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Stimulated Emission Originated from Collision of Long Life Zn*

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Under the resonant excitation of Zn atomic $4p\,^3P_1$ state a strong stimulated emission at 481.0 nm is produced, which corresponds to the transition between $5s\,^3S_1$ and $4p\,^3P_2$ of Zn. If N_2 molecular gas is used as buffer gas instead of rare gas He or Ar, the stimulated emission may be enhanced by several orders of magnitude. The dependences of intensity on density of atomic Zn and N_2 pressure are studied experimentally. The mechanism which causes population inversion is proposed.

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The possibility of populating high-lying atomic states by collisional pooling of lower excited states has been studied for the past years.[1,2] In our previous paper[3] the pooling process of $4p\,^3P_1$ Zn atoms was demonstrated, in which rare gases played a significant role. Here we report a new result that the stimulated emission(SE) at wavelength of 481.0 nm is generated when He or Ar is replaced by N_2 molecule. It is a resonant SE from $5s\,^3S_1$ state to $4p\,^3P_2$ of atomic Zn.

The experimental set up is similar to that in Ref. 3. Briefly, the output of a pulse YAG pumped R110 dye laser is frequency doubled by BBO crystal and is tuned in the range of 305.0–310.0 nm. An unfocused UV beam with energy between 0.1 and 2 mJ and pulse duration of about 15 ns passes through a Zn heat-pipe oven made by stainless steel and filled with He or N_2 as buffer gas. The oven temperature varies from 400°C to 600°C, thus the corresponding density of Zn atoms is changed from 1.07×10^{15} to $1.23 \times 10^{17} \mathrm{cm}^{-3}$. The power density of laser in the operation range estimates to be $0.2 - 4 \times 10^6 \mathrm{Wcm}^{-2}$, that should make the saturated excitation from ground state $4s\,^1S_0$ to $4p\,^3P_1$ state. The fluorescences and SE are collected in backward direction and detected by a photomultiplier following 1 m grating spectrometer. The output signal is fed to a boxcar averager and then processed by a microcomputer.

At lower density of atomic Zn with either He or N_2 as buffer gas, under the excitation of resonant laser at 307.6 nm, the emissions at 481.0, 472.2 and 468.0 nm, which come from the transitions of same upper state $5s\,^3S_1$ to three fine-structure states of $4p\,^3P_{2,1,0}$, are observed. Their intensities are comparable. Other emissions at 636.2, 518.2, 328.2, 330.3 and 334.5 nm could be seen, which originate from higher singlet and triplet states of Zn, populated through collisional energy transfer from occupied $4p\,^3P_1$ state. The relevant energy level scheme is shown in Fig. 1. However, increasing density
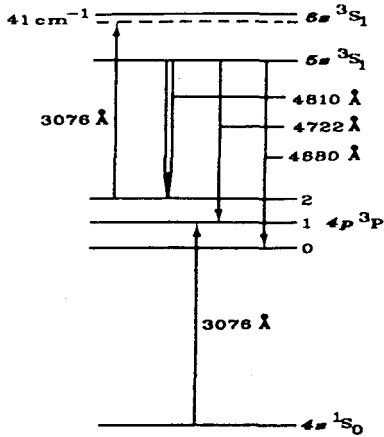
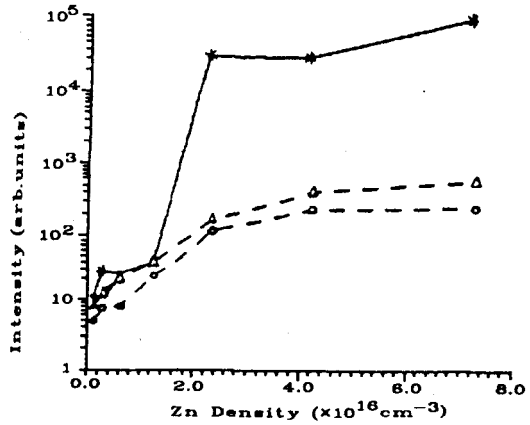Fig. 1: Simplified lower levels of atomic Zn and related transitions. Pump laser is at 307.6 nm.

Fig. 2: Variation of emission intensity with density of atomic Zn.   *: 481.0 nm, Δ: 472.2 nm, o: 468.0 nm.
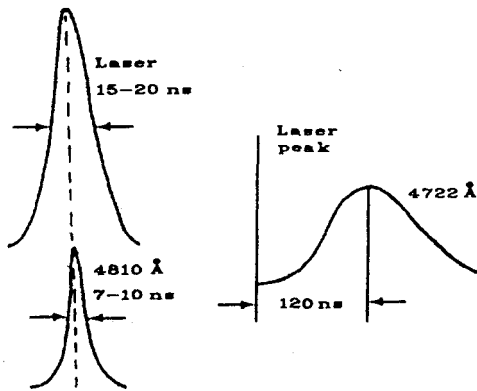

Fig. 3: Pulse shapes of laser, SE at 481.0 nm and long fluorescence of 472.2 nm.
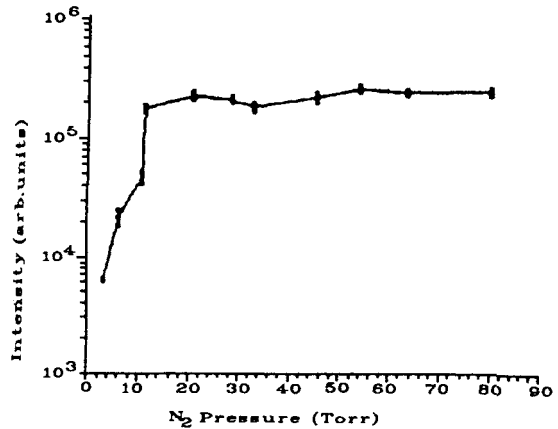

Fig. 4: Intensity of SE as a function of N_2 pressure.

of Zn to $1.4 \times 10^{16} \text{cm}^{-3}$ with laser energy higher than 1 mJ and N_2 as buffer gas, the emission intensity at 481.0 nm goes up abruptly. It is about 10^3 times higher than that at 472.2 and 468.0 nm. In this case the pulse duration becomes obviously different from that at 472.2 and 468.0 nm. The trace on oscilloscope for 481.0 nm exhibits a sharp pulse with FWHM of less than 10 ns and its maximum coincides with the laser peak, while the pulse at 472.2 nm looks very wide and the peak of intensity delays by about 120 ns (See Figs. 2 and 3). Meanwhile the color of fluorescence line in oven changes from weak reddish violet to bright blue and the radiation becomes directional. These indicate that the dominant decay process of $5s\,^3S_1$ becomes SE at 481.0 nm rather than

spontaneous emission.

At a fixed Zn density of $4.3 \times 10^{16} \text{cm}^{-3}$ with laser energy of 1 mJ the dependence of intensity at 481.0 nm on the pressure of N_2 is measured. The results are drawn in Fig. 4. It can be found that in the region of 3–12 Torr of N_2 pressure the intensity enhances rapidly. However, the reproducibility of signals of emission is relatively poor, which may not be explained by the laser instability only. Beyond 12 Torr of N_2 the resulting measurements of 481.0 nm emission become insensitive to the pressure and the reproducibility is somewhat improved.

To fully understand the effects of N_2 molecular gas we compare the dependences of intensities at 481.0 nm on density of Zn atoms for two cases, in which He or N_2 of 60 Torr is filled into oven respectively. In Fig. 5 it is shown that if density is less than $1 \times 10^{16} \text{cm}^{-3}$ the intensities are comparable, while with further increasing density of Zn atoms, N_2 molecules become much more effective for inducing the emission at 481.0 nm.
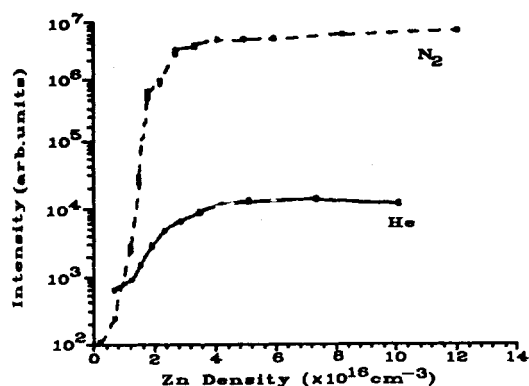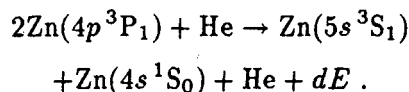
As mentioned previously the high-lying states can be populated by the collision of atoms at lower excited states. In our case the upper state of 481.0 nm emission, $5s\,^3S_1$, is populated by the pooling process of laser pumped long life $4p\,^3P_1$ state, i.e.

$$2\text{Zn}(4p\,^3P_1) + \text{He} \rightarrow \text{Zn}(5s\,^3S_1)$$
$$+\text{Zn}(4s\,^1S_0) + \text{He} + dE \ .$$

But besides $5s\,^3S_1$, the lower state of 481.0 nm transition $4p\,^3P_2$ should be collisionally occupied as well, or even more easily. Fortunately, according to the energy level structure of Zn the transition wavelength between $4p\,^3P_2$ and $6s\,^3S_1$ is 307.2 nm, which is close to 307.6 nm of the pump laser wavelength

Fig. 5: Intensity of SE vs density of atomic Zn for N_2 or He as buffer gas.

with an energy defect of 41cm^{-1} (See Fig. 1). The off-resonant excitation of $6s\,^3S_1$ due to pump laser may simultaneously deplete the lower state of 481.0 nm. So the population inversion is expected to be established during the laser pulse. After laser pulse the population inversion should disappear, consequently the stimulated emission can no longer be generated and only the normal fluorescence remains. This agrees with our observation on the time behavior of SE in Fig. 2. The delay of peak position of 472.2 nm emission might be attributed to the cascade transitions of remainder from $6s\,^3S_1$ through $5p\,^3P$ to $5s\,^3S_1$.

The collisional characters of rare gas He and molecular gas N_2 are quite different. In the case of He, He atoms can only transfer their translation energy to Zn atoms because of very high excited energy of He. While if N_2 is selected as buffer gas, some new

mechanism might operate. For example, molecular vibrational-rotational energy of N_2 could transfer to Zn atoms to compensate the energy defect. Considering the depletion process of $4p\,^3P_2$, the pump laser of 307.6 nm is not sufficient to make deexcitation resonantly. We notice that the energy difference between the vibrational-rotational level $J = 4$ and $J = 0$ of N_2 molecular ground state $X\,^1\Sigma_g^+$ is 40.2 cm^{-1} and that the molecular rotational level $J = 4$ can be surely populated under the working temperature of 600°C. The $V \rightarrow E$ energy transfer would compensate the energy defect, and so make the deexcitation of $4p\,^3P_2$ more effective though the translation energy transfer of both He and N_2 to Zn may compensate the energy defect in principle. This could be the reason why SE at 481.0 nm in N_2 is stronger than that in He gas. In a sense, this suggestion is similar to that about the mechanism for quenching by molecules, where near resonant electronic to vibrational-rotational, $E \rightarrow V$, energy transfer was considered as the reason for the relaxation of certain excited atomic state.[4] The difference between their mechanism and ours is that photon excitation with vibrational-rotational to electronic, $V \rightarrow E$, energy transfer is involved in the near resonant deexcitation of $4p\,^3P_2$ state in our case.

At higher oven temperature the density of Zn atoms as well as the excited Zn atoms increases. The formation rate of Zn_2 molecules created through three-body collision, e.g. $2Zn^* + He \rightarrow Zn_2 + He + dE$, goes up. This has been verified in the intensity measurement of Zn_2 molecular emission bands centered at 385 and 424 nm.[5] It limits the population in high-lying state of atomic Zn, and makes the SE at 481.0 nm consequently saturated.

In summary, we have observed stimulated emission of 481.0 nm in atomic Zn vapor. Its upper state is populated by energy pooling and lower state is emptied by the radiation of 307.6 nm with the aid of rotational energy transfer. The results have shown that in near-resonant excitation or deexcitation properly selected molecular gas can play a great role.

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