Coherent population trapping in ^{87}Rb atoms induced by the optical frequency comb excitation

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The excitation of room-temperature four-level ^{87}Rb atoms by a train of ultrashort pulses is investigated theoretically in the conditions when the pulse repetition period is shorter than the characteristic atomic relaxation times. It is shown that coherent accumulation of excitation leads to coherent population trapping and electromagnetically induced transparency of the excitation pulses when the pulse repetition rate is a subharmonic of the ground-state hyperfine splitting. It is illustrated how the judicious choice for the frequency comb parameters can provide a means to effectively control the degree of coherence between the ground-state hyperfine levels for selective atomic groups, and even transfer the whole atomic distribution to the dark state with up to 95% efficiency.

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Coherent preparation of the quantum states of atoms by laser light can lead to quantum interference in the amplitudes of optical transitions. The optical properties of a medium can be dramatically modified in this way, opening new possibilities in nonlinear optics [1] and quantum information science [2,3]. Coherent population trapping (CPT) and electromagnetically induced transparency (EIT) recently attracted a lot of attention due to their ability to suppress linear absorption and enhance the nonlinear response of a resonant medium [4,5]. In EIT, the atomic medium is made transparent to a resonant probe field by means of a coupling field tuned to a linked transition. The two excitation laser fields create destructive interference between excitation pathways and a dark superposition state is formed, with the population reduced in the upper state and trapped within the two ground states [4]. Following the early work of Harris *et al.* [6], EIT has been demonstrated in various experiments: in continuous and pulsed regimes [7,8], with room-temperature atomic gasses [9] or cold atoms [10], with solids doped by rare-earth-metal ions [11] and semiconductor quantum wells [12], and for wavelengths ranging from microwaves [13] to γ rays [14].

It was theoretically predicted by Kocharovskaya and Khanin [15] that a train of ultrashort pulses may also induce CPT when interacting with a three-level Λ system. They showed that if the pulse repetition frequency is matched to a subharmonic of the frequency splitting of the ground state, a sufficiently intense pulse may excite coherence and the medium becomes transparent to the laser field. EIT in resonant rubidium atomic vapor using a mode-locked diode laser (generating a train of picosecond pulses) was reported in Ref. [16]. Arissian and Diels [17] reported on the repetition rate spectroscopy of a dark-line resonance in rubidium. They performed spectroscopy of a Λ system in ^{87}Rb with a mode-locked laser and observed a dark line, resulting from population trapping between the hyperfine levels, when the repetition rate is $1/57$th of the hyperfine splitting. More recently, Soares and de Araujo [18] theoretically investigated EIT of an ultrashort pulse train interacting with a degenerate three-level Λ system in the conditions when the pulse repetition period is shorter than the excited-state lifetime.

This Brief Report theoretically investigates EIT of a train of femtosecond pulses interacting with the realistic, four-level ^{87}Rb system (Fig. 1), in the conditions when the pulse repetition period is shorter than the characteristic atomic relaxation times. A special case is considered, when the pulse repetition frequency f_{rep} is a subharmonic of the hyperfine splitting of the ground state, $\omega_{12} = 2\pi f_{12}$. A Λ-type excitation scheme typical for EIT experiments is formed in this way, with two different modes within the frequency comb in resonance with two transitions that share a common excited hyperfine level. For the case of ^{87}Rb room-temperature vapor a double Λ-type excitation scheme can be achieved, corresponding to two excited-state hyperfine levels. An iterative analytic solution for the time dynamics of the ^{87}Rb four-level atoms excited by an ultrashort pulse train, presented in detail in Ref. [21], is utilized to study coherent accumulation of excitation in the system. This approach enables the calculation of complete time dynamics of ^{87}Rb atoms subject to optical frequency comb excitation taking into account the velocity distribution of atoms (as in room-temperature vapor), thus enabling a realistic prediction of the experimental results.

Coherent accumulation of excitation occurs as a result of the specific atomic time dynamics when the atomic relaxation times are longer than the pulse repetition period [21–27]. Since the atoms cannot completely relax between two consecutive pulses, they accumulate excitation in the form of coherence (inset in Fig. 2) and excited-state population [21,22]. As seen in Fig. 2 for $f_{\text{rep}} = f_{12}/85$, the system finally reaches a stationary state close to the full coherence ($|\sigma_{12}| = 0.5$) after a large number of pulses, with a substantial increase in the final ground-state coherence with respect to the single-pulse excitation [21,22]. In the frequency domain this corresponds to interaction of atoms with the frequency comb rather than with the spectrum of a single pulse. A strong reduction of the stationary state coherence is observed when f_{rep} is detuned from $f_{12}/85$ by 10 kHz. In general, significant ground-state coherence can only be induced if f_{rep} is equal to (or $<$10 kHz detuned from) a subharmonic of the ground-state hyperfine splitting.

The accumulation of ground-state coherence is followed by the reduction of absorption of the pulses and is not specific to the case of $f_{\text{rep}} = f_{12}/85$. As seen in Fig. 3, a series of dark lines is obtained as f_{rep} is scanned, with each of the dark lines corresponding to a particular subharmonic of the ground-state hyperfine splitting (as indicated by the top x axis in the figure). Indeed, the dark line is a general coherence feature observed

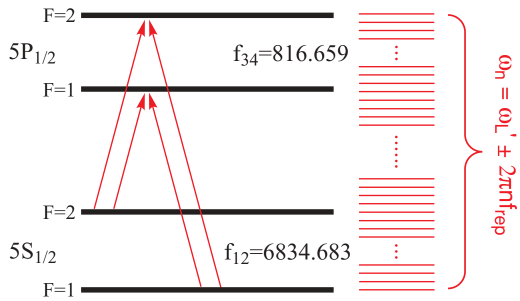

FIG. 1. (Color online) Schematic ^{87}Rb energy-level diagram for the Λ-type system induced by the frequency comb excitation with $f_{\rm rep}$ tuned to a subharmonic of the hyperfine splitting of the ground state. Hyperfine frequency splittings are indicated in megahertz [19,20].

when the repetition frequency of the pulse train equals a submultiple of the ground-state hyperfine splitting [17].

$\overline{\rho_{33}}$ and $\overline{\rho_{44}}$ populations were calculated by taking the contributions of all atomic velocity groups and are proportional to the fluorescence induced by the pulses. Doppler broadening does not affect the dark-line feature, which is in accordance with the results of Ref. [17]. $\overline{\rho_{33}}$ and $\overline{\rho_{44}}$ populations are also independent of the frequency shift of the comb modes (i.e., frequency shift of ω_L'), since it induces only the shift of ρ_{33} and ρ_{44} atomic velocity distributions without the change in the shape of the distributions [21,28]. Therefore there is no need for the offset frequency stabilization in the dark-line measurements [17]. As typically observed in EIT experiments, the width of the dark line increases when the laser power is increased.

The results of Fig. 3 can be understood by invoking a so-called dark state, a linear combination of the ground-state levels which is decoupled from the excitation pulses. A bright state, driven by the excitation pulses, is also introduced. By following Ref. [16] and considering the Λ system related to the ^{87}Rb $5P_{1/2}$ $F=2$ excited level, bright and dark states are defined as $|B\rangle = (|1\rangle + |2\rangle)/\sqrt{2}$ and $|D\rangle = (|1\rangle - |2\rangle)/\sqrt{2}$, where $|1\rangle$ and $|2\rangle$ are ^{87}Rb $5S_{1/2}$ $F=1$ and $F=2$ ground levels, respectively. Bright and dark state populations are then given by $\rho_B = (\rho_{11}+\rho_{22})/2 + \mathrm{Re}(\sigma_{12})$ and $\rho_D = (\rho_{11}+\rho_{22})/2 - \mathrm{Re}(\sigma_{12})$, and they essentially follow the ground-state coherence (since $\rho_{33}, \rho_{44} \ll 1$ and $\rho_{11}+\rho_{22} \approx 1$). Both states start as equally populated ($\rho_B = \rho_D = 0.5$) and upon excitation by a sequence of pulses the population is optically pumped from the bright to the dark state. Coherent accumulation of excitation eventually leads to the complete population transfer to the dark state. Simultaneously, the population of the excited state goes toward zero, the pulses are no longer absorbed, and EIT takes place.

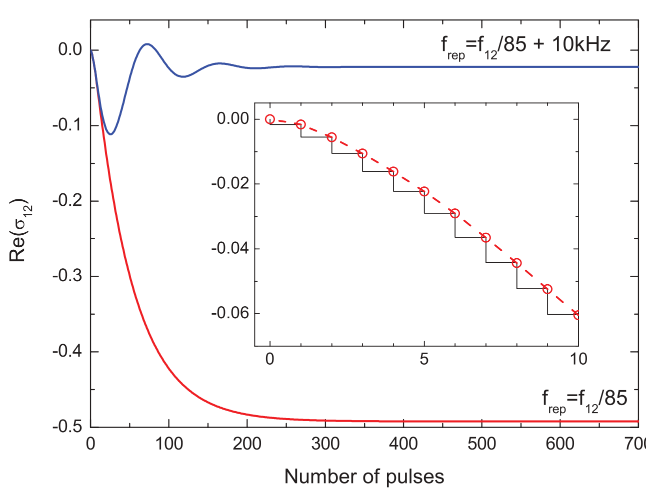

FIG. 2. (Color online) Accumulation of coherence between the ^{87}Rb ground-state hyperfine levels induced by the resonant frequency comb excitation. Analytical solutions for $f_{\rm rep} = f_{12}/85$ and $f_{\rm rep} = f_{12}/85 + 10$ kHz are shown. Inset: Comparison of the numerical (solid line) and analytical (points, dashed line) short-time-scale solution for $f_{\rm rep} = f_{12}/85$. The calculations were performed for a $v=0$ group of atoms with the following pulse train parameters: pulse peak amplitude of $\mathcal{E}_0 = 1 \times 10^6$ V/m, pulse duration of $\tau_p = 100$ fs, and a laser central frequency ω_L' in resonance with the ^{87}Rb $5S_{1/2}$ $F=2 \rightarrow 5P_{1/2}$ $F=2$ transition.

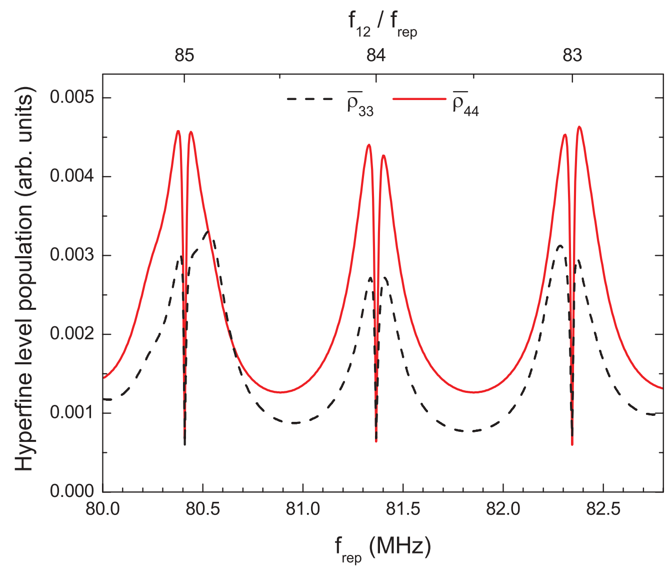

FIG. 3. (Color online) ^{87}Rb $5P_{1/2}$ $F=1$ (ρ_{33}, dashed line) and $F=2$ (ρ_{44}, solid line) excited-state populations, averaged over the atomic velocity ($\overline{\rho_{33}}, \overline{\rho_{44}}$), as a function of pulse repetition frequency. Pulse train parameters used were the same as in Fig. 2, with $\mathcal{E}_0 = 2 \times 10^6$ V/m.

The velocity distribution of dark state population [Fig. 4(a)] follows the general periodic feature typical for frequency comb excitation of room-temperature atoms [21–24,26], with the period equal to $f_{\rm rep}$ (i.e., the frequency separation of the comb modes). The velocity-averaged dark state population as a function of $f_{\rm rep}$ [Fig. 4(b)] shows alternate minima and maxima appearing approximately with every eighth subharmonic of f_{12}. This behavior is independent of ω_L' (i.e., carrier-envelope offset frequency), in the same way as $\overline{\rho_{33}}$ and $\overline{\rho_{44}}$ are independent of ω_L'. The effects of ω_L' only occur for $f_{12}/f_{\rm rep} < 20$, when $f_{\rm rep}$ is comparable to the Doppler width.

Comparison of the results for ρ_D and $\overline{\rho_D}$ shows that maxima in $\overline{\rho_D}$ correspond to ρ_D distributions with almost negligible modulations. The minima in $\overline{\rho_D}$ on the other hand correspond to ρ_D distributions with the most pronounced modulations. Two typical representatives of ρ_D distributions that correspond to the minima and maxima in $\overline{\rho_D}$ are shown in Fig. 5. The ρ_D

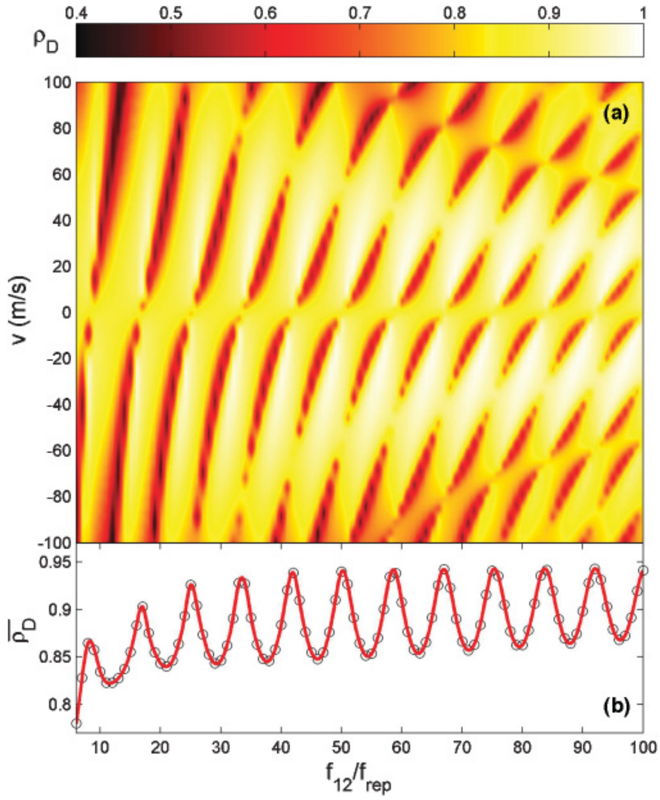

FIG. 4. (Color online) Stationary dark state population as (a) a function of atomic velocity and (b) averaged over the atomic velocity for different f_{rep} given as subharmonics of the ground-state hyperfine splitting. For clarity, only the central part of the velocity distribution is shown. $\mathcal{E}_0$ was scaled to correspond to the same-laser time-averaged power, given by $\mathcal{E}_0 = 2 \times 10^6$ V/m at $f_{\text{rep}} = f_{12}/85$. ω_L' was set in resonance with the ^{87}Rb $5S_{1/2}$ $F = 2 \rightarrow 5P_{1/2}$ $F = 1$ transition for all f_{rep}.

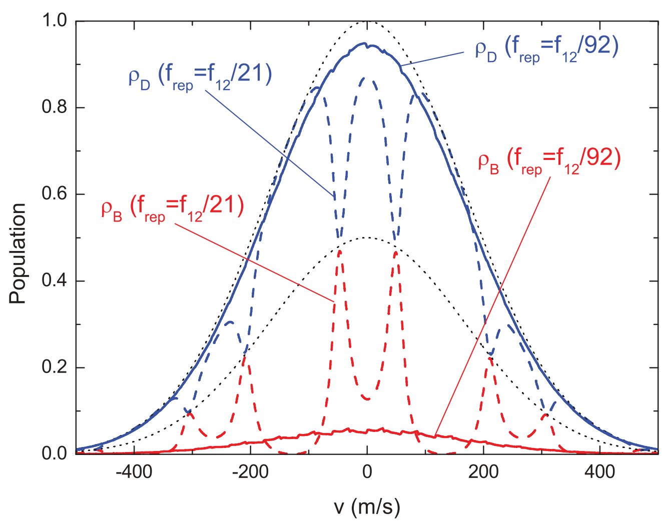

FIG. 5. (Color online) Stationary bright and dark state populations as a function of atomic velocity for $f_{\text{rep}} = f_{12}/21$ (dashed line) and $f_{\text{rep}} = f_{12}/92$ (solid line). Velocity distributions of the initial and maximum dark state populations are indicated as a reference (dotted line).

population distribution for $f_{\text{rep}} = f_{12}/92$ completely follows the initial Gaussian velocity distribution of atoms, with almost all of the population (95%) transferred to the dark state. A similar behavior is observed for all f_{rep} that produce a maximum in $\overline{\rho_D}$ (e.g., $f_{\text{rep}} = f_{12}/84, f_{12}/75, f_{12}/67, f_{12}/59, \ldots$; see Fig. 4). For these f_{rep} a double-Λ-type excitation scheme (see Fig. 1) is achieved for a particular velocity group of atoms, which can only be obtained when f_{rep} is simultaneously a subharmonic of the ground- and excited-state hyperfine splitting ($f_{12}/92 \approx f_{34}/11$). The periodic features in $\overline{\rho_D}$ as a function of f_{rep} are therefore related to the ratio of the ground- and excited-state hyperfine splittings ($f_{12}/f_{34} \approx 8.4$).

The physical mechanism responsible for the obtained dark state distribution can be clarified by inspecting the time evolution of the dark state formation [Fig. 6(b)]. Upon excitation by discrete optical frequencies of the frequency comb, the ρ_D distribution initially (short time scale) shows comblike structure, which corresponds to the buildup of ground-state coherence for the resonant velocity groups of atoms. Quite surprisingly, as the number of excitation pulses is increased, the ground-state coherence builds up also for the nonresonant atomic velocity groups, which eventually leads to the Gaussian-shaped dark state population distribution.

Qualitatively different dark state distributions are obtained when $\overline{\rho_D}$ minima are considered. The ρ_D population distribution for $f_{\text{rep}} = f_{12}/21$ [Figs. 5 and 6(a)] exhibits strong

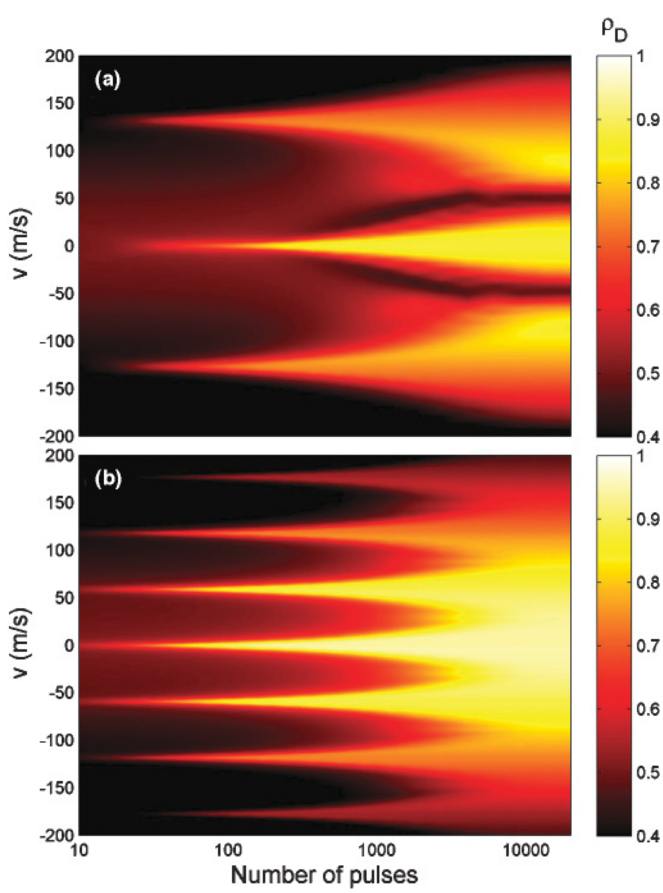

FIG. 6. (Color online) Time dynamics of the dark state population as a function of atomic velocity for (a) $f_{\text{rep}} = f_{12}/21$ and (b) $f_{\text{rep}} = f_{12}/92$.

modulations with the efficient population transfer to the dark state for the resonant velocity groups. Similar behavior is observed for all f_{rep} that produce minima in $\overline{\rho_D}$ (e.g., $f_{\text{rep}} = f_{12}/29, f_{12}/38, f_{12}/46, f_{12}/54, \ldots$; see Fig. 4). For these f_{rep}, when the Λ-type excitation scheme related to the $5P_{1/2}$ $F = 1$ level is in resonance with a particular velocity group of atoms, it is simultaneously as far from resonance as possible ($f_{\text{rep}}/2$) with respect to the Λ-type excitation scheme related to the $5P_{1/2}$ $F = 2$ level, and vice versa. Since the shape of the modulations in ρ_D distributions is generally independent of ω'_L, adjustment of the carrier-envelope offset frequency can be used as the means to control the level of ground-state coherence for a selected velocity group of atoms. As illustrated in Fig. 5, the full range from no to full ground-state coherence can be induced for the selected atomic velocity group by changing the carrier-envelope offset frequency of the frequency comb.

In conclusion, a realistic atomic four-level model was used to study the interaction of room-temperature ^{87}Rb vapor with a train of ultrashort pulses. The accumulation of coherence between ground-state hyperfine levels was shown to occur in the special case when the pulse repetition frequency is a subharmonic of the ground-state hyperfine splitting (Fig. 2), leading to the formation of a dark state. The process effectively leads to electromagnetically induced transparency of the excitation pulses. Calculated room-temperature ^{87}Rb vapor fluorescence spectra reveal a series of dark lines as f_{rep} is scanned, with each corresponding to a subharmonic of the ground-state hyperfine splitting (Fig. 3). These resonances can be exploited to stabilize the repetition frequency of the frequency comb since the dark-line spectral features are independent of the exact comb mode frequencies.

The results illustrate how the judicious choice of the frequency comb parameters can provide means to effectively control the degree of coherence between the ground-state hyperfine levels for selective velocity groups of atoms (Fig. 5). Moreover, it was demonstrated that the full atomic velocity distribution of room-temperature ^{87}Rb atoms (up to 95% of atoms) can be transferred to the dark state by adjusting the pulse repetition frequency to be simultaneously a subharmonic of the ground- and excited-state hyperfine splitting (double-Λ-type system).

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