

# Pumping of atomic alkali lasers by photoexcitation of a resonance line blue satellite and alkali-rare gas excimer dissociation

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Photoassociation of alkali-rare gas atomic collision pairs provides an alternative approach to optically pumping atomic alkali lasers. Lasing on the  $6^2P_{1/2} \rightarrow 6^2S_{1/2}$  ( $D_1$ ) transition of Cs has been observed when the blue satellite of the  $6^2P_{3/2} \leftarrow 6^2S_{1/2}$  ( $D_2$ ) transition, peaking at  $\sim 837$  nm for mixtures of Cs vapor, Ar, and ethane, is pumped by a pulsed dye laser. For 50% output coupling, laser threshold with respect to absorbed pump energy is  $\sim 40$   $\mu\text{J}$  at  $T=435$  K and the slope efficiency approaches 10%. The measured spectral breadth for the blue satellite ( $\geq 5$  nm at 410 K) will accommodate cw pumping of the Cs laser with conventional semiconductor lasers. © 2009 American Institute of Physics. [DOI: 10.1063/1.3151854]

Spectral satellites associated with resonance transitions of the atomic alkalis were observed in alkali-rare gas mixtures early in the last century.<sup>1</sup> Their origin remained a subject of debate until 1972 when Hedges *et al.*<sup>2</sup> interpreted far wing line broadening, and the blue satellite of the  $n^2P_{3/2} \leftarrow n^2S_{1/2}$  alkali absorption lines ( $n \geq 6$  for Cs), in terms of optical transitions between electronic states of the alkali-rare gas excimer. The reduced absorption coefficient  $\alpha(\lambda)$ , expressed in units of  $\text{cm}^5$ , was described by the relation

$$\alpha(\lambda) \propto A(R) \left| \frac{d(V_B - V_X)}{dR} \right|^{-1}, \quad (1)$$

where the Einstein  $A$  coefficient is a weak function of the interatomic separation  $R$ , and the upper and lower diatomic state potentials  $V_B(R)$  and  $V_X(R)$ , respectively, are related to the absorption wavelength  $\lambda$  by the Franck–Condon relation,

$$V_B(R) - V_X(R) = \frac{hc}{\lambda}. \quad (2)$$

Consequently, blue (and red) satellites are attributed to a pair of alkali-rare gas interatomic potentials that are essentially parallel (i.e.,  $d/dR [V_B(R) - V_X(R)] \rightarrow 0$ ) within a specific region of internuclear separation. As illustrated in Fig. 1, for example, dipole-allowed transitions of Cs–Ar collision pairs from the thermal and vibrational continua of the ground state ( $X^2\Sigma_{1/2}^+$ ) to the dissociative  $B^2\Sigma_{1/2}^+$  potential of CsAr (correlated with  $\text{Cs}(6p^2P_{3/2}) + \text{Ar}(3p^6^1S_0)$  in the separated atom limit)<sup>3</sup> give rise to a satellite lying to the short wavelength side of the Cs  $6p^2P_{3/2} \leftarrow 6s^2S_{1/2}$  ( $D_2$ ) transition.<sup>4</sup> The position and spectral profile of the satellite specify the window in internuclear separation in which the transitions occur.

In this letter, blue satellites of the alkali principal series are shown to provide a pathway for optically pumping atomic lasers on either the  $D_1$  ( $np^2P_{1/2} \leftarrow ns^2S_{1/2}$ ) or  $D_2$  ( $np^2P_{3/2} \leftarrow ns^2S_{1/2}$ ) transitions of the atom. Lasing on the  $D_1$  line of Cs at 894.3 nm has been observed<sup>5</sup> when the blue satellite of the  $D_2$  transition, peaking at  $\sim 837$  nm in Cs/Ar/

ethane mixtures, is pumped with a pulsed dye laser. Since 2003, several laboratories<sup>6–8</sup> have demonstrated lasing on the  $D_1$  lines of Cs, Rb, and K by exciting the  $D_2$  transition directly with Ti:sapphire or semiconductor diode lasers. This three level laser system is characterized by quantum efficiencies above 90% but requires pump bandwidths of typically 10 GHz or less. The laser excitation approach reported here relies on the photoassociation of ground state Cs–Ar collision pairs in the  $\sim 836 \text{ nm} \leq \lambda \leq 850 \text{ nm}$  spectral interval, thereby populating the  $B^2\Sigma_{1/2}^+$  state of the CsAr excimer. Rapid dissociation of the excited molecule generates Cs  $6p^2P_{3/2}$  atoms, and at no time during the pumping process is the Cs–Ar complex in a bound state. Inserting the transient alkali-rare gas species into the pumping scheme serves primarily to shift and broaden the absorption spectrum so that the linewidths of conventional, high power semiconductor lasers can now be accommodated directly. Another benefit of this excitation scheme is the ability to specify, through the choice of rare gas perturber and the alkali/rare

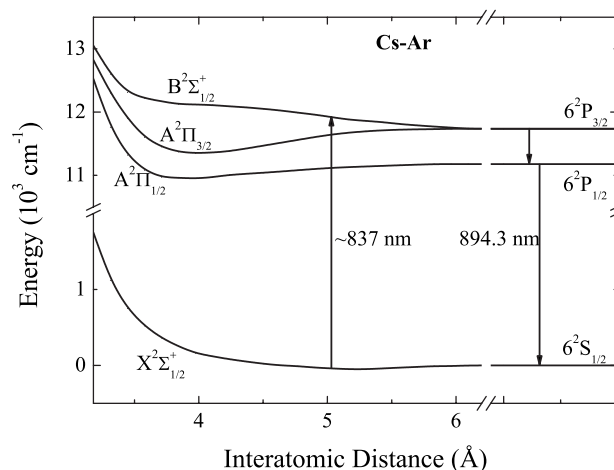


FIG. 1. Interaction potentials for the ground and lowest excited states of the CsAr molecule (after Pascale and Vandeplanque, Ref. 3). Also illustrated is photopumping of the Cs( $6^2P_{3/2} \leftarrow 6^2S_{1/2}$ ) blue satellite, peaking at  $\sim 837$  nm in Cs–Ar mixtures, so as to excite Cs–Ar collision pairs to the dissociative  $B^2\Sigma_{1/2}^+$  state of the CsAr excimer.

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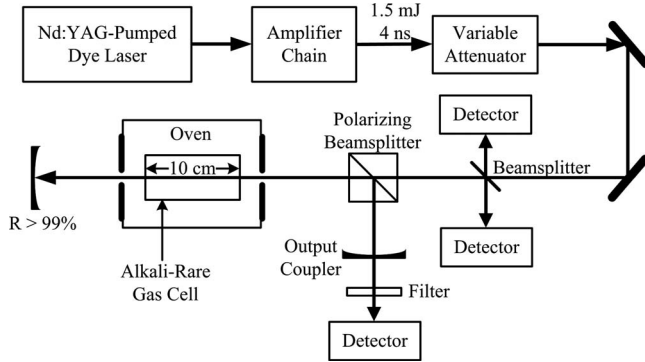


FIG. 2. Schematic diagram of the experimental arrangement for demonstrating lasing on the  $D_1$  line of Cs.

gas number densities, the peak absorption coefficient and the bandwidth of the blue satellite. Both are distinct assets when scaling the power and volume of alkali atomic laser systems.

A schematic diagram of the experimental arrangement for demonstrating the Cs 894.3 nm laser is given in Fig. 2. Single pulses having an energy and spectral linewidth of  $\leq 1.5$  mJ and  $0.2$   $\text{cm}^{-1}$  ( $\sim 7$  GHz), respectively, are generated by a neodymium-doped yttrium aluminum garnet (Nd:YAG)/dye laser system at a repetition frequency of 10 Hz. Mixing two laser dyes permits continuous tuning over the  $\sim 819$ – $856$  nm spectral range. A polarizing beamsplitter couples the pump beam into the optical cavity, which consists of two 3 m radius of curvature mirrors—one high reflector and an output coupler having a transmission of 50% at 894 nm. The optical cell has a diameter and length of 2.5 and 10 cm, respectively, and is equipped with plane parallel windows. Throughout the cell, the pump beam cross-section was slightly elliptical (major and minor axis lengths of 7 and 5 mm, respectively). For all of the experiments described here, the gas/vapor mixture in the cell comprised natural abundance Cs (saturated vapor), 100 Torr of ethane, and 500 Torr of Ar. As described previously,<sup>5–7</sup> ethane serves the purpose of collisionally relaxing a portion of the Cs  $6p^2P_{3/2}$  population into the lower-lying fine structure level,  $6p^2P_{1/2}$ .

When tuning the pump dye laser through the blue satellite of the  $D_2$  line in Cs–Ar– $\text{C}_2\text{H}_6$  mixtures, lasing on the  $D_1$  transition of Cs is observed readily and Fig. 3 presents laser excitation spectra obtained at three values of the optical cell temperature (410, 434, and 450 K), which correspond to Cs number densities of  $[\text{Cs}] = 1.2 \times 10^{14}$   $\text{cm}^{-3}$ ,  $3.6 \times 10^{14}$   $\text{cm}^{-3}$ , and  $7.2 \times 10^{14}$   $\text{cm}^{-3}$ , respectively. Obtained by recording the relative  $D_1$  laser intensity as the pump wavelength ( $\lambda_p$ ) was scanned, the excitation spectra show clearly that lasing is, indeed, attributable to photopumping of Cs–Ar collision pairs. Over the entire range of alkali number densities investigated in these experiments, the laser excitation spectra retain virtually all of the prominent characteristics of the blue satellite observed in absorption (illustrated in Fig. 3 for  $T = 471$  K). These include the steep slope between  $\sim 834$  and 836 nm, the local maximum at  $\sim 837$  nm, and a distinct, gradually rising plateau extending from  $\sim 842.5$  to  $\sim 850$  nm. Note that the spectral breadth of the satellite is  $\geq 5$  nm, and the rise in the laser intensity with increasing temperature for  $835$  nm  $\leq \lambda_p \leq 848$  nm is approximately linear in  $[\text{Cs}]$ .

The  $\lambda_p \sim 852$  nm portion of the excitation spectra of Fig. 3 reflects lasing at 894.3 nm driven by direct photoex-

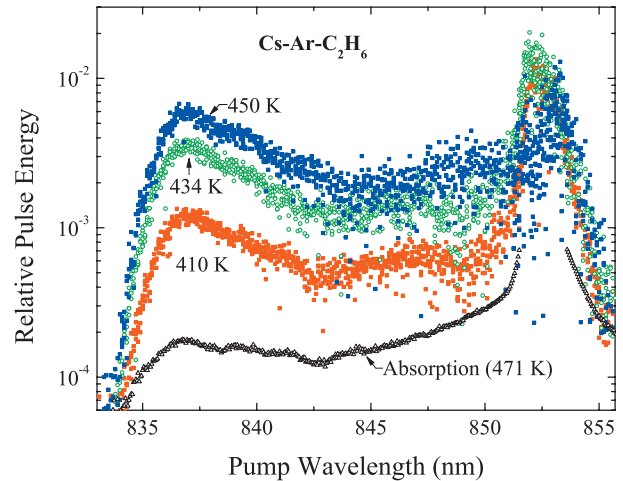


FIG. 3. (Color) Excitation spectra acquired by recording the 894.3 nm relative laser intensity as the pump laser wavelength was scanned over the  $\sim 832$ – $856$  nm interval. Data were obtained for three cell temperatures [410 K (red), 434 K (green), and 450 K (blue)], corresponding to  $[\text{Cs}] = 1.2 \times 10^{14}$   $\text{cm}^{-3}$ ,  $3.6 \times 10^{14}$   $\text{cm}^{-3}$ , and  $7.2 \times 10^{14}$   $\text{cm}^{-3}$ , respectively. The absorption spectrum recorded at 471 K over the same wavelength interval is also given but, for the sake of clarity, data in the  $\sim 851.5$ – $853.0$  nm region are not shown. Note that the ordinate is logarithmic.

citation of the Cs  $D_2$  transition. Laser output energy grows more slowly here than does the satellite ( $834$  nm  $\leq \lambda_p \leq 843$  nm when  $[\text{Cs}]$  is increased from  $1.2 \times 10^{14}$  to  $7.2 \times 10^{14}$   $\text{cm}^{-3}$ ). Indeed, the onset of collapse of the 450 K excitation spectrum (blue circles) in the vicinity of the  $D_2$  line suggests the influence of superradiance, parasitic oscillations (owing to the plane parallel windows of the Pyrex cell), and multiphoton ionization of the Cs vapor when pumping on resonance. With respect to the latter, tuning onto the  $D_2$  line resonantly enhances the three photon ionization cross-section. Since the oscillator strengths for the first  $n^2P_{1/2,3/2} \leftrightarrow ^2S_{1/2}$  doublet in all of the alkalis are known to be large ( $f \sim 1$ ),<sup>9</sup> this three photon process is presumably saturated for the peak pump intensities characteristic of these experiments. Before leaving Fig. 3, it should be mentioned that the shoulder evident on the red side of the  $D_2$  line suggests that the 894.3 laser can also be pumped by photoexciting Cs–Ar pairs to large R portions of the shallow  $A^2\Pi_{3/2}$  state of CsAr.

Figure 4 compares measurements of the optical conversion efficiency for the Cs 894.3 nm laser when pumped at 836.7 nm (blue satellite) or excited directly on the  $D_2$  line at 852.1 nm. Both sets of experiments, conducted under the same conditions with the cell temperature fixed at  $435 \pm 2$  K, entailed recording the dependence of the laser output energy  $E_o$  (monitored by calibrated pyroelectric detectors) on the absorbed pump pulse energy  $E_a$ . Considerable care was devoted to ensuring the calibration and linearity of the detectors, as well as determining accurately the absorbed pump pulse energy in the double pass arrangement of Fig. 2. Given the available dye laser pulse energy, the limit for the single pulse energy absorbed in two passes by the Cs/Ar/ethane mixture at  $\lambda_p = 836.7$  nm was found to be  $\sim 160$   $\mu\text{J}$ . From the data of Fig. 4, the slope efficiency for the excimer-pumped Cs atomic laser approaches 10% and its threshold pulse energy is  $\sim 40$   $\mu\text{J}$ . Both values are virtually identical to those for the three level,  $D_2$  line-pumped system. We conclude that, on a per photon absorbed basis, the laser thresh-

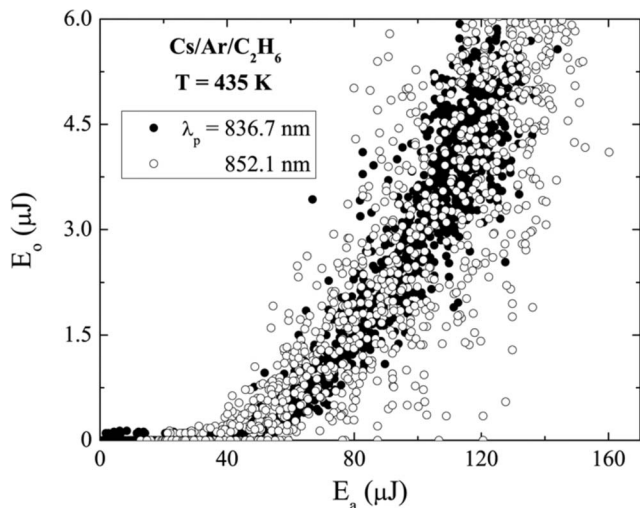


FIG. 4. Dependence of the 894.3 nm laser pulse energy ( $E_o$ ) on the absorbed pump energy ( $E_a$ ) for dye laser excitation wavelengths of 836.7 (●) and 852.1 nm (○). All data were recorded at a temperature of  $435 \pm 2$  K, which corresponds to  $[Cs] = (3.8 \pm 0.4) \times 10^{14} \text{ cm}^{-3}$ .

old is identical when pumping the CsAr excimer or photoexciting the Cs atom directly. With the ultimate performance of cw, diode-pumped alkali lasers in mind, the critical point to be made is that populating the lowest  $^2P_{1/2,3/2}$  states of the alkalis by dissociating alkali-rare gas diatomics yields slope efficiencies that are also the same (to within experimental uncertainty) as those associated with direct photopumping of the alkali  $^2P_{3/2}$  level.

In summary, lasing on the  $D_1$  transition of Cs has been realized by photoexciting the blue satellite of the  $D_2$  line, peaking at  $\sim 837$  nm in Cs/Ar/ethane mixtures. Furthermore, the slope efficiency and threshold pump pulse energy for the excimer-based pumping process are the same as those measured (with respect to the absorbed pump energy) for the conventional atomic three level scheme.<sup>6,7</sup> Satellites similar to those of interest here have also been observed in other atoms (Tl, In, and the alkaline earths, for example)<sup>10-12</sup> when

perturbed by the rare gases, suggesting that a family of efficient cw and pulsed lasers pumped in this manner is feasible. It is also anticipated that lasing on the  $D_2$  line of Cs at 852.1 nm will be realized in the near future.

The laser pumping mechanism reported here is intriguing because a transient diatomic molecule is the intermediate species in populating the upper state of an atomic laser. The blue satellite associated with an alkali-rare gas pair specifies a Franck-Condon region that is the vehicle for accessing excited states of the alkali while bypassing the constraints imposed by photopumping a resonance line of the atom. The spectra of Fig. 3 demonstrate that the bandwidth of at least the  $n=6$  satellite for Cs-Ar (and other alkali-rare gas pairs as well) is more than adequate to accommodate the linewidths of conventional, high power diode arrays.

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<sup>1</sup>S.-Y. Ch'en and M. Takeo, *Rev. Mod. Phys.* **29**, 20 (1957).

<sup>2</sup>R. E. M. Hedges, D. L. Drummond, and A. Gallagher, *Phys. Rev. A* **6**, 1519 (1972).

<sup>3</sup>J. Pascale and J. Vandeplanque, *J. Chem. Phys.* **60**, 2278 (1974).

<sup>4</sup>C. L. Chen and A. V. Phelps, *Phys. Rev. A* **7**, 470 (1973).

<sup>5</sup>J. D. Readle, C. J. Wagner, J. T. Verdeyen, D. L. Carroll, and J. G. Eden, *Electron. Lett.* **44**, 1466 (2008).

<sup>6</sup>W. F. Krupke, R. J. Beach, V. K. Kanz, and S. A. Payne, *Opt. Lett.* **28**, 2336 (2003).

<sup>7</sup>B. V. Zhdanov, T. Ehrenreich, and R. J. Knize, *Opt. Commun.* **260**, 696 (2006).

<sup>8</sup>Y. Wang, T. Kasamatsu, Y. Zheng, H. Miyajima, H. Fukuoka, S. Matsuoka, M. Niigaki, H. Kubomura, T. Hiruma, and H. Kan, *Appl. Phys. Lett.* **88**, 141112 (2006).

<sup>9</sup>R. B. Miles and S. E. Harris, *IEEE J. Quantum Electron.* **QE-9**, 470 (1973).

<sup>10</sup>S. Y. Ch'en and A. T. Lonseth, *Phys. Rev. A* **3**, 946 (1971).

<sup>11</sup>S. Y. Ch'en, M. R. Atwood, and T. H. Warnock, *Physica (Amsterdam)* **27**, 1170 (1961).

<sup>12</sup>D. E. Johnson and J. G. Eden, *J. Chem. Phys.* **82**, 2927 (1985).