Blue five-level lasing-without-inversion system in rubidium

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We demonstrate continuous coherent blue laser light production using a five-level lasing-without-inversion scheme in rubidium vapor. Two low-power lasers at 780 nm and 776 nm induce a 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 a coherent state between four levels. The coherent blue output is enhanced through 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 due to optical pumping, and optimal phase-matching. We report experimental findings and compare with results from a semiclassical Maxwell-Bloch model. © 2005 Optical Society of America

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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 (EIT), and lasing without inversion (LWI),[2] provide a remarkably rich "playground" for nonlinear optics, leading to an explosion of developments in recent years, e.g. in spectroscopy,[3] atomic frequency standards,[4] quantum information processing and memory,[5] controlling chemical reactions,[6] magnetometry and other sensing,[1] and both slow and fast light pulse propagation.[7] 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 mixing[8] and high-harmonic generation.[9] Here we investigate a specific frequency upconversion process using the rubidium two-color cascade system.

We demonstrate continuous coherent blue laser light production using a five-level lasing-without-inversion scheme in ^{85}Rb vapor (see Fig. 1). Two low-power (20 mW) lasers at 780 nm and 776 nm induce a 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 5.5 μm radiation is amplified through conventional population inversion of the 6P–5D levels, and on the 420 nm transition through lasing-without-inversion; that is, because coherent coupling of the 5S ground-state with the 5P and 5D states effectively reduces the 5S population.

Our scheme extends earlier demonstrations of frequency upconversion in Rb,[10] by coupling both ground-state hyperfine levels ($F = 2, 3$) to the 5P and 5D states, effecting a four-level coherent state. Substantially higher coherent blue output was obtained, up to $P_{420} = 40\,\mu$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. Finally, 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 to the results from a semiclassical Maxwell-Bloch model including four, five, and sixteen levels.

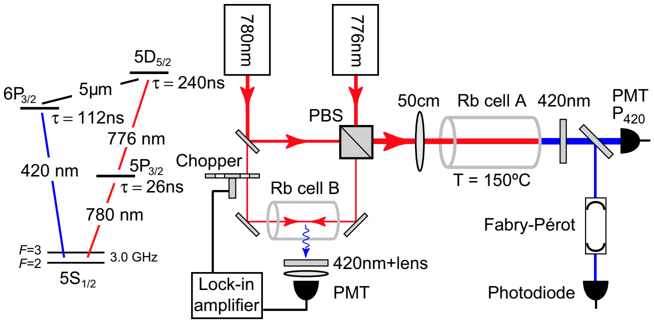

Fig. 1. (Color online) Experimental arrangement. Two lasers (780, 776 nm) excite ^{85}Rb on the 5S-5P-5D transitions, and coherent 420 nm radiation is emitted. PBS polarizing beam splitter, PMT photomultiplier tube.

Figure 1 shows a schematic of the experimental arrangement, and the relevant energy levels of rubidium. Two low-power infrared laser[11] beams were co-propagated through rubidium vapor cell A ($T = 100$ to 200°C, length 8 cm), with natural isotopic abundance of Rb and no buffer gas. The power in each laser beam was around 20 mW, focused to a beam waist of diameter 0.3 mm with a $f = 50$ cm lens.

With the lasers tuned to the two-step excitation resonance, strong blue fluorescence was observed in the primary 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 using 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 vapor cell B (70°C), with counter-propagating 780 nm and 776 nm light beams. We define the net detuning of both lasers $\delta_{780} + \delta_{776} \equiv 0$ when the two-step excitation is exactly on the 5S $F = 3 \rightarrow$ 5D resonance, indicated by the blue fluorescence intensity.

Blue fluorescence was observed in the primary vapor cell A for a very 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 both spatially and temporally coherent, verified with two-slit diffraction which showed high-contrast interference fringes (Fig. 2 inset), and from the linewidth measured with a confocal Fabry-Pérot (FP) etalon.

Figure 2 shows the 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 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. 10, or when $\delta_{780} = +3\,\text{GHz}, \delta_{776} = 0$ ($F = 2$ ground state), both maximum at temperature $T \approx 150°\text{C}$.

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 $\delta_{780} \approx 2\,\text{GHz}$ and $T \approx 200°\text{C}$ we found a maximum of approximately 40 μW, three times greater than observed previously[10] for similar input power.

Using the FP etalon (1.5 GHz FSR) we investigated the spectral properties of the blue output for several specific detunings (Fig. 3). Spectra A,B have net detunings $\delta_{780} + \delta_{776} = 0$, for which we expect blue output to the $F = 3$ ground state, while C,D have $\delta_{780} + \delta_{776} = 3\,\text{GHz}$ (i.e. $F = 2$). Structure due to the 6P hyperfine splittings ($F = 4, 3, 2, 1$; separations 40, 20, 10 MHz[12]) is evident from the double peaks in spectra A,B and the broad peaks of C,D which suggest overlap of several lines of similar strength.

The apparent frequency shifts of the spectra are modulo the 1.5 GHz FSR of the etalon (and no correction has been made for drifts of up to a few hundred MHz due to temperature and pressure variations in the etalon). Curve B is shifted by $|\delta\nu \mod 1.5\,\text{GHz}| = 1\,\text{GHz}$ which is also consistent with a shift of, for example, -0.5 or -2 GHz. The large shifts of the non-resonant cases B, C are attributed to ac Stark shift of the dressed ground-state level.

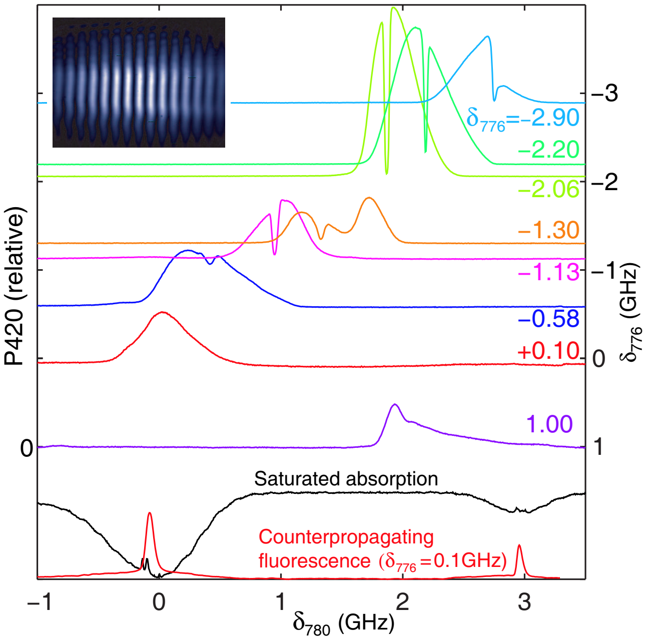

Fig. 2. (Color online) Blue output power for different pump laser detunings. The maximum output was observed when the 780 and 776 nm input fields were orthogonally polarized, with $\delta_{780} \approx 2$ GHz. The lowest two curves show frequency reference spectra, and the inset shows two-slit diffraction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

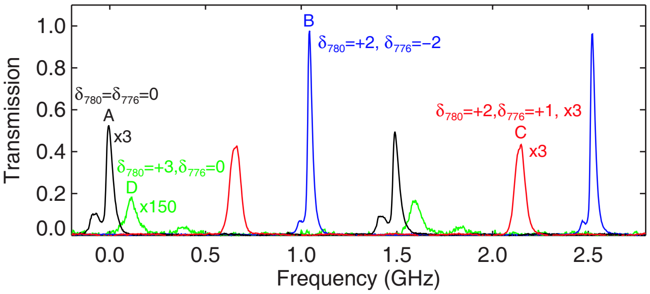

Fig. 3. (Color online) Blue output spectra from Fabry-Pérot (1.5 GHz FSR), normalized to spectrum B.

We have modeled the system using a semiclassical Maxwell-Bloch approach, with slowly varying envelope and phase approximations.[2,13,14] The four plane-wave classical fields are described by Maxwell's wave equation linking the electric field $\mathbf{E}$ to the induced polarization $\mathbf{P}$, where $\mathbf{P}$ is given by the appropriate off-diagonal element of the 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 include 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 16-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, j, i.e. $(\mathcal{H}_{\rm int})_{ij} \equiv \hbar\,\Omega_{ij}$. We transform 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, $\frac{\partial}{\partial z}\Omega_{ij} \propto {\rm Im}\,(\rho_{ij})$. We are particularly interested in gain on the 420 nm transitions, $\rho_{\rm 5S(F=2,3),6P}$.

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-Pérot 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 $\delta_{780} = 0$.

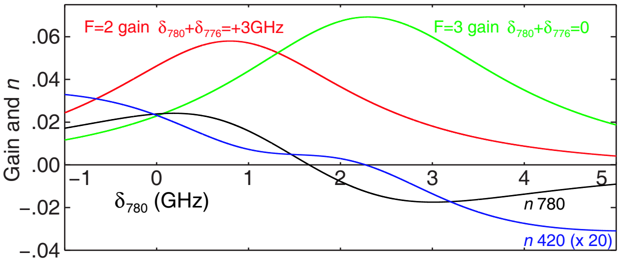

Fig. 4. (Color online) Gain and refractive index dependence of blue transition on δ_{780} in the five-level model. Gain and refractive index are proportional to ${\rm Im}\,(\rho_{ij})$ and $\sum_F {\rm Re}\,(\rho_{{\rm 5S}_F,j}) \equiv n$ respectively, where $F = 2, 3$ and j is 6P (420 nm) or 5P (780 nm).

Phase matching depends on the refractive index, calculated from the real part of the off-diagonal density matrix elements. In the 5-level model, these are small and slowly varying around 2 GHz, reducing phase mismatch for both the 780 nm pump and 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 due 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, which also inherently minimizes losses due to optical pumping between hyperfine states. Coupling both ground-state levels induces a five-level coherence, 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). 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 mis-match 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 challenging, there are many interesting phenomena to explore, such as the apparent resonance in the blue light near optimum conditions, which suggests an EIT-like coherence effect, and the possibility of gain enhancement with an external cavity.

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