Velocity Selective Optical Pumping of Rb Hyperfine Lines Induced by a Train of Femtosecond Pulses

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We present direct observation of the velocity-selective optical pumping of the Rb ground state hyperfine levels induced by $5S_{1/2} \rightarrow 5P_{1/2}$ femtosecond pulse-train excitation. A modified direct frequency comb spectroscopy based on the fixed frequency comb and a weak cw scanning probe laser was developed. The femtosecond pulse-train excitation of a Doppler-broadened Rb four-level atomic vapor is investigated theoretically in the context of the density matrix formalism and the results are compared with the experiment.

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Mode-locked, phase-stabilized femtosecond (fs) lasers with high repetition rates produce stabilized wide-bandwidth optical frequency combs [1,2]. It is possible to directly reference [3,4] the comb spacing and position to the microwave cesium time standard, thereby determining the absolute frequencies of all comb lines. Such a series of secondary reference lines that may be extended across the optical spectrum brought revolutionary advances in metrology [5], optical frequency synthesis [6,7], and spectroscopy [8–11].

The phase-stabilized optical frequency combs have been used as a bridge between the fields of high-resolution spectroscopy and ultrafast dynamic [5,12,13]. The coherent accumulation processes in two- and three-level atoms excited by a train of ultrashort pulses, in the case where the atomic relaxation times are greater than the laser repetition period, were observed and theoretically modeled [14,15]. This enabled the measurement of absolute one- and two-photon atomic transition frequencies in laser-cooled ^{87}Rb [16]. It was also shown that the pulse-train effects play an important role in coherent control [17,18].

In this work we present observations of velocity-selective population transfer between the Rb $5S_{1/2}$ hyperfine ground levels. It is achieved by the Rb $5S_{1/2} \rightarrow 5P_{1/2}$ fs excitation, through the optical pumping process. Femtosecond pulse-train excitation leads to population and coherence accumulation effects, which give rise to a velocity-dependent excitation of a Doppler-broadened atomic system, therefore resulting in an efficient velocity-selective optical pumping mechanism. We developed a modified direct frequency comb spectroscopy, using a fixed frequency comb and a weak cw laser which probes the hyperfine ground state population. The interaction of the inhomogeneously broadened (room temperature) four-level ^{85,87}Rb atoms with the fs pulse train was calculated. Excellent agreement between the theoretical and experimental results was obtained.

In the experiment a Tsunami mode-locked Ti:sapphire laser with pulse duration of ~100 fs and pulse repetition of 80 MHz was used. The frequency comb was kept fixed during the measurements. The fs laser positioned on the Rb $5S_{1/2} \rightarrow 5P_{1/2}$ transition at 795 nm (maximum average power up to 700 mW) was focused into the center of the glass cell containing rubidium vapor at room temperature. The cell was 5 cm long with the outer diameter of 2.5 cm. The ^{85,87}Rb $5S_{1/2}$ hyperfine ground state populations were probed with a weak cw diode laser (~2 μW/mm^2), which was propagating nearly collinearly with the fs laser beam, intersecting it under a small angle in the center of the cell. The probe laser (TOPTICA DL 100, ECDL at 780 nm) provided a continuous single-mode tuning range of up to 15 GHz, with a linewidth of the order of 1 MHz. The probe laser frequency was scanned across the Doppler-broadened ^{85,87}Rb $5S_{1/2} \rightarrow 5P_{3/2}$ hyperfine transitions at 0.3 GHz/ms scanning rate. The probe laser transmission was measured with a Hamamatsu Si photodiode and the signal was fed into a digital oscilloscope (Tektronix TDS5104).

Theoretical modeling of the interaction of the fs pulse train with the Doppler-broadened four-level rubidium atoms was carried out utilizing standard density matrix formalism [15,19], starting from the Liouville equation, with appropriate modifications to take into account repopulation of the ground states due to spontaneous decay from the excited states. Additionally, the collisional mixing term is included in the model, allowing the thermalization of the hyperfine levels [19]. The four-level atomic system comprises two $5S_{1/2}$ hyperfine ground levels ($F_g = 2, 3$ for ^{85}Rb, $F_g = 1, 2$ for ^{87}Rb) and two $5P_{1/2}$ hyperfine excited levels ($F_e = 2, 3$ for ^{85}Rb, $F_e = 1, 2$ for ^{87}Rb). The pulse-train electric field is given by

$$E_T(t) = \left[\sum_{n=0}^{N} \varepsilon(t - nT_R) e^{\mathrm{in}\Phi_R}\right] e^{i\omega_L t}, \qquad (1)$$

where N is a large integer (order of 10^6), $\varepsilon(t - nT_R)$ the slowly varying envelope of the nth hyperbolic-secant laser pulse, Φ_R the round-trip phase acquired by the laser within the cavity, T_R the laser repetition period, and ω_L the laser frequency. 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 nth mode frequency is given by $\omega_n = \omega_L + \Phi_R/T_R \pm 2\pi n/T_R$. The Hamiltonian of the system is $H = H_o + H_{\text{int}}$, where H_o is the Hamiltonian of the free atom and $(H_{\text{int}})_{kl} = -\mu_{kl}E_T(t)$ $(k, l = 1, 2, 3, 4)$ represents the interaction of the atom with the pulse-train electric field. μ_{kl} is the dipole moment of the electronically allowed ($\Delta F = 0, \pm 1$) transitions, deduced from Ref. [20]. A system of 10 coupled differential equations for the slowly varying density matrix components is obtained. The atomic level populations are given by the diagonal density matrix elements, whereas off-diagonal elements represent the coherences. The hyperfine excited level lifetimes, which enter the equations through the excited level relaxation terms and ground level repopulating terms, are calculated from the $5P_{1/2}$ lifetime $T = 27.7$ ns [21]. $5P_{1/2}$ coherence lifetime at low vapor density is equal to $2T$ (55.4 ns). The collisional mixing term is given by the product of the collision cross section [22], average atom velocity, and the atomic number density. In our experimental conditions it is about 8 kHz, which is much smaller than the relaxation and repopulation rates. On that account a significant optical pumping of the hyperfine ground levels can be achieved.

Typical time evolution of the ^{87}Rb$(5S_{1/2}, 5P_{1/2})$ hyperfine level populations are shown in Fig. 1. The calculations were performed for the electric field amplitude 1.5×10^6 V/m, $T_R = 12.5$ ns, $\Phi_R = 0$, and ω_L equal to ^{87}Rb $F_g = 1 \rightarrow F_e = 2$ transition frequency. Because of the pulse repetition period T_R, which is smaller than the relaxation times of the system, the system can never completely relax between two consecutive laser pulses. Therefore, the atoms accumulate excitation in the form of coherence and excited state population, as observed in the inset of Fig. 1(a) (ρ_{44}). A stationary state is achieved on a time scale of 10 μs with the $5P_{1/2}$ population of less than 1%. This is a result of the optical pumping process in the four-level system without optically closed transitions and the relatively weak electric field. The significant difference in the fractional hyperfine ground state populations, due to the optical pumping, is clearly seen in Fig. 1(b). Time $\tau = 1.25$ μs indicated in Fig. 1 represents the average interaction time of the atoms with the fs laser (calculated from the fs laser beam diameter and the most probable speed of the Rb atoms). Therefore, in the analysis of experimental results we use the populations at time τ as level populations, rather than stationary state populations. In the case of ^{85}Rb the populations exhibit similar time evolutions, with the $5P_{1/2}$ excited state population of about 1%.

For the rubidium vapor at room temperature, the inhomogeneous Doppler broadening (~500 MHz) is significantly larger than the homogeneous broadening. Therefore, the atomic transition frequency ω_{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

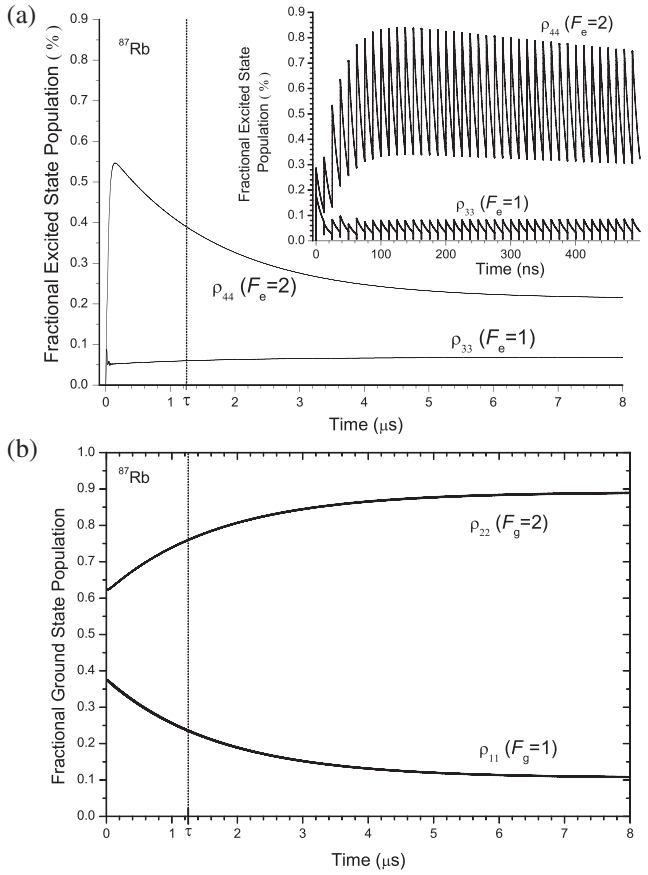

FIG. 1. Typical time evolution of the ^{87}Rb hyperfine state populations, presented as a time average per laser repetition period. $\tau = 1.25$ μs represents the average interaction time. (a) ^{87}Rb$(5P_{1/2})$ populations. Inset: The results of the direct numerical integration. (b) ^{87}Rb$(5S_{1/2})$ populations.

ω_n and a given $5S_{1/2}(F_g) \rightarrow 5P_{1/2}(F_e)$ hyperfine transition there is a velocity group (δ_n detuning) which satisfies the $\omega_n = \omega_{ge}{}'$ 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 velocity groups with detuning $\delta = \delta_n \pm 2\pi m/T_R$, where m is a positive integer, corresponding to resonance with different laser modes. Therefore, different velocity groups are in different situations with respect to the excitation (accumulation) process, which leads to the velocity-selective optical pumping. The variations of ρ_{11} and ρ_{22} fractional ground state populations for different atomic velocity groups (different detuning δ), weighted by the Doppler profile, are shown in Fig. 2. The level populations vary with an 80 MHz period, which is a direct consequence of the comb frequency spectrum. For the ^{87}Rb, $F_g = 1$ ($F_g = 2$) population in one period exhibits two maxima (minima) separated by 12 MHz and one minimum (maximum). This is a consequence of the $5S_{1/2}$ and $5P_{1/2}$ hyperfine splittings and relative transition probabilities [inset of Fig. 2(a)]. Two maxima in ρ_{11} come from $F_g = 2 \rightarrow F_e = 1, 2$ transitions, separated by

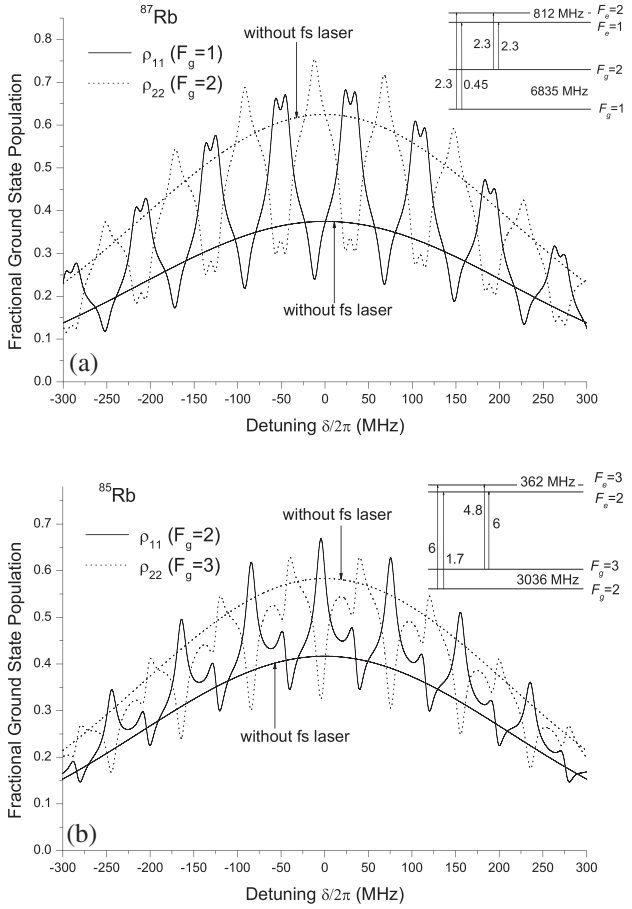

FIG. 2. The variations of the ground state populations ρ_{11} (solid line) and ρ_{22} (dotted line) for different atomic velocity groups with and without fs laser excitation. (a) ^{87}Rb. (b) ^{85}Rb. Insets: Schemes of the $5S_{1/2}$-$5P_{1/2}$ four-level system with the corresponding relative transition probabilities.

812 MHz. However, because of the 80 MHz laser mode separation in the comb spectrum, the resonance condition for these two transitions is obtained for $\delta = 12$ MHz. The minimum in ρ_{11} corresponds to the $F_g = 1 \rightarrow F_e = 2$ transition. Because of the small relative transition probability of the $F_g = 1 \rightarrow F_e = 1$ transition, there is no significant optical pumping effect connected to this excitation. The $F_g = 2(F_g = 3)$ populations for ^{85}Rb [Fig. 2(b)] exhibit in one period two maxima (minima) and one minimum (maximum). The $5S_{1/2}$ hyperfine splitting is 3036 MHz, nearly a multiple of 80 MHz. Therefore, for one velocity group, when the laser is resonant with the $F_g = 2 \rightarrow F_e = 3$ transition (minimum in ρ_{11}), it is 4 MHz off resonance with respect to the $F_g = 3 \rightarrow F_e = 3$ transition (smaller maximum in ρ_{11}). The second maximum in ρ_{11} comes from the $F_g = 3 \rightarrow F_e = 2$ transition, whereas the influence of the $F_g = 2 \rightarrow F_e = 2$ transition is not observed due to the small transition probability.

The velocity-selective observation of hyperfine ground state populations is achieved by monitoring the weak cw probe laser transmission during continuous scan across all four Doppler-broadened $5S_{1/2} \rightarrow 5P_{3/2}$ absorption lines at 780 nm. In the weak field approximation and linear absorption regime, the measured optical thickness is directly proportional to the ρ_{11} and ρ_{22} ground state populations. The absorption spectrum consisting of four lines is obtained in one probe laser scan, two of them resulting from ^{85}Rb absorption and the other two from ^{87}Rb absorption. The lines are presented separately in Figs. 3 and 4. For the theoretical simulation of measured absorption lines, ^{85,87}Rb ρ_{11} and ρ_{22} hyperfine ground state populations shown in Fig. 2 were used. For each hyperfine transition, we calculated the convolution of the velocity distribution of the ground state population with the Lorentzian profile of natural linewidth. One absorption line is calculated by adding the contributions of three hyperfine components [23]. The theoretical results show excellent agreement with the experiment, as seen from Figs. 3 and 4. The modulations in the absorption line profiles are significantly larger for the ^{87}Rb isotope. If the frequency separation of three hyperfine lines forming an absorption line is favorable, as is the case for ^{87}Rb, then the absorption line exhibits stronger modulations. The actual efficiency of the velocity-selective optical pumping is similar for both isotopes (as seen on Fig. 2).

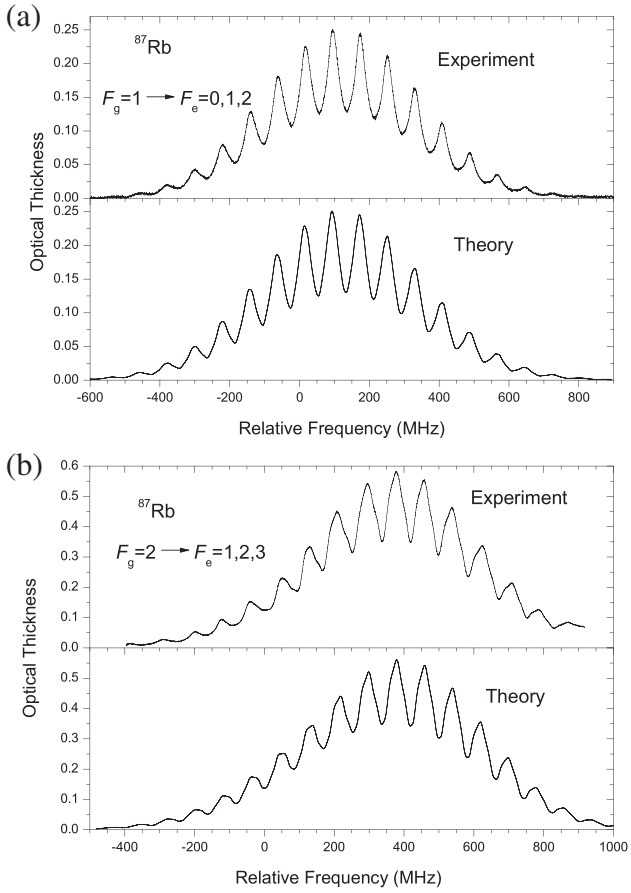

FIG. 3. The comparison of the measured and calculated ^{87}Rb $5S_{1/2} \rightarrow 5P_{3/2}$ hyperfine absorption line profiles: (a) $F_g = 1 \rightarrow F_e = 0, 1, 2$ line; (b) $F_g = 2 \rightarrow F_e = 1, 2, 3$ line.

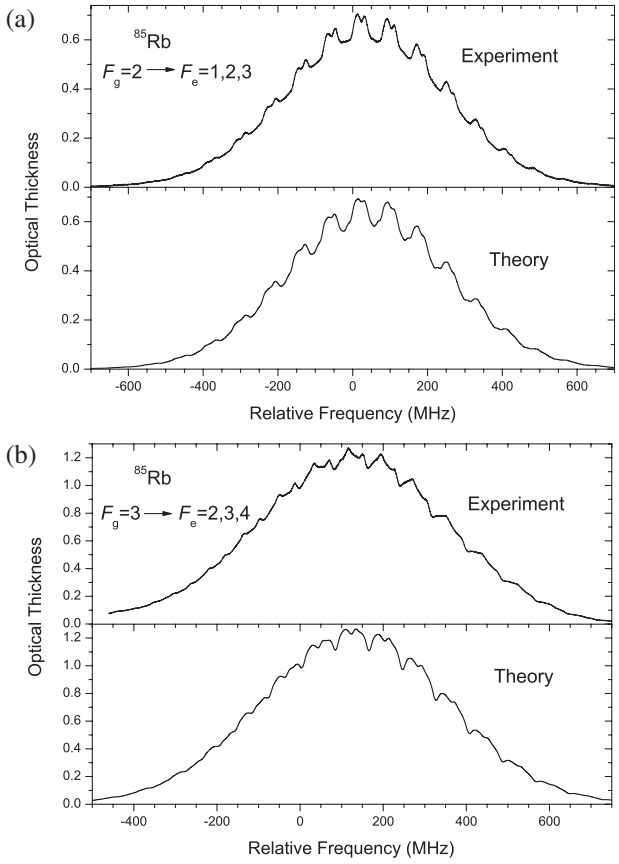

FIG. 4. The comparison of the measured and calculated ^{85}Rb $5S_{1/2} \rightarrow 5P_{3/2}$ hyperfine absorption line profiles: (a) $F_g = 2 \rightarrow F_e = 1, 2, 3$ line; (b) $F_g = 3 \rightarrow F_e = 2, 3, 4$ line.

Our last measurements show that the modulations of the ^{85}Rb absorption line profiles are strongly increased for the fs laser adjusted on the $5S_{1/2} \rightarrow 5P_{3/2}$ resonance transition (780 nm), whereas the modulations of the ^{87}Rb absorption line profiles do not change significantly. Also, the observed modulations exhibit an interesting complex behavior when an external homogeneous magnetic field is applied. These results are currently under thorough analysis and will be reported in a separate publication.

In conclusion, we measured the velocity-dependent Rb hyperfine ground state population transfer induced by $5S_{1/2} \rightarrow 5P_{1/2}$ fs pulse-train excitation. We theoretically modeled the excitation of the Rb four-level system by the fs pulse train and calculated the Rb hyperfine ground state populations. As a result of the velocity-selective optical pumping, these populations exhibit a unique oscillatory structure with the period equal to the fs laser repetition frequency. Based on the calculated hyperfine ground state populations, we performed the simulation of the measured modulations in the $5S_{1/2} \rightarrow 5P_{3/2}$ hyperfine absorption lines. We obtained excellent agreement between theory and experiment.

We foresee an application of the results of this work in the field of spectroscopy of ultracold atoms and the atomic beam experiments. In the systems where Doppler broadening is negligible, by varying the comb optical frequency spectrum it is possible to directly manipulate the fractional populations of hyperfine ground state levels. Our calculations (applicable to our experimental conditions) show that by introducing $\Phi_R = \pi$ pulse phase difference (40 MHz frequency shift), the fractional ^{87}Rb($5S_{1/2}$) ground state population changes by a factor of about 2.5 [see Fig. 2(a)]. We expect that in the optimized experimental conditions this factor could be increased significantly and thus would enable atomic switching effect with possible applications in quantum computing [24–26].

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