Competition effect in atomic-molecular system*

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Abstract

The competition effects among the processes of atomic ionization, optical pumped stimulated radiation(OPSR),four-wave frequency mixing (FWFM) and molecular stimulated diffuse band radiation at the atomic two-photon resonance of $3S \rightarrow 4D$ in $Na_2 - Na$ mixture were observed. The dip at the two-photon resonance in the excitation spectrum for the diffuse-band radiation was interpreted as suppression of population in $4D$ state.

1 Introduction

The generating and utilizing of molecular diffuse-band radiation is an important subject for studying excimer lasers and atomic-molecular physics. The various mechanisms of producing molecular diffuse-band stimulated radiaton were developed, for example, in atomic-molecular system the stimulated radiation from high-lying triple state to low triple state could be obtained by two-photon resonantly exciting atoms then following collision between atoms in high-lying excited state and molecules in the ground state[1-2]. This is an efficient process of producing diffuse-band stimulated radiaton. However there are others processes accompanying process of two-photon resonantly exciting atoms: The photo-ionization process following two-photon resonance,the stimulated radiation starting from high-lying excited state of atoms,four-wave frequency mixing process. The competition effect occuring in above processes resulted in decreasing molecular diffuse- band stimulated radiation. In this paper,we not only finded optimum condition of producing molecular diffuse-band stimulated radiation but also understand clearly the interaction among nonlinear processes through studying the competition effect.

2 Experiment

The laser beam from a Nd:YAG pumped dye laser(Quanta Ray DCR-3D,PDL-2) was focused into the center of the crossed heat-pipe oven by an optical system. Using RD590 dye, the output energy

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of the tunable dye laser was about 40 mJ at the wavelength region from 565 nm to 591 nm with line width about 0.1 cm^{-1} and pulse width of 8ns. The mixture vapor of atomic and molecular sodium was produced by the heat-pipe oven containing pure sodium sample, the densities of atomic and molecular sodium were determained by the temperature of the oven center. An ionization detector was installed in the heat-pipe oven to measure photo-ionization signal when optical signal being detected. The buffer gas was not filled in the oven. The radiation from the forward direction of the oven was received after passing the monochromater, then photo- electric signal was fed into channel B of the BOXCAR. At the same time the ionization signal produced from two-photon resonance three-photon ionization was introduced by a resistance of 10KΩ and sent to channel A of the BOXCAR. The optical and the ionization signals were monitored by oscilloscope 1 and 2 respectively. The BOXCAR and the two oscilloscopes were trigged by a photo-electric detector as receiving a small pulse signal of the laser. Because of the different time decay behavior for optical and ionization signals, the different time delay and gate widths of two gates were chosen to get the higher signal- noise rate of the average value of the signals. All measurements were performed under the condition that the laser energy was stabilized, which was guaranteed through monitored laser energy in the experiment. The error, which is brought about by the fluctuation of the sample temperature, could be reduced with the help of high accuracy of the temperature controller.

3 Results and discussion

The part of energy-level diagram of atomic and molecular sodium is shown in Fig.1. After atomic sodium transition from the ground state $3S$ to $4D$ state produced by two- photon excitation corresponding to laser wavelength of 578.7 nm,there are some possible processes:

(1) The two-photon resonance three-photon ionization through the atoms in the $4D$ state absorbing one more photon.

$$Na(4D) + h\nu \overset{\text{ionization}}{\rightarrow} Na^+ + e^- \tag{1}$$

(2) The optical pumped stimulated radiation owing to population inversion between $4D$ and $3P$ states, $4D$ and $4P$ states.

$$Na(4D) \overset{\text{radiation}}{\rightarrow} Na(4P) + h\nu(2.33\mu m) \tag{2}$$

$$Na(4D) \overset{\text{radiation}}{\rightarrow} Na(3P) + h\nu(568.6nm, 568.8nm) \tag{3}$$

(3) The four-wave frequency mixing by nonliear interaction between pumping wave and optical stimulated radiation wave in the sodium vapor:

$$h\nu_{uv} = 2h\nu_L - h\nu_{IR} \tag{4}$$

where, $\nu_L, \nu_{IR}, \nu_{uv}$ are the frequencies of pumping optical wave,optical pumped stimulated radiation wave and coherent radiation wave respectively

(4) The diffuse-band stimulated radiation generated by transition from high-lying triplet state populated through collision between atoms in $4D$ state and molecules in the ground state.

$$Na(4D) + Na_2(X^1\Sigma_g^+) \overset{\text{collision}}{\rightarrow} Na_2(^1\Lambda_g, 2^3\Pi_g) + Na(3S) \tag{5}$$

$$Na_2(^1\Lambda_g) \overset{\text{collision}}{\longrightarrow} Na_2(2^3\Pi_g) \tag{6}$$

$$Na_2(2^3\Pi_g) \overset{\text{radiation}}{\longrightarrow} Na_2(a^3\Sigma_u^+) + h\nu(around436.0nm) \tag{7}$$

To understand the competition among the reaction processes above under different temperature, the four kinds of signal were measured respectively. The change of excitation spectrum measured with temperature for producing the diffuse-band stimulated radiation of molecular sodium are shown in Fig.2. For the ionization signals,optical pumped stimulated radiation signal, measurements which is similar to Fig.2.were also done and the changes of those signals at different temperatures were also obtained.The result showed clealy:the optimum temperature was different for producing the above signals. For example, the diffuse-band signal gradually approached zero at low temperature.But with increasing of the temperature, it not only increased at the two-photon resonance exciting of $3S \rightarrow 4D$, but also could be observed in the certain wavelength region corresponding to offset of $3S \rightarrow 4D$.When oven temperature arrived 380°C, the diffuse-band radiation signal reached maximum. As the temperature continusly increases ($350 - 370^o$), the diffuse-band signal at the position of atomic resonant excitation weakened. However, it rose on two sides of resonant excitation of atoms. As the temperature was above 410^oC, the peak of atomic resonant excitation disappeared.At 450^oC,the "dip" appeared at the position of atomic resonant excitation. Such a phenomenon has been observed in our provious work about molecular potassium[3−4].

The changes of various signals generated by two- photon resonant excitation of atoms($3S \rightarrow 4D$) with temperature were shown in Fig.3. Within the temperature below 310^oC, there were two processes of atomic ionization and molecular diffuse-band radiation but OPSR and FWFM signals weakened, the ionization signal started to increase at 150^o. It reached the maximum at the 250^oC, but diffuse-band signal decreased; When the temperature was above $250C$, the ionization signal started to decreas, but the diffuse-band signal increased.When the ionization vanished at 340^oC, the diffuse-band signal reached the maximum. Apparently, there was the competition between two-photon resonance three- photon ionization and the collisional population from excited state atoms to molecules.

In the range of $340 - 500^o$, the ionization signal weakened but OPSR and FWFM signals started to increase,at 390^o both of them reached the maximum value, the diffuse-band signals started to fall from the maximum value. When the temperature continused to increase, OPSR and FWFM signals reduced. This fact shown that in the temperature of 340^o to 500^oC, there were apparent competitions among the processes described in eq.(1) to eq.(4b). The presence of OPSR and FWFM depopulated atoms in $4D$ state. This led to decrease the population in high-lying states of molecule. The transmission spectrum of laser light passing the sodium vapor is shown in Fig.4. There was a intense absorption at the two-photon resonance excitation of atom but the diffuse-band radiation was still small. This could also indicated that the population in $4D$ state was suppresed by other reaction processes.

We should notice that with rising of temperature, the density of molecular sodium increased too. So the diffuse-band radiation by two-photon exciting Na_2 in wider rage of pumping wavelength could be produced. This have been proved in our previous paper[5]. In the present experiment, the diffuse-band stimulated radiation could be detected in the excitation wavelength range of 577-580 nm. It increased with temperature as shown apparently in Fig.2. At low temperatures, the diffuse-band radiation signal were composed of the intense signal got by two-photon excitation of Na and the weaken signal got by two-photon excitation of Na_2; with increasing of temperature, the signal of Na_2 also increased. At high tempwerature, the diffuse- band signal produced by two-photon excitation of Na_2 was large. At the position of two-photon resonant absorption of atoms,the possible reason for the appearence of "dip" can be as follows: (1) The two-photon absorption of atomic sodium decreased the excitation of Na_2.(2) After atomic sodium being populated in $4D$ state, the collisional transfer from atoms to molecules was decreased.

4 Conclusion

The competition among the processes in producing diffuse-band by the collisional transfer of energy from atoms to molecules, four-wave frequency mixing and three-photon ionization were studied in experiment. At lower temperatures, there was mainly the competition between diffuse-band stimulated radiation and two-photon resonance three- photon ionization of atoms;At high temperatures, there was the interaction among the diffuse-band stimulated radiation, optical pumped stimulated radiation and four-wave frequency mixing; At further higher temperatures, the "dip" at the position of two-photon excitation of atoms for excitation spectrum of producing diffuse-band radiation resulted from the coherent process of optical pumped stimulated radiation and four-wave frequency mixing supressing the non coherent process of collisional transfer energy from atoms to molecules.

References

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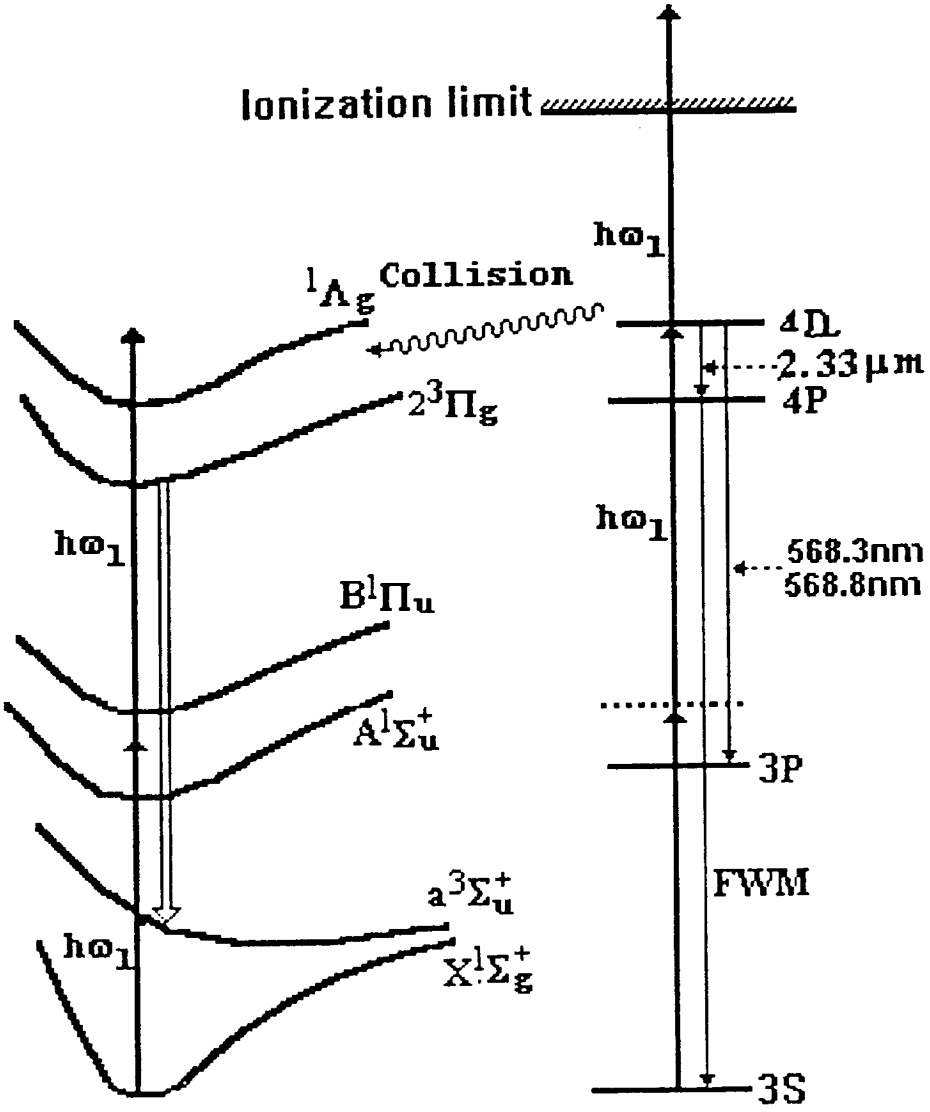
Captions of Figure

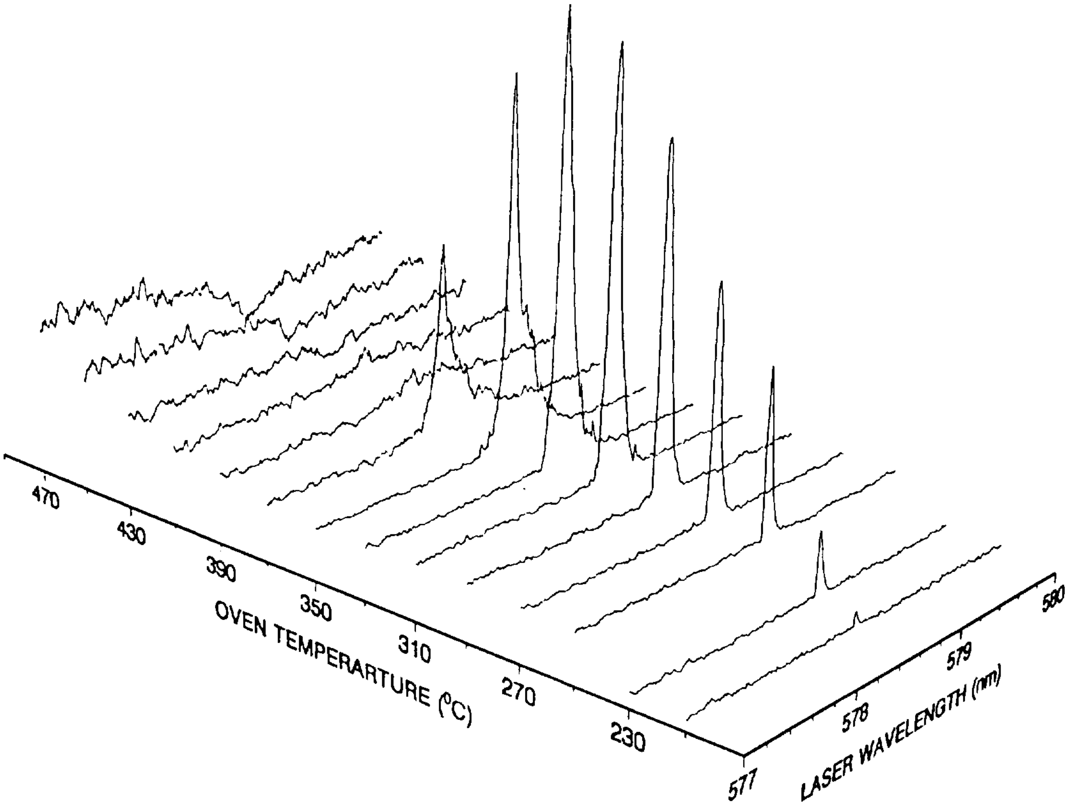
Fig.1. The part of energy-level diagram of Na_2 and Na.

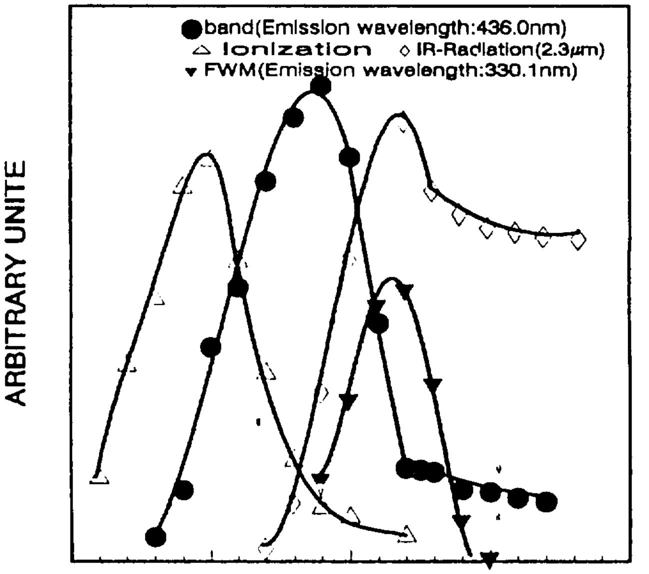
Fig.2. The excitation spectra for generating diffuse band radiation from transition of $2^3\Pi_g \rightarrow a^3\Sigma_u^+$.

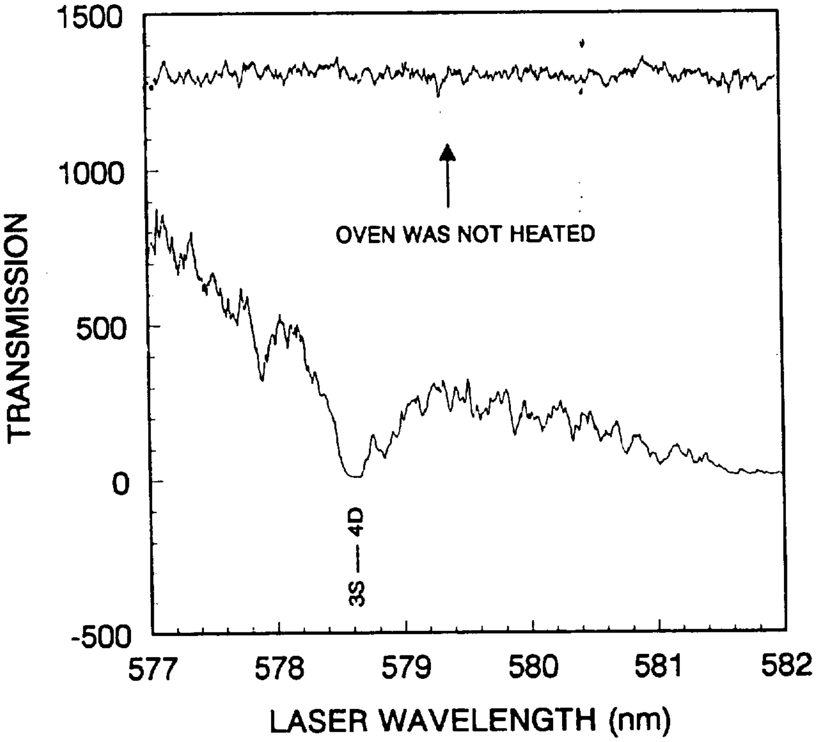
Fig.3. The dependence of four kinds of signal on temperature for two-photon transition $3S \rightarrow 4D$.

Fig.4. Transmission spectrum in sodium vapor at 450^oC.


Ionization limit
hω1
$^1\Lambda_g$ Collision
4D
2.33μm
4P
$2^3\Pi_g$
hω1
hω1
568.3nm
568.8nm
$B^1\Pi_u$
$A^1\Sigma_u^+$
3P
FWM
$a^3\Sigma_u^+$
hω1
$X^1\Sigma_g^+$
3S


470
430
390
350
310
270
230
OVEN TEMPERARTURE (°C)
577
578
579
580
LASER WAVELENGTH (nm)


●band(Emission wavelength:436.0nm)
△ Ionization ◇ IR-Radiation(2.3μm)
▼ FWM(Emission wavelength:330.1nm)
ARBITRARY UNITE
150
250
350
450
550
TEMPERATURE (C°)


1500
1000
500
0
-500
TRANSMISSION
577
578
579
580
581
582
LASER WAVELENGTH (nm)
OVEN WAS NOT HEATED
3S — 4D