Excitation mechanism of hydrogen Balmer lines in a fast plasma-mixing device

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The emission of hydrogen Balmer series and the laser oscillation of H_β at 486.12 nm and $H\gamma$ at 434.06 nm in hydrogen-neon discharge operated in a pulsed mode have been reported by Dezenber and Willett [IEEE J. Quantum Electron. **QE-7**, 491 (1971)]. In their work, a "two step" excitation of molecular hydrogen to molecular ionic states, followed by dissociation of hydrogen to excited atomic species, was proposed as the main excitation mechanism. The present work was conducted to study the excitation mechanism of the hydrogen H_α line at 656.28 nm in a flowing, afterglow plasma, by using a crossed-beam plasma-mixing device. It was found that the excitation of the H_α line in this device is due to a direct dissociative energy transfer from neon metastables to molecular hydrogen.

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INTRODUCTION

A near-resonant energy transfer collision, combined with dissociate excitation, can provide an efficient pumping mechanism for the "glow discharge" pumped gas laser. In most cases, energy transfer from atomic metastables or ions to molecules cannot be utilized efficiently in a gas discharge consisting of a mixture of rare gases and molecular gases. This procedure is inefficient primarily due to the fact that a significant fraction of energy is lost through numerous electron collisions which excite the molecular gas into a series of distributed excited energy levels. Consequently, near-resonant energy transfer collisions occur between the metastable atoms and the excited molecular species, generating molecules in some other states which may not be the intended upper level of the laser. A crossed-beam plasma-mixing device has been designed and operated to overcome the previously mentioned disadvantages of the glow discharge pumped lasers using near-resonant dissociative energy transfer processes. Successful operation of the atomic fluorine laser [2] and atomic oxygen laser [3] using the fast plasma-mixing concept was demonstrated by Schaefer and Kirkici. The basic principle of crossed-beam plasma mixing and the operating characteristics of that device have been described in detail in the literature [2,3]. One other advantage of using a flowing plasma-mixing principle compared to a glow discharge technique for a continuous wave (cw) laser oscillation is described as follows. The molecules which undergo collision processes are depleted after the dissociative excitation, thus one must replenish the starting molecules to ensure continuous excitation and emission. This continuous excitation is possible with the plasma-mixing device since the molecular gas is injected into the plasma continuously, and the by-products are pumped out after excitation has occurred.

FAST PLASMA-MIXING DEVICE AND EXPERIMENTS

The basic concept of the crossed-beam plasma-mixing device is as follows. Two different gas beams, one, a neutral molecular gas and the other, an afterglow plasma of a hollow cathode discharge, are formed by two narrow slit type nozzles. The nozzle exit planes are located perpendicular to each other so that the mixing occurs at the junction of these two slit exits. The excitation takes place within the mixing volume, and the volume is continuously pumped out to sustain continuous excitation of the molecular gas [3].

Hollow cathode discharges are known to possess high ion and electron concentrations [4], as well as high metastable densities [5]. Also, the electron energy distribution function has a definite non-Maxwellian form [6] which contains a very high density of low-energy electrons (≈ 2 eV) and a significant population of high-energy electrons (≈ 18 eV). These three distinctive properties of a hollow cathode discharge make it an attractive plasma source for laser systems where near-resonant energy and charge transfer collisions are used to generate population inversion [7]. The afterglow plasma of the hollow cathode discharge of the crossed-beam plasma-mixing device is characterized as a "cold" plasma [8]. The term "cold" indicates that the high-energy electrons responsible for the excitation of a gas only exist in the active region of the hollow cathode (upstream part of the slit nozzle), but not in the flowing afterglow. This characteristic is due to the fact that there is sufficient time available for the relaxation of the electron energy distribution while the generated plasma propagates through the nozzle. However, the high-energy electrons can be generated due to metastable collisions in the afterglow (in the mixing region), if no molecular gas is injected into the afterglow plasma. On the other hand, this high-energy electron generation is interrupted as soon as the molecular beam is injected into the afterglow plasma. Therefore, only the near-resonant energy or charge transfer excitation processes between the neutral molecular gas and metastables or ions in the afterglow plasma occur [9].

In the present work, all experiments were performed with helium or a helium-neon gas mixture as the feed gas for the plasma source (nozzle 1–hollow cathode) and pure molecular hydrogen as the molecular gas beam (nozzle 2). The hollow cathode was operated with the current ranging from 0.1 to 1.6 A and a voltage of approximately 220 V. For laser operation, a near concentric resonator with a beam waist of approximately 100 μm was aligned along the transverse direction to the active volume [3]. The fluorescence intensity and the laser power were measured as a function of different operating parameters, such as outlet pressure, p_{out}, hollow cathode current, I, and the ratio of neon to helium plasma. For the fluorescence measurements, the end mirrors were taken out of the cavity. The device was optimized depending on the system parameters, specifically the input pressure of the plasma source, p_1, the input pressure of the molecular gas beam, p_2, the outlet pressure in the mixing volume, p_{out}, the current, I, and the voltage, V, of the hollow cathode discharge. These parameters were kept at the optimum operating conditions for maximum fluorescence intensity and laser power.

RESULTS

Dezenber and Willett observed emission of H_β at 486.12 nm and H_γ at 434.06 nm in a pulsed neon discharge. In this work, hydrogen was present as an impurity in the discharge tube. A "two step" excitation of molecular hydrogen, given by the following relations, was proposed as the main excitation mechanism of the Balmer series and the laser oscillation [1]:

$$Ne^{*m}+H_2 \rightarrow Ne+H_2{}^{+}({}^2\Sigma_g^+;v)+e \ , \qquad (1)$$

$$H_2{}^{+}({}^2\Sigma_g^+;v)+e \rightarrow H^*(n=3,4,5)+H+E_{KE} \ . \qquad (2)$$

Experiments with the fast plasma-mixing device using a helium-neon mixture as the plasma and hydrogen as the molecular gas showed strong fluorescence of the hydrogen Balmer α line at 656.28 nm. The other lines of the series were not observed. We did not detect emission from any other atomic or molecular hydrogen systems. Also, when the plasma source was operated with pure helium (no neon admixture in helium plasma), and molecular hydrogen was injected into the afterglow plasma, emissions of H_α ceased and no other lines of the hydrogen Balmer series were observed. The emission spectrum showed only the helium atomic or molecular emission lines. In another series of experiments, the plasma source was operated with 1% hydrogen in pure helium. Also, no molecular side flow was present. The emission spectrum was studied and the observed lines were H_α at 656.28 nm, H_β at 486.12 nm, and H_γ at 434.06 nm with decreasing intensity, respectively. This result was expected, because in the active region of the hollow cathode discharge, helium ions, metastables, and high-energy electrons are present. A possible scenario for the excitation of these lines could be a "two step" excitation of hydrogen, similar to the one given by Dezenber and Willett [1]. However, we did not study the excitation mechanism of this operating mode, because we were primarily interested in the excitation mechanism in the mixing volume not in the active plasma generation region of the slit hollow cathode. Also, in comparison, the intensity of H_α, the most intense line of the series in the He-H_2 plasma, was much smaller than the intensity observed when molecular hydrogen was injected into the He-Ne afterglow plasma, showing a low efficiency of the two step hydrogen excitation in the active plasma channel.

Feld *et al.* [10] proposed that direct dissociation of hydrogen by near-resonant energy transfer from a neon metastable could be utilized to populate the upper level of the Balmer α line. In this work, "hot" hydrogen atoms were produced in the $n=3$ state by the reaction

$$Ne^*+H_2 \rightarrow Ne+H+H^*(n=3)+0.05 \text{ eV} \ . \qquad (3)$$

The cross section of this process is $\sigma=1.4\times10^{-15}$ cm^2 [10]. However, the gain of their system was not sufficient to obtain laser oscillation. We believe that the strong emission intensity of the H_α line in our plasma device is also due to the reaction given above.

The operating conditions in the crossed-beam plasma-mixing device are significantly different than in any type of gas discharge [8]. Plasma is generated in a separate channel before it is mixed with the molecular hydrogen. The metastables and ions are the main energy carriers at the exit of the slit hollow cathode. Also, the majority of the electrons reach the mixing volume as low-energy electrons, and they do not contribute significantly to the excitation of molecular gas injected into the afterglow plasma. Consequently, the excitation of molecular gas is due to the near-resonant energy transfer collisions in the mixing volume. In this case, since the plasma source is operated with a mixture of He-Ne gas, the majority of the expected energy carriers are the neon metastable and/or ions. Upon comparison of the energy of neon metastables (16.6 and 16.7 eV) and the dissociation energy (4.47 eV) of a hydrogen molecule, one can conclude that only the states $n\leq3$ can be excited by the direct dissociative energy transfer process. As a result, it is concluded that the excitation mechanism of the H_α line in the plasma-mixing device is due to the direct dissociative energy transfer from neon metastable given by reaction (3).

Figure 1 shows the laser intensity of the H_α line versus the neon percentage in a helium-neon plasma at optimum

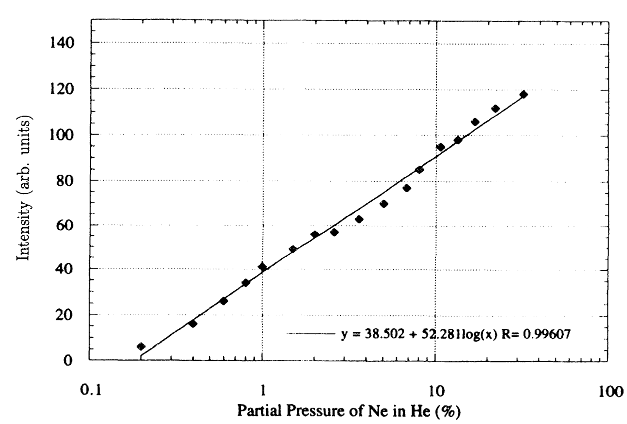

FIG. 1. Intensity of H_α line at 656.28 nm as a function of neon admixture in helium plasma at optimum operating conditions ($p_1=520$ mbar and $p_2=310$ mbar, $p_{out}=40$ mbar, the current $I=1$ A, and the voltage $V=220$ V).

operating conditions: input pressures $p_1 = 520$ mbar and $p_2 = 310$ mbar, the outlet pressure $p_{out} = 40$ mbar, the current $I = 1$ A, and the voltage $V = 220$ V. It is seen that the laser power increases logarithmically with increasing percentage of neon in helium plasma. The neon percentage threshold in the plasma was approximately 0.2%. When the neon ratio in the helium plasma was increased beyond 20%, the discharge became unstable, and some "hot spots" appeared at the slit exit. At this point, the data acquisition was terminated. The optimum neon ratio for an efficient operation of the hollow cathode was determined to be 8%.

Contrary to the strong fluorescence at 656.28 nm, the laser oscillation at the H_α line was very weak. The maximum laser power observed was on the order of 0.1 mW. The hydrogen $n = 3$ excited state is also the upper level of the Lyman β line at 102.57 nm. The Einstein coefficients of these two levels, $3p\ ^2P^o$ (L_β line) and $3d\ ^2D$ (H_α line), are of the same order of magnitude (5.5×10^7 sec^{-1} and 4.41×10^7 sec^{-1}, respectively [11]), and the energy difference between these levels is approximately 10^{-4} eV. If the $3d\ ^2D$ state is populated through dissociative excitation, the $3p\ ^2P^o$ state should also be populated. Considering the dissociative resonant energy transfer reaction given by Eq. (3), one would also expect excitation of the $n = 2$ state, as well as $n = 3$, and hence a consequent emission of L_β at 102.57 nm and L_α at 121.57 nm and H_α at 656.28 nm. The Einstein coefficient of $2p\ ^2P^o$ (L_α line) is 6.3×10^8 sec^{-1} [11], which means that the emission of the L_α line should also be readily present. However, we were not able to observe any emission of the Lyman series, because they lie outside the range of the equipment used in the experiments for spectroscopic investigation. If the velocity distribution of the upper level ($n = 3$ states) is broader than the lower level ($n = 2$ state), as was proposed by Feld *et al.* [10], the population inversion and the laser efficiency then depend on the velocity distribution of both lines. This might be a reason for the low laser power level of the system.

The pump rates required to reach the threshold for laser action depend on the wavelength and the gain profile of the system. The total pump rate required to reach laser threshold at 650 nm was calculated to be on the order of 7.5×10^{17} cm^{-3}s^{-1}; we assumed that the line width $\Delta\lambda$ for the hydrogen system is 0.05 nm, and a branching ratio into the upper H_α line is 30%. For the calculations, it was also assumed that no cavity losses were present. Also, a minimum gain of 0.1% cm^{-1} was considered. The minimum pump rate in the mixing volume, in terms of the density of the excited species or ions in the plasma beam, was then calculated to be on the order of 7.5×10^{11} cm^{-3}. This was based upon the flow velocity of the energy carriers, metastable and ions, of $v = 500$ m/s (a helium plasma beam which flows through a subsonic nozzle), and a mixing volume cross section, over which the energy exchange collision occurs, of $d \approx 0.5$ mm (which is on the order of a few mean free paths for a gas at a pressure of 10 mbar) [8]. The electron density in the plasma was measured by measuring the Stark broadened Balmer β line. It was determined to be on the order of 10^{14} cm^{-3}. If one assumes a 1% efficiency of neon metastable production in the plasma, then the expected metastable density is approximately 10^{12} cm^{-3}. This is the limiting case of the calculated value given above. This may explain why the laser intensity was very low in contrast to a very strong fluorescence intensity.

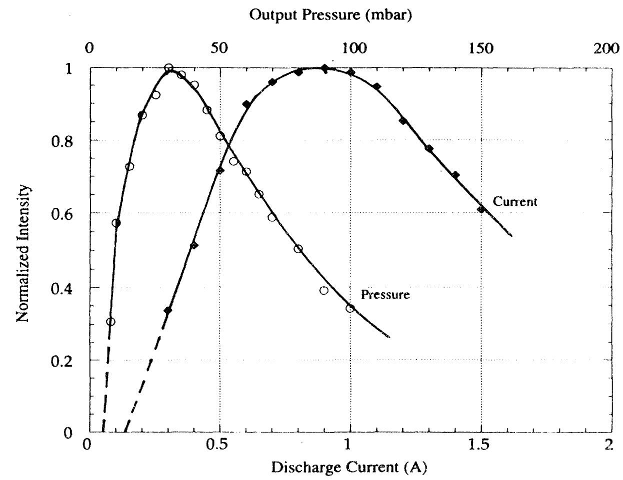

FIG. 2. Normalized laser intensity of H_α line at 656.28 nm as a function of discharge current and as a function of output pressure at optimum operating conditions.

Figure 2 shows the normalized laser intensity as a function of discharge current and as a function of output pressure. The linear increase of power level as current increases indicates that the excitation mechanism of the hydrogen is due to near-resonant energy transfer from neon metastables. The linear increase of the metastable production is a characteristic of the discharge chamber [3]. Also the metastable production reaches a steady state value at approximately 1 A, then starts to decrease as the discharge current increases. At this high current value, the ion production becomes dominant. These ionic species, therefore, are not contributing to be the excitation of the H_α line. The current threshold for the laser oscillation is approximately 0.15 A, and the pressure threshold is 8 mbar. The optimum current value is between 0.8 and 1.0 A, and the optimum pressure is 40 mbar. At pressures higher than that, the density of the collision partners is too high for an efficient energy transfer collision process.

CONCLUSIONS

A crossed-beam plasma-mixing device was utilized to study the excitation mechanism of the hydrogen H_α line at 656.28 nm in a flowing afterglow plasma of a helium-neon gas mixture. Contrary to the work by Dezenber and Willett, the hydrogen H_α line was the only visible line observed in the present work. Near-resonant dissociative energy transfer from neon metastables to neutral hydrogen molecules was proven to be the excitation mechanism of the emission. It was also determined that this excitation mechanism was a very efficient process in the crossed-beam plasma-mixing device. Contrary to the very strong fluorescence emission observed in the present work, a relatively low level of laser power was obtained. The reasons for this phenomenon are concluded to be the low density of neon metastables in the plasma, the high cavity losses, and the velocity distribution of the excited hydrogen atom after the dissociation process. With a high gain laser cavity and increased neon metastable density in the mixing volume, the laser intensity should increase. Further studies on the emission spectra are required to determine the presence of L_β at 102.57 nm and L_α at 121.57 nm. Should these lines be measured, their presence will further support the conclusion of high selectivity of direct dissociative energy transfer from neon metastables to neural molecular hydrogen in a crossed-beam plasma-mixing device.

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