

Lasing in Cs at 894.3 nm pumped by the dissociation of CsAr excimers

J.D. Readle, C.J. Wagner, J.T. Verdeyen, D.A. Carroll and J.G. Eden

Lasing on the $6^2P_{1/2} \rightarrow 6^2S_{1/2}$ (D_1) resonance transition of atomic Cs at 894.3 nm has been realised in mixtures of Ar, ethane, and Cs vapour by the photoexcitation of ground state Cs–Ar collision pairs and subsequent dissociation of diatomic, electronically-excited CsAr molecules (excimers). The blue satellites of the alkali D_2 lines provide a pathway for optically pumping atomic alkali lasers on the principal series (resonance) transitions with broad linewidth ($\gtrsim 2$ nm) semiconductor diode lasers.

Introduction: Atomic alkali lasers, optically excited on the D_2 resonance line ($n^2P_{3/2} \leftarrow n^2S_{1/2}$, $n = 5$ and 6 for Rb and Cs, respectively) and operating on the D_1 transition ($n^2P_{1/2} \rightarrow n^2S_{1/2}$) [1–3], are attractive for several reasons, one of which is the quantum efficiency (typically $>90\%$) associated with this pump/lasing cycle. Furthermore, the first principal series of K, Rb, and Cs lie in a spectral region (766–894 nm) that is accessible to high-power semiconductor lasers, and diode-pumped Cs and Rb oscillators have been demonstrated [4, 5] with output powers reported to date approaching 50 W [5]. One significant drawback of the three-level laser system pursued previously, however, is the requirement that the linewidth of the optical pump source be comparable to the spectral breadth of the atomic alkali absorption line which, when broadened by ~ 1 atm of rare gas, is in the order of 10 GHz. The necessity of narrowing the linewidth of conventional high-power laser diodes by ~ 2 orders of magnitude and electronically locking the pump source onto the $2^2P_{3/2} \leftarrow 2^2S_{1/2}$ transition significantly raises the complexity of the overall system.

We report an alternative approach to optically pumping the resonance transitions of the alkali atoms. By photoexciting the blue satellite associated with the D_2 absorption line in alkali-rare mixtures, atomic alkali–rare-gas collision pairs in the ground state are pumped to a dissociative, excited state interaction potential for the alkali–rare-gas excimer. The spectral width of the satellite is typically several nanometres, thus permitting conventional high-power laser diodes to serve as the pump source. The experiments described here entailed the photoexcitation at 837 nm of the blue satellite of the Cs D_2 transition in Cs–Ar–ethane mixtures and the observation of lasing at 894.3 nm on the $6^2P_{1/2} \rightarrow 6^2S_{1/2}$ transition of Cs.

Experiments: A pulsed dye laser, driven by the second harmonic of an Nd:YAG system, served as the pump source for these experiments. Generating ~ 4 – 5 ns FWHM pulses with energies as large as 6 mJ, the dye laser had a pulse repetition frequency and nominal linewidth of 10 Hz and ~ 0.2 cm^{-1} (~ 7 GHz), respectively, and was tunable over the ~ 819 – 862 nm spectral interval with a mixture of two dyes. After entering the laser cavity by means of a polarising beam splitter, the pump radiation propagated through a quartz cell having a length and diameter of 6.0 and ~ 2.5 cm, respectively. The dye laser beam throughout the cell was slightly elliptical in cross-section with major and minor axis lengths of 7 and 5 mm, respectively. Equipped with Brewster angle windows, the cell contained a mixture of Cs vapour, 500 torr Ar, and 100 torr ethane. As in the experiments of [1–5], for example, ethane served the purpose of relaxing Cs $2^2P_{3/2}$ atoms into the lower-lying ($J = 1/2$) fine structure level of the $6p$ state. The optical cavity comprised two 3 m radius of curvature mirrors, separated by ~ 1 m. One was a high reflector ($R > 99\%$) and the other transmitted 10 or 50% at 894 nm.

Fig. 1 shows absorption spectra for the Cs/Ar/ethane mixture, recorded in the 820–900 nm wavelength interval at two temperatures (371 and 471 K) which correspond to Cs number densities of $1.3 \times 10^{13} \text{ cm}^{-3}$ and $1.7 \times 10^{15} \text{ cm}^{-3}$, respectively. Associated with the Cs D_2 line (852.1 nm in air) is a blue satellite that peaks at ~ 837 nm [6] and has an approximate spectral width of 6 nm FWHM. Hedges *et al.* [7] showed in 1972 that such satellites are the result of optical transitions between two Cs–rare-gas interaction potentials that are parallel to one another in a specific region of internuclear separation. In the present situation, pumping the blue D_2 satellite of Fig. 1 photoexcites Cs–Ar collision pairs, moving along the ground ($X^2\Sigma^+_{1/2}$) interaction potential,

to the $B^2\Sigma^+_{1/2}$ state of the CsAr excimer that is dissociative and correlated with Cs ($6p^2P_{3/2}$) + Ar ($3p^6\ ^1S_0$) in the separated atom limit.

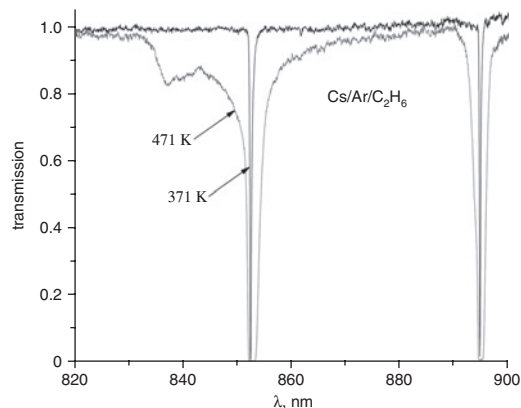


Fig. 1 Absorption spectra for Cs/500 torr Ar/100 torr ethane mixtures at 371 and 471 K, which correspond to Cs number densities of $1.3 \times 10^{13} \text{ cm}^{-3}$ and $1.7 \times 10^{15} \text{ cm}^{-3}$, respectively

Lasing at 894.3 nm is observed when the pump dye laser is tuned through the blue satellite of Fig. 1, and Fig. 2 is an excitation spectrum obtained by recording the relative 894.3 nm laser pulse energy as the pump wavelength is scanned over the 830–858 nm region. Throughout the data acquisition process, the energy of each pulse of the Cs atomic laser was normalised to that for the pump dye laser and the output coupling of the optical cavity was fixed at 10%. Note in Fig. 2 that the excitation spectrum in the region of the blue sideband has a width of ~ 3 nm FWHM and falls gradually on the red side of the peak at ~ 837 nm. In addition to the laser excitation spectra, the temporal history of the Cs laser pulses was recorded with a silicon avalanche photodiode (peak spectral response at ~ 850 nm) and a 1 GHz bandwidth oscilloscope. Representative pump and 894 nm laser pulse waveforms are shown in Fig. 3 by the black and grey curves, respectively. Recorded for a dye laser (pumping) wavelength of 837 nm, a cavity output coupling of 50%, and the cell temperature maintained at 423 K ($[Cs] = 2.2 \times 10^{14} \text{ cm}^{-3}$), the waveforms have been normalised to unity for convenience, and the Cs laser waveform illustrated in Fig. 3 is the average of 30 pulses. To minimise the influence of scattered pump radiation, baffling was installed in the optical system and a band-pass filter was inserted into the path of the photodiode when 894 nm laser waveforms were recorded. From Fig. 3, it is evident that peak 894 nm laser output occurs ~ 8 ns following maximum pump intensity and the temporal width of the Cs laser pulse is ~ 1.8 ns, or roughly one-half that of the 837 nm pump pulse. Note also that the zero-to-peak risetime of the Cs laser output is ~ 1.3 ns. Detailed measurements of the 894 nm laser power are in progress but experiments to date show output pulse energies of at least several microjoules and a weak dependence of the threshold pump pulse energy on cavity output coupling. The latter observation suggests a significant role for Dicke super-radiance or amplified spontaneous emission for the pumping intensities characteristic of these experiments.

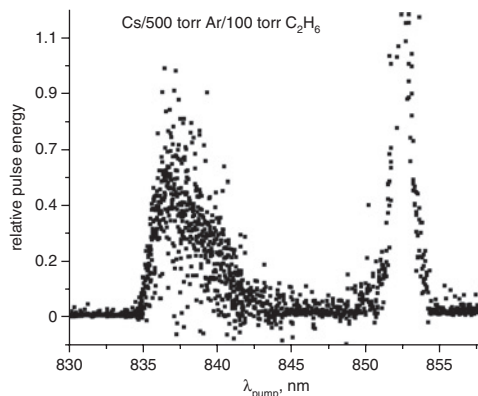


Fig. 2 Laser excitation spectrum, acquired at cell temperature of 423 K ($[Cs] = 2.2 \times 10^{14} \text{ cm}^{-3}$) by recording relative Cs 894.3 nm laser pulse energy as dye laser wavelength is scanned

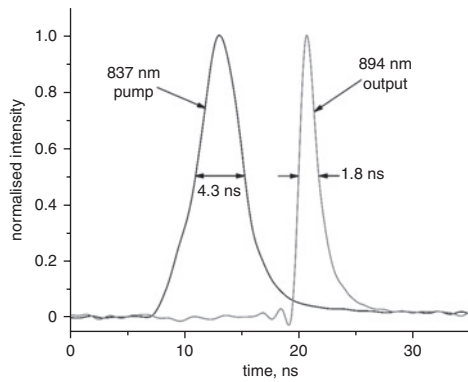


Fig. 3 Normalised pump (black curve) and Cs laser (grey curve) waveforms observed when photoexcitation wavelength is 837 nm

894 nm (Cs) laser waveform is average of 30 pulses. Note that delay in time between maximum dye laser pumping intensity and peak 894 nm laser output is ~8 ns, and risetime (zero-to-peak) of Cs laser pulse is 1.3 ns

Conclusions: An atomic Cs laser at 894.3 nm ($6p^2P_{1/2} \rightarrow 6s^2S_{1/2}$ transition) has been demonstrated by photoexciting the blue satellite, peaking at ~837 nm, that is associated with the $6p^2P_{3/2} \leftarrow 6s^2S_{1/2}$ (D_2) line of Cs in Cs–Ar mixtures. The spectral position and absorption coefficient for this (and all) alkali–rare-gas satellites are dependent on the identity of the rare gas (or other perturber), a characteristic that is advantageous for controlling the coupling of the pump to the gain medium in large volume lasers. The measured spectral breadth of the Cs–Ar blue satellite is fully compatible with the linewidths of high-power semiconductor lasers, thereby eliminating the necessity of dramatically reducing the diode pump bandwidth. Inserting alkali–rare-gas excimers into the pumping process as an intermediate, transient species preserves the well-known quantum efficiency for the alkali atomic lasers but also exploits the alkali–rare-gas interaction potentials so as to realise pumping bandwidths of several nanometres, as opposed to the tens of gigahertz linewidths available directly from the pressure-broadened D_2 line itself. It is clear that the excimer-based pumping approach reported here, yielding an excimer-pumped atomic laser

(which we denote as XPAL), will be of broad utility for all of the alkalis and other atoms as well.

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