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

Irina Novikova, David F. Phillips, Alexander S. Zibrov,* and Ronald L. Walsworth

Harvard-Smithsonian Center for Astrophysics and Department of Physics, Harvard University, Cambridge, Massachusetts 02138

Aleksei V. Taichenachev and Valeriy I. Yudin

Institute of Laser Physics SB RAS and Novosibirsk State University, Novosibirsk 630090, Russia

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We demonstrate that first-order light shifts can be canceled for an all-optical, three-photon-absorption resonance (*N*-resonance) on the D_1 transition of ^{87}Rb. This light-shift cancellation facilitates improved frequency stability for an *N*-resonance clock. For example, by using a tabletop apparatus designed for *N*-resonance spectroscopy, we measured a short-term fractional frequency stability (Allan deviation) of $\simeq 1.5 \times 10^{-11}\, \tau^{-1/2}$ for observation times of $1\text{ s} \leqslant \tau \leqslant 50$ 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 of $\sim 10^{-12}$ or better. Significant progress toward this goal has been achieved by using coherent population trapping (CPT) resonances in atomic vapor.[1] 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.[2,3]

Recently our group demonstrated that a three-photon-absorption resonance (known as an *N*-resonance) is a promising alternative for small atomic clocks.[4] 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. By employing such light-shift cancellation in a simple tabletop apparatus, we observed promising short-term frequency stability ($\approx 1.5 \times 10^{-11}\, \tau^{-1/2}$) for an *N*-resonance on the D_1 transition of ^{87}Rb vapor. We expect that superior frequency stability will be possible in a small *N*-resonance clock designed for good thermal control, low phase noise, etc.

Figure 1(a) shows the *N*-resonance interaction scheme.[4,5] A probe field Ω_P and drive field Ω_D are in two-photon Raman resonances with the ground-state hyperfine levels $|b\rangle$ and $|c\rangle$, with Ω_P nearly resonant with the optical transition $|c\rangle \rightarrow |a\rangle$ and Ω_D red detuned from this optical transition by the ground-state hyperfine splitting ν_0 ($\simeq 6.835$ GHz for ^{87}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 Ω_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 δ (i.e., the detuning from ν_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:

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

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 Δ. Thus near the extrema, $\Delta = \pm \gamma/2$, the total *N*-resonance light shift has only a quadratic dependence on the probe field detuning:

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

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

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

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(c). We phase modulated the output of a free-running New Focus external cavity diode laser by using an electro-optical modulator (EOM), which produced two optical sidebands separated by $\simeq 6.835$ 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 Ω_P) was tuned

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close to the $5S_{1/2}\ F=2 \rightarrow 5P_{1/2}\ F'=2$ transition of ^{87}Rb; the carrier-frequency field then served as the drive field Ω_D. The probe–drive field intensity ratio was set by the EOM phase-modulation index. The laser beam was circularly polarized by using a quarter-wave plate and weakly focused to a diameter of 0.8 mm before entering the Rb vapor cell.

We employed a 7.5 cm long cylindrical Pyrex cell (diameter 2.5 cm) 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.[6] 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 from the magnetic-field-sensitive $m_F=\pm 1$ transitions. The strong drive field and the lower-frequency sideband were filtered from the light transmitted through the cell by using a quartz, narrowband Fabry–Perot etalon (free spectral range of 20 GHz, finesse of 30),

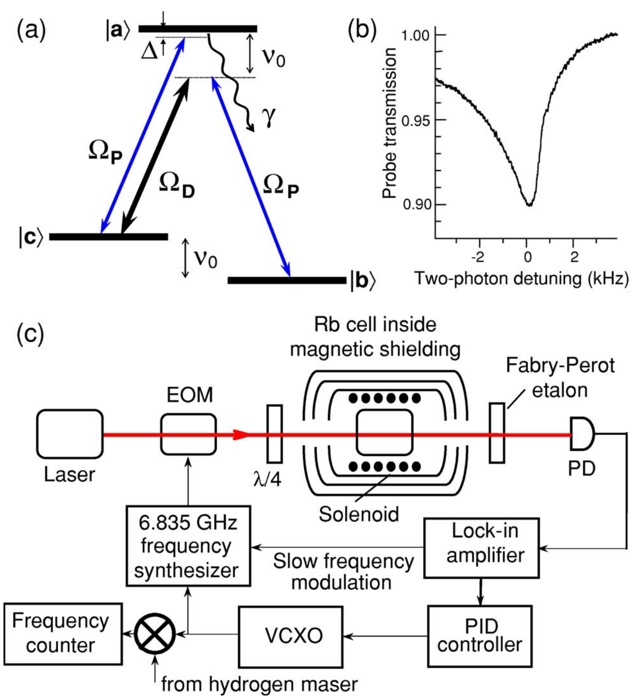

Fig. 1. (Color online) (a) N-resonance interaction scheme. Ω_P and Ω_D are the probe and drive optical fields that create and interrogate the N-resonance, respectively; ν_0 is the hyperfine splitting of the two lower energy levels, $|b\rangle$ and $|c\rangle$; γ is the collisionally broadened decoherence rate of the excited state $|a\rangle$; and Δ is the one-photon detuning of the probe field from resonance with the $|c\rangle$ to $|a\rangle$ transition. (b) Sample N-resonance spectrum. Probe transmission is normalized to unity away from the two-photon resonance. Zero two-photon detuning corresponds to an EOM modulation frequency of 6.8 346 896 GHz, which includes an $\approx$7 kHz buffer gas collision shift. (c) Schematic of the experimental setup. See text for abbreviations.

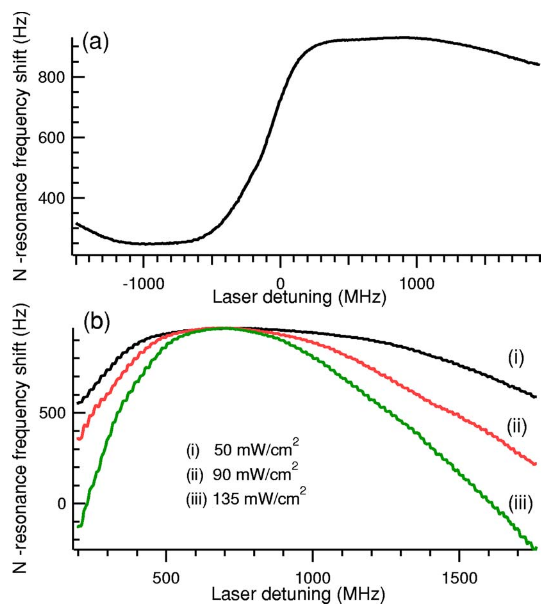

Fig. 2. (Color online) Measured detuning light shift, i.e., N-resonance frequency shift as a function of the detuning Δ of the probe field from the $5S_{1/2}\ F=2 \rightarrow 5P_{1/2}\ F'=2$ transition in ^{87}Rb vapor. (a) Example of the light shift's dispersive-like dependence on Δ; total laser intensity $\approx$30 mW/cm^2. (b) Insensitivity of the light shift to variations in the probe field frequency and the total laser intensity near the optimized probe field detuning ($\approx$700 MHz); the probe/drive intensity ratio is $\simeq$11%, to cancel the intensity light shift (see Fig. 3).

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.

Figures 2 and 3 show examples of the measured dependence of the N-resonance frequency on laser detuning, intensity, and probe–drive field intensity ratio. Consistent with Eq. (1), we observed two extrema in the detuning light shift: one below and one above the probe field resonance frequency; see Fig. 2(a). As illustrated in Fig. 2(b), we found that the probe field detuning at the light-shift maximum ($\Delta \approx 700$ MHz) is effectively independent of the total laser intensity, as expected from Eq. (2), when the probe/drive field intensity ratio is set to make the total light shift independent of the laser intensity. We determined this intensity light-shift cancellation ratio ($|\Omega_P|^2/|\Omega_D|^2 \simeq 11\%$, given by EOM phase-modulation index $\simeq$0.22) from the measurements shown in Fig. 3. Note that the experimentally determined ratio is in reasonable agreement with the prediction of $|\Omega_P|^2/|\Omega_D|^2 \simeq 9\%$ given by Eq. (3) for our experimental conditions. For small deviations of the probe/drive field intensity ratio from the cancellation value, the light shift increases linearly as shown in Fig. 3(b), which imposes restrictions on phase-modulation power stability.

We next characterized the frequency stability of a crude N-resonance clock, i.e., the VCXO locked to the ^{87}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 $\Delta \simeq 700$ MHz, probe–drive intensity ratio $\simeq 11\%$) with total laser power $\simeq 140\ \mu$W (intensity $\simeq 30\ \text{mW/cm}^2$). Under such conditions the N-resonance linewidth $\simeq 1400$ Hz (FWHM) and contrast $\simeq 8\%$, which implies a shot-noise-limited short-term frequency $\simeq 5 \times 10^{-14}\ \tau^{-1/2}$ (Ref. 3). Figure 4 shows the measured N-resonance clock fractional frequency stability (Allan deviation). The short-term stability $\simeq 1.5 \times 10^{-11}\ \tau^{-1/2}$ for observation times of $1\ \text{s} \leq \tau \leq 50$ s. At longer times the stability degrades due to uncontrolled temperature and mechanical variations in our tabletop apparatus, as well as long-term drifts of the laser frequency. Despite this nonoptimal 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 a vapor cell volume of $\sim 1\ \text{mm}^3$), because of promising N-resonance characteristics at high buffer gas pressure.[4]

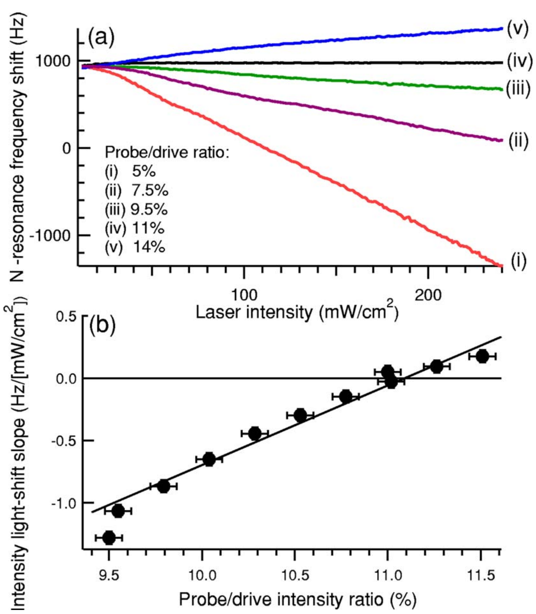

Fig. 3. (Color online) (a) Measured intensity light shift, i.e., N-resonance frequency shift as a function of laser intensity for different ratios between the probe and drive field intensities; the probe field is detuned to the light-shift maximum ($\Delta \approx 700$ MHz). (b) Fitted linear slopes for the measured light-shift variation with laser intensity for probe/drive field intensity ratios near the intensity light-shift cancellation value. The solid diagonal line is a linear fit to the plotted slope values.

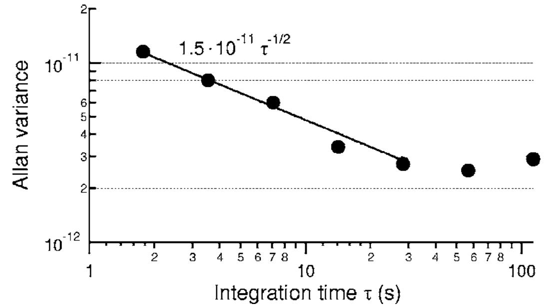

Fig. 4. Measured frequency stability of a 100 MHz crystal oscillator locked to the ^{87}Rb N-resonance, relative to a hydrogen maser. The solid line is a fit to the early time measurements.

In conclusion, we have demonstrated cancellation of first-order light shifts for an all-optical, three-photon-absorption N-resonance on the D_1 line of ^{87}Rb vapor. By employing this light-shift cancellation in a tabletop apparatus that was not engineered for stable clock performance, we nonetheless observed N-resonance frequency stability comparable to or better than that of 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.

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I. Novikova's e-mail address is i.novikova@osa.org.

*Also at the Lebedev Institute of Physics, Moscow, Russia.

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