Two-photon frequency comb spectroscopy of the 6*s*–8*s* transition in cesium

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We report a new absolute frequency measurement of the Cs 6*s*–8*s* two-photon transition measured using frequency comb spectroscopy. The fractional frequency uncertainty is 5×10^{-11}, a factor of 6 better than previous results. The comb is derived from a stabilized picosecond laser and referenced to an octave-spanning femtosecond frequency comb. The relative merits of picosecond-based frequency combs are discussed, and it is shown that the AC Stark shift of the transition is determined by the average rather than the much larger peak intensity. © 2007 Optical Society of America

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Frequency combs are important tools for metrology and spectroscopy.[1–3] Phase coherence between successive laser pulses enables high-accuracy, high-resolution optical frequency measurements. High peak powers make it possible to extend high-resolution spectroscopy into spectral regions that are not accessible by using continuous lasers,[4] enabling a broad range of new measurement possibilities.

In this Letter, we demonstrate the use of a stabilized picosecond (ps) mode-locked laser in conjunction with an octave-spanning femtosecond (fs) comb for precision spectroscopy. While the fs laser is used as an optical clockwork to reference the ps laser to a radio-frequency Cs clock, the modes of the ps laser are used for frequency comb spectroscopy.[5] It has been shown some time ago[6] that this method makes it possible to obtain atomic linewidths limited by the linewidth of a single mode while achieving transition rates determined by the intensity of all modes combined (average intensity). In addition, the AC Stark effect derives from the average laser intensity rather than the peak intensity. In that sense frequency comb spectroscopy combines all positive aspects of cw laser spectroscopy while it allows efficient nonlinear conversion to shorter wavelengths.

Unlike previous approaches we use longer ps pulses for doing spectroscopy rather than fs pulses to provide a greater interaction volume, which is given by the pulse collision region for two-photon Doppler-free excitation with counterpropagating beams. This reduces the time-of-flight broadening, which is inversely proportional to the size of the interaction volume and therefore proportional to the laser bandwidth. The narrower ps spectrum also simplifies the two-photon Doppler-free measurements in Cs and Rb because the comb spectrum does not overlap with one-photon allowed transitions. To demonstrate this we have measured the Cs 6*s*–8*s* two-photon transition improving the accuracy of previous work by a factor of 6.

The ps comb is derived from a Ti:sapphire laser with a 2 ps pulse duration, actively mode-locked with an 82 MHz repetition rate. Two independent lock points are required to stabilize its offset frequency f_{ceo} and the repetition rate f_{rep}.[1] The center frequency is locked to an optical reference cavity[7] feeding back on a piezo mounted folding mirror of the laser. The reference cavity is made from an 18 cm long ultralow-expansion glass spacer horizontally mounted in a temperature-controlled vacuum system with a free spectral range of $10\times f_{rep}$ and a finesse of 1000. The locking system stabilizes a combination of f_{ceo} and f_{rep} but leaves room for small adjustments of one of those frequencies that we use for a second control loop to phase-lock f_{rep} to a Cs-clock-referenced local oscillator. In order to maintain the intrinsic short-term (<1 ms) stability of the ps laser we use a low-bandwidth (<100 Hz) feedback loop for f_{rep}. The long-term drift was measured with the fs comb to be less than 50 Hz/s. The frequency instability of a single comb mode was measured to be less than10 kHz in a 1 s measurement time.

A schematic diagram of our experiment is shown in Fig. 1. The ps comb spectrum is scanned by using an acousto-optic modulator (AOM 2) in a double-pass configuration.[8] AOM 3 is used in zero order to stabilize the laser intensity. After passing through the cell, the laser beam is retroreflected by using a mirror placed $c/2f_{rep}=1.8$ m behind the cell. This ensures that two counterpropagating laser pulses collide in the cell. When Cs atoms are excited to the 8*s* state, a fraction of them decay via the 7*p*-state back to the ground state. Photons emitted by the 7*p*–6*s* decay channel are isolated by using an interference filter

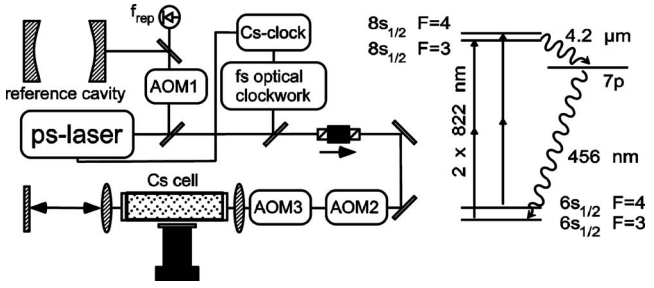

Fig. 1. Experimental setup and partial energy level diagram for Cs. The laser frequency is shifted by using AOM 2, and the intensity is stabilized by using AOM 3. The distance between the mirror and the cell is $c/2f_{rep}$. An optical isolator is used to prevent the backreflected light from going back into the laser.

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and detected by a photomultiplier tube. Not shown in Fig. 1 is a pinhole between the isolator and AOM 2. The counterpropagating laser beam is aligned so that it returns through the pinhole, ensuring that the laser beams are properly aligned and that the wavefronts are matched. Also not shown in the figure is a polarizing beam-splitter cube after the pinhole, which ensures a polarization purity of 1000:1.

The absolute frequency of the laser is measured by using a fs optical clockwork described in detail in Ref. 1. The clockwork consists of a fs laser broadened in a microstructure optical fiber. The offset frequency is detected using an $f-2f$ interferometer. Both the repetition rate and the offset frequency are phase-locked to a Cs atomic clock. A diode laser is phase-locked to one mode in the ps frequency comb. The absolute frequency of the frequency comb is then determined by measuring the beat signal between the laser diode and a mode in the fs-optical clockwork laser.

We measure both the $F'=3$ to $F=3$ and $F'=4$ to $F=4$ hyperfine components of the $6s-8s$ transition (see Fig. 2). For each measurement point, we change the ps comb frequency using AOM 2, allow the system to settle, and simultaneously count fluorescence photons and measure the laser beat frequency. By repeatedly scanning over the atomic transition, we obtain estimates of the statistical uncertainties in our data. For clarity, all frequencies quoted in this paper are measured at the ps comb wavelength of 822 nm.

Because the 1 THz spectral width of the ps comb is greater than the 9 GHz hyperfine splitting, both components appear in Fig. 2(a). The spectrum repeats with a periodicity of $f_{\mathrm{rep}}/2$ because the frequency comb presents a periodic collection of laser modes to excite the spectrum.[6] Doppler-free two-photon absorption occurs every time the frequency of two counterpropagating photons adds up to the total transition energy. This happens when half the atomic frequency coincides with a laser mode, and also when it lies exactly halfway between two laser modes.

An accurate determination of the absolute transition frequency requires careful attention to a number of possible systematic uncertainties such as the pressure shift, Zeeman shift, and the AC stark shift. In addition, one must account for impurities in the vapor cell and understand how experimental artifacts can distort the line shape and add to the overall measurement uncertainty.

The Cs cell is maintained at 22°C to minimize the pressure shift. At this temperature, the Cs pressure is 10^{-6} Torr. Using the previously measured shift of 10 kHz/mTorr for this transition,[9] we expect a shift of 10 Hz. It is also possible for impurities inside the vapor cell to shift the transition frequency. This effect is somewhat reduced due to the gettering action of Cs. The uncertainty stemming from impurities is difficult to estimate, but based on Rb and I_2 cell measurements, it is probably not larger than 10 kHz.[10]

The Zeeman effect can produce a potentially large frequency shift. The double layer of mu-metal shielding on the cell reduces the influence of external fields. We verify this by measuring the transition frequency while applying different magnetic fields. For an applied field of 10 Gauss the line shifted 2 kHz, which is smaller than our measurement uncertainty. When we place a strong rare-earth magnet near the opening in the mu-metal shielding through which we measure fluorescence (2 cm from the interaction region), the line shifts by 20 kHz. Due to the placement of magnetic optical mounts some 10 cm from the cell, we estimate the Zeeman shift to be less than 5 kHz.

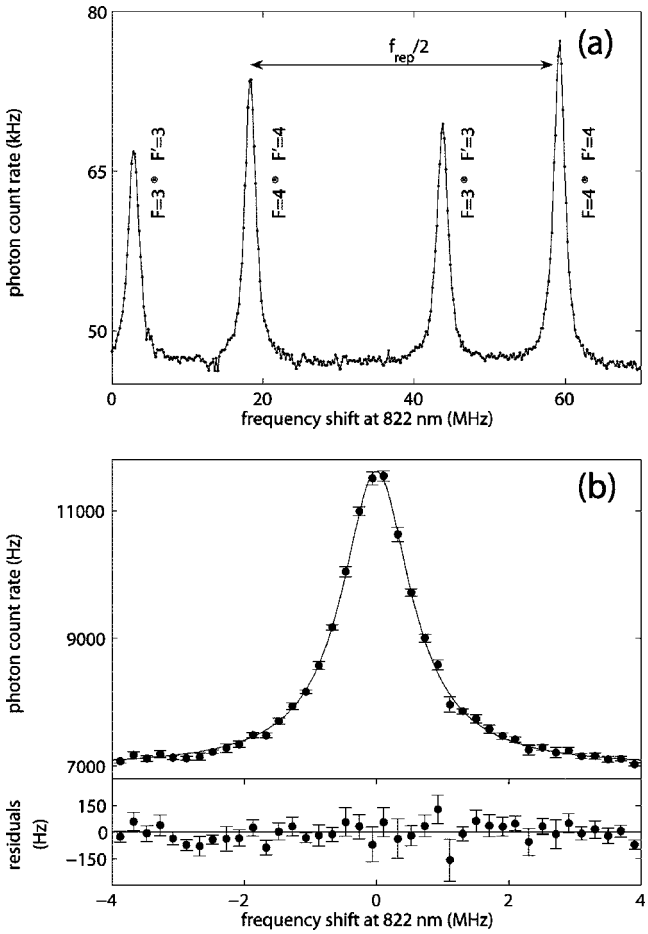

Fig. 2. (a) Fluorescence measurement showing both hyperfine components of the Cs $6s-8s$ transition. Because of the comb structure, the signal repeats itself every $f_{\mathrm{rep}}/2$. (b) $F'=3$ to $F=3$ hyperfine component of the $6s-8s$ transition fitted to a Lorentzian line profile. These data are an average of nine separate scans, each measured at 450 mW laser power, a Gaussian waist of 0.72 mm, and a 1 s integration time per data point. The bottom panel shows the plot residuals as a percentage of the background-subtracted Lorentzian peak height.

We measured the AC Stark effect to shift the $6s-8s$ transition with the average single-beam intensity by $-0.21\ \mathrm{Hz}/(\mathrm{mW/cm^2})$ in agreement with continuous-wave measurements[9] and theoretical values.[11] This demonstrates that the average, rather than the peak power must be used for its evaluation. To minimize this effect, the actual data were taken by using an unfocused laser beam with a Gaussian waist ($1/e^2$ intensity radius) of 0.72 mm. For a laser power of 225 mW incident on the Cs cell, the AC Stark shift is 2.9 kHz. For the data reported here, we measure the transition frequency over a range of laser powers and extrapolate to zero.

At low laser intensities, the line shape is symmetric and is well represented by a Lorentzian [see Fig. 2(b)]. The FWHM of this line is 1.2 MHz (at 822 nm), which is broader than the 920 kHz natural width calculated from theoretical matrix elements.[11] One contribution to this extra broadening is due to the time-of-flight of the Cs atoms through the pulse collision region. This effect may be thought of as having a less than perfect cancelation of the first-order Doppler effect due to the finite linewidth of the ps frequency comb. However, this effect does not shift but broaden the line so that it is more appropriately called time-of-flight broadening. Our detailed line shape model produces a transition linewidth around 960 kHz. The additional broadening is probably due to residual acoustic vibrations of the ps laser cavity that cannot be compensated by the servo systems. A small angle between the counterpropagating beam may cause yet another first-order Doppler effect. Provided that the atomic velocity distribution is isotropic, this should cause no shift but an additional broadening.

Combining these systematic uncertainties with the 10 kHz statistical uncertainty in the line center determination gives an overall uncertainty of 15 kHz, a factor of 6 smaller than previously published results[9]:

$$\nu_{6s-8s}(F=3) = 364\,507\,238\,417\,(15)\ \text{kHz},$$

$$\nu_{6s-8s}(F=4) = 364\,503\,080\,351\,(15)\ \text{kHz}.$$

These values are respectively 97 and 91 kHz higher than previously measured values,[9] but within that measurement's 100 kHz error bar. We measure the splitting between the two hyperfine components to be 4 158 066 (14) kHz, in perfect agreement with Ref. 9 and previous work.[12] Using the accepted value of the ground state Cs hyperfine splitting, we derive a new value for the 8s hyperfine splitting of 876 499 (14) kHz.

In summary, we have measured the absolute frequency of the Cs $6s-8s$ two-photon transition with an uncertainty 6 times smaller than previous work. This is, to our knowledge, the first absolute frequency measurement using a frequency comb derived from a ps mode-locked laser. These lasers can be used to measure narrow atomic transitions and have much higher conversion efficiencies than cw systems, especially as the wavelengths approach the UV cutoff of nonlinear crystals. We anticipate extending this laser system to measure the $1s-3s$ transition in hydrogen at 205 nm to determine the Rydberg constant with greater accuracy, and to provide an important test of QED.[13,14]

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