Laser Original

An Account of Excitation Mechanism of Discharge-Pumped XeCl Laser Using Ne/Ar Diluent

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Comparison of the XeCl laser output performances using various rare gas mixtures as the diluent gas is described. The electron densities in the discharge-pumped XeCl laser using Ne, He, and Ne/Ar diluents are determined by measuring the Stark half-width of the $H\beta$ line. The excitation mechanism of the discharge-pumped XeCl laser using Ne/Ar diluent is discussed from the viewpoint of the electron energy distribution functions. It is shown that for the Ne/Ar diluent the higher electron density and the higher mean electron energy can lead to the efficient lasing at relatively low pressures.

Key Words: XeCl laser, Electron density.

1. Introduction

For the discharge-pumped excimer laser, the diluent gas has an important role to control the electron energy distribution function in the discharge plasma and to form excimers efficiently. Neon has been used as the diluent in most commercial discharge-pumped XeCl lasers, since Sze has shown that lasing is more efficient in the Ne diluent than in the He diluent^{1,2)}. In order to obtain an efficient lasing by using the Ne diluent, however, it is necessary to operate the XeCl laser at high pressures of 4-6 atm. On the other hand, when argon was used as the diluent, the maximum output energy was obtained at the total gas pressure of less than 2 atm, although there was the problem of arc formation in the discharge. Hiramatsu *et al.* have obtained the improved performance of the discharge-pumped XeCl laser below 2 atm by using the Ar/He and Ne/Ar diluents^{3,4)}. The maximum total efficiency of 0.9% has been obtained at the total gas pressure

of 1.8 atm, the fractional concentrations of 65.8% Ne, 30.0% Ar, 4.0% Xe, and 0.2% HCl, and the charging voltage of 18 kV⁵⁾. The Ne/Ar diluent XeCl laser can be operated most efficiently at lower pressures. However, little information is available about its excitation mechanism.

In this paper, for the purpose of interpreting the characteristics of the laser output and understanding the fundamental processes of the Ne/Ar diluent XeCl laser, the effect of the diluent gas on the laser output performance has been systematically reexamined using various rare gas mixtures as the diluent gas (He, Ne, Ar, Kr, and their combinations), and the electron densities in the Ne, He, and Ne/Ar diluent XeCl lasers have been measured by using the Stark-broadening technique⁶⁾. The excitation mechanism of the Ne/Ar diluent XeCl laser has been discussed from the viewpoint of the electron energy distribution function.

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2. Experimental

The laser device used in the experiment was a compact transverse discharge-pumped XeCl laser employing the automatic preionization scheme described in detail elsewhere⁵⁾. It had dimensions of $18.8 \times 16.5 \times 50.0 \text{ cm}^3$. The external circuit powering the device was of the capacitor-transfer type, switched by a nitrogen spark gap. Discharge chamber, primary energy storage capacitor bank C_1 , and spark gap were constructed in a body to obtain a minimum inductance of the charge transfer circuit from C_1 to C_2 and the discharge circuit. The capacitance of the primary energy storage capacitor bank C_1 was 20.4 nF. The capacitance of the secondary energy storage capacitor bank C_2 in the laser chamber was 18.8 nF. The stainless electrodes, 1.5 cm wide and 40 cm long, were separated by 1.25 cm. The laser output beam had dimensions of $1.25 \times 0.5 \text{ cm}^2$ and the active discharge volume was about 22.5 cm^3 . Three stainless studs were connected with the terminal of each capacitor of C_2 , giving the total preionization gaps of 60. The distance between the preionization gaps and the discharge electrodes was 1.2 cm and the interval between two preionization gaps was 1.33 cm.

The electron densities in the XeCl lasers have been determined by adding 0.2% H_2 to the host gases and measuring the Stark half-width of the hydrogen Balmer beta line ($\text{H}\beta$ line)*. The experimental setup for that measurement was the same as that described in our previous paper⁶⁾. The $\text{H}\beta$ line profiles were obtained from

the end-light intensity measurements. All measurements were carried out at the central portion of the discharge imaged onto the monochromator slit.

Under the present experimental conditions, the Stark half-width of the $\text{H}\beta$ line was greater than 0.3 nm. On the other hand, the Doppler width and the collisional width were less than 0.01 nm. The instrumental width in this case was 0.02 nm. Therefore, the error of the obtained electron density was estimated to be about 15%.

3. Results and Discussion

3.1 Comparison of the Laser Output Performance of the Discharge-Pumped XeCl Laser Using the Various Rare Gas Mixtures as the Diluent Gas

In our previous paper, it has shown that a higher laser output energy in the Ne/Ar diluent than in the Ne and He diluents could be obtained at the total gas pressures below 2 atm⁴⁾. In this section, using several rare gas mixtures (Ar/He, Ne/He, Ne/Ar, Kr/Ar, Kr/Ne, and Kr/He), diluent gas effect on the XeCl laser output performance has been systematically investigated.

Figure 1 shows the XeCl laser output energy as a function of the total gas pressure for different rare gas mixture diluents and pure rare gas diluents. In each case, the fractional concentration of the heavier rare gas in the mixture diluent was kept at 50%. The fractional concentrations of Xe and HCl for all mixtures were kept at 1.3% and 0.2%, respectively, which were

* If electrons and singly charged ions are considered in the Stark half-width calculation, the electron density n_e is given by

$$n_e = C(n_e, T_e) (\Delta \lambda_s)^{3/2},$$

where $\Delta \lambda_s$ is the Stark half-width of the $\text{H}\beta$ line, and the coefficient $C(n_e, T_e)$ is only a slowly varying function of the electron density and temperature⁷⁾. Under the present experimental condition, the value of $C(n_e, T_e)$ for the $\text{H}\beta$ line was almost constant at 1.1×10^{16} . Therefore, if the Stark broadening was dominant for the $\text{H}\beta$ line profile, the electron density was determined directly from the measurement of the half-width of the $\text{H}\beta$ line.

Fig. 1. XeCl 308-nm laser output energy as a function of the total gas pressure for various combinations of rare gas mixtures as the diluent gas. The fractional concentration of the heavier rare gas in the mixture diluent is 50% in each case. The fractional concentrations of Xe and HCl for all mixtures are 1.3% and 0.2%, respectively. The charging voltage of the primary energy storage capacitor bank C_1 is 20 kV.

close to the optimum values. The charging voltage of the primary energy storage capacitor bank C_1 was 20 kV. In the range of the total gas pressure region from 1.0 to 2.0 atm, the Ne/Ar diluent showed the best performance. At a total gas pressure of 1.5 atm, the laser output energy for the Ne/Ar diluent was three times as large as that for the Ne diluent. Kr and Ar diluents had the tendency of arc formation in the discharge at high pressures. In particular, the laser output energy for the Kr diluent was low and the laser pulse reproducibility was very poor. The addition of helium or neon to the Kr and Ar diluents helped to reduce the arc formation and resulted in a more uniform discharge.

Figure 2 shows the XeCl laser output energy as a function of the charging voltage of the primary energy storage capacitor bank C_1 for the various diluents. Results in Fig. 2 were obtained at the total gas pressure of 1.5 atm. The fractional concentrations were the same as the values in Fig. 1. The laser output energy of the

Fig. 2. XeCl 308-nm laser output energy for the various diluents as a function of the charging voltage of the primary energy storage capacitor bank C_1 . The total gas pressure is 1.5 atm. The fractional concentrations are the same as in Fig. 1.

Ne/Ar diluent was the largest of all, and increased well with the increase in the charging voltage, which shows that the impedance matching is good for the Ne/Ar diluent.

Figure 3 shows the self-breakdown voltage across the main discharge electrodes using various diluents as a function of the charging voltage of the primary energy storage capacitor bank C_1 . Results in Fig. 3 were obtained at the total gas pressure of 1.5 atm. The fractional concentrations were the same as the values in Fig. 1: The self-breakdown voltage of the Ne/Ar diluent was high, and well increased with the charging voltage. On the other hand, the self-breakdown voltages of the Ne and Ne/He diluents were low and were saturated around 16 kV presumably owing to the impedance mismatching, which shows that high operating pressure is required for the efficient lasing for the Ne and Ne/He diluents.

For the He-mixed diluents (Kr/He diluent was not included), the laser pulse duration was about 10 ns (FWHM). For the Ne/Ar and Ne diluents, the laser pulse duration was about 12 ns and a secondary hump appeared on the laser pulse shape. For the Kr-mixed and Ar diluents,

Fig. 3. Self-breakdown voltage for various diluents as a function of the charging voltage of the primary energy storage capacitor bank C_1 at the total gas pressure of 1.5 atm. The fractional concentrations are the same as in Fig. 1.

on the other hand, the laser pulse duration was fairly short (about 8 ns) and, in addition, the shot-to-shot reproducibility of the laser pulse shape was not good.

From the above results in Figs. 1 to 3, the XeCl laser output performance of the Ne/Ar diluent was shown to be the best at relatively low pressures below 2 atm. As the results of gas optimization, the largest output energy of 40 mJ for the Ne/Ar diluent was obtained at a total gas pressure of 1.8 atm and a charging voltage of 22 kV. The maximum total efficiency of 1% for the Ne/Ar diluent was obtained at a total gas pressure of 1.8 atm and a charging voltage of 18 kV. Detailed investigation about the gas optimization of the Ne/Ar diluent XeCl laser was described in reference⁴). In the following section, determination of electron density and excitation mechanism of the discharge-pumped Ne/Ar diluent XeCl laser are described, in comparison with those of the He and Ne diluents which have been fully investigated by a number of researchers.

3.2 Electron Densities in the XeCl Laser Discharges

Figure 4 shows the measured electron densi-

ties as a function of the total gas pressure for the Ne, He, and Ne/Ar diluents. In each diluent, Xe and HCl fractional concentrations were 0.2%, respectively. For the Ne/Ar diluent, Ar fractional concentration was 20%. The charging voltage of the primary energy storage capacitor bank C_1 was 20 kV. Results in Fig. 4 were obtained at 14 ns after the start of the main discharge, when the $H\beta$ emission reached its peak value. The measured electron densities in the XeCl laser discharges are of an order of 10^{15} cm^{-3} , which are relatively close to those predicted by the simulation analyses⁸⁻¹⁰). The electron density for the Ne/Ar diluent is the highest of those for the three kinds of diluents. The electron densities for the Ne and Ne/Ar diluents increase more rapidly with the increase in the total gas pressure than that for the He diluent does.

Figure 5 shows the time dependence of the electron densities in the Ne, He, and Ne/Ar (Ne:Ar=8:2) diluent XeCl lasers. Stark-broadened profiles of the $H\beta$ line were measured at 4, 14,

Fig. 4. Electron densities as a function of the total gas pressure for three kinds of gas mixtures, Ne:Xe:HCl = 99.6:0.2:0.2 (○), He:Xe:HCl = 99.6:0.2:0.2 (▲), and Ne:Ar:Xe:HCl = 79.6:20.0:0.2:0.2 (●). The charging voltage of the primary energy storage capacitor bank C_1 is 20 kV.

Fig. 5. Time dependence of the electron densities for the XeCl discharges for Ne, He, and Ne/Ar diluents. (○) Ne/Xe/HCl, (▲) He/Xe/HCl, (●) Ne/Ar/Xe/HCl. The time origin corresponds to the moment of the self-breakdown.

25, and 40 ns after the start of the main discharge. In each diluent, Xe and HCl fractional concentrations were 0.2%, respectively. The charging voltage of the primary energy storage capacitor bank C_1 was 20 kV and the total gas pressure was 2.0 atm. The time origin in Fig. 5 corresponds to the start of the main discharge. For all diluents investigated, the electron densities reach the maximum values at about 25 ns after the start of the main discharge. That time almost corresponds to the moment when the 308 nm emission reaches its peak value.

Figure 6 shows the electron density for the Ne/Ar diluent XeCl laser discharge as a function of the Ar fractional concentration in the Ne/Ar/Xe/HCl mixture. Electron densities in Fig. 6 were measured at about 14 ns after the start of the main discharge. In Fig. 6, Xe and HCl fractional concentrations were 0.2%, respectively. The charging voltage of the primary energy stor-

Fig. 6. Electron density for the Ne/Ar diluent XeCl laser discharge as a function of the Ar fractional concentration in the Ne/Ar/Xe/HCl mixture. The fractional concentrations of Xe and HCl are 0.2%, respectively. The charging voltage of the primary energy storage capacitor bank C_1 is 20 kV and the total gas pressure is 1.7 atm.

age capacitor bank C_1 was 20 kV and the total gas pressure was 1.7 atm. The electron density for the Ne/Ar diluent increase slowly with the increase in the Ar fractional concentration. At the Ar fractional concentration of 30%, the electron density for the Ne/Ar diluent is $2.5 \times 10^{15} \text{ cm}^{-3}$, which is larger by a factor of 1.3 than that for the Ne diluent.

3.3 Excitation Mechanism of the Discharge-Pumped XeCl Laser Using Ne/Ar Diluent

In the discharge-pumped excimer lasers, diluent gases are used mainly both in order to control the electron energy distribution function in the discharge plasma and to prevent the constriction of the glow discharge. Figure 7 shows the electron energy distributions for the He, Ne,

and Ne/Ar pulsed discharges. The values of E/N are 6×10^{-17} Vcm² for He, 4×10^{-17} Vcm² for Ne, and 1.2×10^{-16} Vcm² for Ne/Ar, which are close to the optimum values for the He, Ne, and Ne/Ar diluent XeCl lasers, respectively. Results in Fig. 7 were obtained by multiplying the normalized electron energy distribution functions by the measured electron densities. Those normalized electron energy distribution functions were obtained by solving the Boltzmann transport equation numerically in the same way as described in reference¹¹). Although the effects of xenon and hydrogen chloride are not included, it is believed that the addition of a small amount of these gases scarcely affects the shapes of the three curves. As can be seen from Fig. 7, for the He diluent, the electron densities of low energy are very high and the mean electron energy is low. On the other hand, since the electron densities spread toward the high-energy region for the Ne and Ne/Ar diluents, the mean electron energy is high. In addition, the density of electrons below 12 eV for the Ne/Ar diluent is larger

Fig. 7. Electron energy distributions for the He, Ne, Ne/Ar pulsed discharges. The values of E/N for He, Ne, and Ne/Ar diluents are 6×10^{-17} , 4×10^{-17} , and 1.2×10^{-16} Vcm², respectively.

than that for the Ne diluent.

The sequence diagram indicated by the solid lines in Fig. 8 illustrates the XeCl* excimer formation process which has been believed to be dominant in the discharge-pumped XeCl lasers. XeCl* excimers are formed mainly in the three-body ion-ion recombination process:

where M represents the diluent gas atom (He, Ne, Ar)^{9,12}). For the efficient formation of XeCl* excimers, therefore, the large densities of Xe⁺ and Cl⁻ ions at the beginning stage in the main discharge, and the large rate constant for the reaction (1) are required.

The rate constants for reaction (1) reported recently were 1.3×10^{-7} cm³s⁻¹ for the He diluent, 1.4×10^{-6} cm³s⁻¹ for the Ne diluent, and 1.2×10^{-6} cm³s⁻¹ for the Ar/He diluent¹³). For the Ne and Ar diluent KrF lasers, in addition, the three-body ion-ion recombination rate constants for KrF* excimer formation were nearly equal at the total gas pressure around 2 atm[#]. Accordingly, at that pressure the rate constant for reaction (1) for the Ne/Ar diluent is expected to be as large as that for the Ne diluent. As the results, the fast and efficient formation of XeCl* excimers is expectable for the Ne/Ar diluent. Moreover, the large rate constant for reaction (1) leads to the fast reduction of the density of

Fig. 8. Sequence diagram showing the dominant processes for the XeCl* excimer formation in the discharge-pumped XeCl laser. M represents the diluent atom.

Cl^- ions, which are believed to be the dominant absorbers at 308 nm in the XeCl laser¹²⁾.

The direct and stepwise ionization of xenon:

are the dominant processes for the formation of Xe^+ ions. As can be seen from Fig. 7, the densities of electrons above 12 eV, which are required for the direct ionization of xenon by the electron impact, are much larger for the Ne and Ne/Ar diluents than that for the He diluent. In addition, the density of electrons above 4 eV, which are required for the stepwise ionization, is larger for the Ne/Ar diluent than for the Ne and He diluents. Therefore Xe^+ ions can be formed most efficiently for the Ne/Ar diluent.

When Ar/He and Ne/Ar diluents are used, the density of Ar^* excited atoms ($\sim 10^{14} \text{ cm}^{-3}$) is two orders of magnitude larger than that of He^* or Ne^* excited atoms in the He or Ne diluent¹³⁾. Accordingly, for the Ne/Ar diluent, the processes of the energy transfer from Ar^* excited atoms, Ar^+ ions, and Ar^+

$_2$ molecular ions into xenon presumably play the important roles as the additional channels of the formation of Xe^+ ions and free electrons¹³⁾.

The dominant process for the formation of Cl^- ions is the vibrational excitation of HCl followed by the dissociative attachment⁸⁾:

Reactions (4) proceed easily in the low-energy electron region below 5 eV. From Fig. 7, therefore, the Ne/Ar diluent is more advantageous

than the Ne diluent. For the efficient formation of Cl^- ions, the He diluent is the most advantageous, and the density of Cl^- ions for the He diluent was larger by a factor of 2.2 than that for the Ne diluent¹³⁾. However, that too large density of Cl^- ions results in the low efficiency for the He diluent XeCl laser, because Cl^- ions are the dominant absorbers at 308 nm.

For the Ne and Ne/Ar diluents, XeCl^* excimers are formed not only by reaction (1), but also by the ion-ion recombination reaction¹⁰⁾ indicated by the dotted lines in Fig. 8:

In addition, since the following reactions:

are the significant processes in the formation of XeCl^* excimers in the e-beam-pumped XeCl laser using Ar diluent¹⁵⁾, for the discharge-pumped Ne/Ar diluent XeCl laser as well, XeCl^* excimers can possibly be formed also by the above reactions indicated by the broken lines in Fig. 8.

From ref. 14, the rate constants for the three-body ion-ion recombination for the Ne and Ar diluent KrF lasers are saturated around 4 and 1.5 atm, respectively. Considering the similarity of XeCl and KrF lasers, the rate constant for reaction (1) for the Ne/Ar diluent XeCl laser is saturated probably below 2 atm. The saturation of the XeCl laser output energy around 1.5 atm for the Ne/Ar diluent in Fig. 1 is partly due to that saturation of the rate constant for reaction (1). In addition, the Ne/Ar diluent XeCl laser has the tendency of the arc formation in the discharge, which may additionally lead to the

Calculated three-body ion-ion recombination rate constants at 300 K and a total gas pressure of 2 atm for

$\text{Kr}^+ + \text{F}^- + \text{M} \rightarrow \text{KrF}^* + \text{M}$ (M: He, Ne, Ar)

were 3.0×10^{-7} , 2.8×10^{-6} , and $3.0 \times 10^{-6} \text{ cm}^3 \text{ sec}^{-1}$ for He, Ne, and Ar diluents, respectively¹⁴⁾.

saturation of the XeCl laser output energy at high pressures.

XeCl* excimers are quenched dominantly by electron collisions^{9,16)}, and therefore the XeCl* excimer quenching rate for the Ne/Ar diluent with the higher electron density is somewhat larger than that for the He diluent. However for the Ne/Ar diluent the XeCl* excimer formation rate is rather larger than for the He diluent because the density of electrons above 4 eV for reactions (2) and (3) is larger by a factor of 2.3 than for the He diluent, and there are the additional excimer formation channels of reactions (5) and (6). Consequently the Ne/Ar diluent XeCl excimer laser has the larger laser power than the He diluent XeCl laser.

4. Summary

Comparison of the output performances of the gas mixture diluent XeCl lasers has been carried out. The Ne/Ar diluent laser showed the best performance at relatively low pressures below 2 atm. The electron density of the Ne/Ar diluent laser was measured by using the Stark-broadening technique. It has been shown that the higher electron density and the higher mean electron energy for the Ne/Ar diluent can lead to the efficient lasing at relatively low pressures.

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