Lineshape asymmetry for joint coherent population trapping and three-photon *N* resonances

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Received April 16, 2008; revised May 21, 2008; accepted May 27, 2008;
posted June 9, 2008 (Doc. ID 95131); published June 30, 2008

We show that a characteristic two-photon lineshape asymmetry arises in coherent population trapping (CPT) and three-photon (*N*) resonances, because both resonances are simultaneously induced by modulation sidebands in the interrogating laser light. The *N* resonance is a three-photon resonance in which a two-photon Raman excitation is combined with a resonant optical pumping field. This joint CPT and *N* resonance can be the dominant source of lineshape distortion, with direct relevance for the operation of miniaturized atomic frequency standards. We present the results of both an experimental study and theoretical treatment of the asymmetry of the joint CPT and *N* resonance under conditions typical to the operation of an *N* resonance clock. © 2008 Optical Society of America

OCIS codes: 020.1670, 020.3690, 120.3930, 300.6380.

In compact, all-optical atomic clocks employing coherent population trapping (CPT) [1–4], or three photon *N* resonances [5,6,10] interrogating light fields are typically generated by current modulating a single-mode diode laser with a large modulation index. This creates additional optical fields that can also drive atomic resonances. As we show here, both *N* resonances [6] and CPT resonances [7] are typically present. These two processes may then interfere and undergo differential ac Stark shifts, which generally lead to an asymmetric joint resonance. Temporal variation in lineshape asymmetry has been shown experimentally to lead to clock frequency instabilities [8].

An *N* resonance is a three-photon, two-optical-field absorptive resonance. A probe field Ω_1, resonant with the transition between the higher-energy hyperfine level of the ground electronic state and an electronically excited state, optically pumps the atoms into the lower hyperfine level. This probe field, Ω_1, also acts on the lower hyperfine state in combination with a drive field Ω_0 detuned from the probe field by the atomic hyperfine frequency h. Together, Ω_1 and Ω_0 create a two-photon Raman resonance that coherently drives atoms from the lower to the upper hyperfine level. This causes increased absorption of the probe field Ω_1 in a narrow resonance with linewidth $\Delta\nu$, set by the ground-state hyperfine decoherence rate.

A practical *N*-resonance clock creates the fields Ω_0 and Ω_1 using modulation of a single laser source. The laser carrier field Ω_0 is detuned by approximately the ground-state hyperfine frequency below the $F=2 \rightarrow F'=1$ transition, and the modulation frequency is set so that the first sideband Ω_1 is resonant with that transition, leading to an *N* resonance. Additional sidebands such as Ω_2 [see Fig. 1(a)] are also present. These sidebands participate with Ω_0 and/or Ω_1 in CPT resonances, which simultaneously compete with the *N* resonances, leading to an overall lineshape asymmetry. While an ideal CPT resonance produced by two optical fields will not have an associated *N* resonance, additional optical fields are typically present in CPT clocks driven by a modulated laser. At the large modulation index used in CPT clocks [9], higher-order sidebands that drive pairs of *N* resonances are present. For pure phase modulation of the optical field, the asymmetric signals from these *N* resonances cancel. In the presence of amplitude modulation this cancellation is incomplete, and residual asymmetry may become significant.

Our experimental studies of joint *N*+CPT resonances [9–11] used a beam of 795 nm light from a diode laser, tuned near the ^{87}Rb D_1 transition and modulated at the hyperfine frequency by an electro-optic modulator (EOM). The laser light was circularly polarized and sent through a heated ^{87}Rb vapor cell (65 °C, Neon buffer gas of 30 Torr). The vapor cell was housed in high-permeability magnetic shields, inside of which was a uniform longitudinal magnetic field (used to split the Zeeman degeneracy) created by a solenoid. In results presented here, the $\Delta m=0$, magnetic field-independent transition was studied. A

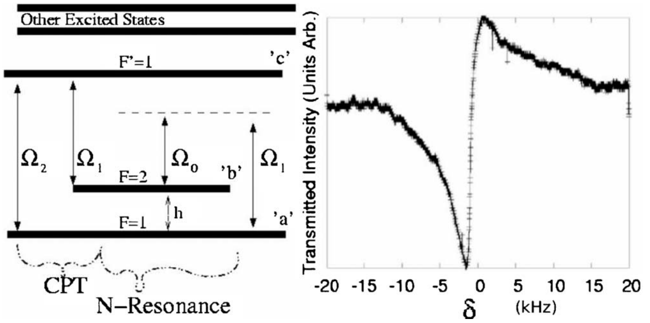

Fig. 1. (a) Simplified level diagram with applied fields Ω_0 (carrier) and Ω_1, Ω_2 (sidebands). (b) Example experimental *N*+CPT joint resonance, illustrating the typical asymmetry of the transmitted probe field Ω_1 lineshape in the presence of the Ω_2 sideband. The x axis is the two-photon detuning. The laser power is 0.088 mW, and the one-photon detuning is 350 MHz.

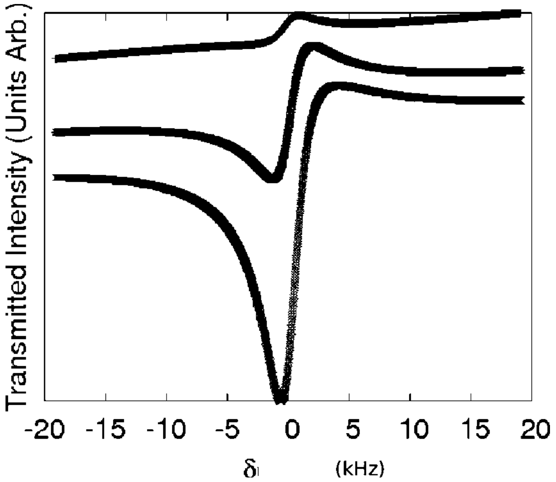

Fig. 2. Numerical calculations of the probe field Ω_1 transmission intensity for (top) CPT, (bottom) N, and (middle) joint N+CPT resonances, vertically offset for clarity. All parameters are for the ^{87}Rb atom with Rabi frequencies typical of experiments and a 100 MHz one-photon detuning (causing the background to not be level). The x axis is two-photon detuning.

temperature-stabilized Fabry–Perot (FP) cavity after the cell selected the +1 sideband, whose intensity was measured with a photodiode. Additionally, for elimination of the +2 sideband before the cell, the laser beam was retroreflected off a second FP tuned to pass only the +2 sideband. Two-photon lineshapes were measured for various laser detunings and powers. In all data presented, the rf (EOM) modulation index was fixed at 0.6. For example, Fig. 1(b) shows a typical measured lineshape for the transmitted probe field Ω_1, illustrating the asymmetry of the joint N+CPT resonance.

To the usual three-state Λ system used to model CPT, we append an additional excited state with dipole coupling to the two ground states. This fourth state is assumed to be far off-resonance and accounts for the nonresonant dipole polarizability to which many excited states may contribute. The optical fields Ω_1 and Ω_2 form the CPT system, whereas Ω_0 and Ω_1 participate in the N resonance [see Fig. 1(a)]. Δ is the (one-photon) laser detuning of Ω_1, and δ is the two-photon detuning of the laser fields for both the CPT and N resonances.

Using our model we find that to leading order in the optical fields, the transmission of the probe field Ω_1 is proportional to $T(\Delta) - Im\left(\rho_{ab}\frac{\Omega_0}{\Omega_1(h-\Delta)}\right)$. Here $T(\Delta)$ is the transmission independent of the CPT and N resonances, h is the hyperfine frequency, and ρ_{ab} is the ground-state coherence. In steady state we find

$$\left(\Gamma - i\left(\delta + \frac{|\Omega_1|^2 - |\Omega_0|^2}{4(h-\Delta)}\right)\right)\rho_{ab} = -i\frac{\Omega_1}{2}\rho_{cb} + i\frac{\Omega_2}{2}\rho_{ac} + \frac{\Omega_1\Omega_0}{4(h-\Delta)}(\rho_{bb} - \rho_{aa}), \quad (1)$$

where Γ is the ground-state depolarization rate and the subscripts refer to the atomic levels, as shown in Fig. 1(a). The RHS of the first line in Eq. (1) is associated with the CPT resonance, and the last line is the contribution of the N resonance. This expression is derived by adiabatically eliminating the nonresonant states contributing to the atomic polarizability. The equation for the ground-state population difference, $\rho_{bb} - \rho_{aa}$, is structurally equivalent to Eq. (1), with an N resonance term proportional to ρ_{ab} and a CPT term linear in ρ_{ac} and ρ_{bc}. Since atomic coherences (e.g., ρ_{cb}) scale as Ω/Δ, the leading order CPT and N resonance driving terms are of the same order at relevant one-photon detunings. Note that for the numerically calculated results presented below, the contribution from the full excited-state manifold is utilized, whereas Eq. (1) is for only a single resonant excited state.

Numerical calculations of the probe field Ω_1 transmission intensity are shown graphically in Fig. 2 in the approximation that the vapor cell is optically thin. The limiting cases of pure CPT resonance ($\Omega_0 = 0$) and ideal N resonance ($\Omega_2 = 0$) are not centered at the same two-photon detuning, because they experience different ac Stark shifts. The relative amplitudes for CPT and N resonances are typical of experimental results.

For comparison between theory and experiment we quantify the lineshape asymmetry by fitting to a skew Lorentzian [12]:

$$I(\delta) = C + D\delta + \frac{A\Gamma + B(\delta - \delta_0)}{(\delta - \delta_0)^2 + \Gamma^2}. \quad (2)$$

Here $I(\delta)$ is the transmitted Ω_1 intensity of the combined N+CPT resonance, δ is the two-photon frequency, and A, B, C, and D are fitting amplitudes. The fitting parameter Γ is proportional to the resonance linewidth, and δ_0 is the two-photon resonance frequency. The amplitudes A and B describe the symmetric (Lorentzian) and antisymmetric (dispersive) components of the lineshape, respectively. We define

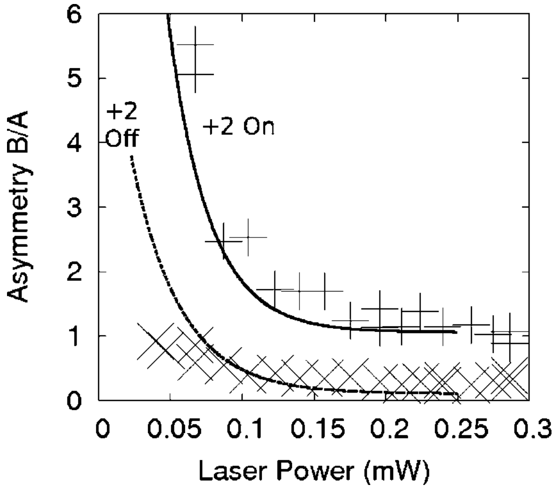

Fig. 3. Lineshape asymmetry B/A for N+CPT resonances on the D_1 transition of ^{87}Rb with and largely without (85% reduced intensity) the +2 sideband. Numerical calculations based on our model are the lines; the "+" and "x" are the associated fitted B/A from experimental data.

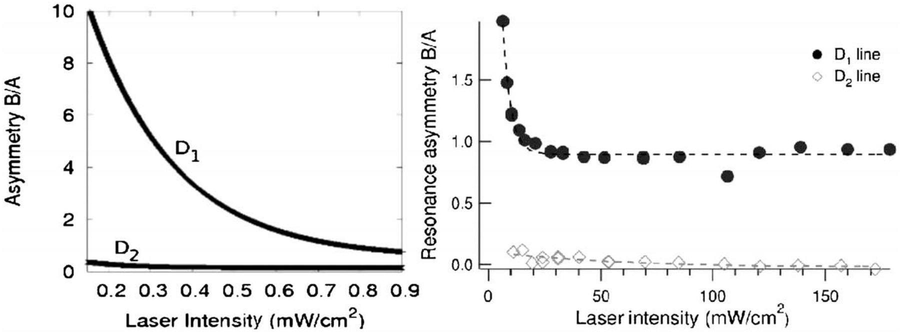

Fig. 4. Comparison of N+CPT lineshape asymmetry for D_1 and D_2 transitions: (a) Numerical calculations and (b) experiment (data from [11]). Note that the experimental regime of [11] is different from that of the experiment of Fig. 3 and is at higher light power than our model can pertubatively accomodate.

the line asymmetry as the dimensionless ratio B/A. As an example, consider the effect of nonresonant light fields; these fields are time varying in the rotating frame and thus contribute ac Stark shifts to each transition. The sign of these shifts depends on the sign of the nonresonant field's detuning. Different ac Stark shifts for the CPT and N resonances force the maximum of the optical response away from $\delta=\delta_0$, creating an overall asymmetric lineshape. The B term, linear in the two-photon detuning $\delta-\delta_0$, accommodates this effect.

We fit the skew Lorentzian [Eq. (2)] to both experimental data and numerical calculations using the model described above with atomic parameters for the ^{87}Rb atom. We find that the lineshape asymmetry B/A decreases as the overall laser intensity increases in both cases (Fig. 3). This reduction of asymmetry with increasing optical power is consistent with our model of the different multiphoton (nonlinear) natures of the CPT and N resonances. As a three-photon process, the N resonance contrast scales faster with laser power than that of the two-photon CPT process [see Eq. (1)]; thus lineshape asymmetry is less pronounced at large total power. Furthermore, the line asymmetry is greatly reduced over the entire range when the +2 sideband is suppressed (in our experiments by approximately 85% in intensity) *before* the light enters the atomic vapor cell. Our model indicates that reduction of the +2 sideband greatly inhibits the CPT resonance. The asymmetry B/A in the two-photon line shape is proportional to the ratio of the Rabi frequencies Ω_2/Ω_0 and decreases with increasing ground-state population difference. This connection between the parameterization of Eq. (2) and the full model explains the qualitative features of Figs. 3 and 4.

In another example, our model explains the difference between the observed lineshape asymmetries of N+CPT resonances on the D_1 and D_2 optical transitions [11]. Under identical conditions the CPT resonance has lower contrast on the D_2 transition as compared to that of D_1 [13], This is a consequence of direct depolarization transitions ($F=2\rightarrow F'=3$) for the D_2 drive and the resultant suppression of optical pumping of the ground state. Under identical conditions, the D_2 N resonance is more symmetrical and has higher contrast than the D_1 N resonance (Fig. 4).

In conclusion, we have shown that a characteristic two-photon lineshape asymmetry arises in CPT and N resonances owing to modulation sidebands in the interrogating laser light. A simple model for the combined effect of these optical fields in the joint N+CPT system explains quantitatively many observed features of the lineshape asymmetry. The effect described here is most relevant for N-resonance-based clock stability but can also contribute to the optical response of CPT-based clocks as well when driving fields are not perfectly balanced.

We acknowledge support from the Office of Naval Research, the Institute for Theoretical Atomic and Molecular Physics, and the Radcliffe Institute for Advanced Studies.

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