Doppler-free two-photon resonances for atom detection and sum frequency stabilization

A M Akulshin, B V Hall, V Ivannikov, A A Orel and A I Sidorov

Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, Hawthorn, Victoria 3122, Australia
E-mail: aakoulchine@.swin.edu.au

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Abstract
We investigate the excitation of the 5D5/2 level in Rb atoms using counter-propagating laser beams, which are nearly resonant to the one-photon 5S1/2 → 5P3/2 and 5P3/2 → 5D5/2 transitions, ensuring that a sum of the optical frequencies corresponds to the 5S1/2 → 5D5/2 transition. The excitation produced by two-photon and stepwise processes is detected via spontaneously emitted fluorescence at 420 nm arising from the 6P3/2 → 5S1/2 transition. The dependences of blue fluorescence intensity on atomic density and laser detuning from the intermediate 5P3/2 level have been investigated. The sensitivity of the frequency-detuned bi-chromatic scheme for atom detection has been estimated. A novel method for sum-frequency stabilization of two free-running lasers has been suggested and implemented using two-photon Doppler-free fluorescence and polarization resonances.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Optical detection of a few atoms relies predominantly on signals generated by spontaneously emitted photons [1]. While a resonant laser radiation facilitates optical excitation, it can also readily produce a large background due to scattering of light from optical elements. If the weak signal and the unwanted scattered light are not spectrally separated then heroic efforts must be undertaken to obtain a reasonable signal-to-noise ratio (SNR) while ensuring the high collection efficiency of spontaneously emitted photons that is crucial for quantum information experiments [2]. This may become unrealistic for fluorescence imaging of single atoms located near surfaces on atom chips or in the microscopic pyramidal magneto-optical traps [3].

An alternative approach is to utilize two-photon excitation to access higher lying atomic states which decay by emitting photons with sufficiently different frequencies. For example, in rubidium atoms the two-photon transition 5S1/2 → 5D can be realized using laser radiation at 778 nm, while subsequent decay via the 6P3/2 and 6P1/2 levels yields spontaneously emitted light at 420 nm, which can be effectively separated from the applied laser radiation using colour or interference filters. The intermediate 5P3/2 energy level, situated nearly half-way between the 5S1/2 and 5D levels, makes this two-photon transition strongest amongst alkali atoms.

The two-photon transition 5S1/2 → 5D5/2 in Rb atoms [4, 5] has found a wide range of applications. Due to a relatively long natural lifetime of the 5D5/2 level (~240 ns), the two-photon transition 5S1/2(F = 3) → 5D5/2(F′′ = 5) in 85Rb, where F and F′′ are the total angular momenta, was recommended as a secondary frequency reference [6]. Also, this transition was suggested to be used for transferring long-term frequency stability to the telecommunication spectral region at 1.5 μm [7]. The two-photon transition was employed for studying the excitation transfer between the Rb 5D fine-structure levels in collisions with ground-state atoms [8]. This scheme was also used to populate the 5D level in a magnetic trap whereby photoionization and subsequent ion detection yielded atom counting capability [9]. The two-photon excitation to the 5D5/2 level was probed, in addition to blue fluorescence, by the polarization rotation of the laser light due to the induced optical anisotropy of the medium [10].
More efficient transfer of atoms from the 5S_{1/2} to 5D_{5/2} levels and, consequently, more intense blue fluorescence can be obtained using a bi-chromatic excitation scheme, which employs radiation resonant to the one-photon transitions 5S_{1/2} \rightarrow 5P_{3/2} and 5P_{3/2} \rightarrow 5D_{5/2}, ensuring that the sum frequency of the two laser fields (\nu_1 + \nu_2) is equal to the two-photon transition frequency. In this case, in addition to the enhanced two-photon excitation rate due to proximity of the intermediate level 5P_{3/2}, there is another process, stepwise excitation, which plays an important role when the frequency detunings of the applied radiation from the one-photon transitions are comparable with inhomogeneous broadening of the transition.

The bi-chromatic excitation scheme has received a lot of attention. If the intermediate state 5P_{3/2} is coupled by both components of the bi-chromatic radiation, a good SNR of the Doppler-free spectra of the Rb 5D_{5/2} level can be obtained even in a room temperature vapour [11]. The bi-chromatic excitation scheme has been used for in situ imaging of an ultracold atom cloud by measuring absorption at 776 nm or by observation of the 420 nm fluorescence [12, 13]. Electromagnetically induced transparency (EIT) in the cascade atomic system, when the absorption of a probe beam resonant to the lower cascade transition is reduced under the action of a coupling beam resonant to the higher transition, has been studied [14].

It is worth noting that the co-propagating instead of the counter-propagating scheme of the bi-chromatic excitation of the Rb 5D_{5/2} level allows an optical field generation at 420 nm, which is both spatially and temporally coherent [15–18]. Taking into account that the detection of the two-photon 5S_{1/2} \rightarrow 5D_{5/2} monochromatic excitation based on the stimulated emission at 776 nm was recently demonstrated [19], possible applications of coherent light at 420 nm for atom detection deserves detailed consideration. This will form the subject of a subsequent paper. In this paper, we study bi-chromatic excitation of rubidium atoms in a vapour cell by nearly resonant laser radiation with a subsequent detection of spontaneously emitted blue fluorescence paying particular attention to the case when the laser frequency detuning is larger than the inhomogeneous broadening of the Rb D_{2} absorption line.

This paper is structured as follows. In section 2, a description of the experimental apparatus is given along with the methods for acquiring and comparing stepwise and two-photon processes. Section 3 presents measurements of the blue fluorescence as a function of laser frequency detunings from single-photon transitions. The fluorescence signal behaviour with laser intensity, polarization and atomic density along with detection capability of this method is discussed. In section 4, a novel sum-frequency locking of two free-running lasers to the 5S_{1/2} \rightarrow 5D_{5/2} transition in Rb atoms is considered. Doppler-free fluorescence resonances are used as frequency references for the sum-frequency stabilization. Dispersive-type polarization resonances have been suggested for modulation-free sum-frequency locking.

2. Excitation scheme and experimental setup

The two-photon excitation of Rb atoms in a vapour cell is produced by a bi-chromatic laser radiation whose components at 780 and 776 nm are near resonant to the one-photon transitions 5S_{1/2} \rightarrow 5P_{3/2} and 5P_{3/2} \rightarrow 5D_{5/2}, respectively. The relevant energy levels and optical transitions are depicted in figure 1(a). After excitation to the 5D_{5/2} level, 65% of Rb atoms spontaneously decay back to the 5P_{3/2} level, while the remaining atoms decay first to the 6P_{3/2} level and then to the ground state emitting blue photons at 420 nm [20]. Counter-propagating geometry of the applied laser beams results in much stronger blue fluorescence because of partial Doppler shift compensation for laser beams with close wavelengths [21]. Blue fluorescence and a signal proportional to the rotation of the polarization of the 776 nm laser beam passed through the atomic media are used for detecting the two-photon excitation.

The scheme of the apparatus is shown in figure 1(b). Two home-built external cavity diode lasers supply output powers of approximately 20 mW at 780 and 776 nm. The absolute frequency and the scan range of the 780 nm laser are evaluated based on Doppler-free absorption spectra obtained from an ancillary Rb cell and transmission resonances of a tunable low-finesse Fabry–Perot cavity (FPC1). The frequency of the 776 nm laser is monitored with another low-drift Fabry–Perot cavity (FPC2) with the free spectral range of 710 MHz and spectroscopic signals from the 6P_{3/2} \rightarrow 5D_{5/2} transition.

The excitation occurs in a vapour cell which contains a natural mixture of both ^{85}Rb and ^{87}Rb isotopes without any buffer gas. The interaction region from which the fluorescence is collected is in the centre of the 50 mm long cell. A specially designed heating/cooling thermoelectric element based on the Peltier effect allows varying the cell temperature from 10 to 70 °C. Thus, atomic density estimated based on the temperature measured in the coldest part of the cell is in the range 2 \times 10^{9} \leq N \leq 7 \times 10^{11} \text{ cm}^{-3} [22].

The counter-propagating laser beams are parallel or focused to waists of approximately 250 \mu m. Control of the polarization and intensity of the laser beams is achieved using polarizing beam splitters and wave plates not shown in figure 1(b) for simplicity. A 420 nm bandpass interference filter and colour filters ensure that only blue fluorescence is...
detected by the Hamamatsu R928 photomultiplier tube (PMT). Spectroscopic signals are recorded and processed using a digital oscilloscope and a computer. Amplitude fluorescence noise uncorrelated with laser frequency is reduced by applying signal averaging over 32 or 64 periods of frequency scans. In order to further increase the SNR, a lock-in amplifier is used. Both amplitude and frequency modulation methods with corresponding modulation frequencies of 700 Hz and 10 kHz are employed.

3. Results and discussion

3.1. Two-photon resonances on the $6S_{1/2} \rightarrow 5D_{5/2}$ transition

To demonstrate the whole spectrum of the blue fluorescence arising from the $6S_{1/2} \rightarrow 5D_{5/2}$ transition in Rb vapour under the nearly resonant bi-chromatic excitation, a wide frequency scan of the 776 nm laser is applied, while the fixed frequency 780 nm laser is tuned to the spectral region between the $85$Rb absorption lines on the $6S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ and $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transitions. Figure 2 shows four resonances of blue fluorescence, which occur when the sum frequency of the two lasers ($v_1 + v_2$) is equal to the frequencies of the transitions started from each ground-state sublevel of both Rb isotopes to the $5D_{5/2}$ levels. The strongest fluorescence resonance corresponds to the excitation of $85$Rb atoms from the $5S_{1/2}(F = 3)$ level. The relative amplitudes of fluorescence resonances depend on the frequency offset of the 780 nm laser from the one-photon transitions within the Rb $D_2$ absorption line. This issue is considered in the following subsection. The inset shows the ten-times amplified fluorescence feature attributed to the excitation of $85$Rb atoms from the $5S_{1/2}(F = 1)$ ground-state sublevel.

The hyperfine structure of the $5D_{5/2}$ level remains unresolved due to the fast wide-range laser frequency scan; however, with a smaller frequency scan every peak of blue fluorescence presented in figure 2 reveals a structure. As an example, figure 3 demonstrates blue fluorescence obtained with $85$Rb atoms excited from $5S_{1/2}(F = 1)$ and $5S_{1/2}(F = 2)$ ground-state sublevels, respectively, as a function of frequency detuning of the 780 nm laser from the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition in $85$Rb. The spectrum of blue fluorescence shown in figure 3(a) is obtained when the fixed frequency 776 nm laser is blue detuned by approximately 5.7 GHz from the $5P_{3/2} \rightarrow 5D_{5/2}$ transition, while figure 3(b) demonstrates the spectral dependence of blue fluorescence with 1.1 GHz red-detuned laser frequency from the same transition.

The selection rule for two-photon transitions ($\Delta F \leq 2$) allows three transitions from the lower ground-state sublevel $5S_{1/2}(F = 1)$ and four transitions from the higher ground-state sublevel $5S_{1/2}(F = 2)$ to the $5D_{5/2}(F'' = 1, 2, 3, 4)$ levels. Figure 3 demonstrates the well-resolved hyperfine structure of the $5^2D_{5/2}$ level. The frequency intervals between...
peaks, measured using a frequency scale that is provided by the saturated absorption spectrum on $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}$ transitions recorded simultaneously in the addylo cell reference, correspond within 10% to the frequency splitting of the $5D_{3/2}$ level [11]. The discrepancy is due to nonlinearity of the laser frequency scan.

The width of the fluorescence resonances depends on frequency detuning from the intermediate $5P_{3/2}$ level. The ultimate linewidth of the Doppler-free resonances produced by the monochromatic two-photon excitation is defined solely by a sum of the initial and final state lifetime $\gamma_S + \gamma_D$, where $\gamma_D \simeq 2\pi \times 0.66 \text{ MHz}$ and $\gamma_S$ is determined by the time of flight of atoms across the interaction region $T = d/v$, where $d$ is the beam diameter and $v$ is the average atomic velocity. The time-of-flight broadening ($\gamma_T \simeq 1/(2\pi T)$) for parallel laser beams is negligibly small ($\approx 22 \text{ kHz}$), while for the focused laser beams to waists of approximately 250 $\mu$m, the broadening ($\approx 250 \text{ kHz}$) may become noticeable. In the bi-chromatic scheme, these resonances are broadened approximately by $\sqrt{\gamma_P^2 + \gamma_D^2}$ due to a mismatch in Doppler shifts arising from the frequency difference of the two optical fields.

If $(v_1 - v_0) \ll \Delta_D$, then the stepwise excitation process through the intermediate $5P_{3/2}$ level contributes to the excitation of the $5D_{3/2}$ level. This process results in larger linewidth because $\gamma_P \gg \gamma_D$, where $\gamma_P$ is the natural linewidth of the intermediate $5P_{3/2}$ levels. Thus, the width of the Doppler-free fluorescence resonances depends on a ratio between two processes. Theoretical modelling of the corresponding excitation in Cs atoms [23] shows that the contribution of the two-photon process prevails at high power. The typical FWHM of experimentally observed fluorescence resonances (figure 3) is approximately 6 MHz, which lies between the two-photon and stepwise limits, taking into account that power broadening is stronger for the stepwise process. Laser linewidth and magnetic field broadening in the unshielded Rb cell also contribute to the observed width. The shape of the peaks is slightly different from the Lorentzian profile, which suggests the presence of distinctive broadening mechanisms.

### 3.2. Blue fluorescence dependence on laser detuning

We use a double-frequency sweeping technique to explore the fluorescence amplitude dependence on frequency detuning from the intermediate $5P_{3/2}$ level. This technique allows quick data acquisition that eases requirements for stability of major experimental parameters. The 780 nm laser is slowly scanned with sweep frequency $f_1$ across a few GHz wide region, while the other laser is swept much faster, with sweep frequency $f_2$ ($f_2 \gg f_1$), relative to the position where the sum optical frequency $(v_1 + v_2)$ is equal to the frequencies of the transitions from the ground state to the $5D_{3/2}$ level. Thus, the two-photon excitation condition is met $f_2/f_1$ times during the 780 nm laser scan. This results in the appearance of a set of fluorescence profiles shifted in frequency of the 780 nm laser. The TDS3012 digital oscilloscope used for data acquisition is capable of recording the whole set of traces.

As an example, figure 4(a) shows the peak fluorescence evolution obtained by the double sweeping technique. The excitation of $^{87}$Rb atoms from the $5S_{1/2}(F = 3)$ level occurs in the spectral region I, which is red-shifted from the Doppler broadened absorption line observed in the axial cell on the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}$ transitions in $^{85}$Rb. The excitation of $^{85}$Rb atoms from the $5S_{1/2}(F = 3)$ level takes place in the spectral region II, which spectrally coincides with the blue-frequency slope of the same $^{87}$Rb absorption line. The spectral position and the width of the regions show that the 776 nm laser is blue detuned with respect to the $5P_{3/2} \rightarrow 5D_{3/2}$ transitions and that the detuning is swept from 0.5 to 1.35 GHz.
The systematic measurements of fluorescence dependence on laser detunings are performed on the $5S_{1/2}(F = 3) \rightarrow 5D_{5/2}$ transitions in $^{87}$Rb. The vapour cell temperature is chosen to be 63 °C ($N \approx 4 \times 10^{11} \text{ cm}^{-3}$). As the laser beams are focused inside the cell, the maximum light intensities at 780 and 776 nm are 6.5 and 0.9 W cm$^{-2}$, respectively. These values are much higher than the saturation intensity for the both one-photon transitions.

The linear fit of experimental points plotted in the log-log scale and shown in the inset of figure 4(b), reveals that the slope is equal to $-1.98$. This means that the fluorescence amplitude varies inversely proportional to the square of the frequency detuning in this spectral region. Despite strong saturation of the single-photon transitions in our experiment, the dependence is in good agreement with well-established theory that describes the spectral lineshape of bi-chromatic two-photon excitation in the low saturation limit [11, 21], which predicts the $(v_{1} - v_{0})^{-2}$ dependence for the excitation rate. At larger frequency detuning, the fluorescence amplitude decays slower, as $(v_{1} - v_{0})^{-\alpha}$, where $\alpha \approx 1.6$.

### 3.3. Atomic density dependence and detection sensitivity

The atomic density dependence and sensitivity of the method for atom detection are investigated by using signals from the PMT, which are directly recorded or preliminary amplified by a lock-in amplifier. In the latter case, 700 Hz amplitude modulation of the laser radiation at 780 nm produced by a mechanical chopper is applied.

This investigation is performed using parallel laser beams. In this case, the interaction region inside the Rb cell is approximately 5 cm long and 0.3 cm in diameter. Blue fluorescence is collected by a cylindrical lens ensuring the collection efficiency of approximately 1.8%.

To ensure predominantly the two-photon nature of the $5D_{5/2}$ level excitation, the 780 nm laser is red detuned from the strongest $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition in $^{87}$Rb atoms. To keep the laser frequency detuning constant during the rather long period of time required for changing the atomic density from $2 \times 10^9$ to $7 \times 10^{11} \text{ cm}^{-3}$, the following procedure has been used. One transmission peak of the FPC1 cavity is tuned to coincide with the Doppler-free absorption resonance on the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition obtained in the auxiliary cell, and then the laser frequency is locked to the next transmission FPC1 peak, which is red detuned by the free spectral range of the cavity, which is 710 MHz. The position of the reference resonance is checked from time to time ensuring that the 780 nm laser frequency is kept detuned by the FSR of the cavity from the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition. The 776 nm laser is scanned across the $5P_{3/2} \rightarrow 5D_{5/2}$ transitions.

Figure 5 demonstrates the derived atomic density dependence of the detected fluorescence when intensities of the 780 and 776 nm beams in the cell are approximately 180 and 300 mW cm$^{-2}$, respectively. The SNR is estimated using the averaged fluorescence signal normalized on the root-mean-square (RMS) voltage recorded for off-resonant laser frequencies.

Using direct detection we are able to detect the blue fluorescence signal with a SNR of approximately unity at a temperature as low as 10 °C at which the atomic density of saturated Rb vapour is $2 \times 10^{9} \text{ cm}^{-3}$. As shown in figure 5, lock-in amplification results in an improvement of the detection capability by a factor of 200 due to the narrower detection bandwidth.

As expected, the fluorescence signals detected directly and using lock-in amplification grow steadily with atomic density. A linear fit to the experimental data presented in the double-logarithmic plot reveals that the fluorescence signal is proportional to atomic density $N$ (SNR $\sim N^\beta$, where $\beta = 1.08 \pm 0.11$). Extrapolating this dependence to the SNR = 1 level provides an estimate for the minimum detectable number density of $3 \times 10^{6} \text{ cm}^{-3}$, as shown by the dotted line in figure 5.

As the amplitude and frequency noise of laser light and noise of the detection system limit the detection capability of the method, signal averaging can increase the SNR in proportion to the square root of the number of averages $k$ if signal and noise are uncorrelated. However, the number of averages is limited by the frequency stability of both lasers. Taking into account the FWHM of fluorescence resonances ($\approx 6 \text{ MHz}$) and the typical drift of the FP cavity reference resonance due to temperature and pressure instability ($\approx 0.4 \text{ MHz} s^{-1}$), we can conclude that averaging the signal over 32 periods of 50 Hz scans of the 776 nm laser is tolerable because during this time interval (0.64 s) the drift of the 780 nm laser stabilized over the FP cavity is less than 5% of the FWHM of the blue fluorescence resonance. Thus, averaging over $k = 32$ periods results in a $k^{1/2} \approx 5.5$-fold reduction of the noise uncorrelated with laser scanning and at the same time does not significantly degrade the amplitude of the signal and distort the width of recorded spectra.

In order to avoid saturation of the PMT signal at high atomic densities and to be in its linear regime for the whole
range of atomic densities, blue fluorescence is recorded at the lowest available voltage (∼380 V) of the Techtron photomultiplier driver. However, increasing the PMT voltage up to 850 V increases the output signal by a factor of 640 while the dark noise level is less than triple, which improves the SNR. At least a 200-fold increase of the SNR has been obtained in our experiment in this way.

Thus, considering that the data presented in figure 5 have been recorded at the lowest possible gain of the PMT and have not been averaged, a combination of signal averaging and higher PMT voltage could result in a 1000-fold increase of the SNR allowing Rb atom detection at $3 \times 10^3$ cm$^{-3}$ atomic density. Taking into account the interaction volume (∼0.35 cm$^{-3}$), we may expect to reach a detection threshold of approximately 1000 atoms.

The detection capability can be further improved by better collection of emitted blue fluorescence, applying higher modulation frequency and longer, up to minutes, signal averaging accessible with better laser stability.

4. Sum-frequency locking of two independent lasers

As Doppler-free fluorescence resonances occur when the sum frequency of the two lasers $(v_1 + v_2)$ is equal to frequencies of the $5S_{1/2} \rightarrow 5D_{3/2}$ transitions, they can be used as a reference for sum-frequency stabilization using a single servo system. A similar idea has been implemented with two independent lasers locked to a narrow CPT-type absorption resonance (A-resonance), which also has the two-photon origin; however, in that case the locking results in differential frequency stabilization [26].

The sum-frequency locking of two lasers can be implemented using a standard locking technique based on the frequency modulation and the lock-in amplification on one of the sub-Doppler fluorescence resonances shown in figure 3. It makes no difference whether the feedback is applied to the 780 or 776 nm lasers and which laser is modulated. The sum frequency $(v_1 + v_2)$ can be stabilized even if one laser is free running. Once the lock is engaged, the servo system compensates random frequency excursions of the free-running laser by changing correspondingly the frequency of the stabilized laser to keep the sum frequency fixed.

To demonstrate the direct sum-frequency stabilization using one servo system, the 780 nm laser is approximately 500 MHz blue detuned from the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition in $^{87}$Rb, while the 776 nm laser is tuned in such a way that excitation of $^{87}$Rb atoms to the $5D_{3/2}$ level occurs.

Figure 6 presents the PMT direct output proportional to the blue fluorescence and the servo system output, which is actually a derivative of the PMT signal, recorded for three different regimes. A slow manual scan of the 776 nm laser in the vicinity of the $5P_{3/2}(F' = 4) \rightarrow 5D_{5/2}(F'' = 1, 2, 3, 4)$ transitions allows a voltage-to-frequency conversion coefficient to be deduced for the servo signal using the hyperfine splitting of the $5D_{5/2}$ level as a scale. The stable fluorescence signal after applying feedback to the 776 nm laser (at $t = 0$) on the level of the strongest resonance in the scanning regime reveals that the sum frequency $(v_1 + v_2)$ is locked to the $5S_{1/2}(F = 2) \rightarrow 5D_{5/2}(F'' = 4)$ transition. Using a PID servo system which provides only slow feedback (from dc to 1 kHz) to the piezo-electric transducer that controls the 776 nm laser cavity, the sum-frequency instability estimated from the RMS of the error signal is approximately 240 kHz. This value corresponds to the relative instability $\delta \nu/(v_1 + v_2)$ of $3.6 \times 10^{-10}$. The sum-frequency instability can be reduced by proper optimization of the reference line and by locking of the free-running laser to a tunable FP cavity. The suggested and demonstrated sum-frequency stabilization could be useful for frequency up-conversion in alkali vapours [15–18] and Rydberg atom excitation experiments [27, 28] and in intervening spectral regions devoid of strong atomic transitions. As the blue fluorescence signal depends on frequency detuning, this can then be used to derive an absolute frequency of the 780 nm laser in the vicinity of the Rb $D_2$ line. It is envisaged that this could form the basis for an additional level locking scheme based on the amplitude of the fluorescence resonance.

4.1. Polarization spectroscopy of the $5P_{3/2} \rightarrow 5D_{5/2}$ transition in Rb

Even small frequency modulation applied for frequency locking results in additional broadening of the laser linewidth, which is often undesirable. A dispersive-shaped polarization resonance is an ideal reference line for the modulation-free frequency locking.

The polarization rotation of resonant monochromatic laser light that excites the two-photon transition was demonstrated in [10], and we implement a polarization spectroscopy setup for the bi-chromatic excitation scheme. Considering that independent setting of polarizations of laser beams is important for maximizing the polarization signals, the bi-chromatic excitation scheme allowing this arrangement possesses an obvious advantage compared to the monochromatic excitation scheme.

In the present experiment, by an analogue to the conventional polarization spectroscopy of one-photon
transitions [24], the 776 nm laser beam analyses birefringence of the Rb vapour induced by the circularly polarized 780 nm light in the vicinity of the 5P3/2 → 5D5/2 transition in 87Rb. Curves (i) and (ii) represent the blue fluorescence signal and the transmitted intensity of the 776 nm light, respectively, as a function of the frequency detuning of the 776 nm laser.

The intensity of the linearly polarized 776 nm laser beam transmitted through the vapour cell and a polarizer is detected by a photodiode. When the polarizer is almost crossed the spectral dependence of the transmitted probe-laser intensity reveals resonances with the well-pronounced dispersive shape. To make this signal suitable for sum-frequency locking a non-resonant dc background should be subtracted using a differential signal from two photodiodes [25]. The steep slope of the polarization signal, shown in figure 7, coincides with the top of the fluorescence resonance recorder simultaneously on the strongest 5S1/2(F = 2) → 5D5/2(F'' = 4) transition for 87Rb atoms. This polarization resonance can be used for modulation-free sum-frequency stabilization.

5. Conclusion

We study two-photon and stepwise bi-chromatic excitations of the 5D5/2 level in 85Rb and 87Rb atoms by monitoring blue fluorescence emitted by the atoms decaying from the level 6P3/2. The double-frequency sweeping technique allows quick data acquisition and relaxes requirements for stability of major experimental parameters. The dependence of blue fluorescence as a function of atomic density is investigated in the temperature range of 10 to 70 °C.

The fluorescence amplitude varies inversely proportional to the square of the frequency detuning from the intermediate 5P3/2 level. Despite strong saturation of the single-photon transitions, the dependence is in good agreement with the theoretical prediction of the two-photon excitation rate obtained for the low saturation case.

The sensitivity of the bi-chromatic scheme for atom detection using direct fluorescence observation and lock-in amplification has been estimated. A novel method for the sum-frequency stabilization of two free-running lasers has been implemented using the Doppler-free two-photon excitation. The estimated value of the relative sum-frequency instability is lower than 4 × 10^{-10}.

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