Cancellation of light-shifts in an $N$-resonance clock

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We demonstrate that first-order light-shifts can be cancelled for an all-optical, three-photon-absorption resonance ("$N$-resonance") on the $D_1$ transition of $^{87}\text{Rb}$. This light-shift cancellation enables improved frequency stability for an $N$-resonance clock. For example, using a table-top apparatus designed for $N$-resonance spectroscopy, we measured a short-term fractional frequency stability (Allan deviation) $\simeq 1.5 \times 10^{-11} \tau^{-1/2}$ for observation times $1 \text{s} \lesssim \tau \lesssim 50 \text{s}$. Further improvements in frequency stability should be possible with an apparatus designed as a dedicated $N$-resonance clock.

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There is great current interest in developing small, economical atomic frequency standards (clocks) with fractional frequency stability $\sim 10^{-12}$ or better. Significant progress toward this goal has been achieved using coherent population trapping (CPT) resonances in atomic vapors. However, the frequency stability of CPT clocks is limited in part by light-shifts, i.e., shifts of the resonance frequency due to the applied electromagnetic fields.

Recently our group demonstrated that a three-photon-absorption resonance (known as an "$N$-resonance") is a promising alternative for small atomic clocks. Here we show that it is possible to cancel first-order light-shifts by optimizing the intensity ratio and frequency of the two optical fields that create and interrogate the $N$-resonance. Employing such light-shift cancellation in a simple, table-top apparatus, we observed promising short-term frequency stability ($\simeq 1.5 \times 10^{-11} \tau^{-1/2}$) for an $N$-resonance on the $D_1$ transition of $^{87}\text{Rb}$ vapor. We expect superior frequency stability will be possible in a small $N$-resonance clock designed for good thermal control, low phase noise, etc.

Fig. 1(a) shows the $N$-resonance interaction scheme. A probe field $\Omega_P$ and drive field $\Omega_D$ are in two-photon Raman resonance with the ground-state hyperfine levels $|b\rangle$ and $|c\rangle$, with $\Omega_P$ nearly resonant with the optical transition $|c\rangle \rightarrow |a\rangle$ and $\Omega_D$ red-detuned from this optical transition by the ground-state hyperfine splitting $\nu_0$ ($\simeq 6.835 \text{ GHz for } ^{87}\text{Rb}$). The two-photon Raman process drives atoms coherently from state $|b\rangle$ to $|c\rangle$, followed by a one-photon transition to excited state $|a\rangle$ via absorption from field $\Omega_P$. Together, this three-photon process produces a narrow absorptive resonance in the probe field transmitted intensity, with a width that is limited by the relaxation rate of the atoms’ ground-state coherence.

For such an idealized three-level $N$-resonance, the light-shift $\delta$ (i.e., the detuning from $\nu_0$ of the difference frequency between the probe and drive fields, as measured by maximum probe field absorption) consists of three leading (first-order) terms: shifts of both ground-states due to interaction with the strong, far-detuned drive field, and a shift of ground-state $|c\rangle$ due to interaction with the near-resonant probe field:

$$\delta \approx -\frac{|\Omega_D|^2}{\nu_0 + \Delta} + \frac{|\Omega_D|^2}{2\nu_0 + \Delta} + \frac{|\Omega_P|^2 \Delta}{\Delta^2 + \gamma^2/4}. \quad (1)$$

Here $\Delta$ is the one-photon detuning of the probe field from resonance, $\gamma$ is the collisionally-broadened decoherence rate of the excited state $|a\rangle$, and $\Delta$ is the one-photon detuning of the probe field from resonance with the $|c\rangle$ to $|a\rangle$ transition. (b) Schematic of the experimental setup. See text for abbreviations.

FIG. 1: (a) $N$-resonance interaction scheme. $\Omega_P$ and $\Omega_D$ are the probe and drive optical fields that create and interrogate the $N$-resonance, $\nu_0$ is the hyperfine splitting of the two lower energy levels $|b\rangle$ and $|c\rangle$. $\gamma$ is the collisionally-broadened decoherence rate of the excited state $|a\rangle$, and $\Delta$ is the one-photon detuning of the probe field from resonance with the $|c\rangle$ to $|a\rangle$ transition.
enence rate of the excited state, and $\Omega_D$ and $\Omega_P$ indicate the probe and drive fields’ Rabi frequencies. The light-shifts due to the far-detuned drive field (the first and second terms in Eq. (1)) are proportional to the drive field intensity, but practically independent of the laser frequency for $\Delta \ll \nu_0$. In contrast, the light-shift due to the near-resonant probe field (the last term in Eq. (1)) has a strong dispersive-like dependence on $\Delta$. Thus, near the extrema, $\Delta = \pm \gamma/2$, the total $N$-resonance light-shift has only a quadratic dependence on the probe field detuning:

$$\delta \approx -\frac{|\Omega_D|^2}{2\nu_0} + \frac{|\Omega_P|^2}{\gamma} + \frac{2|\Omega_P|^2}{\gamma^3}(\Delta \mp \gamma/2)^2. \quad (2)$$

This light-shift can then be cancelled by (i) detuning the probe field to the high-frequency extremum, and (ii) properly setting the intensity ratio of the drive and probe fields:

$$\Delta = \gamma/2, \quad \frac{|\Omega_P|^2}{|\Omega_D|^2} = \frac{\gamma}{2\nu_0}. \quad (3)$$

With such light-shift cancellation, the measured $N$-resonance center frequency should be insensitive (to leading order) to fluctuations of the probe field frequency and total laser intensity. Note that the light-shift cancellation does not depend on the absolute values of either optical field.

To verify these predictions, we measured $^{87}$Rb $N$-resonance light-shifts using the experimental setup shown in Fig. 1(b). We phase-modulated the output of a free-running New Focus external cavity diode laser using an electro-optical modulator (EOM), which produced two optical sidebands separated by $\approx 6.835$ GHz. The EOM was driven by a microwave synthesizer locked to a 100 MHz voltage-controlled crystal oscillator (VCXO). The laser frequency was adjusted such that the high-frequency sideband (serving as the probe field $\Omega_P$) was tuned close to the $5S_{1/2} F = 2 \rightarrow 5P_{1/2} F' = 2$ transition of $^{87}$Rb ($\lambda \approx 795$ nm); the carrier-frequency field then served as the drive field $\Omega_D$. The probe/drive field intensity ratio was set by the EOM phase-modulation index. The laser beam was circularly polarized using a quarter wave plate and weakly focused to a diameter of 0.8 mm before entering the Rb vapor cell.

We employed a cylindrical Pyrex cell containing isotopically enriched $^{87}$Rb and a mixture of buffer gases (15 Torr Ne + 15 Torr Ar + 5 Torr N$_2$) chosen to minimize the temperature dependence of the $^{87}$Rb ground-state hyperfine frequency shift due to buffer gas collisions. Associated collisional broadening of the excited state is estimated to be $\gamma \approx \pi \times 1.2$ GHz. During experiments, the vapor cell was heated to 55°C and isolated from external magnetic fields with three layers of high permeability shielding. A small (\approx 10 mG) longitudinal magnetic field was applied to lift the degeneracy of the Zeeman sublevels and separate the desired $F = 1$, $m_F = 0$ to $F = 2$, $m_F = 0$ clock transition (no first-order magnetic field dependence) from the $m_F = \pm 1$ transitions (first-order Zeeman splitting). The strong drive field and the lower-frequency sideband were filtered from the light transmitted through the cell using a quartz, narrow-band Fabry-Perot etalon (free spectral range of 20 GHz, finesse of 30), which was tuned to the frequency of the probe field and placed before the photodetector (PD).

To lock the frequency of the VCXO (and hence the detuning of the probe and drive fields) to the $N$-resonance, we superimposed a slow frequency modulation at $f_m = 400$ Hz on the 6.8 GHz signal from the microwave synthesizer. We demodulated the photodetector output at $f_m$ with a lock-in amplifier, and used the in-phase lock-in amplifier output as an error signal to feed back to the VCXO. We then monitored the frequency of the locked VCXO (and thus the $N$-resonance center frequency) by comparing it with a 100 MHz signal derived from a hydrogen maser.

Figs. 2 and 3 show examples of the measured dependence of the $N$-resonance frequency on laser detuning, in-
In conclusion, we demonstrated cancellation of first-order light-shifts for an all-optical, three-photon-absorption N-resonance on the $D_1$ line of $^{87}\text{Rb}$ vapor. Employing this light-shift cancellation in a table-top apparatus not engineered for stable clock performance, we nonetheless observed N-resonance frequency stability comparable to or better than existing CPT clocks. Significant improvements in N-resonance frequency stability should be possible in a small device with standard techniques. We note also that similar light-shift cancellation is possible for other N-resonances, e.g., the Rb $D_2$ line ($\lambda = 780$ nm). Currently, diode lasers for the $D_2$ line of Rb and Cs are more easily obtained than for the $D_1$ line.

We next characterized the frequency stability of a crude “N-resonance clock” — i.e., the VCXO locked to the $^{87}\text{Rb}$ N-resonance as described above — relative to a hydrogen maser. For this measurement we tuned our system to the conditions for optimal light-shift cancellation (laser detuning $\Delta \approx 700$ MHz, probe/drive field intensity ratio $\approx 11\%$) with total laser power $\approx 140$ $\mu$W (intensity $\approx 30$ mW/cm$^2$). Under such conditions the N-resonance linewidth $\approx 1400$ Hz (FWHM) and contrast $\approx 7\%$, which implies a shot-noise-limited short-term frequency $\approx 5 \times 10^{-14} \tau^{-1/2}$. Fig. 4 shows the measured N-resonance clock fractional frequency stability (Allan deviation). The short-term stability $\approx 1.5 \times 10^{-11} \tau^{-1/2}$ for observation times $1 \text{ s} \lesssim \tau \lesssim 50 \text{ s}$. At longer times the stability degrades due to uncontrolled temperature and mechanical variations in our table-top apparatus, as well as long-term drifts of the laser frequency. Despite this non-optimal clock apparatus, the short-term N-resonance frequency stability is already better than that provided by many recently-demonstrated CPT clocks $[1,7,8]$. We expect that both the short- and long-term N-resonance frequency stability can be further improved by straightforward optimization of the VCXO lock-loop (to reduce phase noise), temperature stabilization, laser control, etc. We also expect that a high-stability N-resonance clock should be possible in a compact physical package (with vapor cell volume $\approx 1 \text{ mm}^3$), because of promising N-resonance characteristics at high buffer gas pressure $[4]$.

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[1] J. Vanier, Appl. Phys. B 81, 421 (2005).
[2] M. Zhu and L.S. Cutler, in Proceedings of the 32nd Precision Time and Time Interval (PTTI) Systems and Applications Meeting, 2000, edited by L. Breakiron (USNO, Washington, 2001), p.311.
[3] J. Vanier, M. W. Levine, D. Janssen, M. J. Delaney, IEEE Trans. Instrum. Meas. 52, 822 (2003).
[4] S. Zibrov, I. Novikova, D. F. Phillips, A. V. Taichenachev, V. I. Yudin, R. L. Walsworth, and A. S. Zibrov, Phys. Rev. A 71, 011801(R) (2005).
[5] A. S. Zibrov, C. Y. Ye, Y. V. Rostovtsev, A. B. Matsko, and M. O. Scully, Phys. Rev. A 65, 043817 (2002).
[6] J. Vanier and C. Audoin, The Quantum Physics of Atomic Frequency Standards, (Hilger, New York, 1989).
[7] M. Merimaa, T. Lindwall, I. Tittonen, and E. Ikonen, J. Opt. Soc. Am. B 20, 273 (2003).
[8] S. Knappe, P. D. D. Schwindt, V. Shah, L. Hollberg, J. Kitching, L. Liew, and J. Moreland, Opt. Express 13, 1249 (2005).