Observation of net gain in the yellow band of K_2 excimer by electron-beam pumping

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The design of a high-temperature cell appropriate for an electron-beam transverse pump of alkali dimer excimer lasers is described. By using the cell, an amplified spontaneous emission and a maximum net gain coefficient of 3% cm^{-1} of the K_2 yellow band (574 nm) were observed from the e-beam-excited mixture of K/K_2 vapor with argon buffer gas. The dissociative recombination of K_3^+ is discussed as an efficient formation process of the upper state by the electron-beam pumping. © *1999 American Institute of Physics.* [S0003-6951(99)04505-2]

There has been a considerable number of studies on alkali metal vapor dimers, with interest in developing such excimers as possible sources of tunable, efficiency, and high-power lasers.[1–6] Optical pumpings have primarily been used as pumping sources because they are easily compatible with the experimental geometry's which require heated metal vapors. In particular, Bahns and Stwalley[7] have reported an observation of small signal gain in the violet band of Na_2 by Kr^+ laser pumping. Recently, the bound-free emissions ($2^3\Pi_g \rightarrow 1^3\Sigma_u^+$) of Na_2 violet, K_2 yellow, and Rb_2 orange bands have been successfully observed by electron-beam pumping.[8,9]

Electron-beam excitation is an efficient means of conventional rare-gas-involved excimer excitations, and this excitation method is capable with producing high pumping power densities and large pumping volumes. Also, we have confirmed its usefulness in the excitation of various metal vapor excimer transitions.[8–10] In such studies, however, a necessary cold zone which separates the hot vapor zone from the output window of the electron-beam equipment introduces an extra energy loss because of a longitudinal pumping scheme.[8,9] A transverse excitation scheme simplifies the experiment (such as absorption or gain measurement) by avoiding the complications of coupling both the electron beam and the laser probe beams; and observation of laser oscillation by using an optical cavity at the two ends of the cell. Also, this scheme does not require the intense magnetic guiding fields that are necessary for longitudinal excitation. In this letter we report a design of a high-temperature cell appropriate for electron-beam transverse pumping of alkali dimer excimer lasers; and a measurement of small signal gain coefficients of the K_2 yellow band (574 nm) by electron-beam pumping.

A cross-sectional view of the vapor cell coupled to the electron-beam machine is shown in Fig. 1. The cell consists of the electron-beam-pumped region, shaped as a half cylinder with a diameter of 5 cm and 20 cm long, and two water-cooled end tubes separating the pumped region from the windows. The end tubes have an inner diameter of 3 cm and 12 cm long. The half-cylindrical region is welded to a backplate with a rectangular opening for coupling the electron beam into the cell. A steel wick is incorporated in the cell to provide recirculation as in standard heat pipes. To prevent condensation of the alkali metal vapor the cell remains at approximately 30 °C hotter than the alkali reservoir. This is very important to control the uniformity of the alkali vapor density and suppress the cluster formation in the cell. The body of the cell is constructed by a type of 316 stainless steel. It has a sufficiently high tensile strength even at a high-temperature of 700 °C; and a corrosion resistance (particularly against reactants such as alkali metals).

The Inconel foil with 30 μm thickness seals the rectangular-shaped opening (14×2.8 cm) of the half-cylindrical heated region of the cell. The foil has to be capable with high temperatures and pressures, and yet still allow electron-beam penetration into the cell. To solve the difficulty of cell sealing, we designed a knife-edge groove in the backplate around the opening to mate with a copper gas-

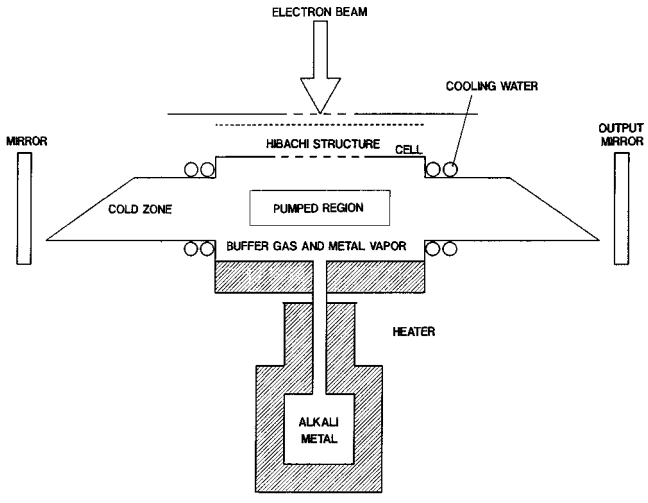

FIG. 1. Simplified cross-sectional view of the hot vapor cell coupled to the electron-beam machine. Heated cell is sealed with Inconel foil, and the electron-beam anode region was sealed with titanium foil or a Kapton foil with a conductive carbon coating.

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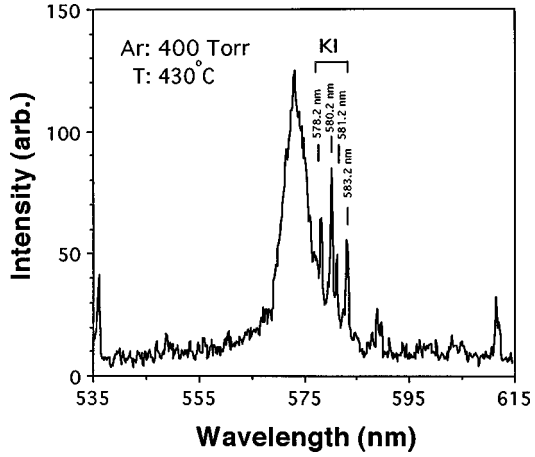

FIG. 2. The emission spectrum of the K_2 yellow excimer band from electron-beam-excited Ar/K/K_2 mixtures. The excitation energy density was about 9 mJ/cm^3.

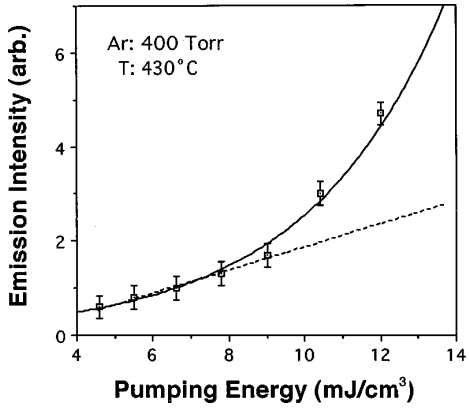

FIG. 3. The pumping density dependence of the 574 nm yellow excimer band. Ar pressure: 600 Torr; Temperature: 430 °C.

ket, and another similar shaped knife-edge groove on the top plate which has a series of holes forming a Hibachi-like structure. The seal is completed by the top plate which is held against the Inconel foil by a set of bolts around the rectangular opening. The Hibachi-like structure provides support for the foil at high inner pressures while allowing the maximum electron-beam deposition inside the cell. Two MgF_2 optical windows at the end of the cell are sealed at a Brostow angle to separate the high pressure in the cell from the atmosphere (or vacuum) in the ultracavity.

The pulsed electron-beam facility used in the experiment has a beam size of 12.5×2.2 cm^2, a peak current of 12 kA, a pulse width of 50 ns (full width at half maximum), and a maximum electron particle energy of 300 keV. The electron beam, confined by an externally applied 0.7 kG magnetic field, passes through the anode foil and a vacuum gap about 2 cm (necessary to isolate the heat loss to the electron-beam diode), and then injected into the heating cell through the Inconel foil and the Hibachi-like structure.

A half-cylindrical heater could be used to heat the cell, but during the experiment we found an easier and better way to reach the operating temperature of the cell. In the scheme we use three heating halogen lamps (20 cm long) to heat the top, bottom, and backside of the half-cylindrical cell. Each lamp was 500 W with gold coating parabolic mirrors. The operating temperature was internally between 400 and 500 °C. This was monitored in five locations at the heated cell wall along the active zone using Chromel–Alumel thermocouples. The measured temperature data showed a coincidence within a 3 °C variation in a 14 cm length of the central part of the cell. The alkali reservoir was heated by a cylindrical heater with a power supply of 1 kW.

The emission from the active zone was introduced into a spectrum multichannel analyzer (SMA) through the window, and a 15 m length quartz fiber with a coupling optics. The resolution of the SMA system was 0.1 nm/channel with a 1200 grooves/mm grating used in the 0.3 m spectrometer.

Figure 2 shows a confirmed K_2 diffuse band fluorescence observed from the e-beam-excited Ar/K/K_2 vapor mixture in the transverse-pumping scheme. The Ar pressure was 400 Torr and the cell temperature was 430 °C. Spectra in the range of 535–615 nm consisted of several sharp potassium atomic lines and a broad continuous molecular band. The band spread from 545 to 580 nm and showed good coincidence with that observed in the optical pumping;[1,5] and the previous electron-beam pumping.[9] The broad emission centered at 574 nm has been identified as the K_2 triplet band $2^3\Pi_g\rightarrow1^3\Sigma_u^+$ bound-free transition.[1] This will be abbreviated as the K_2 yellow band in the remaining part of the letter. The emission spectrum exhibits a 7 nm wide peak and a blue-shaded diffuse feature spreading to 545 nm. Both of these are in good agreement with a previous theoretical calculation[9] using the *ab initio* potentials given by Krauss and Stevens[11] and Jeung and Ross.[12]

The behavior of the 574 nm transition was investigated as a function of the excitation energy density by varying the input electron particle energy. The observed excitation energy dependence of the bound-free 574 nm emission is given in Fig. 3. In the lower pumping energy, the intensity of the yellow band almost linearly increases with the increasing of the pumping energy. An emission enhancement of the 574 nm and a narrowing of the spectrum were observed when the excitation energy density exceeded 10 mJ/cm^3, which showed a phenomenon of amplified spontaneous emission.

The effective gain coefficient of the yellow band at 574 nm was investigated by two methods. The first one used a probe light beam into the cell and directly measured the spectral intensity increase of the output beam.[7] In this case the beam was well collimated and was spectrally broad with a diameter of 5 mm at the center of the cell and a beam divergence of 8 mrad. The second method introduced a collimated back mirror and then measured the intensities of the yellow band spectrum with and without the mirror. For this case the net gain-length product $\gamma L=LN[(I_{\text{with}}/I_{\text{without}})-1]$. Because of thermal convection, a light-scattering loss was presumable at the conjunction parts between the hot and cooling zones. The effective gain-length L should be a little smaller than the length of the pumped zone, which was ~12 cm. In the present experimental conditions, L was estimated to be 10 cm. This was supported by a fitting calculation using the absorption density measured at the K resonance line, as well as the vapor density calibrated by the absorption linewidth at the heat-pipe condition as discussed above.

The results from the two different observing methods show a coincidence within a 20% variation. However, due to the distance of the observing point to the center of the cell

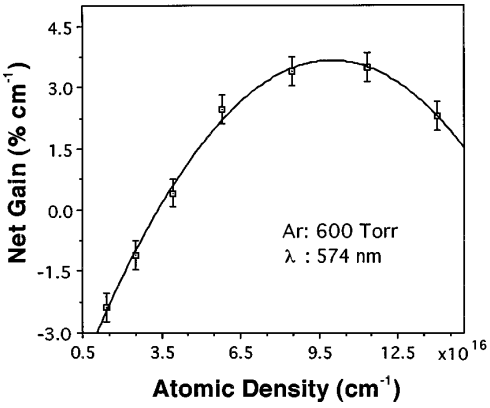

FIG. 4. The potassium density dependence of the gain coefficients of the yellow excimer band. Ar buffer gas: 600 Torr; pumping density: 12 mJ/cm^3.

being much larger than the length of the active zone, the second method was considered to be more accurate under the present experiment conditions.

The maximum net gain coefficient of approximately 3% of the K_2 yellow excimer band was observed from e-beam-excited mixtures of K/K_2 vapor with argon buffer gas. Figure 4 shows a potassium density (or temperature) dependence of the gain coefficients. The buffer gas was 400 Torr Ar, and the pumping energy density was about 12 mJ/cm^3. At the present experiment conditions, the result clearly shows that the net gain changed from negative to positive when the temperature is higher than 375 °C (corresponding to a potassium number density about $4\times10^{16}\,\mathrm{cm}^{-3}$). When the temperature is above 455 °C (about $1.4\times10^{17}\,\mathrm{cm}^{-3}$) the effective gain shows an obvious decrease with increasing temperature. This can be considered to be the increase of the K vapor absorption and the excited-state dissociation caused by the thermal collision.

Figure 5 shows the buffer-gas pressure dependence of the gain coefficients in the He buffer gas. The pumping energy density was about 10 mJ/cm^3. The result shows that the gain of the yellow band did not linearly increase with the buffer gas pressure, and the effective gain showed a saturation after the pressure reached about 1200 Torr in the He buffer gas. This could be considered to result from the increase of the buffer pressure which caused a change in the formation mechanism of the upper state.

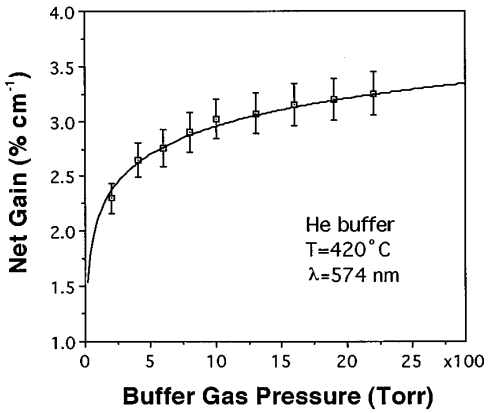

FIG. 5. The buffer-gas pressure dependence of the gain coefficients of the K_2 yellow band. The pumping density about 10 mJ/cm^3. Temperature was 420 °C.

Finally, the dissociative recombination of K_3^+, which is efficiently yielded by the e-beam excitation,[13] is proposed as the main channel for the upper state $(2^3\Pi_g)$ formation of the K_2 yellow band:

$$K_3^+ + e^- \rightarrow K_3^*(\text{unstable}) \rightarrow K + K_2(2^3\Pi_g). \quad (1)$$

The intermediate state K_3^* is expected to be unstable because the reaction energy is greater than the dissociation limit of K_3^* bound sites. Thus, resulting in a paired dissociation to $K_2^* + K$. This reaction provides a mechanism for selective production of a variety of highly excited diatomic states.[13] Process (1) is predicted to have a large cross section[7,13] because it is near resonant in reaction energies [ΔE is 0.28 eV (Ref. 14)].

As discussed elsewhere,[15,16] this mechanism is particularly attractive in that the ionization is not a serious loss but rather an integral part of the pumping, and the predominant ions in the alkali vapors are trimer ions. For example, at 430 °C (about 10 Torr, saturated vapor pressure), $P(K_3^+)/P(K_2^+)/P(K^+) \geqslant 99:1:1$ by a theoretical estimation.[16] Thus, the upper state of such a specific excimer transition may be produced selectively by a dissociative recombination process subsequent to very nonselective ionization. For this reason, the potential alkali dimer laser might be realized by such an efficient excitation process.

In conclusion, we report an observation of optical gain in K_2 yellow diffuse bands by electron-beam pumping. The strong continuum emission centered at 574 is confirmed to be attributed to the $K_2\ 2^3\Pi_g \rightarrow 1^3\Sigma_u^+$ bound-free transitions of K_2. For pumping intensity above 10 mJ/cm^3, an amplified spontaneous emission of the yellow band and a net gain of $0.03\% \pm 15\%\ \mathrm{cm}^{-1}$ was observed in the present experiment. This value should be further increased by the addition of the pumping power density; and by the optimization of the cell geometry and the operating condition.

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