Four level, atomic Cs laser at 852.1 nm with a quantum efficiency above 98%: Observation of three body photoassociation

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Lasing on the D2 (6p 2P3/2 → 6s 2S1/2) transition of atomic Cs at 852.1 nm has been observed with a four level system in which the Cs 6p 2P3/2 state is pumped by the photoassociation and subsequent dissociation of Cs-rare gas collision pairs. Characterized by a quantum efficiency >98%, this laser requires no atomic precursor to the upper laser level and provides oscillation on an alkali transition inaccessible to three level, photopumped alkali laser systems. Measurements of photoabsorption and pump energy threshold in Cs–Ar–Kr mixtures reveal the influence of three body photoassociation.

Quantum efficiency is one measure of the ultimate performance and utility of a laser. In this regard, the three level optically pumped alkali lasers (Cs, Rb, K) (Refs. 1–3) are distinctive in that direct photoexcitation of the lowest np 2P3/2 state (n=4–6 for K, Rb, and Cs, respectively) and subsequent lasing on the D1 (n2P1/2→n2S1/2) transition yield quantum efficiencies above 90%, limited only by the alkali np 2P3/2→np 2P1/2 fine structure splitting. Although this energy defect is sufficiently large in Cs, for example, to restrict the D1 laser quantum efficiency to 95%, a greater drawback of the three level alkali system is the requirement for a relaxant species to collisionally transfer the 2P3/2 population into the 2P1/2 state without quenching, to a significant degree, the pumped 2P3/2 level. Typically a hydrocarbon such as C2H6, the relaxant presents long-term reliability and laser medium optical quality concerns owing to gradual pyrolysis of the relaxant which, in addition to the depletion of the molecule, results in the formation of alkali-hydride particulates and carbon deposits.

This letter reports the observation of lasing on the D2 transition (6p 2P3/2 → 6s 2P1/2) of Cs at 852.1 nm when thermal Cs–rare gas (Ar, Kr) atomic collision pairs are photoassociated in a broad (~3 nm full width at half maximum) wavelength region centered at ~837 nm. Pumped by the dissociation of Cs–rare gas diatomic excimer molecules in the repulsive B 2Σ1/2 state, this four level laser has a quantum efficiency >98% because the lasing transition originates from the atomic state populated by the pump through Cs–rare gas molecular dissociation, terminates on ground, and the Cs–rare gas interaction is weak. It should be emphasized that lasing on the D2 transition of the alkali atoms is not allowed with the three level atomic scheme employed previously.1–3 Oscillation on the Cs D2 transition demonstrates vividly that this Cs–rare gas laser system must be treated on a four level basis, despite the short lifetimes of the intermediate species (the alkali-rare gas complex) in its ground and excited states. Furthermore, the photoassociation of ternary Cs–Ar–Kr ensembles has been observed. Having an 852.1 nm laser excitation spectrum virtually identical to that for Cs–Ar pairs, the photoassociation of Cs–Ar–Kr complexes lowers the pump energy threshold by >25%.

In the semiclassical view, the photoassociation of Cs–rare gas pairs in the thermal continuum of the X 2Σ1/2 ground state interaction potential occurs predominantly in the region of intermolecular separation in which the B 2Σ1/2–X 2Σ1/2 difference potential varies slowly. Such free-free optical transitions of the binary Cs–rare gas complex define the blue satellite of the D2 transition and provide a vehicle by which the atomic laser upper level can be accessed directly with a broadband optical source. Although a similar process sequence has proven successful in demonstrating lasing on the D1 line of Cs (894.3 nm) in Cs–Ar–C2H6 and Cs–Kr–C2H6 mixtures,4–6 the experiments described here demonstrate that neither the Cs 6p 2P1/2 intermediate state nor an auxiliary species in the gas phase, such as the relaxant C2H6, is necessary.

Pump/lasing cycles sharing several characteristics of the scheme described here have been reported previously. In 1979, Chilukuri7 observed super-radiance on the green 2Σ1/2 → 2P3/2 transition (535.1 nm) of atomic thallium in Tl–Hg vapor mixtures when pumping several nm to the red of the 377.6 nm resonance transition. Subsequently, Atamas et al.8 photoexcited Tl 3.9 nm into the violet wing of the 377.6 nm line, pressure-broadened by He. In both sets of experiments, however, the lasing transition terminated on the 2P1/2 metastable atomic state lying ~1 eV above ground.

The experimental arrangement and data acquisition procedures for these studies are similar to those described previously4,5 and will only be reviewed briefly here. A pulsed dye laser, tunable over the ~819–865 nm region and driven by the second harmonic of an Nd:YAG laser system, served as the optical pump source. After two stages of amplification, the pump pulses have an energy ~2 mJ and a temporal width of ~4 ns. The optical cavity is an L-configuration that provides for two passes of the pump beam through the gain medium, and comprises a high reflector (>97% reflectivity at 852 nm) and a ~50% output coupler. Separated by ~80 cm, the mirrors have a radius of curvature of 3 m. Natural abundance Cs and either Ar at a number density of ~1.6 × 1019 cm−3 (500 Torr at room temperature) or an Ar/Kr mixture at the same total pressure were contained in a

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quantum efficiency ($\eta$) of the D$_2$ laser for a given pump wavelength, shows that $\eta>98\%$ for all $\lambda_p > 835$ nm.

Measurements of the variation in the 852 nm laser pulse energy with the pump energy absorbed by the gain medium ($E_a$) are summarized in Fig. 2 for the same optical cell temperatures as those of Fig. 1. Note that the threshold absorbed pump pulse energy for [Cs]$=1.3 \times 10^{15}$ cm$^{-3}$ (464 K) of $\sim 130$ $\mu$J is quite close to the corresponding value recorded for the D$_1$ laser (894.3 nm) operating in Cs–Kr–C$_2$H$_6$ mixtures at a comparable temperature [$E_a$(threshold)=$140$ $\mu$J at 468 K]. This result is somewhat surprising because the degeneracy (2J+1) for the 6$^3$P$_{3/2}$ upper laser level in the present experiments is twice that for the D$_1$ laser, thus suggesting a higher pump energy threshold for the 852.1 nm laser. Nevertheless, the threshold pump energies for the three sets of data in Fig. 2—130 $\mu$J, 175 $\mu$J, and 250 $\mu$J for 464 K, 475 K, and 485 K, respectively—vary linearly with [Cs] as one would expect. It is also evident that, for a fixed value of [Cs], the slope efficiencies of Fig. 2 increase with $E_a$.

Unexpected behavior is observed when experiments identical to those described above were conducted with Cs–Ar–Kr mixtures in which [Ar]=[$Kr]=8 \times 10^{18}$ cm$^{-3}$. Motivated by the recent observation$^5$ that Cs–Kr pair absorption spectra broader than those available with a single rare gas. Spectral engineering of the alkali-rare gas pump band by the superposition of binary alkali-rare gas absorption profiles might well lead to pump acceptance bandwidths $>5$–6 nm. However, as illustrated in Fig. 3 for a constant [Cs]$=1.3 \times 10^{15}$ cm$^{-3}$ (464 ± 2 K), the D$_2$ laser excitation spectrum for Cs–Ar–Kr mixtures (open circles: ○) is indistinguishable from that for Cs–Ar (solid triangles: △). The maximum for both spectra lies at $\sim 836.7$ nm and the Cs–Kr peak at 841.1 nm is extremely weak. Further insight into the dynamics of the D$_2$ laser in multi-rare gas environments is provided by measurements of the 852 nm laser output energy. Panel (a) of Fig. 4 presents data acquired for both Cs–Ar and Cs–Ar–Kr mixtures at 464 ± 2 K. In obtaining...
these results, the laser was photoexcited at 836.7 nm and the output pulse energy ($E_o$) was recorded for incident pulse energies up to ~2.3 mJ. It is clear from Fig. 4(a) that the threshold pump pulse energy for the ternary Cs–Ar–Kr mixture is ~600 µJ, or ~28% lower than that for Cs–Ar alone (~830 µJ). As illustrated in Fig. 4(b), however, the two and three component alkali-rare gas mixtures exhibit laser thresholds with respect to the absorbed pump pulse energy that are, to within experimental uncertainty, identical.

Several conclusions can be drawn from Figs. 3 and 4. Although Cs–Ar–Kr mixtures reduce the D$_2$ laser pump energy threshold significantly (relative to that for the binary Cs–Ar mixture), the partial substitution of Kr for Ar has no discernible effect on the laser excitation spectral profile. It is evident, therefore, that Kr assists the photoassociation of Cs–Ar pairs because it is the Cs–Ar interaction potentials that dominate the excitation spectrum. These considerations suggest that three body photoassociation of Cs–Ar–Kr atomic ensembles has been observed and is responsible for reducing the pump energy threshold by ~25%.

In summary, lasing on the 852.1 nm (D$_2$) transition of Cs has been observed when Cs–Ar and Cs–Ar–Kr mixtures are photoexcited through the blue satellite of the transition. Lasing at this wavelength is not an option with previous three level laser schemes based upon pumping the atomic species directly, and the quantum efficiency for this four level system is ~98%. Comparisons of laser excitation spectra and output pulse energy data for Cs–Ar and Cs–Ar–Kr mixtures suggest that three body photoassociation of the Cs–Ar–Kr collision complex has been observed and this process is effective in accessing the Cs (2P$_{3/2}$) upper laser level. The introduction of Kr to the laser mixture does not influence noticeably the laser excitation spectrum, indicating that Kr increases the reduced absorption coefficient at the pump wavelength by ~5–10% but plays a role subordinate to Ar with respect to the interaction potentials shaping the excitation spectrum.

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