High Contrast Ramsey Fringes with Coherent-Population-Trapping Pulses in a Double Lambda Atomic System

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We report the observation of Raman-Ramsey fringes using a double lambda scheme creating coherent population trapping in an atomic ensemble combined with pulsed optical radiations. The observation was made in a Cs vapor mixed with N_2 buffer gas in a closed cell. The double lambda scheme is created with lin $\perp$ lin polarized laser beams leading to higher contrast than the usual simple lambda scheme. The pulsed trapping technique leads to narrow fringe widths scaling as $1/(2T)$ with high contrasts which are no longer limited by the saturation effect. This technique operates in a different way from the classical Ramsey sequence: the signal is done by applying a long trapping pulse to prepare the atomic state superposition, and fringe detection is accomplished by optical transmission during a short second trapping pulse without any perturbation of the dark state.

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Continuous stimulated Raman resonant excitation [coherent population trapping (CPT)] [1–4] in vapor cells with buffer gas has been investigated in several groups for many years in view of application to atomic clocks. Continuous CPT interrogation techniques have led to narrow linewidths down to 50–25 Hz [5,6] and relative clock frequency stability as low as 1.3×10^{-12} for 1 s integration time with detection of the transmitted light [7] and 3.2×10^{-12} at 1 s for the CPT maser detecting the hyperfine coherence in a microwave cavity [8].

Fundamental limitations for achieving very narrow atomic lines in these experiments come from saturation and optical pumping effects. In a continuous interrogation, the saturation intensity is very weak ($\sim 1\ \mu\mathrm{W/cm^2}$). As a consequence, the low laser intensity required to avoid CPT line broadening leads to a poor signal-to-noise ratio. Circularly polarized coherent radiation realizes the optical Λ scheme connecting two ground states $|F, m_F = 0\rangle \equiv |1\rangle$, $|F' = F + 1, m_F = 0\rangle \equiv |2\rangle$ called clock states and an excited state $|F_e, m_{F_e}\rangle \equiv |3\rangle$ as shown on Fig. 1 (left). On resonance, they create the dark state, which is a particular superposition of the ground states. According to polarized light helicity, two different Λ schemes can be realized with $|F_e, m_{F_e} = -1\rangle \equiv |3\rangle$ or $|F_e, m_{F_e} = +1\rangle \equiv |4\rangle$. For typical values of the static magnetic field ($\sim 10\ \mu$T), the Zeeman splitting between these two upper levels is small (~ 25 kHz) compared to their width which is broadened by collisions in the buffer gas (~ 625 MHz) and makes these levels degenerate. The buffer gas confines the atoms in a region smaller than the microwave excitation field wavelength (Dicke regime [9]) canceling the transit time and Doppler broadening. Such Λ schemes are not closed systems: because of optical pumping effects, there is leakage of atoms towards the extreme Zeeman sublevels reducing the number of atoms trapped in the dark state and therefore the contrast of the CPT line. The effect of optical pumping can be reduced by using a double lambda scheme [10,11]. The idea is based on optically pumping the maximum number of atoms into the dark state out of any other trap state. Ideally, the double lambda scheme consists of two phase-coherent monochromatic beams tuned to optical transitions sharing a common excited level, with a frequency difference equal to the 0-0 resonance frequency. If the beams are parallel to the Zeeman field, with orthogonal linear polarizations, then σ^+ and σ^- transitions are excited, which is our situation [12] described in Fig. 1 (right) where both laser fields $\mathbf{E}_1$ orthogonal to $\mathbf{E}_2$ are tuned, respectively, to the transitions $|1\rangle \leftrightarrow |3\rangle, |4\rangle$ and $|2\rangle \leftrightarrow |3\rangle, |4\rangle$. Equivalent conditions of optical pumping with two waves σ^+ and σ^- have been studied independently in other groups, by using alternating circular polarizations allowing push-pull optical pumping [11] or orthogonal circular polarizations with counterpropagating

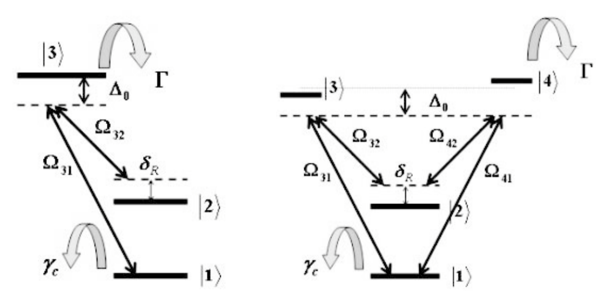

FIG. 1. Three level (simple lambda scheme) and four level (double lambda scheme) systems under two coherent laser fields excitation. δ_R is the Raman detuning and Δ_0 is the optical detuning. $\Omega_{i,j}$ with ($i = 3, 4$) and ($j = 1, 2$) are the Rabi angular frequencies corresponding to the various possible transitions. As mentioned in the text, $|3\rangle \equiv |F_e, -1\rangle$ and $|4\rangle \equiv |F_e, +1\rangle$ are Zeeman sublevels of the same excited hyperfine level. Γ is the decay rate modified by collisions and γ_c is the relaxation rate of the hyperfine coherence due to buffer gas.

waves [13]. If the two laser polarizations are parallel, the CPT phenomenon cannot be observed [14] because the superposition of states for one Λ scheme is orthogonal to the other.

The interaction operator between the atoms and the laser fields is $\mathbf{V} = -\mathbf{D} \cdot (\mathbf{E}_1 + \mathbf{E}_2)$, where $\mathbf{D}$ is the electric dipole moment. In the rotating frame, the secular approximation leads to the following simplified expression of the interaction operator:

$$\tilde{\mathbf{V}} = \frac{\hbar}{2\sqrt{2}}\{-\Omega_{41}|4\rangle\langle 1| + i\Omega_{42}|4\rangle\langle 2| + \Omega_{31}|3\rangle\langle 1| + i\Omega_{32}|3\rangle\langle 2| + \mathrm{H.c.}\} \quad (1)$$

with the Rabi angular frequency $\Omega_{ij} = -\langle i|\mathbf{D}|j\rangle E_j/\hbar$. The Clebsch-Gordan coefficients for the CPT resonance of the D_1 line in alkali atoms are such that for the clock transition $|1\rangle \longleftrightarrow |2\rangle$:

$$\frac{\Omega_{42}}{\Omega_{41}} = -\frac{\Omega_{32}}{\Omega_{31}}. \quad (2)$$

The dark state $|\Psi_{\mathrm{NC}}\rangle$ is not coupled to the laser fields according to $\widetilde{V}|\Psi_{\mathrm{NC}}\rangle = 0$. Using Eqs. (1) and (2), its expression is then

$$|\Psi_{NC}\rangle = \frac{\Omega_{42}}{\sqrt{\Omega_{42}^2 + \Omega_{41}^2}}|1\rangle - i\frac{\Omega_{41}}{\sqrt{\Omega_{42}^2 + \Omega_{41}^2}}|2\rangle. \quad (3)$$

This particular superposition of states is dark for both excited states $|3\rangle$ and $|4\rangle$ due to Eq. (2). A similar relation is quoted in [15]. For the Zeeman sublevels $m_F \neq 0$, the state superposition can be seen as a double Λ scheme with a low leakage. In order to test this particular dark state, we have developed, in our experiment, coherent laser fields produced by two extended cavity laser diodes tuned to the Cs D_1 line and phase locked with a frequency difference tunable around 9.192 GHz. Figure 2 shows the Zeeman spectrum detected through a 5 cm long cell filled with Cs and N_2 buffer gas at 23 Torr. The laser beams diameter is 15 mm. The "contrast" is defined as the on-resonance increase of the photodiode signal divided by the off-resonance signal. The photodiode signal is proportional to the laser intensity transmitted through the cell. The seven CPT resonances between the Zeeman sublevels $|3, m_F\rangle \leftrightarrow |4, m_F\rangle$ allowed by our double Λ scheme are observed. The peak amplitudes are symmetric around the $|F = 3, m_F = 0\rangle \leftrightarrow |F = 4, m_F = 0\rangle$ clock transition, contrary to what is observed with a single Λ excitation. Experimental CPT line shapes observed with continuous interrogation are well described by solving the optical Bloch equations applied only to a closed three level system [2]. With this model, we can derive an analytical expression of the CPT signal linewidth:

$$\Delta\omega = \frac{2\gamma_c}{\sqrt{1+S}}\sqrt{\left(1 + 3S + \frac{\Gamma}{\gamma_c}S^*\right)\left(1 + \frac{\Gamma}{\gamma_c}S^*\right)}, \quad (4)$$

where $\gamma_c/2\pi \sim 30$ Hz is the decay rate of the hyperfine coherence due to collisions with buffer gas, Γ is the excited state relaxation term, $\Gamma/2\pi \sim 625$ MHz. In Eq. (4), we retrieve the classical saturation parameter S coming from the rate equations and the CPT saturation parameter $S^*\Gamma/\gamma_c$ with

$$S = \frac{\Omega_1^2\Omega_2^2}{(\Omega_1^2 + \Omega_2^2)\Gamma^2/4}, \qquad S^* = \frac{(\Omega_1/2)^2 + (\Omega_2/2)^2}{\Gamma^2}, \quad (5)$$

where Ω_1 and Ω_2 are the Rabi angular frequencies associated with the optical coupling between the two ground states and the intermediate excited state. Since $S \ll 1$ and $\Gamma/\gamma_c \gg 1$, then from Eq. (4) the linewidth linearly scales as S^*, which was experimentally observed [2,4]. The light intensities needed to produce CPT resonances with high signal-to-noise ratios cause substantial broadening of the CPT resonances. This drawback can be circumvented by using the Ramsey method of two oscillatory separated fields.

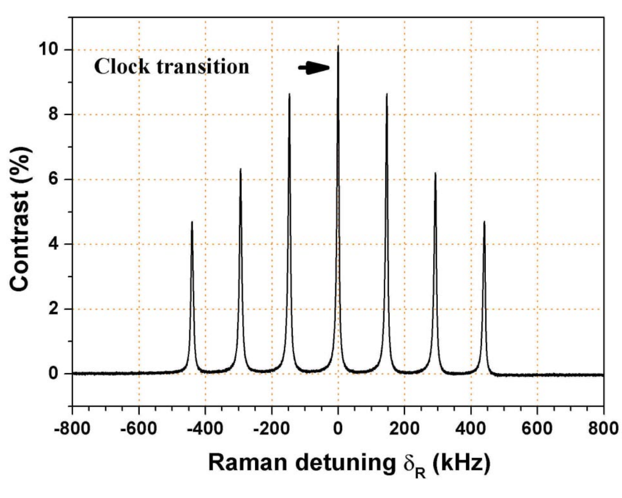

FIG. 2 (color online). Experimental CPT transitions observed in a 20 μT static magnetic field using orthogonal laser polarizations. With total intensity $\sim$1 mW/cm^2, the linewidth is $\sim$12 kHz.

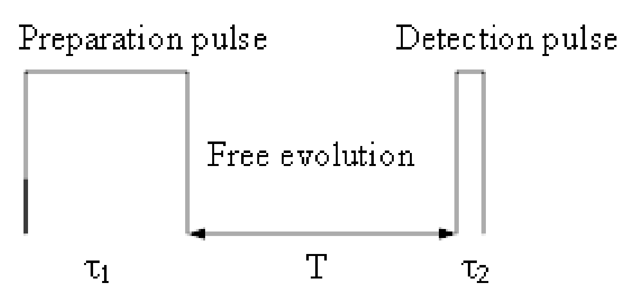

FIG. 3. Raman-Ramsey pulse sequence with duration τ_1 for the first pulse and τ_2 for the second pulse. T is the free evolution time of the Raman coherence between the two pulses.

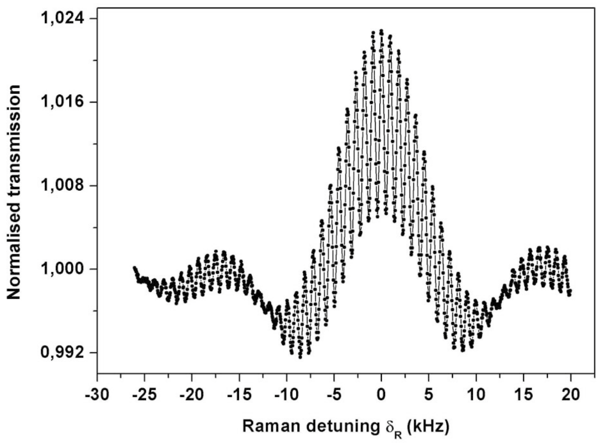

FIG. 4. Experimental Raman-Ramsey fringes with total intensity $\sim 100\ \mu\mathrm{W/cm^2}$. The first pulse duration is $\tau_1 = 80\ \mu$s and the second pulse duration is $\tau_2 = 80\ \mu$s; 8 average scan.

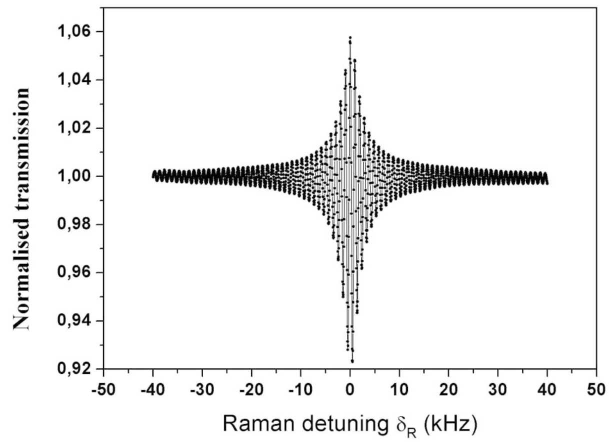

FIG. 6. Experimental saturated Raman-Ramsey fringes with total intensity $\sim 500\ \mu\mathrm{W/cm^2}$. The first pulse duration is $\tau_1 =$ 5 ms and the second pulse duration is $\tau_2 = 5\ \mu$s; 8 average scan.

Historically, coherent pulses were first used to produce Raman-Ramsey fringes with a thermal Na beam interacting with two spatially separated CPT laser fields and realizing the first atomic Raman clock [16]. Raman-Ramsey fringe widths were dependent only on the transit time T between the two interaction zones as $1/(2T)$. The fringes were observed by detecting the fluorescence of the beam in the second interaction zone, i.e., the population of the excited state during the second pulse. This is different from the usual Ramsey fringes where the population of one atomic state is measured after the end of the second interaction zone. In our experiment, the same idea is applied in the time domain. Atoms in a vapor cell are interrogated with a CPT pulse sequence as in Fig. 3. The light pulse durations are controlled by an acousto-optic modulator. The detection technique allows a real time measurement of the optical coherence during the two pulses, without perturbing the coherent process. Moreover, it does not require any difference of population prior to the application of the pulses.

Figures 4–6 show experimental records of the normalized transmission (peak valley of the central fringe divided by the signal level at large Raman detuning) versus the Raman detuning δ_R. Laser fields are optically resonant ($\Delta_0 \sim 0$), and the pulses are separated by $T = 1$ ms. In Fig. 4, the interaction sequence is similar to the usual Ramsey sequence ($\tau_1 = \tau_2 = 80\ \mu$s) where the atomic system is never saturated, excited with low laser intensity and short interaction times. The Rabi resonance line shape (full width 12.5 kHz) is modulated by the Raman-Ramsey fringes. As expected for $T = 1$ ms, the width of the central fringe is $1/(2T) = 500$ Hz, in good agreement with a simple wave function formalism [12]. Figures 5 and 6

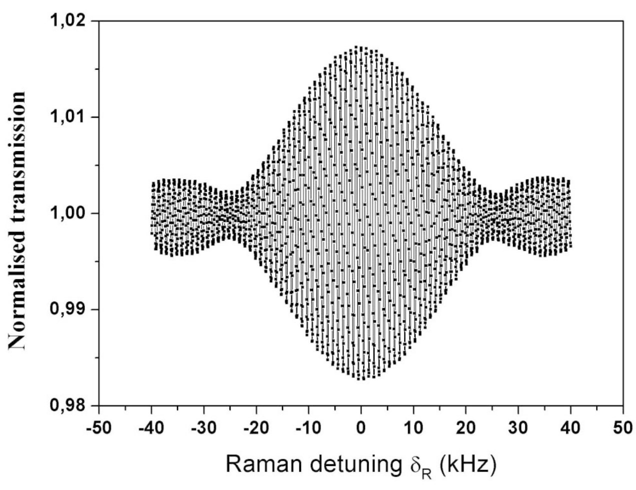

FIG. 5. Experimental Raman-Ramsey fringes with total intensity $\sim 500\ \mu\mathrm{W/cm^2}$. The first pulse duration is $\tau_1 = 40\ \mu$s and the second pulse duration is $\tau_2 = 5\ \mu$s; 8 average scan.

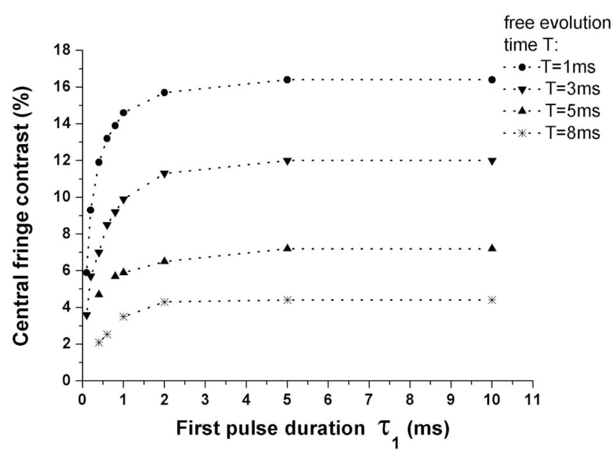

FIG. 7. Central fringe contrast versus the first pulse duration τ_1 for different values of the free evolution time T. The second pulse duration is constant $\tau_2 = 5\ \mu$s. The total intensity is $\sim 500\ \mu\mathrm{W/cm^2}$.

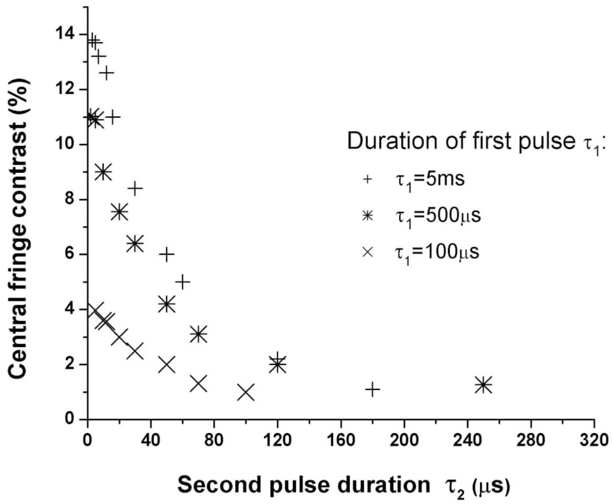

FIG. 8. Central fringe contrast versus the second pulse duration τ_2 for different τ_1 values. The free evolution time is constant $T = 1$ ms and the total intensity is $\sim$500 μW/cm^2.

show that the fringe contrast increases as the Rabi envelope reaches its stationary profile. For the same laser intensities, the contrast increases from 4% with $\tau_1 = 40$ μs in Fig. 5 to about 14% with $\tau_1 = 5$ ms in Fig. 6. Note that the central fringe 425 Hz wide is not broadened by saturation. Under a continuous excitation, the same laser intensities would lead to a linewidth as large as 6 kHz corresponding to the envelope width given by Eq. (4). Our typical Raman-Ramsey sequence differs from the usual Ramsey interrogation by several points. The role of the first interaction is to trap atomic population into the states superposition $|\Psi_{NC}\rangle$. This is done when the pulse duration τ_1 and the parameter S^* are high enough (Fig. 7). Since the dynamical evolution is driven by $\exp[-2\Gamma S^* t]$, the characteristic time for the dark state $|\Psi_{NC}\rangle$ to reach its asymptotic value is about 200 μs in our case. If the pulse τ_2 is small enough ($\sim$1 μs), the relaxation of the Raman coherence under collisions and saturation effects can be neglected during the detection pulse. Thus, the fringe contrast obtained with a short detection pulse ($\tau_2 = 5$ μs) depends only on τ_1 and T as shown in Fig. 7. When the free evolution time is increased, the central fringe narrows as $1/(2T)$ while the contrast decreases as $\exp[-\gamma_c T]$. Figure 8 gives the fringe contrast versus the second pulse duration. If τ_2 is too long, the steady state is reached again and the fringes vanish. The optimum contrast for a typical Raman-Ramsey sequence is obtained for $\tau_1 \gg 1/(2\Gamma S^*) \gg \tau_2$.

We have shown that a double lambda scheme, reducing optical pumping effects which are detrimental for the CPT line contrast, can be simply implemented using two crossed linearly polarized lasers. The possibility to get narrow resonance lines with high signal amplitude using the Ramsey method is also demonstrated. The width of the fringes are then limited only by the free evolution time T between the two pulses. These Raman-Ramsey fringes, obtained without any atomic state preparation, are not equivalent to the usual Ramsey fringes. The contrast optimization requires a long preparation pulse and a short detection pulse. High contrast narrow Raman-Ramsey fringes with a double Λ configuration could open the way to high sensitive magnetometers [17,18] or to high stability clocks in the microwave or optical domain. They could also be applied in experiments using electromagnetically induced transparency (EIT) [19].

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