Direct two-photon resonant excitation and absolute frequency measurement of cesium transitions using a femtosecond comb

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***Abstract*— We measure the optical transition frequencies of the $6s\,^2S_{1/2} \rightarrow 8s\,^2S_{1/2}, 9s\,^2S_{1/2}$, and $7d\,^2D_{3/2,5/2}$ transitions in a ^{133}Cs vapor cell, with an uncertainty < 100 kHz using a femtosecond laser frequency comb.**

Recently, optical frequency metrology using stabilized mode-locked lasers has resulted in the development of new spectroscopic techniques and improved frequency measurements (see e.g. Stowe *et al.* [1]). While many of those experiments have been performed with rather complicated apparatus including MOT's and precision atomic beam sources, we report a relatively simple experimental approach using a frequency comb, to excite a multitude of two-photon transitions in a cesium vapor cell at room temperature. Similar experiments have also recently been reported by Fendel *et al.* [2]. Using this simple apparatus we obtain absolute transition frequencies with uncertainties < 100 kHz, and make marked improvements on the measurement of hyperfine coupling constants. Our apparatus, shown in Fig. 1, is comprised of a cesium vapor cell and a mode-locked femtosecond ring cavity laser based on a Ti:sapphire crystal pumped by a frequency doubled $Nd:YVO_4$, at 532 nm. Each of the $\sim 10^5$, discrete, optical modes in the octave spanning output radiation of the laser is well described by $\nu_n = nf_{rep} + f_0$ [3], where f_{rep} and f_0 are the repetition rate of the laser and the carrier envelope offset frequencies, and n an integer mode identifier. The laser offset frequency is stabilized using the well known self-referencing technique employing a standard $f-2f$ interferometer [4]. The repetition rate of this laser, which is about 1 GHz, is stabilized to a synthesized RF signal using a piezo-electric transducer to control the cavity length. The synthesizer is referenced to a hydrogen maser, allowing for the frequency of each comb mode to be determined with fractional uncertainty $\sim 2\times10^{-13}$. Light from the laser, spanning 600 to 1000 nm, is split by a non-polarizing beam-splitting cube. The beams are then counter-propagated and focussed by $f = 15$ cm lenses to, the cesium vapor cell. Figure 2 summarizes the atomic levels studied in this work, showing the corresponding excitation pathways, and the fluorescence detection channel

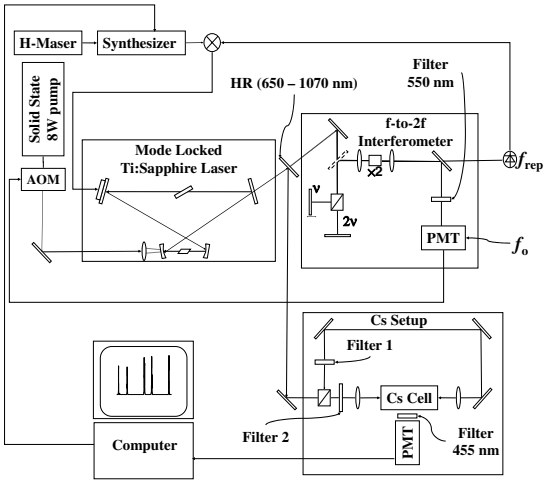

Fig. 1. *The experimental set-up for the cesium two-photon comb spectroscopy.*

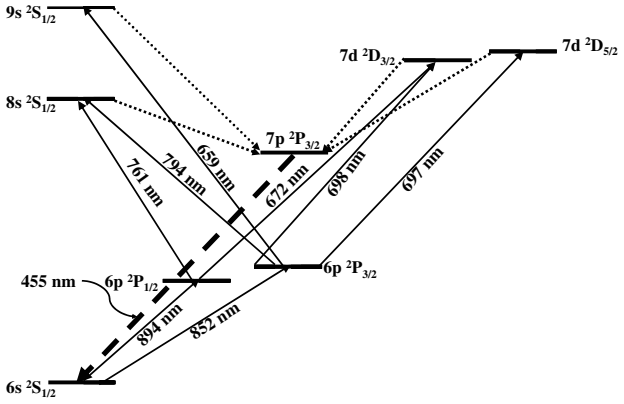

Fig. 2. *The energy level diagram (not drawn to scale) shows the excitation schemes used in this work. Solid lines indicate electric dipole excitation steps, and the dashed and broken lines the decay and the observed fluorescence, respectively. Not shown is the hyperfine splitting of the energy levels.*

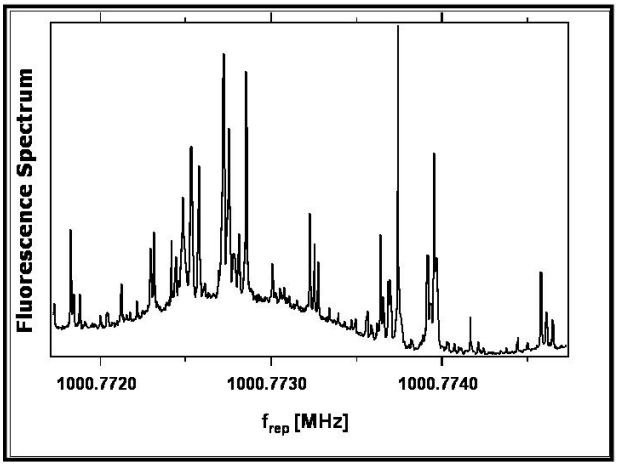

Fig. 3. *The spectrum collected from the two photon excitation with unfiltered light is complicated and has more than 90 peaks. The narrow resonance peaks are a result of a stepwise cascade excitation by counter-propagating light fields, through either of the* 6p *states and the broad Doppler background is two-photon excitation from co-propagating.*

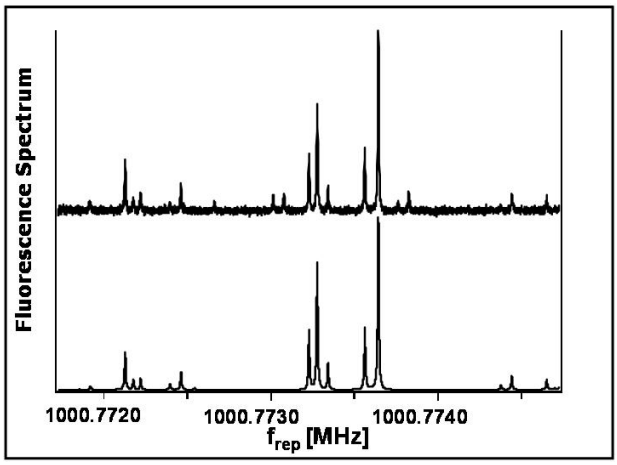

Fig. 4. *Experimental (top) and calculated (bottom) spectra for the* 9S *through the* $6\mathrm{p}\,^2\mathrm{P}_{3/2}$ state.

used for all. Figure 3 shows a spectrum collected by a photo multiplier tube (PMT), adjacent to the cell, which monitored flourescence from the $7\mathrm{p}\,^2\mathrm{P}_{3/2}$ state, at 455 nm. Fluorescence detection at this wavelength minimizes scattering effects on the PMT. The observed spectrum exhibits more than 90 resonance peaks, all of which are a result of the resonant stepwise two-photon excitation of the states $8\mathrm{s}\,^2\mathrm{S}_{1/2}, 9\mathrm{s}\,^2\mathrm{S}_{1/2}$, and $7\mathrm{d}\,^2\mathrm{D}_{3/2,5/2}$ via the $6\mathrm{p}\,^2\mathrm{P}_{1/2,3/2}$ intermediate states. The broad background is a result of two-photon excitation from photons absorbed from a single direction.

We can isolate the excited states and excitation pathways by filtering radiation in the two counter-propagating beams. For example, Fig. 4 shows the 9s spectrum through the $6\mathrm{p}\,^2\mathrm{P}_{3/2}$ state. Also shown is the corresponding calculation using a two-photon formula (see e.g. [5]) averaged over the ground state Maxwellian thermal velocity distribution. In this case, we introduced 850 and 657 nm interference filters with spectral bandwidths of 10 nm into the paths of the counter-propagating beams, thereby dedicating each beam to one step of the $6\mathrm{S} \rightarrow 6\mathrm{P}_{3/2} \rightarrow 9\mathrm{S}$ transitions. The same approach was followed for each of the other states. All the spectra were collected by the scanning of the laser repetition rate (1.000 773 225 GHz) over a 3 kHz range using a 1 Hz step size. The observed variation in signal amplitudes is due to the ground state velocity distribution, the intensity of the relevant comb modes, and the Clebsch-Gordon coefficients for both stages of the excitation.

Lorentzian lineshapes, with linewidths of about 8 Hz in the repetition frequency, were fit to the experimental resonance peaks in all the collected data. These linewidths correspond to ~ 6 MHz in the optical scale and are primarily due to three sources, the two-photon natural width of both the final and intermediate states, and a finite transit time broadening effect. Using non-linear least squares fitting routines we extract the absolute frequencies, and the corresponding hyperfine coupling constants with statistical uncertainties in the $20 \rightarrow 60$ kHz range. We estimate that the final uncertainties including systematic effects, (dynamic Stark shift, Zeeman splitting, beam misalignment and collisional shifts), will be less than 100 kHz [6]. As such, the work reported here could be of benefit to the metrology community, since the two-photon $6\mathrm{s} \rightarrow 8\mathrm{s}$ transition, at 822 nm has been suggested [7] as a secondary standard in the measurement of optical transitions. Experimental uncertainties comparable to the ones obtained using described techniques in this work could be attained by a femtosecond comb referenced to the freely available GPS signals, thus eliminating the need for expensive RF references. Furthermore, there is a need for improved measurements of hyperfine coupling constants which are important for atomic theory [8], and in particular the interpretation of parity non conservation measurements [9], [10].

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