Chapter 6

One-photon absolute frequency measurements

Direct Frequency Comb Spectroscopy (DFCS) can be equally well applied to measure single-photon transitions. However, since we have the ability to dynamically access two-photon transitions (as shown in Chapter 4), we can measure the energy of a given state in two ways. We can either investigate an atomic state as part of a one-photon process or as a two-photon process. We have chosen the 5S-5P transitions in ^{87}Rb to demonstrate this ability. The measurement of 5P states has been carried out directly and also indirectly, via the 5S-5D two-photon transitions by studying their resonant enhancement when comb components are scanned through the intermediate 5P states. We compare the 5P measurements obtained via one-photon and two-photon DFCS and clearly demonstrate the importance of population transfer in working with multilevel systems probed by multiple comb components.

Also, again with the technique of DFCS, we have the advantage of being able to determine absolute atomic transition frequencies anywhere within the comb bandwidth, for one-photon processes.

6.1 Detection and timing scheme for the direct 5P measurements

For the one-photon studies we have investigated both the D_1 and D_2 transitions in ^{87}Rb, namely transitions from the ground state to the $5P_{1/2}$ excited state at 795 nm and to the $5P_{3/2}$ state at 780 nm (see general diagram, in section 4.2).

The three main timing sequences of the magneto-optical trap (MOT), the polarization gradient cooling (PGC), and the probing cycle are similar to those shown in section 4.3. However, here during the probing cycle, we implement a switching scheme for the photomultiplier tube (PMT) and the fs laser.

The main challenge with the direct 5P measurements is to collect the fluorescence emitted by the atoms, while avoiding gathering the light from the fs probe, since they have nearly the same wavelength. In addition, because we use acousto-optic modulators for fast switching of the MOT lasers, there is always some residual light that may affect the measurements. Care was taken to ensure that these lasers did not influence the signal.

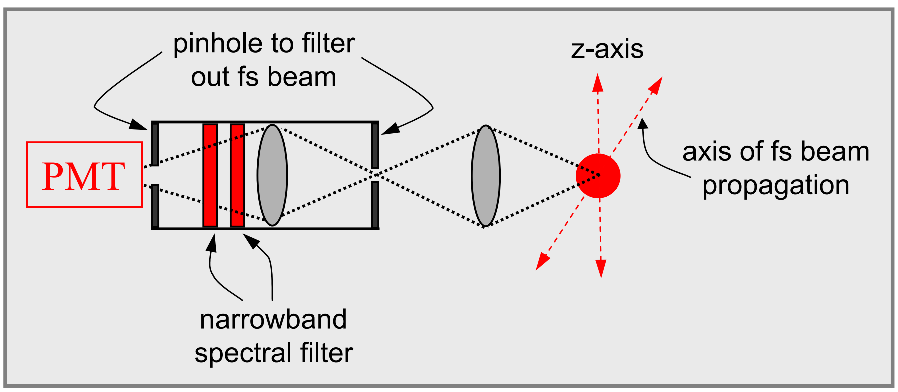

Figure 6.1: Detection assembly for the direct one-photon measurements. Background counts are minimized by using spatial and spectral filters. As before, the PMT signal is sent to a photon counter and the resulting counts are read into a computer.

We directly detect the fluorescence from the two 5P states with a near-infrared PMT coupled with a 3-nm bandwidth interference filter centered at the appropriate wavelength for the transition, as presented in the detection assembly in Fig. 6.1. We use narrow band interference filters reduce the number of comb components that are received by the PMT and thus minimize detection noise. Background counts are further minimized by spatial filtering with two pinholes, the first very small (1 mm), through which the MOT is imaged [Fig. 6.1]. Photon collection during the probe laser-on period is disabled by switching off the PMT.

The loading cycle used for the MOT is 100 Hz, and the sequence of the experiment is similar to the two-photon experiments [Fig. 6.2]: the atoms are loaded in the MOT for 7.8 ms, then the quadrupole magnetic field for the trap is switched off, the atoms are cooled with polarization gradients for 2 ms, then all the MOT beams are extinguished and the femtosecond comb beam is switched on for 200 μs using the Pockels cell (8-ns rise time)

As illustrated in the one-photon timing diagram in Fig. 6.2, during the 200 μs probe window, we have a sequence of short cycles with the probe laser on (200 ns) followed by the PMT on (400 ns) to detect photons from the fast-decaying 5P states. A 2.6 μs interval (PMT switch-off time) is required before initiating the next laser cycle. Both the two-photon and the single-photon signals are averaged over hundreds of 10 ms experiment cycles.

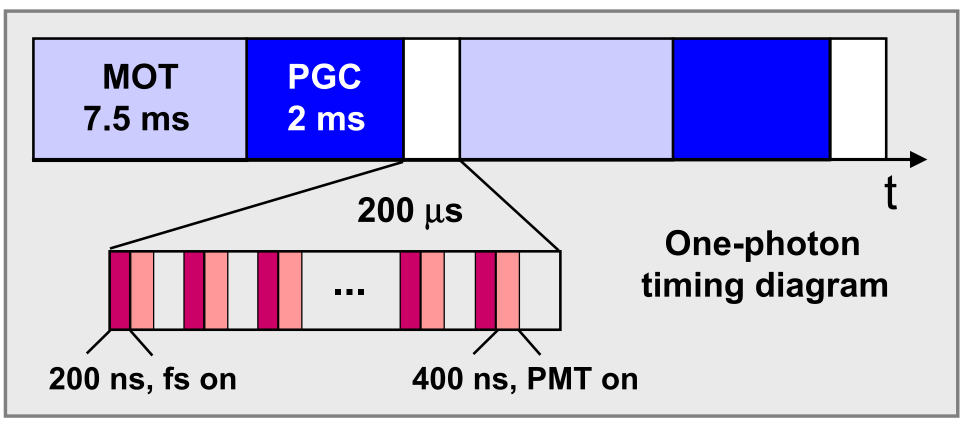

Figure 6.2: Timing scheme for the 100 Hz experiment cycle, where the 200-μs zoom window shows the sequences for the direct one-photon measurements. We switch the PMT off during the laser-on period, to minimize detection noise.

For the indirect 5P measurements, we use results from the theoretical model describing the fs comb interaction with the atoms. As discussed in Chapter 2, the theory accounts for detailed dynamics of population transfer among the atomic states involved in transitions within the comb bandwidth. A numerical scheme is employed to obtain the state of the atomic system after an arbitrary number of pulses [37, 60]. This model is applied to accurately predict the coherent population accumulation in the relatively long-lived 5D, followed by incoherent optical pumping. Especially important for the indirect 5P measurements is the incoherent optical pumping to the ground state hyperfine levels, which depends critically on the 5P state detunings and will be discussed in detail below.

We will now compare one-photon and two-photon DFCS measurements for the 5P state energy levels. The one-photon DFCS employs radiative detection directly from the 5P states (Fig. 6.3, left panel), while the two-photon DFCS studies the 5P states indirectly, via resonant enhancement of the 5S-5D two-photon transitions as a function of the detuning from the intermediate 5P states (Fig. 6.3, right panel).

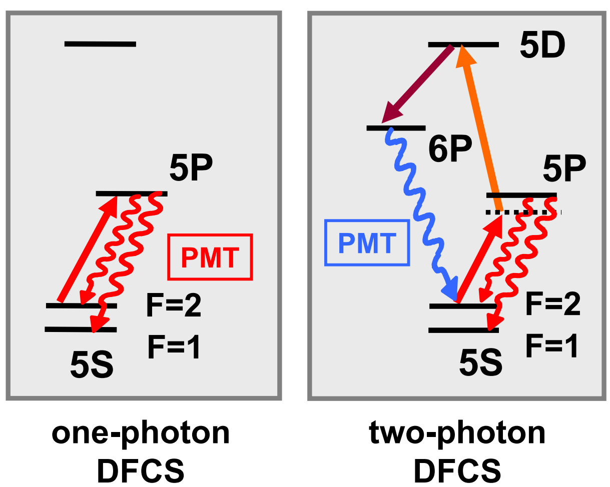

Figure 6.3: Schematic of one- and two-photon DFCS, used for measuring single-photon transition frequencies. The two-photon DFCS here differs from the two-photon process described in Chapter 5 in that the 5D state is kept strictly in resonance. The 5P lineshape is obtained by scanning the detuning.

6.2 $5P_{3/2}$ frequency measurements

First, we measure the $5S_{1/2}$ F=2 → $5P_{3/2}$ F′=3 D_2 transition with one-photon DFCS, with the resulting transition line shown in Fig. 6.4(a) [76]. Frequency scans are carried out similarly to those of the 5S-5D lines, that is, by stepping f_0 continuously while keeping f_r fixed. The absolute optical frequency measured for this transition is 384 228 115 271 (87) kHz, in agreement with a previous measurement done here at JILA [82], which resulted in an absolute frequency value of 384 228 115 203 (7) kHz. Once again, all the measurement errors reported in this chapter, and actually in this thesis, are statistical errors (one standard deviation of the mean).

For the two-photon DFCS, we use a set of different pairs of f_r and f_0 specifically chosen to have varying detunings from the 5P state for each data point shown in Fig. 6.4(b), while at the same time satisfying strictly the 5S-5D two-photon resonance ($\delta_{SD} = 0$). In this case, we monitor once again the 420 nm fluorescence from the 5D states. Such a set of f_r and f_0 is given as an example in Table 6.1. We immediately see that f_r must be known within 10 mHz. To obtain this level of accuracy, we always reference the f_r frequency counter to the cesium clock in our lab. As mentioned earlier, this is necessary because the f_r excursions get multiplied by $\sim 8 \times 10^6$. In contrast, f_0 must only be stable within 100 Hz.

The lineshape in Fig. 6.4(b) is retrieved by detecting the 420 nm signal as a function of 5P state detuning and the optical frequency measured by this two-photon DFCS is 384 228 115 309 (63) kHz, in agreement with the result obtained from one-photon DFCS within the standard deviation. Although the two methods for measuring this 5P state, direct and indirect, agree well, the indirect one is more cumbersome for two main reasons. Firstly, f_r must be changed by such a large amount as to require the output coupler to be placed on a translation stage and thus, the laser box to be opened for each detuning value. Secondly, this required hand-tuning adds significant

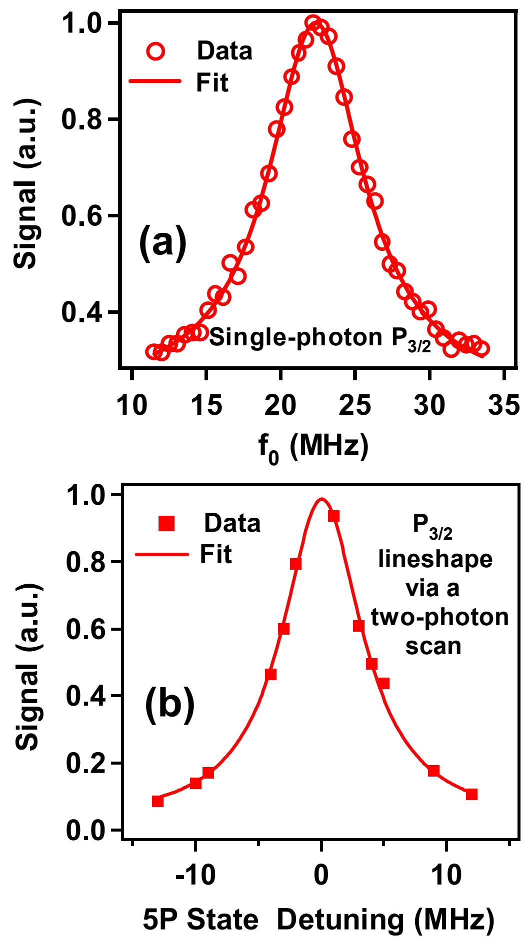

Figure 6.4: (a) Lineshape of the $5S_{1/2}$ F=2→ $5P_{3/2}$ F′=3 transition obtained from a scan of f_0 for a fixed value of f_r, by one-photon DFCS. (b) Same lineshape as in (a), retrieved using its resonant enhancement of the $5S_{1/2}$ F=2→ $5P_{3/2}$ F′=3→ $5D_{5/2}$ F″=4 closed two-photon transition, as a function of the detuning from the intermediate state.

time for retrieving a lineshape, so it is important that the data be taken under the same conditions and thus, it all has to be acquired on the same day. The direct method, though requiring an extra (switched) PMT, allows for a fast and smooth scan of the 5P states.

It is also important to mention that for this scan we take advantage of the $5S_{1/2}$ F=2 → $5P_{3/2}$ F′=3 → $5D_{5/2}$ F″=4 being the only 5S-5D closed transition. As shown in the theory plots in Fig. 6.5, this closed transition ensures that most of

5P detuning (MHz)	f_r (MHz)	f_0 (MHz)
-13	99.89260605	-16.4737
-10	99.92066423	-22.4503
-9	99.93474869	-16.2599
-4	99.92482030	-9.9189
-3	99.93890785	-11.1280
-2	99.91518350	-21.3653
+1	99.94325356	-24.2433
+3	99.86747878	-22.2673
+4	99.88155032	-24.0872
+5	99.89562280	-14.1944
+9	99.84803930	-11.4370
+12	99.84303590	-24.3973

Table 6.1: Example of a set of f_r and f_0 pairs used for the indirect scanning of the intermediate $5P_{3/2}$ F′=3 state. All the detunings are given with respect to this P state. The values for f_r and f_0 are chosen to ensure that the F″=4 state is always resonant for this indirect scan.

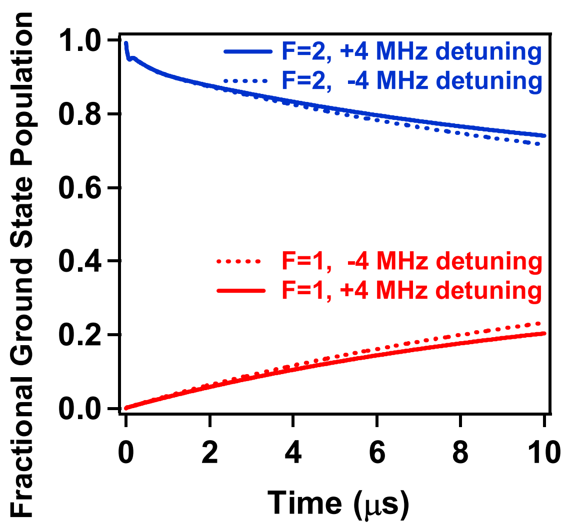

Figure 6.5: Theoretical plot of the time evolution of the ground state populations for two (symmetric) detuning values in Fig. 6.4(b), showing that (i) most of the atoms remain in the initial F=2 ground level for this closed two-photon transition and (ii) the ground state populations are largely insensitive to the sign of the detuning.

the atoms initially starting out in the F=2 ground-state hyperfine level remain in that level, while ∼20% of the atoms fall into the dark F=1 ground state due to optical pumping and hence do not contribute to the signal. In addition, the probe laser power is sufficiently reduced for the two-photon DFCS experiments to further decrease optical pumping effects.

6.3 $5P_{1/2}$ frequency measurements

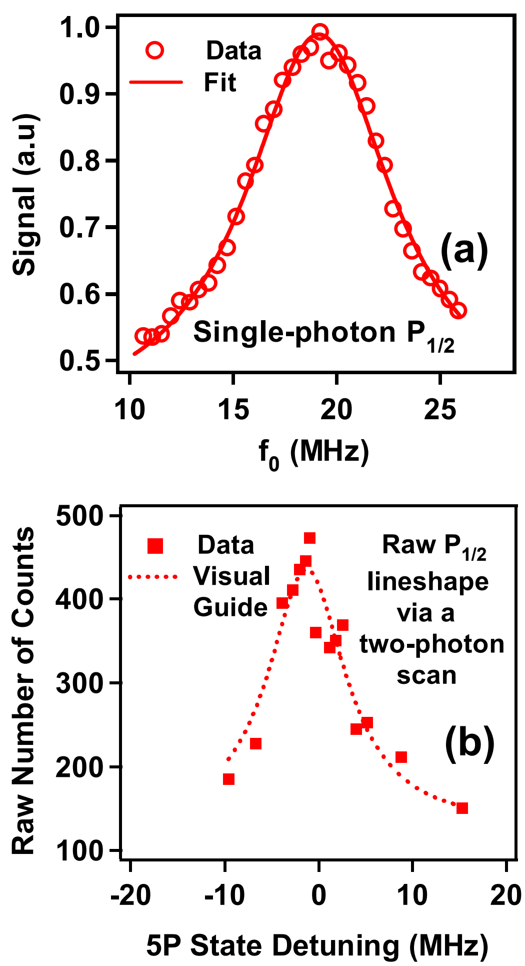

Figure 6.6: (a) Lineshape of the $5S_{1/2}$ F=2 → $5P_{1/2}$ F′=2 transition obtained from a scan of f_0 for a fixed value of f_r, by one-photon DFCS. (b) Raw counts for the same lineshape as in (a) by two-photon DFCS, along with a visual guide for the data.

Next, we employ DFCS to study another single-photon transition in the D_1 manifold, $5S_{1/2}$ F=2 → $5P_{1/2}$ F′=2, as shown in Fig. 6.6(a). Again, f_0 is scanned while f_r is stabilized to a convenient value. The absolute optical frequency for this transition is determined to be 377 105 206 563 (184) kHz, in agreement with a previous wavelengthbased measurement [83], which gave an absolute frequency value of 377 105 206 705 (400) kHz for this transition.

For the corresponding two-photon DFCS experiment we map the $5S_{1/2}$ F=2 $\rightarrow$ $5P_{1/2}$ F′=2 $\rightarrow$ $5D_{3/2}$ F″=3 two-photon transition in the same manner employed for Fig. 6.4(b). Figure 6.6(b) shows the raw data yielded by the (f_r, f_0) pair selections, along with a visual guide for the data. The apparent linewidth is significantly broader than that associated with the 5P state.

Unlike the previous two-photon DFCS measurement reported in Fig. 6.4, the pairs of f_r and f_0 used to obtain each point in Fig. 6.6(b) lead to substantially different detunings of the other 5P states and subsequently, varying optical pumping to the F=1 ground state. Indeed, the theory model applied to the actual experiment conditions predicts significantly different ground state population transfer dynamics. As shown in Fig. 6.7, the asymptotic values of the F=2 ground-state population are not the same for symmetric detunings from the intermediate state.

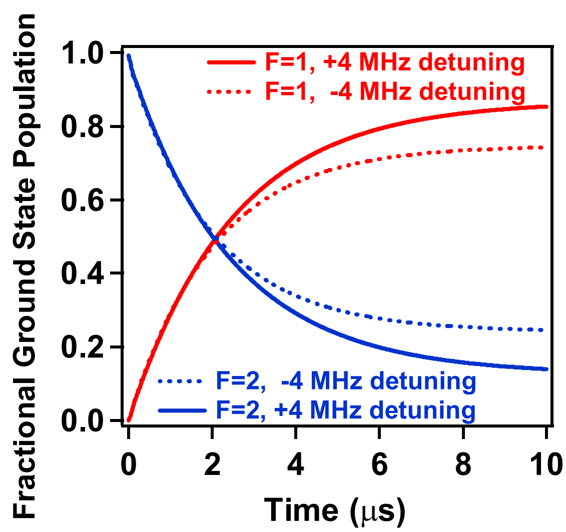

Figure 6.7: Theoretical plot of the time evolution of the ground state populations for two (symmetric) detuning values in 6.6(b), showing a significant difference in the population transfer between the ground state levels due to optical pumping caused by varying detunings from the other 5P states.

Figure 6.8(b) presents the Lorentzian lineshape resulting from the normalization of the raw data shown in Fig. 6.8(a) with respect to the theoretical value of (1 - $\rho_{F=1}$), where $\rho_{F=1}$ is the fractional ground state population in F=1, as shown in Fig. 6.7. After we implement this normalization, the optical frequency for the transition measured by the two-photon DFCS is 377 105 206 939 (179) kHz, which is within the error bars of the corresponding one-photon DFCS result.

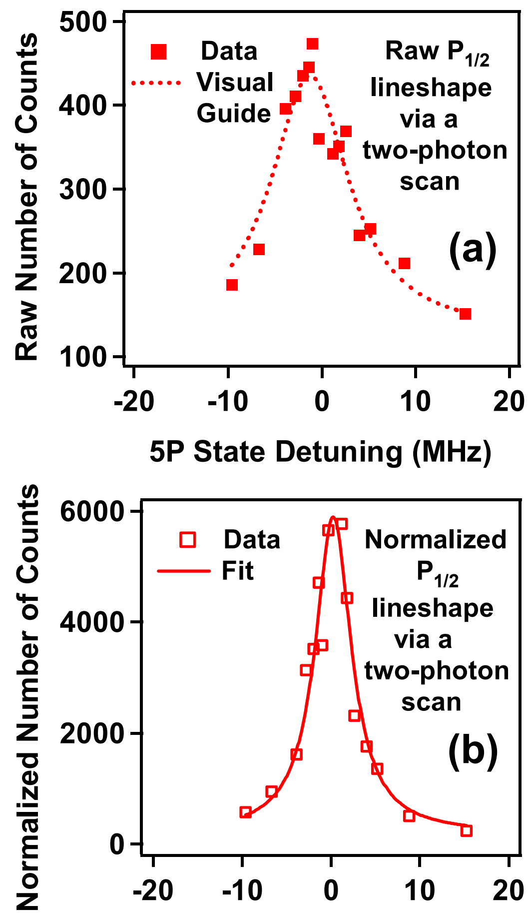

Figure 6.8: Raw data (a) and normalized lineshape (b) obtained by using results from the theory simulation in Fig. 6.8 accounting for optical pumping.

We note that for all measurements reported in this chapter, the statistical errors (one standard deviation of the mean) associated with $5P_{3/2}$ are significantly smaller than those associated with $5P_{1/2}$. This is due to the stronger transition strength and less severe optical pumping effects for $5P_{3/2}$ F'=3 (part of a closed transition), leading to larger signal-to-noise ratios.

Direct frequency comb measurements of one-photon transitions in atoms other than ^{87}Rb have been recently reported. In Leo Hollberg's group at NIST, they probed Cs in an atomic beam with DFCS, but using a Ti:S laser with a 1 GHz repetition rate [84]. Employing a 1-GHz fs laser has several advantages over a lower f_r laser, as stated in Chapter 7.

In conclusion, a phase-stabilized femtosecond comb has been used as an effective tool to perform direct spectroscopy of one-photon transitions in cold ^{87}Rb atoms. We have demonstrated that DFCS can be successfully applied to one-photon studies, by measuring $5S_{1/2} \rightarrow 5P_{1/2,3/2}$ transitions both directly and indirectly, via their resonant enhancement of the 5S-5D two-photon transitions. Additionally, we have shown the importance of including the dynamic population changes arising from pulse-accumulated population transfer in this indirect one-photon measurement.