

Observation of the UV Emission in Sodium Vapor  
under Tunable Dye Laser Pumping

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The output from a Quanta-Ray dye laser using basic Kiton red 610 dye solution is used to pump the sodium vapor contained in a heatpipe. Optical signals are averaged to find the 330.2nm UV emission is an ideal case of a two photon resonance excitation process at low input power and low temperature. Also an amplified spontaneous emission (ASE) from the dye laser is found effective in generating this sodium 4p-3s transition radiation.

## I. INTRODUCTION

Nonlinear optical processes in metal vapors have attracted many interests in recent years.<sup>1,2,3,4</sup> In sodium, 330nm emission is a very distinct example. It has an above 5% conversion efficiency at 578 nm pumping.<sup>5</sup> At higher temperatures, molecular process will dominate the generation of this emission as reported by Chen et al.<sup>4</sup> and Dinev et al.<sup>6</sup> in the present study, we used a different dye solution in a Quanta-Ray PDL-2 dye laser with lasing wavelength ranged from 572nm to 586nm for pumping sodium vapor. Also we used a microprocessor in averaging the photomultiplier tube output to improve the signal to noise ratio and find the ideal quadratic power dependence and linear density dependence on the sodium 330.26 nm emission. An effective pumping of this 330nm UV emission by the ASE from the dye solution with dye laser tuning sitting at 594nm is also observed.

## II. EXPERIMENTAL SET-UP:

The instrument arrangement is shown in Fig. 1. Light source is a Quanta-Ray Nd:YAG pumped dye laser (DCR-2 and PDL-2) with the Kiton-Red 610 basic dye solution which

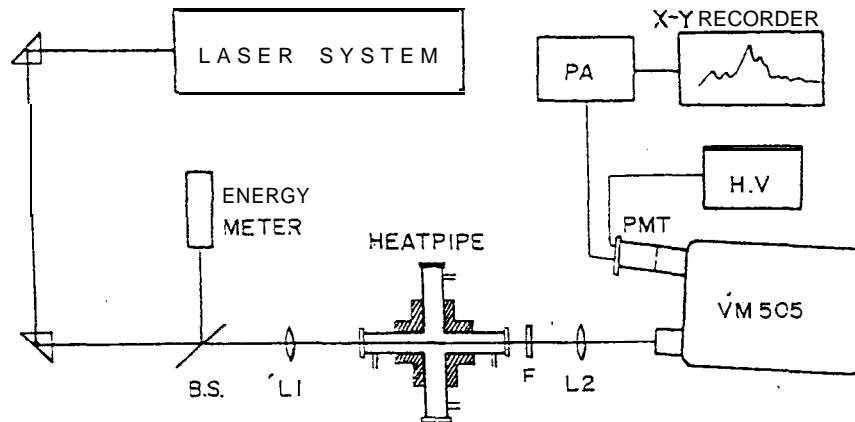


Fig. 1 Instruments arrangement. Details are explained in the text.

has a peak output about 24 mJ/pulse at 576nm. The pulse width is 7nsec and the linewidth is about 0.06nm. Laser wavelength around 578.7nm is used in this study with a 10 Hz repetition rate. The dye solution is found lasing within 572nm and 586nm range. At 594 nm setting of the dye laser wavelength selector we get ASE only with a bandwidth broader than 4.1 nm centered around 578nm and its intensity is below 10% of the laser line intensity at 578nm which is about 22mJ/pulse. This ASE intensity gets much lower within the dye lasing wavelength region.

Sodium vapor is confined in a crossed heatpipe with a 30cm heated length. The temperature profile is shown in Fig. 2. This heatpipe, temperature is controlled by an

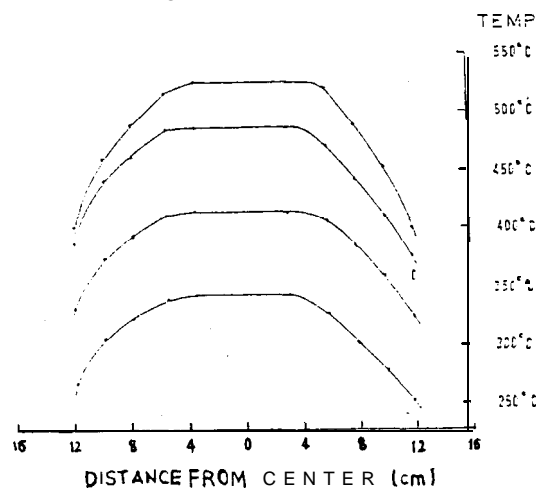


Fig. 2 Heatpipe temperature profile.

Eurotherm temperature controller 093, programmer 125 and Lilliput Thyristor unit 032. Total of twelve Omega K-type thermocouples are used to monitor and measure the temperature profile in this heatpipe. The temperature controller is able to keep the temperature within  $0.01^{\circ}\text{C}/1\text{ Omin}$ . The saturated vapor density is determined from the equilibrium temperature and calculated according to the table given by Nesmeyanov.<sup>7</sup> Argon buffer gas pressure is kept within 5 to 30 Torr through all the experiments so far. We have repeatedly heated the pipe up to  $500^{\circ}\text{C}$  and found no traces of sodium condensation on the quartz windows.

Laser beam is directed through a slide glass plate B.S. for beam splitting. The weaker beam from the beam splitter is fed to the energy meter (Laser Precision RjP7200) while the stronger beam is focused into the heatpipe by a 50cm focusing lens L1. The exit light after passing through sodium vapor is filtered by an Oriel band pass filter F (Oriel #5 1800, transmission in 270nm-400nm and 620nm-IR are allowed) and then collected by a 20cm focusing quartz lens L2 to be fed into the 0.5m vacuum monochromator (Acton Research Corp. VM505). Signals are detected by a Hamamatsu R666 photomultiplier tube PMT operating at -500V to -700V. Output is amplified by a Keithley 414A picoammeter and recorded by a HP701 5B X-Y recorder.

Monochromator scanning system is calibrated by an Oriel Ne spectral lamp (Oriel# 6032) and Hg-Ar lamp (Oriel#6035) and optimized on collimating and focusing by adjusting the optics in VM505 to minimize spectral linewidth measured.

### III. RESULTS AND DISCUSSION

#### 3-1 Emission spectrum

The observed 330nm ultraviolet emission is shown in Fig. 3 and it has been reported

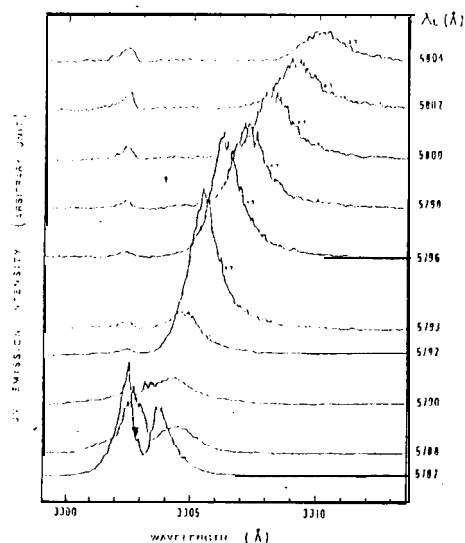


Fig. 3 330nm emission from sodium vapor as a function of laser wavelength dial setting.

by several authors already<sup>4,5,6</sup> The doublet is clearly resolved in the present study and we could see the emission peaks shift as the laser wavelength tuning in accord with the suggestions given by Chen et al.<sup>4</sup> and Hartig.<sup>5</sup> The strong peak at 578.7nm laser wavelength corresponds to the two photon resonance of sodium 3s-4d atomic states. In Fig. 3, obviously the stationary 330.2nm emission is enhanced by parametric process under 578.7nm and 578.8nm excitation elsewhere there is a competition among the population of 4p state and the parametric process, as shown in the figure, evidenced by the increase of 330.2nm intensity as the tunable line emission intensity is getting down.

Fig. 4 shows the emission spectrum of 330nm under ASE pumping at different tem-

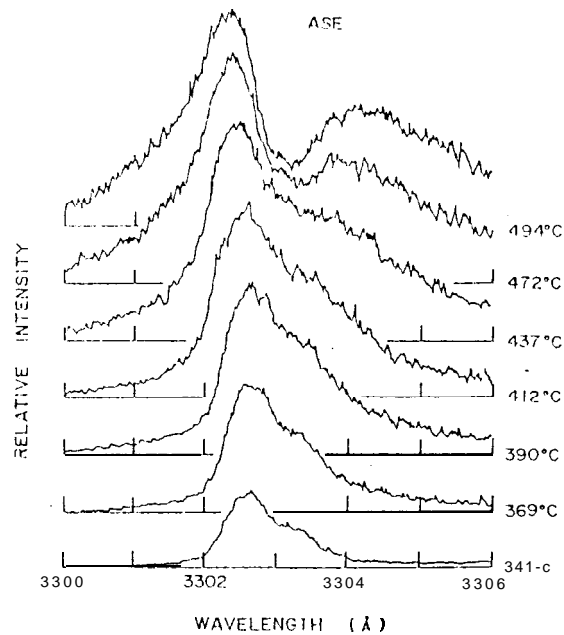


Fig. 4 330nm emission from sodium vapor under ASE pumping at different temperatures.

peratures. The fact that the emission peaks get higher and broader at higher temperatures indicates the sodium molecules are involved in generating this UV emission. The dip at 330.3nm might be due to the atomic absorption of the 3s-4p transition.

### 3-2 Excitation function

Excitation function is taken at the interested wavelength of 330.26nm and 330.34nm both with a monochromator bandwidth adjusted to be within 0.008nm and 0.042nm. This is capable of resolving two peaks shown in Fig. 3. The dye laser output is scanned from 594nm to 569nm at a 0.5nm/sec or slower rate. Fig. 5a(5b) is the excitation function of 330.26nm(330.34nm) at different heatpipe temperatures. Fig. 6 is the blown-up display of the excitation peak at 578.72nm of Fig. 5.

The sharp peaks in Fig. 5a and 5b are discussed before.<sup>4,5,6</sup> They are due to the sodium atomic 3s-4d two photon resonance absorption to populate the 4d level then cascades down to the 4p state which finally gives these 330nm emissions. The broad band

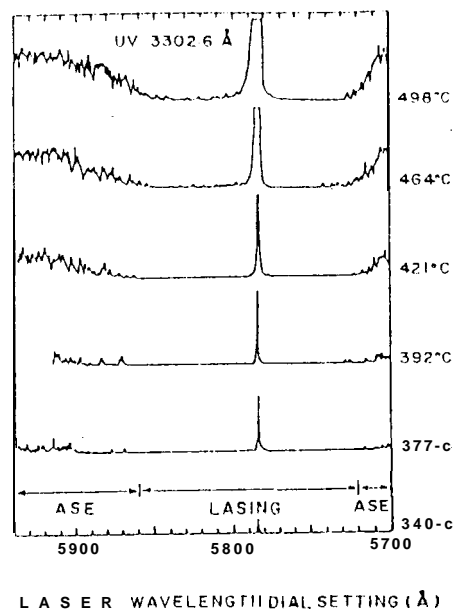


Fig. 5a Excitation function of sodium 330.26nm emission at different temperatures. Lasing wavelength is between 572nm and 586nm.

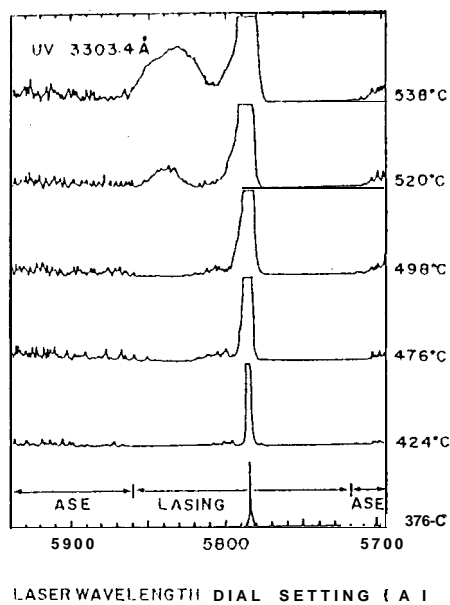


Fig. 5b Excitation function of sodium 330.34nm emission at different temperatures. Lasing wavelength is between 572nm and 586nm.

in Fig. 5b rises above  $500^{\circ}\text{C}$  and is suggested by Chen et al.<sup>4</sup> and Dinev et al.<sup>6</sup> as sodium molecule two-photon absorption and stimulated electronic Raman scattering (SERS) respectively. This broad peak at 583nm excitation will dominate the 330.34nm emission generation at higher temperatures.

We could not find the sharp excitation peaks reported by Dinev et al.<sup>6</sup> but else find

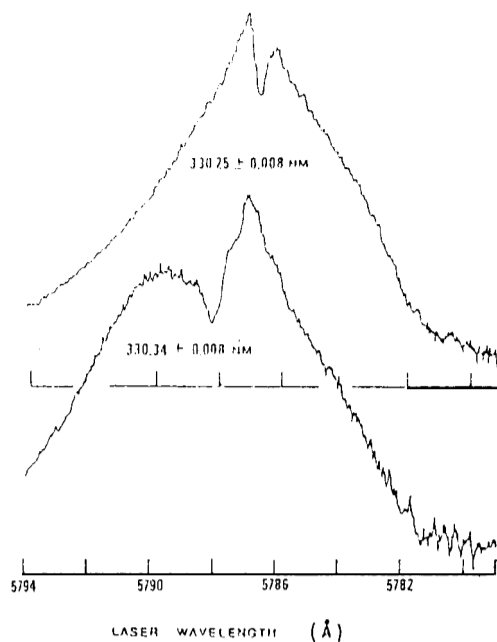


Fig. 6 Excitation functions of sodium 330.25nm and 330.34nm emission under around 578 nm laser pumping.

new excitation mechanism in this UV emission. As laser wavelength tuned beyond 586nm and below 572nm we could get a weak 330.26nm and 330.34nm radiations even the heat-pipe temperature is lowered down to  $377^{\circ}\text{C}$  (corresponding to sodium atom density  $2.91 \times 10^{16}\text{cm}^{-3}$  and molecule density  $5.14 \times 10^{13}\text{cm}^{-3}$ ). Actually, in these wavelength regions the basic Kiton Red 610 dye solution we had is not lasing. Pump beam under this condition is an amplified spontaneous emission (ASE) from the laser dye solution and has a linewidth broader than 4.1 nm. We suggest the sodium electrons are excited from the ground state of the sodium molecule  $X^1\Sigma_g^+$  to the first excited state  $A^1\Sigma_u^+$  and then pumped again by the ASE to an upper state. The pumping mechanism is similar to that of sodium 436nm diffused violet band reported by Wu et al.,<sup>7</sup> Woerdman<sup>9</sup> and Allegrini et al.<sup>10</sup> This upper state then dissociates to 4d or 4p atomic state which finally gives the 330nm emission.

The difference among the present study and those reported in the literatures<sup>4,6,10</sup> is the optical field in the 594nm region is not very strong compared with a laser field. So a nonlinear multiphoton process is not likely to happen. Argon buffer gas in the heatpipe might help the energy relaxation through collisional energy transfer with sodium dimers but why this up-conversion process is so intense (compared with incoherent light source pumping) is not clear yet.

### 3-3 Power dependence

In measuring the power dependence or the density dependence, the output of the picoammeter is fed to an HP3478A digital multimeter and the digitized data are fetched and

analyzed by an Apple II microcomputer through IEEE-488 bus. Each data point is averaged from one hundred microcomputer acquisitions. This increases the signal to noise ratio and improves the results.

Fig. 7 shows the power dependence of the 330.26nm emission has a typical quadratic

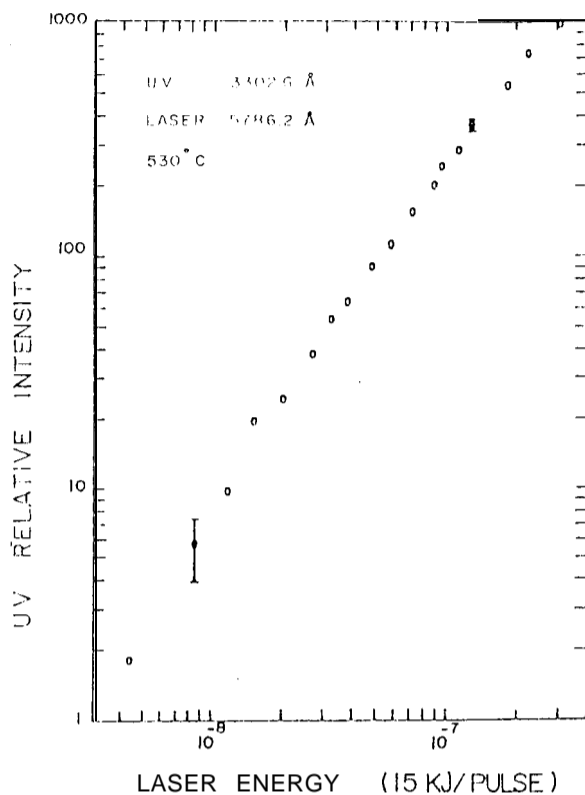


Fig. 7 Power dependence of sodium 330.26nm with 578.62nm excitation.

dependence at low input laser energy. At higher laser energy input, the 330.26nm output deviates from the ideal case might due to the competing SERS and the ionization of sodium atoms and molecules involved.<sup>4,6,11-14</sup> Similar behavior happens in 330.34nm emission as shown in Fig. 8.

Fig. 9 compares the 330.26nm emissions in power dependence at different temperatures under ASE pumping. Both of them indicate a quadratic dependence on input laser energy at the low end. This behavior is ideal for two-photon absorption processes. The 330.26nm emission threshold gets lower as temperature of the heatpipe rises could be ascribed to the increased molecule density at higher temperatures.

#### 3-4 Density dependence

The density dependence of 330.26nm and the 330.34nm emissions are shown in Fig. 10 under 578.72nm laser excitation. The 330.26nm emission shows a linear dependence on the sodium atom density is ideal for the case of two photon process. Also this emission has a lower threshold and a higher saturation output intensity than the 330.34nm emis-

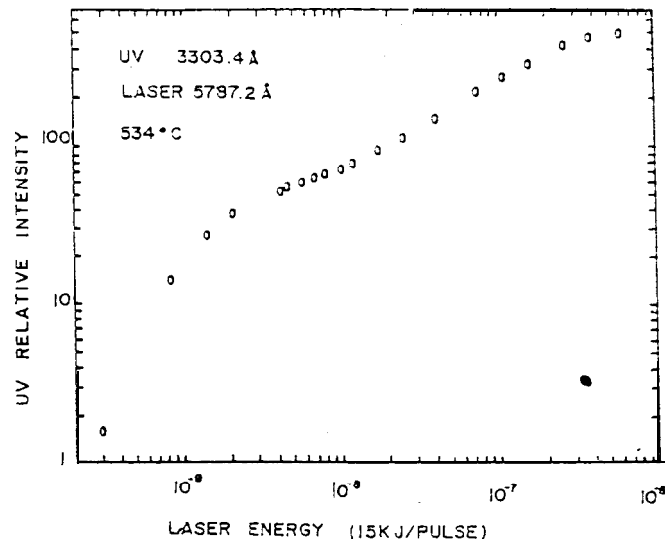


Fig. 8 Power dependence of sodium at 330.34nm emission with 578.72nm excitation.

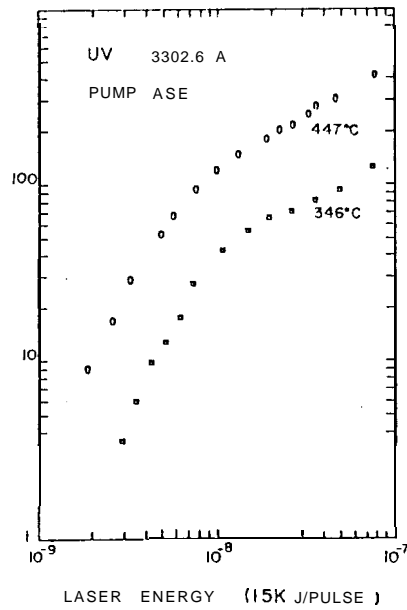


Fig. 9 Power dependence of sodium 330.26nm emission with ASE excitation at different temperatures.

sion does. The source of the distinct decrease of 330.34nm emission intensity at higher temperature is not clear yet.

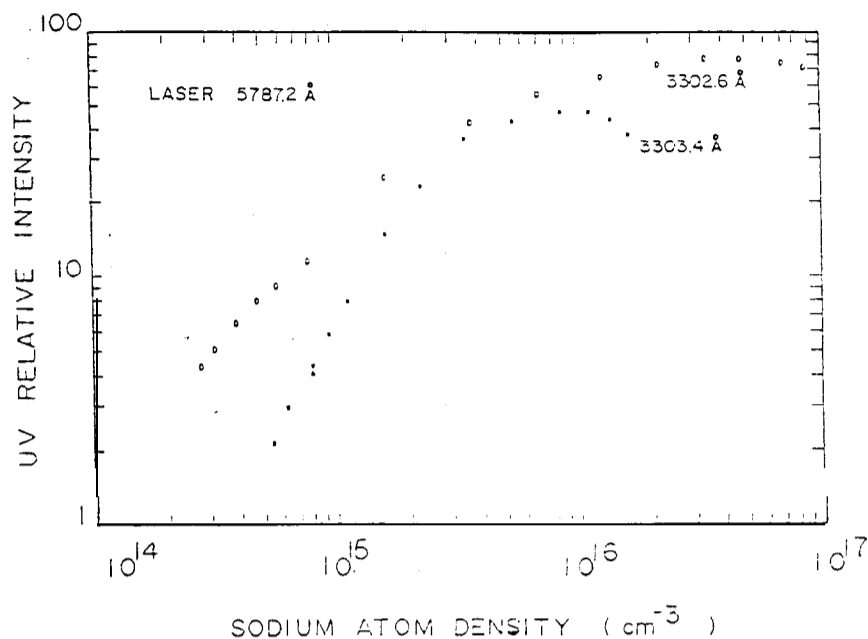


Fig. 10 Density dependence of sodium 330.26nm and 330.34nm emissions under 578.72 nm excitation

#### IV. CONCLUSION

We repeated earlier works done by Chen et al.<sup>4</sup> Hartig<sup>5</sup> and Dinev et al.<sup>6</sup> with a different dye solution and found the 330.26nm emission is a part of an ideal two photon resonance process. Also we have observed a strong UV 330nm emission from sodium vapor under ASE pumping. This emission shows a quadratic dependence on the input ASE power. Detailed study is mandatory to understand this effective ASE pumping up-conversion mechanism.

#### V. ACKNOWLEDGEMENT

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