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# Treating cesium resonance lines with femtosecond pulse train

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We present results of our recent experiments where cesium vapor was illuminated by the femtosecond laser frequency comb centered at either D2 line at 852 nm or D1 line at 894 nm (Figure 1.). This action changed the usual Doppler profile into very peculiar profile with periodic structure that can be observed by cw laser scanning probe. The periodic structure reflected the frequency spectrum of the pulse train consisting of a series of fringes separated by the pulse repetition rate. In cesium system the atomic coherence relaxation time is longer than the laser pulse repetition period. Cs atoms interact with the spectrum of the pulse train, and not with the spectrum of a single pulse. This opens up a possibility for the high resolution spectroscopy [1,2], where the observed linewidths are much less than the Fourier transform limit of the individual pulse in the train.

In our recent papers [3,4] we presented the observation of the velocity selective population transfer between the Rb ground state hyperfine levels induced by fs pulse train excitation. We developed a modified direct frequency comb spectroscopy (DFCS) which uses a fixed frequency comb for the $^{85,87}$Rb $5^2S_{1/2}\rightarrow 5^2P_{1/2,3/2}$ excitation (Tsunami mode locked Ti:sapphire laser with pulse duration of <100 fs and pulse repetition rate of 80 MHz) and a weak cw scanning probe (ECDL at 780 nm) for ground levels population monitoring. The Rb($5^2P_{1/2,3/2}$) excited atomic levels have the relaxation times greater than the fs laser repetition period.

Similar experimental setup was used in this work (see Figure 1.). Weak probe laser (ECDL at 852 nm) scanned the population of the ground hyperfine levels in the 5 cm long pure cesium cell. The cell was enclosed in the thermally controllable (with Peltier elements) container enabling both heating and cooling of the sample. The frequency comb of the fs laser was kept fixed during the measurements. The output wavelength of the fs laser was positioned either at the Cs 6 $^2S_{1/2}\rightarrow 6\ ^2P_{1/2}$ transition at 894 nm or at Cs 6 $^2S_{1/2}\rightarrow 6\ ^2P_{3/2}$ transition at 852 nm. The maximum average power used was up to 500 mW and a spectral full width at half of the pulse maximum around 10 nm. It was focused onto the center of the glass cell containing cesium vapor at room temperature with a $f$ =1 m lens producing beam waist of about 300 μm.

Theoretical modeling of the fs-pulse train interaction with the Cs atoms was carried out utilizing standard density matrix analysis. Our starting point is the Liouville equation for the density-matrix elements $\rho_{kl}$:

**Equation 1.** $$\frac{d\rho_{kl}}{dt} = -\frac{i}{\hbar}\langle k|[H,\rho]|l\rangle - \frac{1}{T_{kl}}\rho_{kl},$$

where $H$ is the Hamiltonian of the system and $T_{kl}$ is the relaxation time of the $\rho_{kl}$ density-matrix element. The Hamiltonian of the system is $H=H_0+H_{\text{int}}$, where $H_0$ is the Hamiltonian of the free atom and $(H_{\text{int}})_{kl}=-\mu_{kl}E_T(t)$ represents the interaction of the atom with the pulse train electric field. $\mu_{kl}$ is the dipole moment of the electronically allowed $(F_g\rightarrow F_e=F_g,F_g\pm 1)$ transitions. The pulse train electric field is given by

**Equation 2.** $$E_T(t) = \left[\sum_{n=0}^{N}\varepsilon(t-nT_R)e^{in\Phi_R}\right]e^{i\omega_L t} = \varepsilon_T(t)e^{i\omega_L t}$$

where $N$ is a large integer (order of $10^6$), $\varepsilon(t-nT_R)$ is the slowly varying envelope of the $n$th hyperbolic-secant laser pulse, $\Phi_R$ is the round-trip phase acquired by the laser within the cavity, $T_R$ is the laser repetition period, and $\omega_L$ is the central laser angular frequency. $\varepsilon_T(t)$ is the slowly varying envelope of the pulse train. The pulse train frequency spectrum consists of a comb of N laser modes separated by $1/T_R$ and centered at $\omega_L+\Phi_R/T_R$. The $n$th-mode angular frequency is given by $\omega_n=\omega_L+\Phi_R/T_R\pm 2\pi n/T_R$.

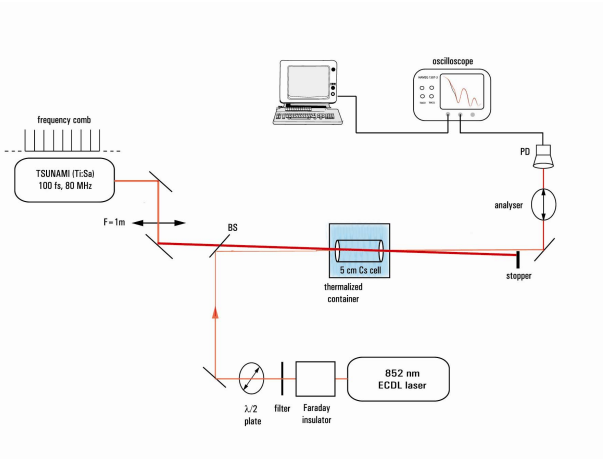


**Figure 1.** Simplified experimental scheme: BS-beam splitter, PD- photodiode.

From Eq. 1 a system of coupled differential equations for the slowly varying density-matrix elements was obtained. Additional terms were included in the equations to account for the repopulation of the ground states due to spontaneous decay from the excited states (repopulation terms) and thermalization of the hyperfine

levels (collisional mixing term). The population of the *k*th atomic level is given by the diagonal density-matrix element $\rho_{kk}$, whereas off-diagonal elements $\sigma_{kl}$, where $\sigma_{kl} = \rho_{kl} e^{-i\omega_L t}$, represent the slowly varying envelope of the coherences.

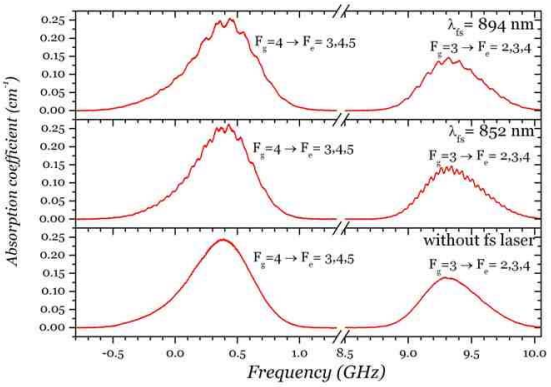


**Figure 2.** Experimentally evaluated absorption coefficient for the Cs 6 $^2S_{1/2}$→6 $^2P_{3/2}$ transition at 852 nm with fs laser at D2 (852 nm) or D1 (894 nm) resonance exhibiting comb-like structure. Absorption without the fs laser influence is also shown on this figure.

For the cesium vapor at room temperature, the inhomogeneous Doppler broadening of about 500 MHz is significantly larger than the homogeneous broadening. Therefore, the atomic transition frequency $\omega_{ge}$ must be replaced with $\omega'_{ge} = \omega_{ge} + \vec{k} \cdot \vec{v}$ where $\vec{k}$ is the laser wave vector and $\vec{v}$ is the atomic velocity. Different velocity groups correspond to different detuning, $\delta = \vec{k} \cdot \vec{v}$, so for a given $\omega_n$ and a given 6 $^2S_{1/2}(F_g)$→6 $^2P_{1/2,3/2}$ ($F_e$) hyperfine transition there is a velocity group ($\delta_n$ detuning), which fulfills $\omega_n = \omega'_{ge}$ resonance condition. Since the pulse train frequency spectrum consists of a comb of laser modes separated by $1/T_R$ (80 MHz), the resonance condition is also satisfied for the velocity groups with detuning $\delta = \delta \pm 2\pi k / T_R$, where $k$ is positive integer. Therefore, different velocity groups are in different situation with respect to the excitation (accumulation) process, which leads to the velocity-selective optical pumping of ground hyperfine levels and velocity selective population in excited hyperfine levels.

To resume, this work shows that it is possible to directly manipulate the fractional populations of hyperfine ground state levels by varying the comb optical frequency spectrum. This could lead to the interesting applications in the systems where Doppler broadening is negligible—for example, ultracold atoms and atomic beam experiments.

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