, due to the significantly shorter wavelength resulting in a larger line-width. In view of this, laser mirrors present a serious problem from the technical point of view for obtaining 210 nm laser oscillation. It is evident that for this aim a hollow cathode laser tube capable of higher gains than can be reached in our tube is necessary. Recent work on high voltage variants of the hollow cathode have shown that they are superior to the conventional hollow cathode for charge transfer pumped lasers [13]. The use of high voltage hollow cathodes in a 15 cm active length He-Zn laser has resulted in an increased gain ($\sim 100\%/m$) at the 492.4 nm Zn II line [14]. It would therefore seem that, if this kind of discharge tube were to be applied, it might well facilitate the task of achieving laser operation at the 210 nm line.

5. Conclusions

The strong increase of the spontaneous 210 nm Zn II transition and the ceasing of the 492.4 nm laser oscillation observed on adding Ne to the He-Zn discharge offers further evidence of the excitation of the $4d^2D_{5/2}$ level by charge transfer collisions with Ne ions. A rate equation model, which takes into account cascade excitation of the 492.4 nm line and charge transfer excitation of the $4d^2D_{5/2}$ level was developed. On the basis of this model, the cross-section for the Ne ion-Zn atom charge transfer collisions was found to be $3.5 \times 10^{-15} \text{ cm}^2$, which is in acceptable agreement with the value $2.3 \times 10^{-15} \text{ cm}^2$ measured in [5]. Considerations taking into account possible excitation of the $4p^2P_{3/2}^0$ lower level of the 210 nm transition suggest the possibility for population inversion and laser oscillation at this line.

Acknowledgement

This work was supported by the Hungarian Science Foundation OTKA, grant T-25941.

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