

Direct Frequency Comb Measurements of Absolute Optical Frequencies and Population Transfer Dynamics

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A phase-stabilized femtosecond laser comb is directly used for high-resolution spectroscopy and absolute optical frequency measurements of one- and two-photon transitions in laser-cooled ^{87}Rb atoms. Absolute atomic transition frequencies, such as the $5S_{1/2} F = 2 \rightarrow 7S_{1/2} F'' = 2$ two-photon resonance measured at 788 794 768 921 (44) kHz, are determined without *a priori* knowledge about their values. Detailed dynamics of population transfer driven by a sequence of pulses are uncovered and taken into account for the measurement of the $5P$ states via resonantly enhanced two-photon transitions.

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Phase-stabilized optical frequency combs based on mode-locked femtosecond lasers form a powerful connection between the fields of precision measurement and ultrafast science [1–3], with a wide range of applications including measurements of absolute optical frequencies [4–6] and the development of optical atomic clocks [7,8]. Recent work has demonstrated that optical frequency combs are a highly efficient tool for precise studies of atomic structure [9,10]. One advantage of using a femtosecond laser is that multiple atomic states may be directly excited and the subsequent dynamics probed, by tuning the appropriate comb lines into resonance. Furthermore, given that the comb has two independent degrees of freedom, it is always possible to simultaneously satisfy two-photon as well as one-photon resonance conditions. Direct frequency comb spectroscopy (DFCS) has been performed on the $5S$ - $5D$ two-photon transitions in ^{87}Rb , permitting high-resolution spectroscopy of all the transitions within the comb bandwidth. Additionally, this approach enables precise studies of time domain dynamics, coherent accumulation and interference, and quantum control [10]. An extension of frequency comb metrology to the deep-ultraviolet spectral region was recently achieved by using the 4th harmonic of a femtosecond laser [11].

In this work, we apply DFCS to determine absolute atomic transition frequencies anywhere within the comb bandwidth, for one- and two-photon processes. By determining the previously unmeasured absolute frequency of the $5S$ - $7S$ two-photon transitions in ^{87}Rb , we show that prior knowledge of atomic transition frequencies is not essential for this technique to work, and indicate that it can be applied in a broad context. When resonant enhancement is enabled by a comb component tuned near an intermediate $5P$ state, we observe two-photon transitions occurring between initial and final states that differ by one unit of the total angular momentum ($\Delta F = \pm 1$), which are absent for far-detuned intermediate states. This capability of accessing adjacent excited hyperfine levels from the

same ground state allows for direct measurements of hyperfine splittings. Additionally, we demonstrate that DFCS can be equally well applied to measuring one-photon transitions, such as the $5S$ - $5P$ transitions in ^{87}Rb . The measurement of $5P$ states has also been carried out 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.

The spectrum of a mode-locked femtosecond laser has a set of discrete optical frequencies that are described by the relation $\nu_N = Nf_r + f_0$, where N is a large integer on the order of 10^6 . The lines are spaced by f_r , the pulse repetition rate, and have a common offset f_0 , the carrier-

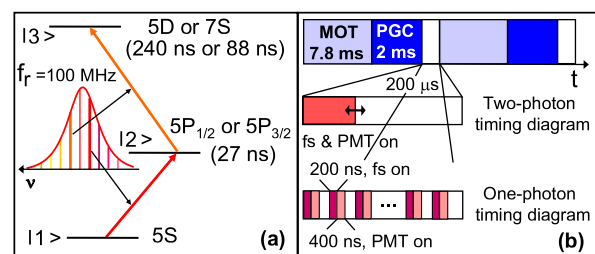


FIG. 1 (color online). (a) Schematic of the ^{87}Rb energy levels participating in the $5S$ - $7S$ two-photon transitions and $5S$ - $5P$ one-photon transitions. For the two-photon studies, the measured $5S$ - $7S$ transitions were resonantly enhanced by the $5P$ states shown. For the $5P$ state measurements, the $5S$ - $5P$ transitions are studied both directly and indirectly, using the $5P$ enhancement of the $5S$ - $5D$ two-photon transitions. (b) Timing scheme for the 100 Hz experiment cycle where the 200 μs zoom window shows the different experiment sequences for the one- and two-photon measurements. Signals in both measurements are averaged over hundreds of 10 ms experiment cycles. All MOT-related fields are turned off while probing with the femtosecond laser.

envelope offset frequency. Both parameters f_r and f_0 are precisely controlled and stabilized to low-noise optical or radio frequency oscillators [2]. In this work, the output of a 20 fs, 100 MHz repetition rate Ti:Sapphire laser with a full width at half maximum of ~ 55 nm is used to directly interrogate a sample of laser-cooled ^{87}Rb atoms. For the two-photon case, we study the transitions from the ground $5S_{1/2}$ state to the excited $7S_{1/2}$ state, as shown in Fig. 1(a). The two independent parameters f_r and f_0 allow for resonant signal enhancement via the intermediate $5P_{1/2}$ and $5P_{3/2}$ states. The relevant lifetimes are 88 ns for the $7S$ states [12] and 27 ns for the $5P$ states. The excited state population is determined from the $7S$ - $6P$ - $5S$ radiative cascade, with the $6P$ - $5S$ decay fluorescence at 420 nm. These blue photons are detected with a photomultiplier tube (PMT) and are counted and subsequently time binned with a multichannel photon counter. The typical loading cycle used for the magneto-optical trap (MOT) is 100 Hz and the sequence of the experiment is as follows [Fig. 1(b)]: 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 further cooled with polarization gradients (PGC) for 2 ms, then all the MOT beams are extinguished, the femtosecond comb beam is switched on for 200 μs using a Pockels cell (8 ns rise time), and, finally, the 420 nm fluorescence is detected.

For the one-photon studies we have investigated 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 [Fig. 1(a)]. 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. Background counts are further minimized by spatial filtering, while photon collection during the probe laser-on period is disabled by switching off the PMT. As can be seen in the one-photon timing diagram of Fig. 1(b), 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.

To null the stray magnetic fields, we apply bias-coil compensation in three orthogonal directions and make use of the two-photon signal itself: Zeeman-shifted spectra are obtained with right and left circularly polarized femtosecond comb light along each direction and the zero-frequency shift point is determined within ± 20 kHz. Two other dominant sources of systematic frequency shifts are both associated with the femtosecond comb. The first is the radiation pressure of the probe laser on the atoms and the second is the AC Stark shift [10]. For the present two-photon experiment, the line-center values are extrapolated to zero interrogation time to suppress shifts associated with photon-momentum transfer and the transitions are probed on optimal resonance (i.e., zero detuning for both the

intermediate state and the final state) to minimize the AC Stark shifts.

A theoretical model describing the interaction of the femtosecond comb with the atoms accounts for detailed dynamics of population transfer among the atomic states involved in transitions within the comb bandwidth. The density matrix for the state of the atomic system is calculated starting with the Liouville equation, with radiative relaxation included via phenomenological decay terms. Impulsive optical excitation followed by free evolution and decay is used to model the interaction with each pulse in the train. The density matrix equations are solved to a fourth order perturbative expansion in the electric field and an iterative numerical scheme is employed to obtain the state of the atomic system after an arbitrary number of pulses [10,13]. This model is applied to accurately predict the coherent population accumulation in the relatively long-lived $5D$ or $7S$ states, 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 begin by discussing the $5S$ - $7S$ two-photon measurements. Shown in Fig. 2(a) is a typical $7S_{1/2}$ $F'' = 2$ Lorentzian line shape, generated by stepping f_0 for a fixed value of f_r , and recording the subsequent blue fluorescence corresponding to the coherently accumulated $7S$ population. Alternatively, this line shape is retrieved by sweeping f_r with f_0 fixed at some convenient value. In general, sweeping f_r has the advantage of yielding all the transitions within the laser bandwidth in only a ~ 26 Hz scan. This is due to the fact that the ratio of one-photon optical transition frequencies (participating in stepwise two-photon transitions) to f_r is $\sim 4 \times 10^6$, so that for a change in f_r of ~ 26 Hz the resonant enhancement is repeated by the next neighboring comb component. The correct mode

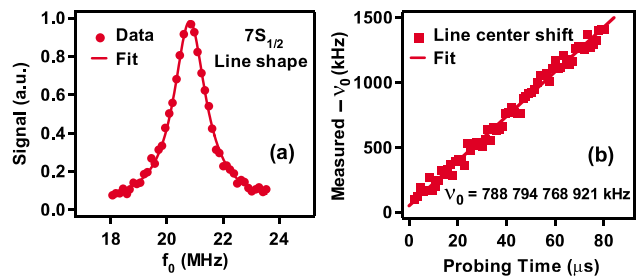


FIG. 2 (color online). (a) Typical $5S_{1/2}$ $F = 2 \rightarrow 7S_{1/2}$ $F'' = 2$ two-photon Lorentzian line shape obtained from a scan of the offset frequency f_0 for a fixed value of the repetition rate f_r . For each data point, the 80 ns-binned counts arising from the fluorescence are integrated over 2.4 μs . (b) Frequency shift of the transition line center vs probing time resulting from the momentum transferred by the femtosecond laser to the cold ^{87}Rb atoms. Extrapolation to zero interrogation time gives absolute atomic transition frequencies, as well as roughly the natural linewidth, free of radiation pressure effects.

number N associated with each transition remains now to be determined. If the optical frequency is already known to within $f_r/2$, this is a straightforward task. For the case of the $5S$ - $7S$ two-photon transitions, where the optical frequencies are not known *a priori* ($\nu_{\text{opt}} = (N_1 + N_2)f_r + 2f_0$), we scan the resonances for two different values of f_r and unambiguously deduce the sum of the two associated integers ($N_1 + N_2$) [11,14]. In our case, the two repetition rates used are separated by 600 kHz to eliminate possible uncertainties arising from estimations of the f_0 value corresponding to the peak of the resonance.

After identifying the associated comb numbers and reducing AC Stark shifts, the remaining error from radiation pressure is suppressed by extrapolating to zero interrogation time, as shown in Fig. 2(b). We determine for the $5S_{1/2}$ $F = 2 \rightarrow 7S_{1/2}$ $F'' = 2$ and the $5S_{1/2}$ $F = 1 \rightarrow 7S_{1/2}$ $F'' = 1$ two-photon transitions in ^{87}Rb the absolute optical frequencies of 788 794 768 921 (44) and 788 800 964 199 (122) kHz, respectively. The excited state hyperfine interval of 639 404 (130) kHz agrees very well with a previous differential measurement performed with a picosecond pulsed laser [15]. The transition spectra reported in [15], as well as a continuous wave (CW)-based scan [16], indicated that the $F = 2 \rightarrow F'' = 2$ and the $F = 1 \rightarrow F'' = 1$,

i.e., $\Delta F = 0$ transitions, were the only allowed $5S$ - $7S$ transitions in ^{87}Rb . However, we observe additional lines, as the resonant intermediate $5P$ state also enables the $F = 2 \rightarrow F'' = 1$ and $F = 1 \rightarrow F'' = 2$, i.e., $\Delta F = \pm 1$, two-photon transitions. Similar $\Delta F = \pm 1$ S - S transitions have been previously observed in Na in a two-step excitation experiment employing two tunable CW dye lasers [17].

DFCS also works well for absolute frequency measurements of one-photon transitions. We now compare one-photon and two-photon DFCS measurements of the $5P$ state energy levels. The one-photon DFCS employs radiative detection directly from the $5P$ states [Fig. 3(a), 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. 3(a), right panel]. First, we measure the $5S_{1/2}$ $F = 2 \rightarrow 5P_{3/2}$ $F' = 3$ transition with one-photon DFCS, with the resulting transition line shown in Fig. 3(b). Frequency scans are carried out similarly to those of the $5S$ - $7S$ 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. 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

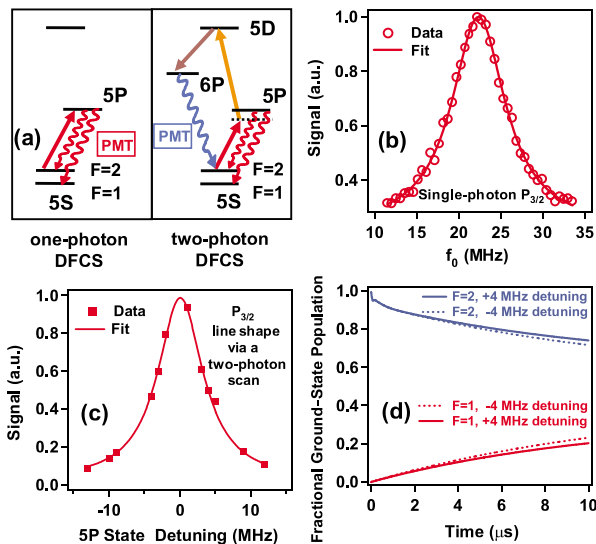


FIG. 3 (color online). (a) Schematic of one- and two-photon DFCS, used for measuring single-photon transition frequencies. (b) Line shape of the $5S_{1/2}$ $F = 2 \rightarrow 5P_{3/2}$ $F' = 3$ transition obtained from a scan of f_0 for a fixed value of f_r , by one-photon DFCS. (c) Same line shape as in (b), retrieved using its resonant enhancement of the $5S_{1/2}$ $F = 2 \rightarrow 5P_{3/2}$ $F' = 3 \rightarrow 5D_{5/2}$ $F'' = 4$ closed two-photon transition, as a function of the detuning from the intermediate state. (d) Theoretical plot of the time evolution of the ground-state populations for two (symmetric) detuning values in (c), 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.

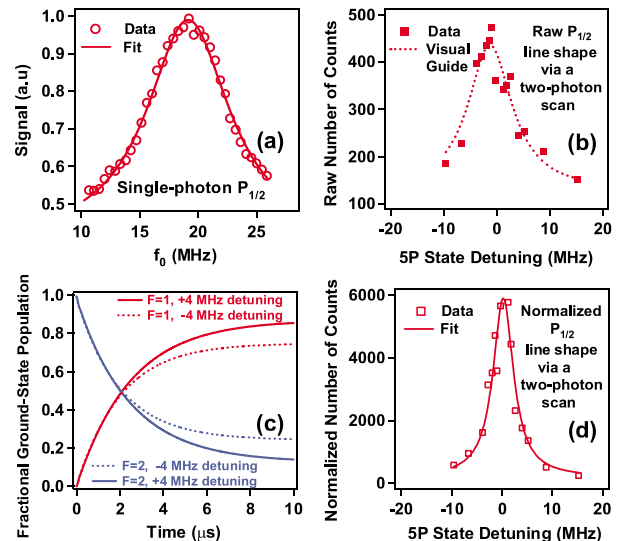


FIG. 4 (color online). (a) Line shape of the $5S_{1/2}$ $F = 2 \rightarrow 5P_{1/2}$ $F' = 2$ transition obtained from a scan of f_0 for a fixed value of f_r , by one-photon DFCS. (b) Raw counts for the same line shape as in (a) by two-photon DFCS, along with a visual guide for the data. (c) Theoretical plot of the time evolution of the ground-state populations for two (symmetric) detuning values in (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. (d) Normalized line shape corresponding to the raw data in (b), obtained by using results from the theory simulation in (c) accounting for optical pumping effects.

shown in Fig. 3(c), while at the same time satisfying the $5S$ - $5D$ two-photon resonance (these transitions have the same decay channels as the $5S$ - $7S$ transitions and are also monitored via the 420 nm fluorescence). The line shape in Fig. 3(c) 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. It is important to mention that for this scan we take advantage of the $5S_{1/2} F = 2 \rightarrow 5P_{3/2} F' = 3 \rightarrow 5D_{5/2} F'' = 4$ being the only $5S$ - $5D$ “closed transition.” As shown in the theory plots in Fig. 3(d), this closed transition ensures that most of the atoms initially starting out in the $F = 2$ ground-state hyperfine level remain in that level, while $\sim 20\%$ 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.

Next, we employ DFCS to study another one-photon transition, $5S_{1/2} F = 2 \rightarrow 5P_{1/2} F' = 2$, as shown in Fig. 4(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 wavelength-based measurement [18]. 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. 3(c). Figure 4(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. 3, the pairs of f_r and f_0 used to obtain each point in Fig. 4(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. 4(c), the asymptotic values of the $F = 2$ ground-state population are not the same for symmetric detunings from the intermediate state. Figure 4(d) presents the Lorentzian line shape resulting from the normalization of the raw data shown in Fig. 4(b) 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. 4(c). After implementing this normalization, the optical frequency for the transition measured by the two-photon DFCS is 377 105 206 939 (179) kHz, within the error bars of the corresponding one-photon DFCS result. We note that for all measurements reported in the paper, the statistical errors (1 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.

In conclusion, a phase-stabilized femtosecond comb has been used as an effective tool to perform direct spectroscopy of one- and two-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. Finally, we have demonstrated that by using DFCS, the absolute value of the $5S_{1/2} \rightarrow 7S_{1/2}$ two-photon transitions in ^{87}Rb is conclusively determined, with no *a priori* knowledge about their optical frequency.

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Note added.—CW laser-based measurements of the $5S$ - $7S$ transitions in Rb were reported recently [19]. V. Gerginov *et al.* recently made direct frequency comb measurements of one-photon transitions in Cs [20].

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