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Demonstration of Raman–Ramsey fringes using time delayed optical pulses in rubidium vapor

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ABSTRACT

We report observation of high contrast Raman–Ramsey fringes using time delayed optical pulse pairs in a rubidium vapor cell. The width of these fringes are not limited by saturation and provides a simpler means to produce narrow atomic linewidths using a thermal vapor medium for compact atomic clock applications. We also demonstrate phase-scanned Raman–Ramsey fringes, with potential application to sensitive detection of trace vapors.

For many applications, there is a need to develop a very compact atomic clock with high accuracy. For example, such a clock could be built into next-generation GPS (Global Positioning System) receivers in order to enhance the navigation accuracy. The conventional approach for an atomic clock employs the excitation of hyperfine resonances in alkali atoms such as Cs or Rb using microwave fields enhanced by a cavity. The size of such a cavity represents a fundamental limitation to the volume and the weight of the clock. However, this problem can be circumvented by exciting the same resonance indirectly, using a two-photon, resonant-Raman transition that corresponds to the effect popularly known as Coherent Population Trapping (CPT) [1]. The CPT based atomic clock was first demonstrated by the group of Shaoul Ezekiel at MIT, using a dye laser, bulk acousto-optic modulators, and a sodium atomic beam [2–4]. These experiments report observations of Ramsey fringes by using resonant Raman transitions in two separated regions of an atomic beam. Small, transit-time limited linewidths are obtained using a large separation (upto 30 cm) between the exciting zones. Subsequently, a directly modulated semiconductor laser was used by the same group to demonstrate the CPT clock using a Cs atomic beam [5]. A clear, quantitative analysis of the improvement in contrast obtained by optical Ramsey fringes in various traveling-wave interaction geometries can be found in an early paper by Borde et al. [6]. Since then, significant developments have taken place in making the CPT clock more compact and robust [7,8].

Of particular significance in this development is the use of vapor cells to replace atomic beams. In order to overcome the transit time induced broadening of the CPT resonance, these cells are typically loaded with a buffer gas, leading to sub-kHz linewidths. However, there remains a key difference with the atomic beam (AB) based approach, which limits the accuracy of the vapor-CPT clocks. In the AB approach, the atoms interact with the optical fields in two separate zones, which is the Raman version of Ramsey's separated fields excitation scheme [9]. The resulting Raman–Ramsey fringe pattern has a linewidth determined primarily by the time-separation between the two zones. The advantages of this approach are as follows. First, the CPT process can be saturated in the first interaction zone in order to maximize the signal amplitude, without power-broadening the Raman–Ramsey fringes. Second, the saturation in the first zone also suppresses the process of shifting the clock transition frequency due to the AC Stark effect [4]. In contrast, the vapor-CPT approach uses a single-zone excitation, so that the linewidth of the CPT signal is limited by power-broadening, and the AC Stark effect is not suppressed effectively.

These problems can be circumvented by using a pair of time-delayed optical pulses in order to produce Raman–Ramsey fringes in a vapor cell. Here, we report on the observation of such fringes using a rubidium vapor cell. In our experiment, Raman transition is produced between the long-lived hyperfine ground states of the D2 line in ^{85}Rb vapor. An earlier experiment by Salour and Cohen-Tannoudji [10] explored a similar approach using short optical pulses (nanosecond) and rapidly decaying atomic states (~lifetime

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5×10^{-8} s) of sodium for high-resolution spectroscopy application. This involved practical difficulties in generating time-delayed, phased-locked short pulses from the laser amplifiers. In contrast, the Ramsey excitation reported here is carried out using long optical pulses, since it involves a Raman transition between long-lived metastable ground states. A theoretical proposal by Dubetsky et al. [11] considered generating stimulated echo and Ramsey fringes sensitive to small changes in atomic velocity by using a sequential excitation of atoms by two counterpropagating traveling waves. A recent experiment [12] has reported a similar approach for observing high contrast fringes in thermal Cs vapor using delayed pulses, using a double-lambda scheme. Such a scheme requires a four-level system, which in this case is created using Zeeman sublevels of the same hyperfine excited level. In our scheme, a simple three-level Λ-type scheme has been shown to produce high-contrast fringes both by changing the Raman detuning as well as by scanning the phase of the interrogating pulse without perturbing the dark state. The phase scan technique can be used in sensing applications where, for example, the interrogating pulse can be sent through a trace medium for determining the trace concentration.

Ramsey fringes are observed experimentally in the amplitude of the time-delayed second probe pulse by varying the frequency detuning between the pump and the probe away from resonant Raman excitation. An electronic time-gated measurement has been used to observe the amplitude oscillation of the second pulse. The width of observed Ramsey fringes is limited only by the inverse of the time delay between the pulse pairs. In practice, this width can be made very small compared to the resonance linewidth observed during continuous excitation, if the medium dephasing time can be made long either by using buffer gas [13] or by using wall coating [14] to preserve the coherence. We also used an alternative approach to Raman–Ramsey oscillations by independently scanning the phase of the second probe pulse with respect to the first pulse using a piezo-actuated mirror, while keeping the difference frequency matched close to the two photon resonance condition. In all these experiments, no special care (such as adding buffer gas or paraffin coating) is taken to prevent rapid medium decoherence, which leaves room for significant improvements.

The inset in Fig. 1 shows a three level Λ-scheme used in the D2 line of ^{85}Rb using two coherent laser fields, ω_1 and ω_2. In the experiment, states $5S_{1/2}$ ($F = 2$) and $5S_{1/2}$ ($F = 3$) are used as two long-lived ground states and the hyperfine excited state $5P_{3/2}$ ($F = 3$) is used as the excited upper level for the Λ-system. Coherent Raman excitation using the resonant optical fields on the two atomic transitions produces a dark superposition state [15], $|-\rangle = [\Omega_2|1\rangle - \Omega_1|2\rangle]/\sqrt{\Omega_1^2 + \Omega_2^2}$, where Ω_1, Ω_2 are the Rabi frequencies associated with the Raman fields, that exhibits CPT in such a Λ-system. When the atom is put in this particular superposition state, it remains in the dark state, which is uncoupled from the excited state. As a result, the medium appears transparent after the dark state is established. In our experiment, the linewidth of the CPT resonance is observed in continuous excitation by sweeping the two-photon (or difference) detuning δ between the two (pump and probe) laser fields. For the copropagating fields in the vapor medium, the resonant two-photon linewidth is unaffected by the Doppler broadening, and is determined by other sources such as light intensities, transit time of atoms and the decay rate between ground states. In experiments using the continuous excitation, the linewidth of resonance is primarily governed by the saturation caused due to light intensities.

The contrast of the resonant signal is determined by the fraction of total number of atoms trapped in the dark state. If orthogonal, circular polarizations are used for the pump and the probe, a large fraction of atomic population get trapped in Zeeman sublevels due to optical pumping caused by laser fields via resonant and off-resonant excitations. This causes significant reduction in the contrast of the CPT signal. In order to circumvent this problem, we have used orthogonal, linear polarizations for the pump and the probe beams [12,16,17], as discussed further later on.

Fig. 1a shows the schematic of our experimental setup used in both continuous and pulsed Raman excitations. A frequency stabilized Ti: Sapphire laser is used to produce resonant laser fields at frequencies ω_1 and ω_2. These are obtained by using acousto-optic modulators (AOMs) driven by microwave voltage controlled oscillators (VCOs). For the two photon Raman resonance, the frequency difference between ω_1 and ω_2 is matched to the ground state splitting (3.0357 GHz) of ^{85}Rb. Fig. 1c shows the energy diagram used for the CPT excitation in Rb^{85} atoms. This is achieved by using two high frequency AOMs made by the Brimrose corporation (center frequency (f_o) = 1.5178 GHz. bandwidth (Δf) = 200 MHz, diffraction efficiency ~25%) in the paths of the probe and pump beams, the probe frequency being upshifted and the pump frequency being downshifted from the laser frequency. The orthogonally polarized beams are combined using a polarizing beam splitter. They are sent through a piece of polarization maintaining optical fiber in order to produce a good spatial mode. The use of the optical fiber also ensures perfect co-propagation between the pump and the probe. This is particularly important for observing very narrow Raman–Ramsey fringes [18]. The beams are expanded and collimated to about 5 mm ($1/e^2$ diameter) before sending them through a 10 cm long rubidium vapor cell. The vapor cell is magnetically shielded using a μ-metal box and wrapped with twisted coils to minimize the effect of magnetic field produced due to heating. Raman excitation using the orthogonal, linear polarizations creates dark states as a superposition of the $|F = 2, m_F = 0\rangle$, $|F = 3, m_F = 0\rangle$ levels of the ground states for both the σ^+ and σ^- components of the linearly polarized beams. This is because the ratios of Clebsch–Gordan coefficients for σ polarizations are equal and opposite for the D2 transitions: $|\text{Dark}\rangle_{\sigma^-} = |\text{Dark}\rangle_{\sigma^+}$ *for* $[\Omega_1/\Omega_2]_{\sigma^+} = -[\Omega_1/\Omega_2]_{\sigma^-}$. Experimentally CPT resonance of linewidth as narrow as 400 KHz has been observed for the probe power ~0.25 mW and a pump four times stronger than the probe.

To observe Raman–Ramsey fringes, we used a repeated sequence of time-delayed optical pulse pairs in the vapor medium. These optical pulses are generated by sending Rf pulses from a pulse generator to the high-frequency AOMs. Both the pump and the probe beams are pulsed simultaneously using the independent AOMs shown in Fig. 1a. During the observation of the probe transmission, the pump pulse is filtered out using a polarizer. The duration T_1 of the first pulse is chosen to be sufficiently long in order to cause efficient excitation of atoms into the dark state. The atoms are then probed using a second pulse of duration T_2. The second pulse is kept short and relatively weak, in order not to perturb significantly the dark state produced by the longer and stronger first pulse. In the experiment, T_1 and T_2 are chosen to be 500 ns and 100 ns, respectively. The fringes are observed in the transmitted amplitude of the second pulse by slowly varying the difference frequency δ while using a repeated sequence of pulses. The probe amplitude of the second pulse is measured using a gated integrator and boxcar averager.

The conceptual diagram in Fig. 2a[1] shows repeated pulse pairs during a slow δ-scan that spans the single-zone CPT line-width. After each coherent excitation by the first (blue) pulse, the atoms are driven into the dark state and are interrogated by the second (red) pulse. The amplitude of the second probe pulse oscillates as

[1] For interpretation to color in Fig. 2, the reader is referred to the web version of this article.

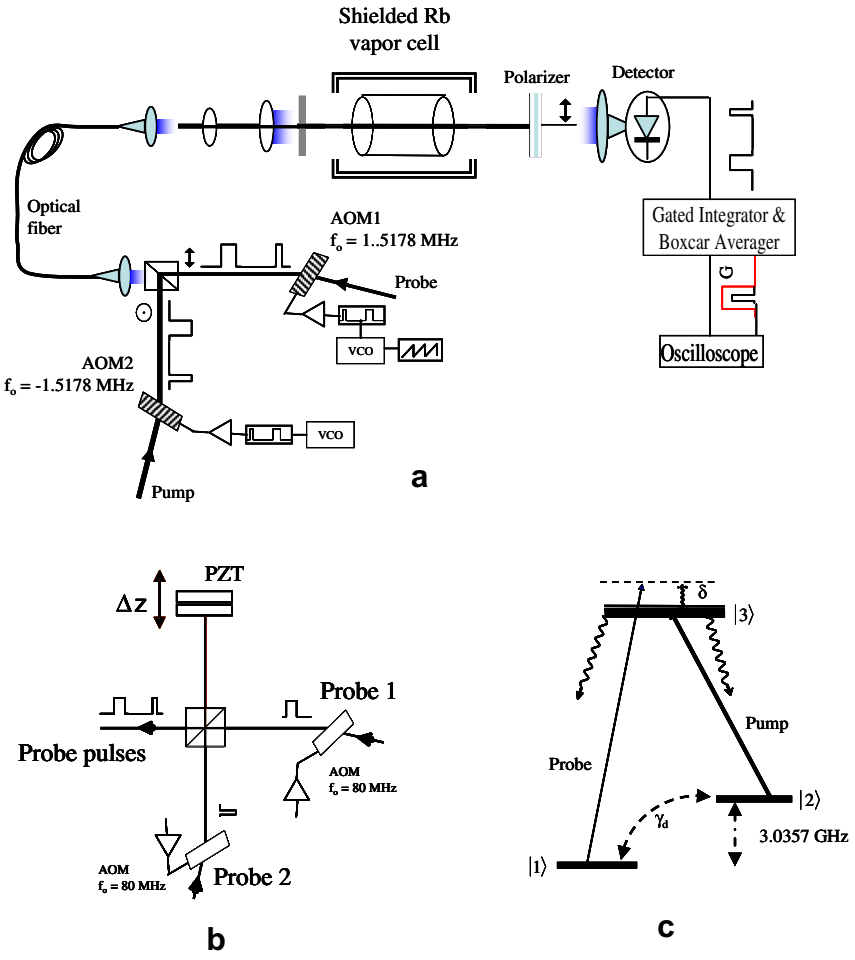

Fig. 1. (a) Layout of the experimental arrangement, f_o corresponds to the center frequency of the AOMs. While observing the CPT signal, a probe power of 250 μW and a pump power ~1 mW have been used. (b) Experimental scheme used to generate independent phase scan of the second probe pulse. (c) Energy diagram for the CPT excitation in rubidium (^{85}Rb D2 line) vapor.

a function of δ as shown in Fig. 2b, giving rise to the Raman–Ramsey oscillations [19]. The Raman–Ramsey oscillations occur as long as the delay T between the pulse pair is smaller than the medium dephasing time. The dephasing time between the ground states in our experiment is measured to be approximately few microseconds, primarily limited due to collision (atom–atom and atom–wall) induced dephasing. However, the relaxation can be improved significantly by adding inert buffer gases [14].

In the experiment, the delayed pulse pairs are repeated after every 100 μs interval. While sending repeated pulse pairs, the difference frequency δ between the probe and pump frequencies is scanned over a range of 4 MHz around the two photon resonance ($\delta = 0$, which corresponds to $\omega_1 - \omega_2 = 3.0357$ GHz) at a slow rate of 10 Hz. This allows us to sample the Raman–Ramsey oscillations with nearly 1000 pulses using the gated boxcar averager. The integration mode in the boxcar also allows us to observe extremely small amplitude oscillations in the second pulse. Fig. 3a and b shows experimentally observed Ramsey fringes using different time-delays T between the first and second optical pulse. The traces in red color indicate the fringes observed in the amplitude of the second probe pulse, whereas the ones in blue show the corresponding amplitude variation of the first probe pulse. The amplitude variation of the first pulse essentially corresponds to a single-pulse, transit-time limited CPT resonance. The delay T for evolution of the coherent dark state is varied in order to observe the smallest width of the fringes. In our case, high contrast interference fringes are observed for delays upto 3 μs. The fringe width at the center varies $1/2T$, as expected, and is significantly narrower than the CPT line-width. Away from the center, the amplitude of oscillation decreases as the two photon resonance condition ($\delta = 0$) is not exactly satisfied.

In an alternative approach, we used a phase scan technique instead of the δ-scan to observe the Raman–Ramsey fringes. This is done by slowly (at a rate of 10 Hz) and continuously scanning the phase of the beat-note between the pump and the probe in the second pulse with respect to the first pulse during the repeated pulse sequence. For this purpose, two additional AOMs (with same frequency shifts, 80 MHz) are used (Fig. 1b) in the probe beam to generate independently the two delayed probe pulses. The diffracted beams from both these AOMs are then combined, before they are sent through a common AOM (1.4378 GHz). Using a piezo-actuated mirror (PM) in one of the AOM beam paths allows us to continuously scan the phase of the second pulse (generated by that AOM) using a voltage ramp. Fig. 3c shows the Ramsey oscillations observed in the amplitude of the second pulse when the difference frequency between the pump and probe is set close to the two photon resonance peak, i.e., $\delta = 0$. The number of oscillations corresponds to the phase cycle of the second pulse, which in our case is limited by the motion of the piezo-actuator. The phase Φ as a function of the scan time t is given by $(\omega_1/c)d_{\max}f_{\text{scan}}t$, where $d_{\max}$ is the maximum translation of the actuator and f_{scan} is the rate of scan. Fig. 3c shows oscillations for two different time separa-

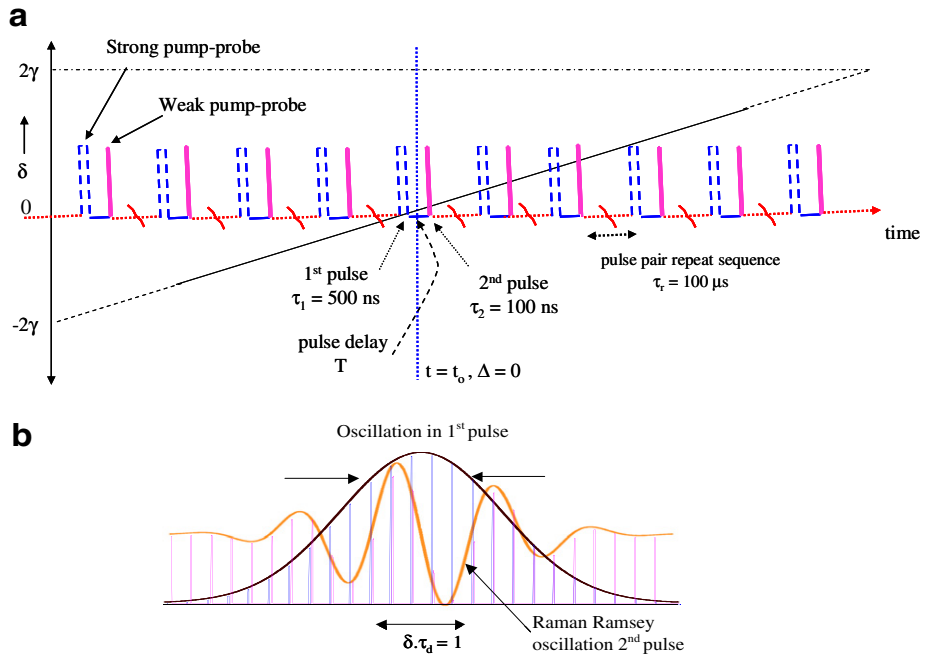

Fig. 2. Illustrative diagrams showing (a) the repeated pulse sequence, where the straight line ramp represents a gradual scan of the pump and probe difference frequency around the two-photon resonance condition ($\delta = 0$), and (b) Raman–Ramsey oscillations in the second pulse amplitude.

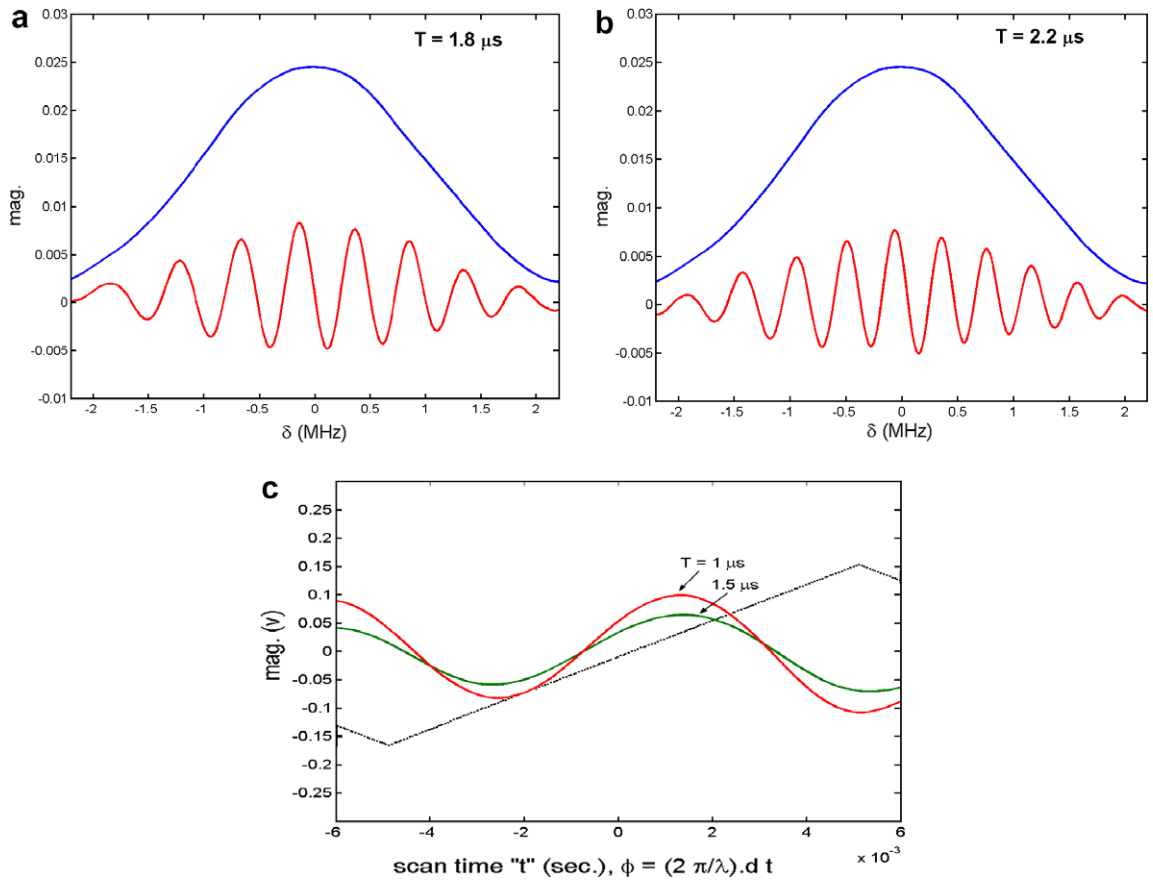

Fig. 3. Experimentally observed Raman–Ramsey oscillations with time delays (a) $T = 1.8$ μs (b) 2.2 μs. The blue trace indicates the amplitude of the first pulse and the red indicates the Ramsey oscillations in amplitude of the second pulse. The width of the central fringe in (a) ~280 KHz, (b) ~230 KHz, (c) Ramsey oscillations observed using the phase scan for pulse time delays $T = 1$ μs and 1.5 μs. The dashed line correspond to the voltage ramp applied to the piezo-actuator to produce the phase scan at 10 Hz rate.

tions between the pulse pairs. The coherence produced by the first pulse is preserved during the free time evolution, which is probed by the second pulse that carries a different phase to produce the Raman–Ramsey fringes. It is possible to phase lock these oscillations using a reference signal generated by a separate interferometer. This can yield a high signal to noise ratio for phase sensitive detection, with possible application to detection of trace vapors.

The primary motivation for the results reported here is the enhancement of the stability of a CPT-based Rb vapor clock. In order to demonstrate the actual enhancement achievable, it is necessary for us to use a buffer gas filled cell and preferably use a diode laser for exciting the CPT transition. Efforts are currently underway to perform such an experiment.

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