Frequency evaluation of collimated blue light generated by wave mixing in Rb vapour

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Abstract
An evaluation of the absolute frequency and tunability of collimated blue light (CBL) generated in warm Rb vapour excited by low-power cw laser radiation at 780 nm and 776 nm has been performed using a Fabry–Perot interferometer and a blue diode laser. For the conditions of our experiments, the CBL tuning range is more than 250 MHz around the resonant frequency of the $^{85}\text{Rb} \, 5S_{1/2} (F = 3) \rightarrow 6P_{3/2} (F' = 4)$ transition. A simple technique for stabilizing the power and frequency of the CBL to within a few per cent and 10 MHz, respectively, is suggested and demonstrated.

1. Introduction
The technique of frequency conversion of low-power cw radiation of diode lasers into collimated blue and far-infrared light in Rb and Cs vapours, first demonstrated by Zibrov \textit{et al} \cite{1}, continues to be an active area of research \cite{2–7}. Potential applications of this approach include tunable coherent light sources, quantum information processing \cite{8} and underwater communication \cite{9}. Key spectral characteristics of collimated blue light (CBL) such as linewidth and tuning range are important for most of these applications. Previous investigations demonstrated high temporal coherence of the CBL using a Fabry–Perot interferometer and two-slit diffraction \cite{1, 2}, but did not determine the absolute frequency in detail.

CBL results from four-wave mixing in atomic media with a diamond-type energy level configuration. An alkali vapour driven by laser radiation tuned close to strong optical transitions in a ladder-type configuration, which in the case of Rb atoms are the $5S_{1/2} \rightarrow 5P_{3/2}$ and $5P_{3/2} \rightarrow 5D_{5/2}$ transitions (figure 1(a)), can produce an optical field at 5.23 μm through amplified spontaneous emission on the $5D_{5/2} \rightarrow 6P_{3/2}$ transition. Mixing of this field and the laser fields produces optical radiation at 420 nm in the direction satisfying the phase-matching relation $k_1 + k_2 = k_{\text{IR}} + k_{\text{BL}}$, where $k_1$, $k_2$, $k_{\text{IR}}$ and $k_{\text{BL}}$ are the wave vectors of the radiation at 780, 776, 5230 and 420 nm, respectively. The wave mixing origin of CBL is supported by observations of the unidirectional generation of the CBL \cite{3} and the transfer of the combined orbital angular momentum of the applied laser fields to the coherent blue light \cite{7}. But the possibility of incoherent optical pumping to the $6P_{3/2}$ level contributing to the observed CBL should not be discounted, particularly given the variations in a number of experimental parameters such as pumping rates and atomic densities made in the present study.

In contrast to conventional optical parametric oscillation, the new field generation occurs without an optical cavity. Rb atoms provide not only high Kerr nonlinearity, but also set the resonant conditions for the blue and far-IR radiation.

From the phase matching condition, which is crucial for the parametric FWM and CBL generation, for co-propagating 780 nm and 776 nm beams the blue light frequency is

\[ \nu_{\text{BL}} = \frac{(n_1 \nu_1 + n_2 \nu_2 - n_{\text{IR}} \nu_{\text{IR}})}{n_{\text{BL}}}. \]
Figure 1. (a) The $^{85}\text{Rb}$ atom energy levels involved in CBL generation and the absolute CBL frequency evaluation. (b) Schematic diagram of setup for collimated blue light generation. (c) Schematic diagram of setup for CBL frequency evaluation.

where $n_1$ and $n_2$ are the refractive indices seen by the light fields at 780 nm and 776 nm, $n_{\text{IR}}$ and $n_{\text{BL}}$ are the refractive indices at 5.23 $\mu$m and 420 nm, respectively, while $n_c$ is the corresponding frequency of the optical field at each wavelength. It is clear that the absolute frequency of the CBL must lie close to the $5S_{1/2}(F = 3)\rightarrow 6P_{3/2}$ transition frequency. This is readily confirmed by the observation of isotropic blue fluorescence from Rb vapour excited only by CBL in a heated auxiliary cell. However, given that the Doppler broadening of the transition is approximately 1 GHz, this observation does not preclude a substantial frequency detuning of the CBL from the $5S_{1/2}(F = 3)\rightarrow 6P_{3/2}$ transition. Taking into account the unknown absolute frequency of the far-IR radiation and that the refractive indices depend on the optical frequencies, intensities and polarizations of the applied laser fields, as well as the atomic density $N$, the precise CBL frequency is a complex function of all these parameters. In this paper, we undertake an experimental study of the absolute frequency and frequency tuning range of the CBL generated by parametric wave mixing in Rb vapour.

2. Absolute frequency evaluation

We evaluate the absolute frequency and tuning range of the CBL generated in $^{85}\text{Rb}$ vapour with high resolution by measuring the frequency difference between the CBL and reference radiation of known frequency. An obvious source of suitable reference radiation is a narrow-linewidth extended-cavity blue diode laser locked to a sub-Doppler feature of the $5S_{1/2}\rightarrow 6P_{3/2}$ transition produced by a standard nonlinear Doppler-free spectroscopic technique [10]. This would readily provide a frequency accuracy of approximately 1 MHz and we could measure the frequency difference between the CBL and this radiation by comparing the spectral positions of Fabry–Perot interferometer (FPI) transmission peaks of the laser light and the CBL. Unfortunately the output power of the available diode laser at 420 nm is insufficient to allow a standard saturation absorption spectroscopy arrangement and we instead implement a modified scheme for sub-Doppler spectroscopy of Rb atoms on the $5S_{1/2}\rightarrow 6P_{3/2}$ transition.

While in a conventional scheme of Doppler-free spectroscopy both the probe and saturating beams are produced from the same laser, we use the 420 nm radiation as a probe while strong optical pumping radiation is provided by a 795 nm laser tuned to the Rb D1 absorption line. This radiation produces velocity selective ground-state optical pumping of the atoms. Distinctive features of Doppler-free spectroscopy have previously been observed when using independent lasers tuned to different absorption lines [11–13].

Dramatic changes in the efficiency of CBL generation by simultaneously driving the D1 transition have been demonstrated in [6], and here we use a similar idea for producing a sub-Doppler frequency reference for the blue laser.

3. Experimental set-up

Our experimental setup, schematically illustrated in figure 1, is similar to that used in our previous experiments [3, 6]. Radiation from extended cavity diode lasers (ECDLs) at 780 and 776 nm drives stepwise and two-photon excitation of Rb atoms. The frequency of the 780 nm ECDL can be swept across the $D2$ absorption line or stabilized using a modulation-free technique based on a sub-Doppler dispersion shaped polarization resonance obtained on the $^{85}\text{Rb} 5S_{1/2}(F = 3)\rightarrow 5P_{3/2}(F = 4)$ transition in an auxiliary Rb cell. The 776 nm laser is also swept across the $^{85}\text{Rb} 5P_{3/2}\rightarrow 5D_{3/2}$ transition or side-locked to a low-finesse tunable confocal etalon. In the frequency stabilized regime, typical values of the standard deviation of the error signal correspond to frequency fluctuations in the range 200–300 kHz over a 1 s time interval for both lasers. The laser linewidth estimated from optical heterodyning of two lasers is approximately 1 MHz.

Radiation from both ECDLs is combined to form a bichromatic beam. The powers of the components at 780 and 776 nm are typically 12 and 6 mW, respectively. The bichromatic beam is weakly focused into a 5 cm long heated Rb cell containing a natural mixture of Rb isotopes with no buffer gas. The cross section of the beam inside the cell is about 0.5 mm$^2$. The temperature of the cell is set within the range 50–100 °C meaning that the density $N$ of Rb atoms varies from about 1.5 $\times$ 10$^{11}$ to 6 $\times$ 10$^{12}$ cm$^{-3}$.

An additional Rb cell heated to 60 °C and two ECDLs tuned to the $^{85}\text{Rb} D1$ line and to the $5S_{1/2}\rightarrow 6P_{3/2}$ transition at 420 nm are employed to determine the absolute frequency reference for the CBL. Colour filters with optical density approximately 0.5 and 4.0 at 420 nm and 780 nm, respectively, are used to spectrally select the CBL and isotropic blue fluorescence, both of which are detected by photomultipliers. A $\mu$-metal shield is used to reduce the ambient magnetic field in the Rb cell to a few milligauss.

The CBL spectral purity and linewidth are explored using a tuneable concave mirror FPI of length $L = 14.5$ cm and having high finesse in the blue spectral region. The
spatial distribution of the blue light transmitted through the interferometer is monitored with a CCD camera to ensure that the radiation is mainly coupled into the fundamental TEM$_{00}$ interferometer mode by a combination of lenses.

4. Results

4.1. Spectral properties of the CBL

Figure 2(a) shows CBL spectral profiles as a function of the 776 nm laser frequency taken at different atomic densities in the cell with the 780 nm laser locked to the $5S_{1/2}(F = 3)\rightarrow 5P_{3/2}(F' = 4)$ transition. Just above the threshold atomic density, which is about $N \approx 1.5 \times 10^{11} \text{ cm}^{-3}$ in these experiments, the profile has a shape close to Gaussian. At higher Rb density the CBL doublet structure, which is both laser intensity and frequency dependent becomes evident. This doublet structure has been discussed previously [3], but is still not properly understood. The frequency range of the 776 nm laser over which the CBL generation occurs is less than the Doppler width of the $5P_{3/2} \rightarrow 5D_{3/2}$ transition at this temperature.

Figure 2(b) demonstrates the blue light transmission through the scanned FPI. Narrow, high-contrast transmission resonances for various FPI tunings over the entire CBL profile confirm the single-frequency narrow-linewidth spectrum of the CBL. The spectral interval between the two highest peaks, which are due to coupling of the CBL to TEM$_{00}$ interferometer modes, corresponds to the free spectral range (FSR) of the interferometer, approximately 1034 MHz. Smaller transmission peaks are due to coupling to higher order transverse modes of the FPI and provide a convenient finer frequency scale. A single TEM$_{00}$ mode transmission resonance is shown in figure 2(c). The resonance is fitted by a Lorentzian profile of width 2.4 MHz (FWHM). This value is quite insensitive to both the applied laser intensity and the atomic density. Results of our investigation of the temporal coherence of the CBL will be presented elsewhere.

From the phase matching condition (1), frequency tuning of the 776 nm laser causes tuning of the CBL, but it is important not to assume the two frequencies will change identically. Reasons for a deviation from a direct one-to-one correspondence in the detunings ($\delta\nu_1$ and $\delta\nu_2$ for the 776 nm laser and $\delta\nu_{BL}$ for the CBL) include dispersion in the warm Rb vapour, and frequency variations of the far-IR radiation at 5.23 $\mu$m, which cannot be detected in our experiment. Spectrally selective refractive index changes have been reported in warm Rb vapours due to light-induced coherences ($\Delta n \sim 10^{-4}$ for vapours with complete transparency due to ground-state EIT [14] and $\Delta n \sim 0.1$ in cascade configuration at high atomic density $N \sim 10^{15} \text{ cm}^{-3}$ [15]). Simultaneous observations of the CBL intensity variations and FPI transmission resonances as a function of the 776 nm laser frequency detuning such as in figure 3(a), allow a quantitative relation between $\delta\nu_1$ and $\delta\nu_{BL}$ to be established. For this purpose, $\delta\nu_2$ is estimated using the low-finesse confocal etalon, while $\delta\nu_{BL}$ is measured using the frequency scale provided by the higher order modes of the blue FPI. The length of the blue FPI is adjusted so that transmission peaks of its fundamental axial modes approximately coincide with one of the intensity maxima of the CBL profile. With this technique we find that $\delta\nu_{BL}/\delta\nu_2 \approx (1.00 \pm 0.03)$. The precision is estimated from the nonlinearity in the 776 nm laser frequency tuning, so that within this...
atomic density dependent just above the CBL threshold \((N < 6 \times 10^{11} \text{ cm}^{-3})\). We have noted previously that there is an atomic density threshold for generation of CBL as well as a strong CBL intensity dependence on atomic density [3]. The atomic density directly influences parameters that are important for parametric FWM such as how far the various beams propagate through the cell before being absorbed. This makes a theoretical estimation of the CBL tunability difficult. Under present experimental conditions the atomic density dependence saturates at \(N > 7 \times 10^{11} \text{ cm}^{-3}\), while the dependence on the 780 nm laser power reveals almost constant growth.

### 4.2. Blue laser frequency evaluation

We now describe how the velocity-selective optical pumping technique can be used in evaluating the absolute frequency of the blue ECDL (figures 1(a), (c)).

If the blue laser with fixed frequency \(v_L\) is tuned to the peak of the inhomogeneously broadened fluorescence line on the \(^{85}\text{Rb} 5\text{S}_1/2(F = 3)\rightarrow 6\text{P}_3/2 (F = 2, 3, 4)\) transitions, then the main contribution to the isotropic fluorescence comes from atoms excited on the \(F = 3\rightarrow F' = 4\) cycling transition and having longitudinal velocity \(v_z = 2\pi (v_L - v_{34})/k_{BL}\), where \(v_{34}\) is the resonant frequency of the cycling transition. Two other resonant velocity groups which interact on the weaker open transitions \(F = 3\rightarrow F' = 2, 3\) are significantly depopulated due to spontaneous decay to the \(5\text{S}_1/2(F = 2)\) level.

The population of the \(v_z\) velocity group in the \(5\text{S}_1/2(F = 3)\) level could itself be modified by hyperfine optical pumping. If, for example, counter-propagating radiation at 795 nm having frequency \(v_{D1}\) is detuned by \(\delta v_{D1} = (v_{2j} - v_{34}) = k_{D1}v_z/2\pi\) from either transition from the \(^{85}\text{Rb} 5\text{S}_1/2(F = 2)\) level, where \(v_{2j}\) is the frequency of the \(5\text{S}_1/2(F = 2)\rightarrow 5\text{P}_1/2(F = j)\) transition, this results in a sub-Doppler width enhancement of the blue fluorescence. Through a comparison of the spectral positions of the sub-Doppler fluorescence peak and saturated absorption resonances observed simultaneously on the \(5\text{S}_1/2(F = 3)\rightarrow 6\text{P}_3/2\) and \(5\text{S}_1/2(F = 2)\rightarrow 5\text{P}_1/2\) transitions in the auxiliary Rb cell as the 795 nm laser is scanned, the frequency \(v_L\) of the blue laser can be evaluated. Due to the different Doppler shifts the detunings differ by a factor equal to the ratio of the wavelengths:

\[
\delta v_{D1} = (v_L - v_{34}) (k_{BL}/k_{D1}) \approx 1.89 \times \delta v_L. \tag{2}
\]

Figure 4 shows normalized fluorescence at 420 nm produced by the fixed-frequency blue laser at \(v_L\) and plotted as a function of the optical pumping laser frequency \(v_{D1}\) as it is swept across the inhomogeneously broadened \(^{85}\text{Rb} 5\text{S}_1/2(F = 2)\rightarrow 5\text{P}_1/2(F = 2; 3)\) transitions. In figure 4(a) the fluorescence is enhanced above the no-optical pumping level at four different frequencies of the pumping laser rather than the expected two. The two smaller peaks arise from optical pumping produced by 795 nm light back-reflected from the cell window. The D1 line saturated absorption resonances observed in the auxiliary Rb cell provide absolute frequency references for the optical pumping laser. The blue laser detuning \(\delta v_L\) is estimated using equation (2) from the offset of the fluorescence resonances from these saturated absorption resonances. For

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**Figure 3.** (a) The CBL profile (i) and blue FPI transmission resonances (ii) as a function of the frequency detuning of the 776 nm laser from the CBL minimum, while the 780 nm laser is locked to the \(^{85}\text{Rb} 5\text{S}_1/2(F = 3)\rightarrow 5\text{P}_3/2(F = 4)\) transition. The FPI length has been adjusted so that the TEM\(_{00}\) transmission peak coincides with one of the CBL maxima. (b) Width of the CBL frequency tuning range measured at the level of 20% as a function of the atomic density. The 780 nm laser is locked to the \(F = 3\rightarrow F' = 4\) transition, while the 776-nm laser is scanned. (c) Width of the CBL tuning range at the 20% level as a function of the laser power at 780 nm, for atomic density \(N \approx 7 \times 10^{11} \text{ cm}^{-3}\).
Curves transition at optical pumping intensities of 25 and 8 mW cm$^2$ able to confirm that the CBL frequency is centred on the around their corresponding transitions. In this way we are by tuning the frequencies of the 776 and 780 nm lasers described above. The separation of the peaks is easily changed by tuning the frequencies of the blue laser normalized on the fluorescence signal without optical pumping: (a) large frequency detuning ($\sim 50$ MHz) of the blue laser from the $^{85}$Rb $5S_{1/2}(F' = 4)$ transition at an optical pumping intensity of 25 mW cm$^{-2}$; (b) blue laser tuned to the $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ transition at optical pumping intensities of 25 and 8 mW cm$^{-2}$. Curves (ii) are saturated absorption profiles on the $^{85}$Rb $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}$ transition obtained in the axillary Rb cell kept at $\sim 50^\circ$C.

For example, the frequency offset shown in figure 3(a) suggests that the blue laser is tuned approximately 60 MHz above the $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ transition frequency. If the blue laser is tuned precisely on resonance, the large and small fluorescence peaks merge and spectrally coincide with the D1 line saturated absorption resonances, as shown in figure 4(b). In this case the 420 nm laser is resonant with the atoms that have zero axial velocity. Using this method the frequency of the blue laser can be tuned to the $^{85}$Rb $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ transition with approximately $\pm 5$ MHz accuracy. Figure 4(b) also illustrates power broadening of the sub-Doppler resonances. It is possible to obtain better resolution with lower pumping power.

Figure 5 shows FPI transmission resonances for the blue laser and CBL with all laser frequencies fixed while the FPI is scanned. The blue laser (broad peaks at zero detuning) is tuned to the $^{85}$Rb $5S_{1/2}(F = 3) \rightarrow 6P_{3/2}(F' = 4)$ transition with an accuracy of $\pm 5$ MHz. The different positions for the CBL peaks (narrower peaks) result from different fixed detunings of the 776 nm laser.

4.3. CBL frequency stabilization

For the CBL to be used effectively as a narrow-band light source, it is desirable to have a means of stabilizing its frequency and amplitude. As has been discussed above, the frequency of the CBL depends primarily on the frequencies of the two applied laser fields. Assuming the other factors are not varying, stabilizing the frequencies of the two applied laser fields can stabilize the CBL frequency and to some extent the CBL power. While the 780 nm laser can be locked routinely to the strong $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ cycling transition with standard Doppler-free spectroscopic techniques, this is not the case for the 776 nm laser as it is tuned to a transition between excited states. Although one could stabilize this laser to a reference cavity with high long-term stability, a more elegant solution is to lock the 776 nm laser directly to the CBL profile. Here we demonstrate that both peak and side locking can be used to control the step-wise excitation of Rb atoms and, consequently, the CBL frequency and intensity, with the error signal derived from the CBL intensity itself.

For the peak locking, the dither voltage at 10 kHz from a lock-in amplifier modulates the 776 nm laser frequency. Curve (i) in figure 6(a) shows the intensity profile of the CBL as the 776 nm laser is scanned across the $5P_{3/2} \rightarrow 5D_{5/2}$ transition, with the 780 nm laser locked to the $^{85}$Rb $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition. Curve (ii) represents the lock-in amplifier output, which is proportional to the first derivative of the CBL spectral profile, and is used as an error signal.

Active stabilization of the CBL is shown in figure 6(b). The 776 nm laser is initially scanned manually, then tuned to the higher peak of the CBL profile and the locking loop closed at $t = 0$. The increased error signal in the $2s < t < 5s$ interval is due to servo system gain adjustments while searching for optimum values of the locking parameters.

Assuming that in the locked mode the error signal fluctuations correspond to CBL frequency variations, we estimate that the relative frequency stability after locking is.
The CBL frequency is found to be centred on the $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F=4)$ transition, and can be tuned over a range of more than 250 MHz around this frequency by tuning the frequencies of the driving lasers. Atomic density and laser power dependences of the CBL tuning range have been examined.

Finally, we have proposed and demonstrated a method for stabilizing the power and frequency of the CBL, in which an error signal is derived from the CBL profile itself and used to lock the frequency of the 776 nm laser involved in the wave mixing process.

5. Conclusion

We have investigated the absolute frequency of collimated blue light generated by a parametric four-wave mixing process in atomic Rb vapour. The frequency of the CBL has been compared with that of a blue diode laser using a tunable Fabry–Perot interferometer. The blue laser frequency has, in turn, been determined using a modified sub-Doppler spectroscopy technique where velocity-selective pumping radiation has been applied on the Rb D1 line. This has allowed the absolute frequency of the CBL to be evaluated with approximately ± 5 MHz precision, limited primarily by the precision with which we are able to determine the frequency of the laser. The CBL frequency is found to be centred on the $5S_{1/2}(F=3)\rightarrow 6P_{3/2}(F=4)$ transition, and can be tuned over a range of more than 250 MHz around this frequency by tuning the frequencies of the driving lasers. Atomic density and laser power dependences of the CBL tuning range have been examined.

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