

High-resolution spectroscopy with a femtosecond laser frequency comb

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The output of a mode-locked femtosecond laser is used for precision single-photon spectroscopy of ^{133}Cs in an atomic beam. By changing the laser's repetition rate, the cesium D_1 ($6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2}$) and D_2 ($6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{3/2}$) transitions are detected and the optical frequencies are measured with accuracy similar to that obtained with a cw laser. Control of the femtosecond laser repetition rate by use of the atomic fluorescence is also implemented, thus realizing a simple cesium optical clock.

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The introduction of the femtosecond laser optical frequency comb (FLFC) began a new era of precision frequency metrology. The FLFC has been used as a tool for optical frequency calibration and measurement, atomic clock comparisons, and microwave synthesis.¹ However, in most experiments to date, the actual atomic physics is done with highly coherent cw lasers and the FLFC is the link between these lasers and microwave or other optical frequency standards. In a few cases, the direct output of a mode-locked laser has been used successfully for two-photon spectroscopy, where many pairs of comb lines add appropriately to connect ground and excited states.^{2–5} In contrast, in this Letter we demonstrate high-resolution single-photon spectroscopy that directly employs the few nanowatts present in a single component of a FLFC. With this source we perform parallel multifrequency optical measurements that can successfully compete with measurements done using cw lasers, but with a significantly simplified experimental setup. Specifically, a FLFC is used to excite the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2}$ and $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{3/2}$ transitions in a neutral ^{133}Cs beam and to measure the corresponding optical frequencies. Subsequently, we lock the FLFC to one of the 8 MHz wide Cs optical transitions, thereby creating a grid of absolute optical frequencies in addition to the divided-down microwave signal at the repetition rate of the femtosecond laser. The fractional uncertainty and 1 s instability of the Cs-stabilized FLFC are $\sim 1 \times 10^{-10}$. We expect that the techniques described here could be applied to atoms and molecules with narrower transitions, yielding correspondingly lower uncertainties and instabilities.

We employ a Ti:sapphire FLFC, for which the carrier-envelope offset frequency f_0 and the repetition rate $f_r \approx 1$ GHz of the femtosecond laser are both phase locked to microwave synthesizers.^{6,7} Initially, these synthesizers are referenced to a stable hydrogen maser, which is calibrated by a Cs atomic fountain clock. The fractional frequency uncertainty of

the comb teeth is equivalent to that of a hydrogen maser, $\sim 2 \times 10^{-13}$ at 1 s, and averages down to the 10^{-15} range after a few hours.

Approximately 10% of the infrared part of the femtosecond laser output (see Fig. 1) is sent via a single-mode fiber to a highly collimated atomic beam (12 mrad divergence, density of $\sim 10^{14}$ cm⁻³).⁸ The output of the single-mode fiber is collimated to a 6.4 mm diameter beam that intersects the Cs atomic beam at a right angle above a large-area photodiode. The laser power at the atomic beam is stabilized by an acousto-optic modulator placed in front of the optical fiber. The magnetic field in the interaction region is zeroed with three pairs of Helmholtz coils to better than 2×10^{-6} T.⁸ To reduce the background pressure of Cs inside the vacuum chamber, a liquid-nitrogen trap is used. The chamber pressure is better than 2×10^{-4} Pa.

Fluorescence from the excited Cs atoms is collected by the photodiode, digitized, and stored on a computer as the synthesizer controlling f_r is scanned. Fluorescence signals are readily observed with broadband (i.e., 750–1000 nm) excitation of the Cs atoms; however, interference filters are used to narrow the laser output spectrum to the region of interest. By use of two optical filters, the spectral width of the FLFC is narrowed to 3 nm in the vicinity of the

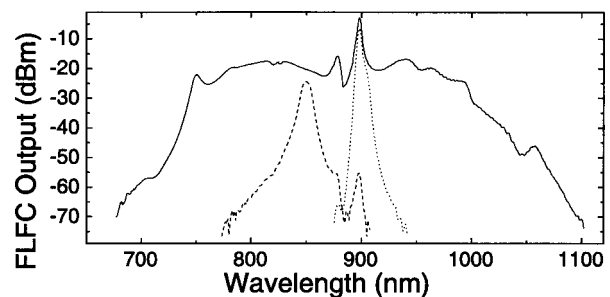


Fig. 1. Femtosecond laser spectrum used for high-resolution spectroscopy of the Cs D_1 and D_2 lines at 895 and 852 nm, respectively.

Cs D_1 line at 895 nm (dotted curve in Fig. 1) and then subsequently to 9 nm for the D_2 line at 852 nm (dashed curve in Fig. 1).

The fluorescence versus f_r for the four different components of the D_1 line is shown in Fig. 2(a) and arises from transitions between $F_g=3,4$ and $F_e=3,4$. Because of the presence of a comb tooth every 1 GHz, the fluorescence signals also repeat every 1 GHz change in optical frequency, corresponding to a change in f_r of ~ 3 kHz. The fluorescence offset is due to scattered laser light from the numerous nonresonant comb components, whereas the nonlinear background is due to the formation of a residual Cs vapor inside the vacuum chamber. The results for the D_2 line are shown in Fig. 2(b). The lower power in the FLFC near 852 nm results in lower peak amplitudes and a lower fluorescence offset. In this case, the observed spectral features are due to transitions between $F_g=3,4$ and $F_e=2,3,4,5$. Because of the particular value of repetition rate f_r , two of the spectral features ($F_3 \rightarrow F_3$ and $F_4 \rightarrow F_4$) overlap, as denoted in Fig. 2(b).

For these measurements we relied on a knowledge of f_0, f_r , the D_2 and D_1 optical frequencies,^{8,9} and relative amplitudes to determine the mode number of the comb component that is resonant with each optical transition. Because of the large repetition rate of the femtosecond laser (1 GHz), such identification is possible knowing the optical frequencies with a precision of several tens of megahertz. By use of multiple measurements with significantly different repetition rates (e.g., a set of N measurements to determine N unknown frequencies), it should be possible to determine the absolute frequencies without previous frequency information.^{10,11}

As an example of the precision that can be obtained, the optical frequency of the D_1 line $F_g=3 \rightarrow F_e=4$ transition (measured in ~ 3 h) is shown in Fig. 3. The model used to determine the line centers has been described in detail in Ref. 8 and is found to be an adequate description of the present line shapes.

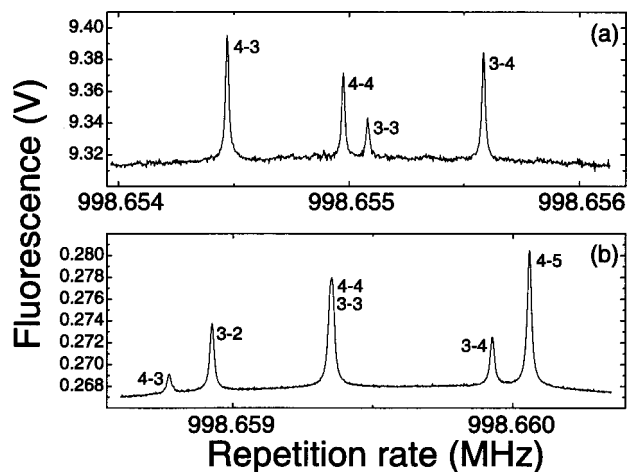


Fig. 2. Fluorescence of the (a) D_1 line and (b) D_2 line measured as a function of the femtosecond laser repetition rate. The laser power is $P=14$ nW per component for (a) the D_1 line and $P=1.5$ nW per component for (b) the D_2 line.

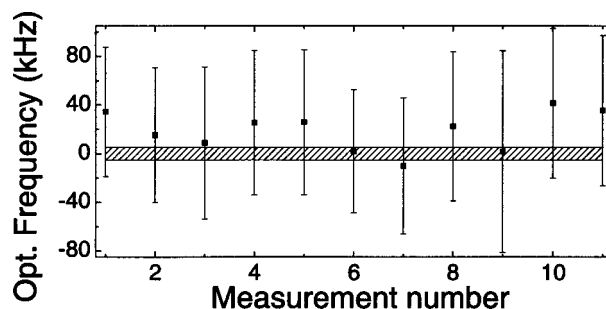


Fig. 3. Absolute frequency of the $6s\ ^2S_{1/2}(F=3) \rightarrow 6p\ ^2P_{1/2}(F=4)$ transition, with the standard deviation for each measurement given as an error bar. (Each measurement requires approximately 15 min to complete.) Previously measured optical frequency⁹ (equal to 335 121 730 483.2 kHz) has been subtracted from the data. The shaded area represents the uncertainty in the previous frequency measurement.

Table 1. Optical Frequencies of $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2,3/2}$ Transitions

Line	$F_g \rightarrow F_e$	Optical Frequency (kHz)	Difference ^a
D_2	3-4	351 730 902 116.9 (34.2)	-16.3
D_2	3-3	351 730 700 766.1 (98.5)	-80.0
D_2	3-2	351 730 549 616.3 (9.7)	-5.2
D_2	4-5	351 721 960 563.5 (4.5)	-22.2
D_2	4-4	351 721 709 471.6 (167.8)	-25.3
D_2	4-3	351 721 508 195.1 (21.7)	-15.4
D_1	3-4	335 121 730 457.3 (16.2)	-25.9
D_1	3-3	335 120 562 753.7 (85.0)	-6.0
D_1	4-4	335 112 537 861.7 (28.0)	7.8
D_1	4-3	335 111 370 121.0 (9.9)	-9.2

^aDifference between these measurements and those given in Refs. 8 and 9.

The measured Lorentzian and Doppler linewidths are consistent with the cw laser experiments.^{8,9} No significant line broadening owing to the comb component linewidth was found. Similar results were obtained for all components of the D_1 and D_2 lines. They are summarized in Table 1. The uncertainty is determined from the fit and the statistics of several measurements. No systematic corrections have been included.

The deviation from the previous values, although not large, is due to several systematic effects. The nonlinear fluorescence background can contribute to systematic shifts by adding an offset to the line centers. The Doppler shift associated with atomic beam-laser beam misalignment is minimized by a simple procedure⁸; however, given the limited signal-to-noise ratio, a residual Doppler shift of several tens of kilohertz may still exist. Finally, extrapolating to zero laser intensity, it was found that the optical frequency values of D_1 components $F_g=3 \rightarrow F_e=4$ and $F_g=4 \rightarrow F_e=3$ would have to be increased by 24.2 and 41.1 kHz, respectively. The uncertainty associated with this correction is of the order of the correction itself owing to a deteriorating signal-to-noise ratio at lower laser intensity. A full evaluation of all system-

atics is beyond the scope of this Letter; however, even with ~ 10 times less data acquired, the present results agree well with the best cw results^{8,9,12} (within the uncertainties) and show great promise for future high-resolution spectroscopy done directly with self-referenced FLFCs.

It is also possible to use one of the optical frequency components as the reference for the f_r of the FLFC instead of the hydrogen maser reference. This is done by stabilizing a mode, $f_N = -f_0 + Nf_r$, of the FLFC to one of the Cs optical resonances f_{opt} . Because all the modes of the FLFC are generated phase coherently, this results in the stabilization of the entire FLFC in addition to $f_r = (f_{\text{opt}} + f_0)/N$, where N is an integer of the order of 300,000. Such an experiment was realized by modulating f_r and using synchronous detection to produce a dispersionlike error signal suitable for steering f_N to match f_{opt} . The signals shown in Fig. 4(a) were measured for the $F_g = 3 \rightarrow F_e = 2$ component of the D_2 line. The error signal was then used to externally change the frequency of the rf synthesizer (which was internally referenced). The frequency of the synthesizer output was then measured against the hydrogen maser, and the fractional frequency uncertainty was computed as shown in Fig. 4(b). The plot shows that a fractional frequency

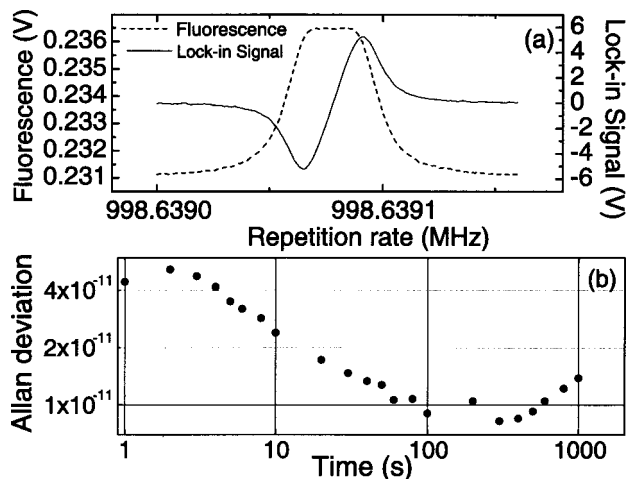


Fig. 4. (a) Error signal used to lock the femtosecond laser repetition rate. A specific FLFC component excites the $F_g = 3 \rightarrow F_e = 2$ transition of the Cs D_2 line. The rf synthesizer frequency controlling the FLFC repetition rate was modulated at a rate of 27 Hz with a 15 Hz depth. Phase detection was used with 5 mV sensitivity and a 1 s time constant. (b) Allan deviation calculated from the data.

instability of better than 1×10^{-11} can be easily achieved in a 100 s integration time. Present systematics limit the uncertainty to 1×10^{-10} (1% of the transition linewidth); however, this should not be viewed as a fundamental limit.

In conclusion, we have demonstrated multifrequency single-photon spectroscopy with a FLFC. Compared to cw laser spectroscopy, this represents a significant experimental simplification while maintaining high precision. Moreover, the entire FLFC has been directly stabilized to a Cs transition, thus providing a convenient, broad, and stable array of absolute optical and microwave frequencies. The achieved uncertainty is already comparable to those of many laboratory standards, but with the advantage of much broader optical and microwave coverage in the frequency output.

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