Controlling the light shift of the CPT resonance by modulation technique

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Abstract. Motivated by recent developments in atomic frequency standards employing the effect of coherent population trapping (CPT), we propose a theoretical framework for the frequency modulation spectroscopy of the CPT resonances. Under realistic assumptions we provide simple yet non-trivial analytical formulae for the major spectroscopic signals such as the CPT resonance line and the in-phase/quadrature responses. We discuss the influence of the light shift and, in particular, derive a simple expression for the displacement of the resonance as a function of modulation index. The performance of the model is checked against numerical simulations, the agreement is good to perfect. The obtained results can be used in more general models accounting for light absorption in the thick optical medium.

1. Introduction
The effect of coherent population trapping (CPT) allows to realize narrow resonances [1, 2, 3] and therefore finds promising applications in frequency standards and quantum metrology. In a conventional setup, a local oscillator is locked to the energy gap between the lower states of the three-level atomic system, see Fig. 1: at an exact resonance the atoms reside in the so-called dark state not absorbing the probing light. Deviations from the resonance are detected by the frequency modulation technique by means of the in-phase response of the slowly modulated probing field. A challenging issue in the design of such atomic standards is the dynamic Stark shift of the atomic levels causing drift and noise in the clock output. There are different remedies for that adverse effect among which we point out the technique [4] where the authors propose to use a combination of both in-phase and quadrature responses to control the deviations from the resonance and simultaneously suppress the light shift. To the best of our knowledge, this technique has never been applied to the CPT-based atomic clocks. Therefore, it makes practical sense to investigate the behavior of the three-level atomic system under conditions used in the FM spectroscopy and, in particular, in the context of the proposal [4]. To be more specific, in the model we introduce below we consider a laser wave interacting with atomic ⁸⁷Rb vapor and resonant to D₁-line transition that is quite conventional in the real CPT-experiments.
2. Model
The internal structure of the Rb atoms interacting with light under the CPT resonance conditions is modeled by a standard Λ-scheme of levels, see Fig. 1. The laser field \( \mathcal{E}_0 \cos(\omega_0 t + a \sin \Phi(t)) \) propagating through the atomic cell is tuned to the optical frequency \( \omega_0 \) such that \( \hbar \omega_0 = (E_a + E_b)/2 \) where \( E_a, E_b, E_c \) are the energies of the respective atomic states. The laser field is frequency modulated due to the term \( a \sin \Phi(t) \) where \( a \) is a modulation index and \( \Phi(t) = \Omega + b \sin \omega_m t \) is the time dependent phase. The instantaneous frequency of modulation, \( \Phi'(t) = \Omega + b \omega_m \cos \omega_m t \), suggests that the modulation is not one-tone at the frequency \( \Omega \). Rather, the microwave frequency \( \Omega \equiv \omega_0/2 + \delta \) is modulated as well at a much lower rate \( \omega_m \) with the modulation index \( b \) and frequency deviation \( b \omega_m \). The rationale behind such nested modulation is simple: the modulation at \( \Omega \) creates the sideband frequencies \( \omega_0 \mp \Omega \) resonant to the transitions \( |a\rangle, |b\rangle \leftrightarrow |c\rangle \) and thus creates the CPT resonance per se whereas the modulation at \( \omega_m \) provides additional spectroscopic signals measured over time against the detuning \( \delta \) from the exact resonance and used for stabilizing frequency of the atomic clocks. We emphasize that the microwave frequency \( \Omega/2\pi \approx 3.4 \text{ GHz} \) differs from the slow modulation rate \( \omega_m/2\pi \sim 0.1 \pm 1 \text{ kHz} \) by 6 to 7 orders of magnitude that makes the direct numerical simulations somewhat expensive but, fortunately, facilitates the analytical treatment of the problem.

3. Approximate solution
To describe the model outlined above, we use the optical Bloch equations for the atomic density matrix \( \dot{\rho}(t) = \sum_{\alpha, \beta = \{a, b, c\}} \rho^{\alpha\beta}(t) |\alpha\rangle \langle \beta| \):

\[
i \left( \frac{d}{dt} + \dot{\Gamma} \right) \dot{\rho} = \left[ \hat{H}, \dot{\rho} \right], \tag{1}
\]

where the Hamiltonian \( \hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \) reads

\[
\hat{H}_0 = \frac{\omega_g}{2} |a\rangle \langle a| - \frac{\omega_g}{2} |b\rangle \langle b|, \quad \hat{H}_{\text{int}} = -V(t) |c\rangle \langle a| - V(t) |c\rangle \langle b| + \text{h.c.}
\]

The interaction Hamiltonian, \( \hat{H}_{\text{int}} \), is written under the resonant approximation with respect to optical transitions \( |a\rangle, |b\rangle \leftrightarrow |c\rangle \) with the time dependent interaction term \( V(t) = d \mathcal{E}_0/2\hbar \propto \exp(-ia \sin \Phi(t)) \) where \( d = d^* \equiv \langle c| \hat{d}| a\rangle = \langle c| \hat{d}| b\rangle \) is a dipole matrix element which is supposed to be real and equal for both transitions.

The operator \( \dot{\Gamma} \) in (1) is

\[
\dot{\Gamma} \dot{\rho} = -\rho^{cc} \{ \gamma_a |a\rangle \langle a| + \gamma_b |b\rangle \langle b| \} + (\gamma_a + \gamma_b) \rho^{cc} |c\rangle \langle c| + \Gamma \left\{ \rho^{ca} |c\rangle \langle a| + \rho^{cb} |c\rangle \langle b| + \text{h.c.} \right\}. \tag{2}
\]
accounts for the incoherent phenomena of two different kinds: (i) pumping of the lower levels due to radiative decay of the upper state at a rate of \( \gamma = \gamma_a + \gamma_b \) (the first and second terms in (2)) and (ii) decay of optical coherences \( \rho_{ca} \), \( \rho_{cb} \) at a rate of \( \Gamma \gg \gamma \) due to other incoherent processes such as collisions of Rb atoms with atoms of the buffer gas (the third term).

The interaction term \( V(t) \) admits the following decomposition:

\[
V(t) = \frac{d\mathcal{E}_0}{2\hbar} e^{-ia\sin\Phi(t)} = \sum_{k=-\infty}^{\infty} \tilde{V}_k(t) e^{-ik\Omega t}, \quad \tilde{V}_k(t) = V_k e^{-ik\varphi(t)}, \quad V_k = \frac{d\mathcal{E}_0}{2\hbar} J_k(a),
\]

where \( \varphi(t) = b \sin \omega_m t + \) and \( J_k(\cdot) \) is the Bessel function of the first kind. As \( \omega_m/\Omega \) is of the order of \( 10^{-7} \div 10^{-6} \), representation (3) means that the interaction can be viewed as a sum of rapidly oscillating harmonics with slowly varying amplitudes. It is therefore tempting to separate the time dependence of different scales for the density matrix as well,

\[
\rho^{\alpha\beta}(t) = \sum_{k=-\infty}^{\infty} \rho^{\alpha\beta}_k(t) \exp(-ik\Omega t), \quad \alpha, \beta = \{a, b, c\},
\]

where the amplitudes \( \rho^{\alpha\beta}_k(t) \) are supposed to change at multiples of the characteristic rate \( \omega_m \). Note also that the spectroscopic signals measured in the experiment are the mean light absorption in the atomic cell, i.e., the mean population of the upper level, \( \langle \rho^{ee}(t) \rangle \), and its Fourier amplitudes at the frequency \( \omega_m \), i.e., \( \rho^{ee}(t) \propto M_S \cos \omega_m t + M_Q \sin \omega_m t \). Obviously, both signals are recorded over times of the order of \( \omega_m^{-1} \) for which the fast oscillations at multiples of \( \Omega \) will be averaged out due to inertia of the experimental setup. As such, it suffices to calculate only the zeroth amplitude, \( \rho^{ee}_0(t) \), i.e., the time average over fast oscillations and use it further to obtain the above mentioned spectroscopic signals.

Projecting equations (1) onto the basis \( \exp(i k\Omega t) \) we get the equations for amplitudes in (4). In doing so, we treat the slow quantities \( \tilde{V}_k(t) \), \( \tilde{V}_k \) as constants neglecting thus small corrections of the order of \( \omega_m/\Omega \approx 10^{-7} \div 10^{-6} \). Below we provide a sketch of the solution to the resulting equations.

First, by nature of the experiment, we seek for a steady-state solution at large times \( \sim \omega_m^{-1} \) neglecting the time derivatives \( d/dt \sim \omega_m \ll \gamma \ll \Gamma \ll \Omega \) wherever possible. Second, the light field is assumed weak, \( d\mathcal{E}_0/2\hbar \ll \gamma \ll \Gamma \ll \Omega \), that holds in the real experiments (the low saturation regime). This implies that there are only three leading amplitudes of the order of unity in series (4), specifically, \( \rho^{aa}_0 \), \( \rho^{bb}_0 \), \( \rho^{ba}_0 \). To demonstrate this, let us consider the equation for \( \rho^{ba}_k(t) \) as an example:

\[
i \left( \frac{d}{dt} + k\Omega + \omega_g \right) \rho^{ba}_k = \sum_s \left( \tilde{V}_{s-k}(t) \rho^{aa}_s - \tilde{V}_{s+k}(t) \rho^{bb}_s \right).
\]

For any \( k \neq 2 \) the term \( k\Omega + \omega_g \) is non-zero and one can neglect the time derivative obtaining thus a steady-state \( \rho^{ba}_k(t) \). This solution is obviously suppressed by the magnitude of the parameter \( |V_k/\Omega| \ll 1 \) and can be neglected. Conversely, if \( k = 2 \), the sum \(-2\Omega + \omega_g = 0 \) can be arbitrarily small, the derivative cannot be omitted, and \( \rho^{ba}_2(t) \) is not necessarily small. The similar reasoning applies to amplitudes \( \rho^{aa}_k \), \( \rho^{bb}_k \) with a leading term at \( k = 0 \), but not to \( \rho^{ca}_k \) and \( \rho^{cb}_k \). The respective equations in the latter case contain an additional imaginary term \( i\Omega \) on the left-hand side that has no counterpart to cancel out for any \( k \). As such, all of the amplitudes \( \rho^{aa}_k \), \( \rho^{bb}_k \) are small.

Note also that though the population of the upper level is small under low saturation, \( \rho^{ee}_0(t) \ll 1 \), it is a quantity of primary interest responsible for the spectroscopic signals mentioned
above. The equation for $\rho_0^{ee}(t)$ is obtained from those for $\rho_0^{aa}(t)$, $\rho_0^{bb}(t)$ by means of the normalization condition $\rho_0^{aa} + \rho_0^{bb} + \rho_0^{ee} = 1$.

The steady-state solution to this equation reads

$$
\rho_0^{ee}(t) = \frac{2\lambda}{\gamma} \left[ 1 - \frac{2\lambda}{\Gamma} \text{Re}\{g(t)e^{-i2\varphi(t)}\} \right],
$$

(5)

where $\lambda \simeq \Gamma (d\mathcal{E}_0/2\hbar \Gamma)^2 J_k^2(a)$ and $g(t)$ is a solution to

$$
d\frac{dg}{dt} + 2[i\Delta + \lambda]g = \Gamma e^{i2\varphi(t)}.
$$

(6)

In equation (6), $\Delta = \delta - \delta_0$, $\delta_0 \simeq 2(a\Omega)^{-1} (d\mathcal{E}_0/2\hbar)^2 J_0(a)J_1(a)$. As $\exp(\pm i2\varphi(t)) = \sum_{k=-\infty}^{\infty} J_k(2b) \exp(\pm ik\omega_m t)$, the steady state solution to (6), $g(t)$, oscillates at multiples of $\omega_m$, and, consequently, so does $\rho_0^{ee}(t)$, see (5). The oscillations at zero frequency yield the mean absorption,

$$
\langle \rho_0^{ee}(t) \rangle = \frac{2\lambda}{\gamma} \left[ 1 - 4\lambda^2 \sum_{k=-\infty}^{\infty} \frac{J_k^2(2b)}{4\lambda^2 + (2\Delta + k\omega_m)^2} \right]
\approx \frac{2\lambda}{\gamma} \frac{\Delta^2}{\lambda^2 + \Delta^2},
$$

(7)

which gets simplified in the instructive case $b \ll 1$ where it has a Lorentzian profile of width $\lambda$ and centered at $\delta_0$. The non-zero constant $\delta_0$ at which the absorption vanishes is due to the light shift of the atomic levels caused by the entire spectrum of the propagating laser field. For some values of modulation index $a$, the light shift is compensated, $\delta_0 = 0$, see Fig. 3, that may be highly beneficial from the experimental standpoint. We also note that the dependence of $\delta_0$ on $a$ was studied in a number of works, e.g., [2], purely numerically.

Two other spectroscopic signals, the in-phase $M_S$ and quadrature $M_Q$ amplitudes, can be obtained by projecting (5) onto $\cos \omega_m t$ and $\sin \omega_m t$:

$$
M_S \propto \sum_{k=-\infty}^{\infty} J_k(2b) \left( \frac{J_{k+1}(2b)}{\lambda^2 + (\Delta' + k + 1)^2} + \frac{J_{k-1}(2b)}{\lambda^2 + (\Delta' + k - 1)^2} \right),
$$

(8)

$$
M_Q \propto \sum_{k=-\infty}^{\infty} J_k(2b) \left( \frac{J_{k-1}(2b)(\Delta' + k - 1)}{\lambda^2 + (\Delta' + k - 1)^2} - \frac{J_{k+1}(2b)(\Delta' + k + 1)}{\lambda^2 + (\Delta' + k + 1)^2} \right),
$$

(9)

where $\Delta' = 2\Delta/\omega_m$, $\lambda' = 2\lambda/\omega_m$ and $M_S$, $M_Q$ are defined up to an irrelevant $\delta$-independent constant. Both the in-phase and quadrature amplitudes turn to zero at $\delta = \delta_0$ whereas $M_Q$ vanishes at two extra points of $\delta$. The latter follows, for example, from expressions (8)-(9) simplified at $b \ll 1$:

$$
M_S \propto \frac{\Delta'}{[\lambda^2 + (\Delta' - 1)^2][\lambda^2 + (\Delta' + 1)^2]}, \quad M_Q \propto \frac{\Delta'[3\lambda^2 + 1 - \Delta'^2]}{[\lambda^2 + (\Delta' - 1)^2][\lambda^2 + (\Delta' + 1)^2][\lambda^2 + \Delta'^2]}.
$$

4. Numerical versus analytical results

In this section we compare the results of the preceding section to those obtained by integrating equations (1) numerically. We employ a Fortran multi-thread ODE solver, yet a typical computational time is as long as 2 to 20 hours depending on the ratio $\omega_m/\Omega$ that crucially affects the time step. We also check the results against a simplified 3-wave model where we keep only three terms in the spectral representation (3): the carrier, $k = 0$, and the first sidebands, $k = \pm 1$. One can easily show that the respective formulae under the 3-wave approximation
### Figure 2. The CPT resonance line, arbitrary units along the y-axis.

### Figure 3. Detuning $\delta_0$ as a function of modulation index $a$.

### Figure 4. The in-phase and quadrature amplitudes $(8)-(9)$, arbitrary units along the y-axis.

The parameters used throughout Figs. 2-4: $\Gamma/2\pi = 0.265$ GHz, $dE_0/(2\hbar \Gamma) = 1/1400$, $\omega_m/2\pi = 129$ Hz, $b = 1/2$, $a = 1.5$ (except for Fig. 3).

coincide with those of Section 3 but the displacement of resonance minimum in this case reads:

$$\delta_0 = \sum_{k=1}^{2} \frac{k\Omega V_k^2}{(k^2\Omega^2 + \Gamma^2)}.$$

In Figs. 2-4 we plot the CPT resonance line, the displacement of resonance, $\delta_0$, versus modulation index $a$ and the profiles of the in-phase and quadrature amplitudes. We see that the full-spectrum model, the 3-wave approximation and the direct numerical integration are in a very good agreement with one another. The 3-wave model, however, overestimates the displacement $\delta_0$ due to lack of the higher sidebands which effectively compress the energy distance between the lower states $|a\rangle$ and $|b\rangle$.

### 5. Conclusion

In summary, we proposed a theoretical framework for the FM spectroscopy of the CPT resonance and attested it using numerical simulations. In particular, we quantified the influence of the light shift of the atomic levels on the location of resonance that may be important for selecting the zero light shift regime making the atomic clocks less sensitive to variations of light intensity.

We would like to add that our results are in good agreement with a recent experiment [5]
on the FM spectroscopy of absorption in Rb vapor. However, the experimental in-phase and quadrature amplitudes vanish at two different points of $\delta$ as opposed to formulae (8)-(9) which predict a common zero of $M_S$ and $M_Q$ (we mean the central zero for the latter).

In that regard, we also refer to the technique [4] where a combined use of the in-phase and quadrature signals for stabilizing atomic frequency standards relies on the difference between the zeros of the respective amplitudes. Under that approach, the in-phase response is used to lock the local oscillator to the atomic resonance whereas the difference between the zeros of the in-phase and quadrature signals allows to control the light shift of the atomic levels. The above mentioned splitting of zeros may be attributed to the absorption of laser field in the optically thick atomic cell [4]. Therefore, we feel a need to generalize the proposed model onto a thick optical layer case. We plan to address this important problem in the nearest future.

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