

Blue five-level frequency-upconversion system in rubidium

T. Meijer, J. D. White,* B. Smeets, M. Jeppesen, and R. E. Scholten

School of Physics, University of Melbourne, Victoria 3010, Australia

Received November 28, 2005; accepted December 22, 2005; posted January 12, 2006 (Doc. ID 66262)

We demonstrate production of continuous coherent blue laser light by using a five-level system in rubidium vapor. Two low-power lasers, at 780 and 776 nm, induce strong atomic coherence in the $5S$ – $5P$ – $5D$ states. The atoms decay to the $6P$ excited state, from which stimulated emission produces a coherent blue (420 nm) beam. We have coupled both ground-state hyperfine levels, effecting coherence between four levels. The coherent blue output is enhanced by several mechanisms, including stronger coupling to a larger fraction of the atomic population, operation at a detuning such that the vapor is nominally transparent to the 780 nm pump field, reduced losses owing to optical pumping, and optimal phase matching. We report experimental findings and compare them with results from a semiclassical Maxwell–Bloch model. © 2006 Optical Society of America

OCIS codes: 270.1670, 190.7220.

Nonlinear interactions of light and atoms can be enhanced dramatically through control of the quantum state of the atoms, in particular, through coherent superposition of the atomic eigenstates.¹ Atomic coherence effects, including coherent population trapping, electromagnetically induced transparency,² and lasing without inversion,³ provide a remarkably rich playground for nonlinear optics, leading to a profusion of developments in recent years, e.g., in spectroscopy,⁴ atomic frequency standards,⁵ quantum information processing and memory,⁶ controlling chemical reactions,⁷ magnetometry and other sensing,¹ and both slow and fast light-pulse propagation.⁸ One of the most important applications of conventional nonlinear optics is frequency conversion, and atomic coherence can allow substantial enhancements, for example, in four-wave mixing⁹ and high-harmonic generation,¹⁰ to be made. Here we investigate a specific frequency-upconversion process that uses a rubidium two-color cascade system.

Continuous coherent blue laser light was produced in ^{85}Rb vapor by a five-level scheme (see Fig. 1). Two low-power (20 mW) lasers, at 780 and 776 nm, induce strong atomic coherence in the $5S$ – $5P$ – $5D$ states. The atoms decay via the $5D_{5/2}$ – $6P_{3/2}$ (5.5 μm) and $6P_{3/2}$ – $5S_{1/2}$ (420 nm) transitions. The 420 nm radiation is amplified by coupling to the $5S$ – $6P$ transition, where coherent coupling of the $5S$ ground states with the $5P$ and $5D$ states effectively reduces the $5S$ population.

Our scheme extends earlier demonstrations of frequency upconversion in Rb (Ref. 11) by coupling both ground-state hyperfine levels ($F=2, 3$) to the $5P$ and $5D$ states, inducing coherence among four levels. Substantially higher coherent blue output was obtained, up to $P_{420}=40 \mu\text{W}$. With both hyperfine ground states involved, we have stronger coupling to a larger fraction of the atomic population. The coupling is obtained for large detunings of the 780 nm pump laser, such that single-photon absorption is small. Since both hyperfine levels are coupled, optical pumping into a dark hyperfine level is minimized. Fi-

nally, cancellation of the optical phase shifts for the two ground states optimizes phase matching for the upconversion process at the appropriate detuning. Measurements of the blue output power and detuning with variations in frequency of the driving lasers were obtained and are compared with the results from a semiclassical Maxwell–Bloch model including four, five, and sixteen levels.

Figure 1 shows a schematic of the experimental arrangement, and the relevant energy levels of rubidium. Two low-power infrared laser¹² beams were copropagated through Rb vapor cell A ($T=100$ – 200°C ; length, 8 cm), with natural isotopic abundance of Rb and no buffer gas. The power in each laser beam was ~ 20 mW, focused to a beam waist of 0.3 mm diameter with an $f=50$ cm lens.

With the lasers tuned to the two-step excitation resonance, strong blue fluorescence was observed in vapor cell A, indicating significant excitation to the upper $5D$ level. Atoms can decay via the $6P$ intermediate state and then to the ground state, emitting blue fluorescence.

The 780 nm laser detuning δ_{780} was determined by saturated-absorption spectroscopy in a separate vapor cell (not shown), relative to the $5S F=3 \rightarrow 5P F=4$ resonance. The 776 nm laser frequency was found from the blue fluorescence spectrum in Rb vapor cell

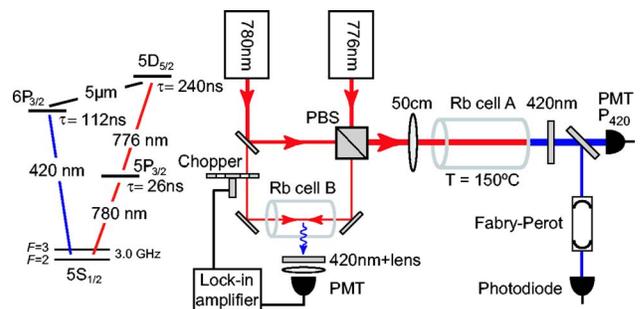


Fig. 1. (Color online) Experimental arrangement. Two lasers (780 and 776 nm) excite ^{85}Rb on the $5S$ – $5P$ – $5D$ transitions, and coherent 420 nm radiation is emitted. PBS, polarizing beam splitter; PMT, photomultiplier tube.

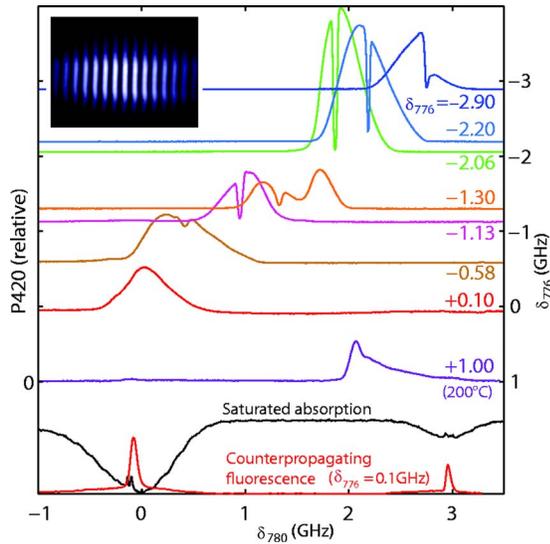


Fig. 2. (Color online) Blue output power for several pump laser detunings at $T \approx 150^\circ\text{C}$. The maximum output was observed when the 780 and 776 nm input fields were orthogonally polarized, with $\delta_{780} \approx 2$ GHz. The two lowest curves show frequency reference spectra, and the inset shows two-slit diffraction (slit separation, 0.25 mm; width 0.04 mm) from the 420 nm beam.

B (70°C), with counterpropagating 780 and 776 nm light beams. We define the net detuning of both lasers, $\delta_{780} + \delta_{776} = 0$, when the two-step excitation is exactly on the $5SF=3 \rightarrow 5D$ resonance, indicated by the blue fluorescence intensity.

Blue fluorescence was observed in primary vapor cell A for a broad range of conditions. For specific detunings of the two lasers, a bright blue beam was produced, exiting the cell in the direction of propagation of the two input pump beams. The output was linearly polarized and was both spatially and temporally coherent, as verified with two-slit diffraction, which showed high-contrast interference fringes (Fig. 2, inset), and from the linewidth measured with a confocal Fabry–Perot etalon.

Figure 2 shows relative output power P_{420} of the coherent beam as a function of the 780 nm laser detuning. Output was observed near each of the single-photon resonances of the two ground-state hyperfine levels, which are separated by 3.0 GHz, provided that the 776 nm laser was tuned such that the sum frequency of both IR lasers was close to the two-photon transition. That is, coherent blue output was generated when $\delta_{780} = \delta_{776} = 0$ ($F=3$ ground state), as demonstrated in Ref. 11 (maximum at $\approx 150^\circ\text{C}$), and also when $\delta_{780} = +3$ GHz, $\delta_{776} = 0$ ($F=2$ ground state) at a higher temperature (200°C ; not shown).

We also observed much stronger output when the 780 nm laser was detuned far from either single-level resonance; that is, between the two ground-state hyperfine levels. At $\delta_{780} \approx 2$ GHz and $T \approx 200^\circ\text{C}$ we found a maximum of approximately $40 \mu\text{W}$, three times greater than observed previously¹¹ for similar input power.

Using the Fabry–Perot etalon (1.5 GHz free spectral range), we investigated the spectral properties of

the blue output for several specific detunings (Fig. 3). Spectra A and B have net detunings $\delta_{780} + \delta_{776} = 0$, for which we expect blue output to the $F=3$ ground state, whereas spectra C and D have $\delta_{780} + \delta_{776} = 3$ GHz (i.e., $F=2$). Structure that is due to the $6P$ hyperfine splittings [$F=4, 3, 2, 1$; separations 40, 20, and 10 MHz (Ref. 13)] is evident from the double peaks in spectra A and B and the broad peaks of C and D, which suggest overlap of several lines of similar strength.

The apparent frequency shifts of the spectra are modulo the 1.5 GHz free-spectral range of the etalon (and no correction has been made for drifts of up to a few hundred megahertz that were due to temperature and pressure variations in the etalon). Curve B is shifted by $|\delta\nu \bmod 1.5 \text{ GHz}| = 1$ GHz, which is also consistent with a shift of, for example, -0.5 or -2 GHz. The large shifts of nonresonant spectra B and C scale approximately with the square root of the 780 nm pump power and are attributed to ac Stark shifts of the dressed ground-state level.

We have modeled the system by using semiclassical Maxwell–Bloch approach, with a slowly varying envelope and phase approximations.^{2,3,14} The four plane-wave classical fields are described by Maxwell’s wave equation that links electric field \mathbf{E} to induced polarization \mathbf{P} , where \mathbf{P} is given by the appropriate off-diagonal element of atomic density matrix ρ , and ρ is determined by solution of the Bloch equations. We have calculated the steady-state atomic density matrix with four, five, and sixteen levels. In the simplest case we included the four primary levels ($5S, 5P, 5D, 6P$). Both hyperfine ground states ($5S, F=2, 3$) were included in a five-level model; and all hyperfine levels of the four primary states, in the sixteen-level calculation. Magnetic substates were neglected in all cases.

The steady-state ρ was determined using the dipole approximation for the interaction Hamiltonian, which we quantify in terms of the Rabi frequency for a given transition between levels i and j , i.e., $(\mathcal{H}_{\text{int}})_{ij} \equiv \hbar \Omega_{ij}$. We transform the Hamiltonian to the interaction basis, make the rotating-wave approximation, and find steady-state solutions via standard linear algebraic matrix methods. The Rabi frequency of a propagating field grows with the appropriate off-diagonal density matrix element, $(\partial/\partial z)\Omega_{ij} \propto \text{Im}(\rho_{ij})$. We are particularly interested in gain on the 420 nm transitions, $\rho_{5S(F=2,3),6P}$.

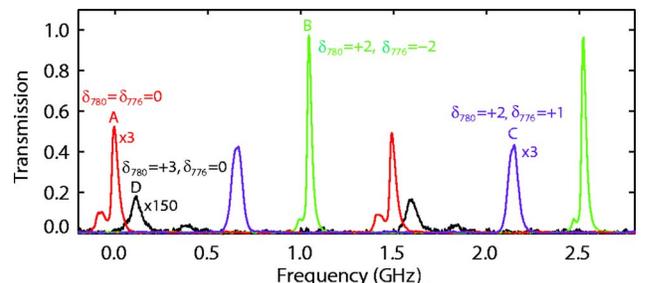


Fig. 3. (Color online) Blue output spectra from a Fabry–Perot etalon (1.5 GHz free spectral range) normalized to spectrum B.

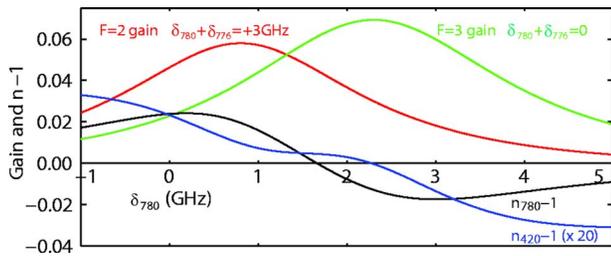


Fig. 4. (Color online) Relative gain and refractive-index dependence of blue transition on δ_{780} in the five-level model, calculated from $\text{Im}(\rho_{ij})$ and $\sum_F \text{Re}(\rho_{5S_{F,j}}) \equiv n-1$, respectively, where $F=2, 3$ and j is $6P$ (420 nm) or $5P$ (780 nm).

Figure 4 shows the relative 420 nm gain for the two hyperfine ground states in the five-level model, with Rabi frequencies applicable to our experimental conditions. The gain is greatest for detunings between the two ground states rather than when resonant with either. The maximum gain to the upper $F=3$ ground state is predicted for $\delta_{780}=+2.1$ GHz, which is consistent with measurements (Fig. 2). For the $F=2$ ground state, maximum gain is at $\delta_{780}=+0.8$ GHz, but the gain is also strong for $\delta_{780}, \delta_{776}=+2, +1$ GHz, corresponding to Fabry–Perot spectrum C (Fig. 3). Sixteen-level calculations including the $5P$ and $5D$ hyperfine levels also predict two gain regions at similar detunings, while in the four-level model there is only a single gain peak around $\delta_{780}=0$. Note that in the calculations the sum detuning is fixed, whereas in the experiments δ_{780} is varied for a given δ_{776} , thus showing Doppler-broadened peaks.

Phase matching depends on the refractive index, calculated from the real part of the off-diagonal density matrix elements. In the five-level model, these are small and slowly varying around 2 GHz, reducing phase mismatch for both the 780 nm pump and the 420 nm output fields (Fig. 4). The zero-crossing and maximum-gain frequencies are sensitive to the intensities of the pump laser fields. Since the latter are strongly varying radially, and also axially owing to absorption, these simple models provide only qualitative guidance.

In conclusion, our results show that significant enhancement of the frequency-upconversion process is possible with careful detuning of the pump lasers. With the 780 nm pump laser detuned between the hyperfine ground states, we couple equally both $F=2$ and $F=3$ population fractions of $5/12$ and $7/12$; the coupling also inherently minimizes losses owing to optical pumping between hyperfine states. Coupling both ground-state levels induces coherence among four levels, indicated by strong off-diagonal elements of the calculated density matrix.

Detuning the 780 nm laser well outside the single-photon Doppler width also increases transmission of the pump along the optically dense vapor, and operation at increased temperature is then possible. At the higher temperature (200°C), we observed that the length of the blue fluorescence along the cell was notably shorter for $\delta_{780} \approx 0$ than off resonance ($\delta_{780}=+2$ GHz). Off resonance, two-photon 780 and 776 nm absorption occurs for a greater length along the cell, at greater density, thus increasing the active population. Finally, phase mismatch is reduced for detuning between the ground-state hyperfine levels.

Further work is needed to develop a quantitative model incorporating atomic magnetic substates, the atomic velocity distribution, and spatial modes of the laser fields. While the problem is challenging, there are many interesting phenomena to explore, such as the apparent resonance in the blue light near optimum conditions, which suggests an electromagnetically induced transparency-like coherence effect, and the possibility of gain enhancement with an external cavity.

We thank A. Akulshin for helpful discussions. This work has been supported by the Australian Research Council and the Australian International Science Linkages program. R. E. Scholten's e-mail address is r.scholten@physics.unimelb.edu.au.

*Permanent address, Juniata College, Huntingdon, Pennsylvania 16652.

References

1. M. Fleischhauer, A. Imamoglu, and J. P. Marangos, *Rev. Mod. Phys.* **77**, 633 (2005).
2. M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge U. Press, 1997).
3. J. Mompert and R. Corbalán, *J. Opt. B Quantum Semiclassical Opt.* **2**, R7 (2000).
4. E. Arimondo, in *Progress in Optics*, E. Wolf, ed. (Elsevier, 1996), Vol. XXXV, pp. 259–354.
5. P. R. Hemmer, G. P. Ontai, and S. Ezekiel, *J. Opt. Soc. Am. B* **3**, 219 (1986).
6. C. H. van der Wal, M. D. Eisaman, A. André, R. L. Walsworth, D. F. Phillips, A. S. Zibrov, and M. D. Lukin, *Science* **301**, 196 (2003).
7. K. Bergmann, H. Theuer, and B. W. Shore, *Rev. Mod. Phys.* **70**, 1003 (1998).
8. L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, *Nature* **398**, 594 (1999).
9. S. E. Harris, *Phys. Rev. Lett.* **85**, 4032 (1990).
10. K. Hakuta, L. Marmet, and B. P. Stoicheff, *Phys. Rev. Lett.* **66**, 596 (1991).
11. A. S. Zibrov, M. D. Lukin, L. Hollberg, and M. O. Scully, *Phys. Rev. A* **65**, 051801R (2002).
12. C. J. Hawthorn, K. P. Weber, and R. E. Scholten, *Rev. Sci. Instrum.* **72**, 4477 (2001).
13. E. Arimondo, M. Inguscio, and P. Violino, *Rev. Mod. Phys.* **49**, 31 (1977).
14. R. E. Scholten, T. J. O'Kane, T. R. Mackin, T. A. Hunt, and P. M. Farrell, *Aust. J. Phys.* **52**, 493 (1999).