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Solar-Powered Alkali Metal Vapor Lasers

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Abstract:

The emission spectrum of the $A^1 \Sigma_0^+ - X^1 \Sigma_9^+$ band of Na₂ has been recorded following excitation by monochromatic radiation in the region of X-A and X-B absorption. The spectral profile has been investigated as a function of excitation wavelength, sodium vapor temperature and buffer gas pressure. Additionally, gain measurements were made for the "satellite" of the A-X band as a function of the sodium vapor temperature and buffer gas pressure.

1. Introduction

Absorption and emission of radiation by alkali metal vapor is studied frequently because of its practical importance and the variety of phenomena that can be studied in a single system. For a simple molecule like Na2 a large number of the electronic states are known (1). In sodium vapor, which consists mainly of Na atoms and Na2 molecules, many of the electronic states can be coupled by collisions. Energy transfer between electronic states can occur in small molecules as a result of the interaction of the molecule with its environment. Electronic energy transfer is known to occur in the alkali dimers from the observation of A-X band fluorescence in both Li₂ and Na₂ following laser excitation into the B state (2,3). At present, two energy transfer processes for the transformation of the B state excitation of Na₂ molecules into A state excitation have been proposed. Based on A-X band intensity measurements of single frequency, Ar⁺ laser excitation of a supersonic expansion of sodium vapor, Hulsman and Willems (4) conclude that the transformation of B state excitation into A state excitation proceeds mainly via atomic excitation. The molecular B state excitation is transformed into atomic excitation by collision with a ground-state atom:

 $Na_2(B^1\Pi | v J') + Na(3s) \rightarrow Na_2(X^1\Sigma_0^+ v J'') + Na(3p),$

then the atomic excitation is transformed into a molecular A state excitation by collision with a ground-state molecule:

Na (3p) + Na₂(X¹ Σ_{4}^{+} |v "J") \rightarrow Na (3s) + Na₂(A¹ Σ^{+} |v 'J').

On the other hand, Hussein et al (5) and Effantin et al (6) have shown that radiative transitions from the $(2)^1\Sigma_{a}^{\dagger}$ state in the infrared are also responsible for populating the A state. Astill et al (7) based on a detailed study of the $A^1\Sigma_g^+ \cdot X^1\Sigma_g^+$ fluorescence in Na₂ following excitation into the $B^1\Pi_u$ state using lines from an A_r^+ laser found that their results could be interpreted through a populating mechanism involving collisional transfer from the $B^1\Pi_u$ to the $(2)^1\Sigma_g^+$ state followed by radiation transfer to the $A^1\Sigma_g^+$ state. The collision partner was assumed to be ground sodium atoms, Na (3s), rather than excited state sodium atoms, Na (3p), proposed by Hussein, et al (5).

In this report we present results of a study on the vapor density dependence of the $A^{1}\Sigma_{u}^{+}$ — $X^{1}\Sigma_{0}^{+}$ fluorescence in Na₂ following excitation into the $B^{1}\Pi_{u}$ state. We also present results of gain measurements of the A-X band head of heads for the same vapor densities.

2. Experimental

The experimental arrangement is shown in Fig. 1. A Cermax 300 W xenon arc lamp (ILC Technology, Inc. model 300) and a 0.25m monochromator (Jarrell-Ash model 82-410) were used to excite the sodium vapor. The sodium vapor was contained in a stainless steel cell fitted with four viewing ports arranged perpendicular to each other. The optical cell is based on a commercially available five-way cross vacuum fitting with one of the five arms attached to the vacuum port and the separately heated sodium cell. Heating of the optical cell is achieved using four commercial 400W watt-flex cartridge heaters (Dalton Electric Heating Co.) inserted externally into a stainless steel block consisting of three pieces machined to fit around the five arms of the optical cell. A chromel/alumel thermocouple is placed in the block to provide a temperature measurement. The separately heated sodium cell is based on a commerically available three-way cross





Fig. 1. The experimental set-up.

vacuum fitting in which one of the three arms is attached to the vacuum port and optical cell. Heating of the sodium cell is achieved using two commercial 400 W cartridge heaters inserted externally into a stainless steel block machined to fit around the three arms of the sodium cell. Temperature measurements of the sodium cell were made using a chromel/alumel thermocouple placed in the block. A stainless steel chimney placed inside the central column connecting the sodium cell to the optical cell was used to introduce the sodium vapor into the optical cell. The advantage of this cell design over the cells previously used is that the sodium vapor stability is increased since the sodium refluxes within the chimney of the connecting arm, reducing condensation on the colder surfaces of the vacuum walls increasing the time between refills. Additionally, it was found that the cell could be operated at very low pressures of the buffer gas and maintain a central column of vapor in the optical cell. Buffer gases were introduced into the cell at each of the four windows of the optical cell. Further, this cell design was found to reduce the contamination of the sapphire windows, and no measurable decrease in the transparency of the windows was observed for data presented in this report.

The fluorescence from the excited Na₂ molecules was collected at right angles to the xenon lamp propagation direction and focused on the entrance slit of a grating monochromator. (Instruments SA Inc. model HR 320). A cooled photomultiplier tube (Hamamatsu model R943-02, GaAs photocathode) was placed at the exit slit of the monochromator. The output of the photomultiplier tube was measured using a photon counter (EMI model C-10) or a micromicroammeter (Keithley Instruments model 414-17). The output from the photon counter or micro-microammeter was measured on a recorder (Lineis Inc. model L6512-1). The intensity and form of the emission band has been investigated as a function of buffer gas

(Ar) pressure from 0 to 40 Torr, cell temperature in the range of 380 to 540 C, and the X-B excitation wavelength using the excitation monochromator.

3. Results and discussion

Fig. 2 shows a typical spectrum obtained for the white light excited sodium vapor. Depending on the temperature of the cell, and consequently on the sodium atomic and molecular density, the Na D lines are seen either as sharp emission features or as absorption on the A-X Na₂ emission background. The molecular spectra show extensive rovibrational structure over the range of A-X and B-X bands. To the red of the A-X band profile, sharp "satellite" structure is visible around 805.7 nm. This structure appears at the turning point of the potential difference curve and is characteristic of the A-X band. (7)

The intensity and form of the A-X band emission has been investigated as a function of the wavelength by scanning the excitation monochromator through the white light continuum from 450 to 720 nm with the emission spectrometer set on the peak of the "satellite" emission at 805.7 nm and by setting the excitation monochromator at wavelengths in the absorption regions of both the A-X and B-X bands.

The emission spectra was also investigated as a function of cell temperature. When the band profiles of the Na₂ A-X and B-X emission were investigated as a function of cell temperature, they were found to be essentially independent of temperature, while the intensity of the bands were found to be dependent on the Na (and hence the Na₂) pressure. The Na partial pressure is found be reference to the tables on vapor pressure against temperature (8). It is also noted this observation is consistent with the data obtained by Pardo et al. (9) and Astill et al., (3) who found that the B-X



Fig. 2. The emission spectrum of sodium vapor in the region 500 - 840 nm excited by the continuum of a 300 watt high pressure xenon lamp.

fluorescence was directly proportional to the intensity in the A-X band when measured as a function of increase or decrease of the laser intensity, and hence concentration of Na_2 . It is also important to stress the fact that the Na_2 A-X emission for excitation of the sodium vapor for wavelengths in the region of B-X absorption is present in the absence of all buffer gas.

Fig 3 shows the emission intensity of the "satellite" (850.7 nm) of the A-X band, for a sodium vapor temperature of 425C, excited by radiation from 450 to 720 nm. The emission intensity of the "satellite" is found to be dependent on the absorption of radiation corresponding to the X-A and X-B transitions of Na₂. For sodium vapor temperatures from 380 to 540°C, the emission intensity of the "satellite" of the A-X band excited by radiation from 450 to 720 nm were obtained. At a sodium vapor temperature of 400 C, the emission intensity of the satellite is found to be the same for the X-B absorption or the X-A absorption. The decrease of the emission intensity for radiation absorbed in the region of the X-B transition compared to radiation absorbed in the region of the X-A transition of Fig. 3 is observed to continue with the increase of the sodium vapor temperature until the "satellite" emission is no longer observable for the absorption of radiation in the region of the X-B transition of Na₂.

To verify that the absorption of radiation in the region of the X-B transition of Na_2 contributes to the observed A-X band emission, the excitation monochromator was set at a wavelength in the region of the X-B band absorption, 470 to 520 nm, and the emission spectrometer scanned from 470 to 840 nm. Fig 4 shows the emission spectrum resulting from the excitation of the sodium vapor at a temperature of 400 C for radiation peaking at 486 nm with the background emission of the sodium vapor also presented for the same wavelength region. The emission spectrum from 470 to



Fig. 3. The emission intensity of the 805.7 nm band of the sodium dimer for continuous scan of the excitation monochromater for wavelength in the region 450-720 nm.

840 nm (resulting from the absorption of radiation peaking at 486 nm) were also obtained for sodium vapor temperatures from 380 to The emission intensity of the "satellite", which is 540 C. comparable to the emission intensity of B-X band in Fig. 4, is observed to decrease as compared to the B-X band emission intensity as the sodium vapor temperatures is increased until the "satellite" emission is no longer observable at 530 C. The observed emission intensity of A-X compared to the emission intensity of the B-X resulting from the absorption of radiation peaking at 486 nm for sodium vapor temperatures of 480 C, were compared to the relative fluorescence quantum yield in the A-X and B-X bands of Na, following single mode excitation of the X-B transition of Na₂ for a cell temperature of 477 C reported by Astill et al. (3). Within the limits of the experimental set up, i.e. a 0.25m monochromator and accepting the broader band width as compared to that of a single mode or multimode Ar⁺ laser, our observations are consistent with those of Astill et al (3) who reported a relative fluorescence quantum yield of ~1% at a sodium vapor temperature of 477 C. It would be beneficial for measurements similar to those described by Astill et al. (3) to be conducted at sodium vapor temperatures of 400 to 430C.

Gain measurements of the "satellite" were also made for sodium vapor temperatures from 380 to 480C. The direction of the pulsed dye laser through the sodium cell is along the spectrometer axis of Fig. 1. The wavelength adjustments for the dye laser module could be checked by observing the output of the photomultiplier tube at the exit slit of the spectrometer. The energy loss of the cell was made for a cell pressure of 2×10^{-8} Torr and a temperature of 23C. Using DOTC dye, the average energy for 10 pulses throught the evacuated cell was found to be 10.14 µJ. Based on the results of the measurements of energy for sodium vapor temperatures from 360 to 480 presented in Table I, and the observed emission intensity of



Fig. 4. The emission spectrum of sodium vapor in the region 500-840 nm excited by monochromatic radiation peaking at 486 nm.

the A-X band satellite, it may be possible to observe lasing for sodium vapor in the 400-440C temperature range, but not at any higher sodium vapor temperature. Results presented in this report have been repeated using different cells with the shortest interval between the data of about two months, and in some cases the results compared are a year apart.

TABLE I

Pulsed optical radiation measurements of the A-X band satellite

Date	Sodium Vapor Temperature, C	Average of 10 Pulses, μJ
8/20/88	480	9.86
9/20/88	480	9.80
9/24/88	360	10.12
9/25/88	420	10.61
9/27/88	420	10.37
10/1/88	440	10.46
10/3/88	480	8.68

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