Analytical theory of dual-frequency sub-Doppler spectroscopy of alkali-metal atoms

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Introduction:

In works [1, 2] a new approach to the creation of miniature optical quantum frequency standards was proposed and investigated. To obtain a reference resonance in these works, colliding bichromatic laser beams with orthogonal linear polarizations are used. It was shown in [2] that, in such a configuration, it is possible to observe sub-Doppler resonances with a high contrast. Numerical simulations of these resonances were carried out in [3, 1]. In this work we present some analytical results that provide a deeper understanding of the physics of the observed effects.

1.Brazhnikov D. et al. Dual-frequency sub-Doppler spectroscopy: Extended theoretical model and microcell-based experiments //Physical Review A. – 2019. – Т. 99. – №. 6. – С. 062508.
2.Brazhnikov D. V. et al. Two-frequency sub-Doppler spectroscopy of the caesium D1 line in various configurations of counterpropagating laser beams //Quantum Electronics. – 2020. – Т. 50. – №. 11. – С. 1015.
3.Hafiz M. A. et al. High-contrast sub-Doppler absorption spikes in a hot atomic vapor cell exposed to a dual-frequency laser field //New Journal of Physics. – 2017. – Т. 19. – №. 7. – С. 073028.

Coherent population trapping (CPT):

Here, we use a simplified spectroscopic model based on a three-level Λ-scheme. The scheme is shown in figure 1. Λ-scheme interacts with a dual-frequency laser field composed of two counter-propagating, linearly polarized plane waves. Expression for the light field with two frequency components ω_1 and ω_2 :

$$E(z,t) = E_1\left(e^{-i(\omega_1 t - k_1 z)} + e^{-i(\omega_2 t - k_2 z)}\right) + E_2\left(e^{-i(\omega_1 t + k_1 z + \varphi_1 + \varphi)} + e^{-i(\omega_2 t + k_2 z - \varphi + \varphi_2)}\right) + \mathrm{C.C.}$$

The angle φ is the mutual angle between linear polarizations of counterpropagating waves. The angles $\varphi_{1,2}$ is the spatial phase shifts.

The field E_1 pumps atoms with velocity -*v* into the non-coupled (dark) state |NC1⟩ while the field E_2 pumps those of velocity +*v* into the non-coupled state |NC2⟩ :

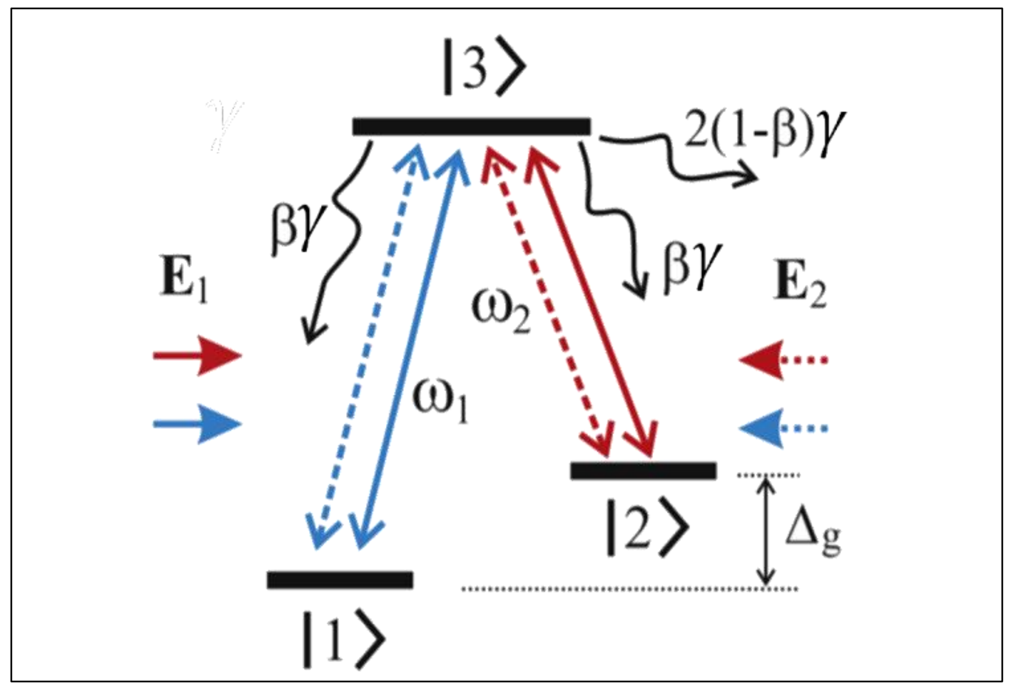

Fig. 1 Λ – scheme of atomic energy levels. Solid vertical arrows denote transitions induced by the light-wave components with vectors $k_{1,2}$, , while dashed vertical arrows are attributed to the waves with - $k_{1,2}$. Wavy black arrows denote spontaneous decay processes. γ – is relaxation rate, β – is the openness coefficient.

$$|NC_1\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle - e^{i\omega_{12}t + ik_{12}z}|2\rangle\right), \qquad |NC_2\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle - e^{i\omega_{12}t - i(k_{12}z + \varphi_{12} - 2\varphi)}|2\rangle\right).$$

Now, we consider the situation where the fields E_1 and E_2 start to interact with the same atoms. There are two main cases in this configuration:

(1) $|NC_1\rangle$ and $|NC_2\rangle$ interfere destructively, i.e. $\langle NC_1|NC_2\rangle = 0$;

(2) the 'competition' between $|NC_1\rangle$ and $|NC_2\rangle$ does not destroy the CPT effect, $\langle NC_1|NC_2\rangle = 1$.

In the first case, atoms start to absorb intensively and scatter energy from the light field since they are no longer in the dark state.

Math model:

Equation for one-atom density matrix in the Wigner representation:

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}\left[\hat{H}_0 + \hat{V}, \hat{\rho}\right] + \hat{R}\{\hat{\rho}\}$$

The Hamiltonian $\hat{H}_0$ of a free atom in the basis of eigenfunctions:

$$\hat{H}_0 = \sum_{n=1}^{3} \mathcal{E}_n |n\rangle\langle n|, \qquad \hat{V} = -\vec{E}\hat{\vec{d}}$$

The system of equations for the density matrix in the case of a linear approximation in the test field. :

$$\rho_{11}^{(0)}\left(\Gamma + 2\gamma_{eg} S_1^{(-)}\right) - \rho_{33}^{(0)}\left(\gamma\beta + 2\gamma_{eg} S_1^{(-)}\right) + \rho_{21}^{(+)} R^2 L^{(-)^*} + \rho_{12}^{(-)} R^2 L^{(-)} = \frac{\Gamma}{2},$$

$$\rho_{22}^{(0)}\left(\Gamma + 2\gamma_{eg} S_1^{(-)}\right) - \rho_{33}^{(0)}\left(\gamma\beta + 2\gamma_{eg} S_1^{(-)}\right) + \rho_{21}^{(+)} R^2 L^{(-)^*} + \rho_{12}^{(-)} R^2 L^{(-)^*} = \frac{\Gamma}{2},$$

$$\left(\rho_{11}^{(0)} + \rho_{22}^{(0)}\right)\Gamma + \rho_{33}^{(0)}\left(\Gamma + 2\gamma(1-\beta)\right) = \Gamma,$$

$$\rho_{11}^{(0)} L^{(-)} R^2 + \rho_{22}^{(0)} L^{(-)^*} R^2 - \rho_{33}^{(0)} R^2\left(L^{(-)^*} + L^{(-)}\right) + \rho_{12}^{(-)}\left(\Gamma + R^2\left(L^{(-)^*} + L^{(-)}\right)\right) = 0,$$

$$\rho_{11}^{(0)} L^{(-)^*} R^2 + \rho_{22}^{(0)} L^{(-)} R^2 - \rho_{33}^{(0)} R^2\left(L^{(-)^*} + L^{(-)}\right) + \rho_{21}^{(+)}\left(\Gamma + R^2\left(L^{(-)^*} + L^{(-)}\right)\right) = 0.$$

Population of the excited level in the approach of a standing wave:

$$\rho_{33} = \frac{2\gamma_{eg}\tau\left(S_1^{(-)} + S_2^{(+)}\right)}{1 + 2\gamma\tau} + \frac{(2\gamma_{eg}\tau)^2}{(1+2\gamma\tau)^2}\left(\left({S_1^{(-)}}^2 + {S_2^{(+)}}^2\right)(4 + 2\gamma\tau(2-\beta)) + S_1^{(-)} S_2^{(+)}(6 + 4\gamma\tau(1-\beta))\right) + 2Re\left(\frac{-(2\gamma_{eg}\tau)^2}{1+2\gamma\tau} S_1^{(-)} S_2^{(+)} e^{-i(\theta + 2k_{12}z)}\right)$$

$S_n^{(\pm)} = R_n^2/({\gamma_{eg}}^2 + (\delta \pm kv)^2)$ – saturation parameters, $L^{(\pm)} = 1/(\gamma_{eg} + i(\delta \pm kv))$ – complex Lorentzian, $R = Ed/\hbar$ – Rabi frequencies, δ – one photon detuning, $\gamma_{eg} = \gamma + \Gamma$ - relaxation rate of optical coherences with Γ the time-of-flight relaxation rate attributed to the finite time of atom–field coherent interaction, $\tau = \Gamma^{-1}$, $\theta = \varphi_1 - \varphi_2 - 2\varphi$.

Results:

Linear approximation in the test field:

In this case, we consider the dependence of the absorption coefficient on the frequency detuning. The expression for the absorption coefficient will take the form:

$$\alpha = \frac{4\pi k N d^2 \gamma}{\left(\gamma^2 + (\delta + x)^2\right)\hbar}\left(\rho_{11} - \rho_{33}\right) + 4\pi k N \frac{d^2}{\hbar}\,\mathrm{Re}\left(L^{(+)*} e^{i\theta} \rho_{21}{}^{(+)}\right)$$

Averaging over the velocities of the thermal motion of atoms, we obtain the curve shown in Fig. 2. The absorption coefficient can be represented as the sum of the Doppler and sub-Doppler parts: $\alpha = \alpha_{\mathrm{Doppler}} + \alpha_{\mathrm{sub-Doppler}}$. Figure 3 shows the graphs for each contribution and the dependence on the total θ phase.

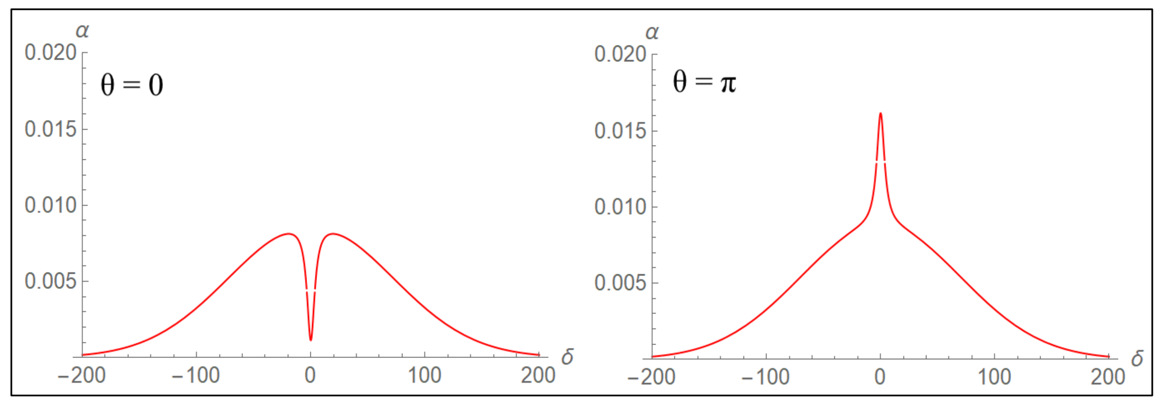

Fig. 2 Dependence of the absorption coefficient on the frequency detuning δ, R=0.5γ, β = 1γ, Γ = $10^{-2}\gamma$, kv_0 = 100γ

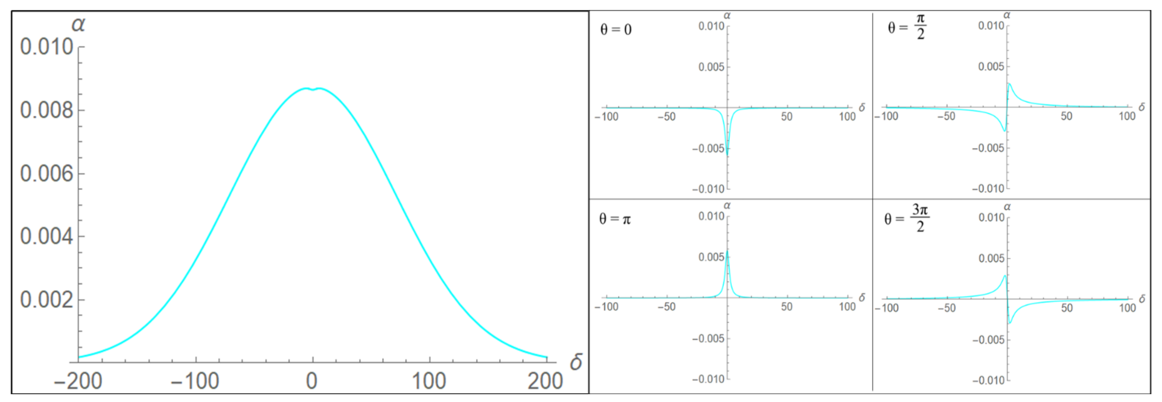

Fig. 3 Doppler (left) and sub-Doppler (right) contributions.

Standing wave mode (Interaction of two weak counterpropagating waves):

In this case, we consider the dependence of the population of the excited level on the frequency detuning. After averaging over the velocities of the thermal motion, the expression for the population of the excited level can be represented as the sum of the linear and non-linear parts. The non-linear part can be thought of as the sum of several different non-linear effects: $\rho_{33}{}^{\text{non-linear.}} = S_{\text{self-saturation}} + S_{\text{CPT}} + S_{\text{res.saturation}} + S_{\text{CPT interference}}$.

$$\frac{S_{\text{CPT}}}{S_{\text{self-saturation}}} \approx \frac{S_{\text{CPT interference}}}{S_{\text{res.saturation}}} \approx \frac{1+2\gamma\tau}{3} \approx \frac{2}{3}\gamma\tau.$$

It can be seen from the presented ratio of the quantities that the CPT effect prevails over the effect from the saturated absorption resonance. If the $\cos(\theta + 2k_{12}z) = -1$ contributions from CPT and SAR have opposite values: SAR always leads to a dip. Since the value of the CPT contribution is larger, in this case a contrast absorption peak is formed in the center of the resonance curve, rather than a dip.

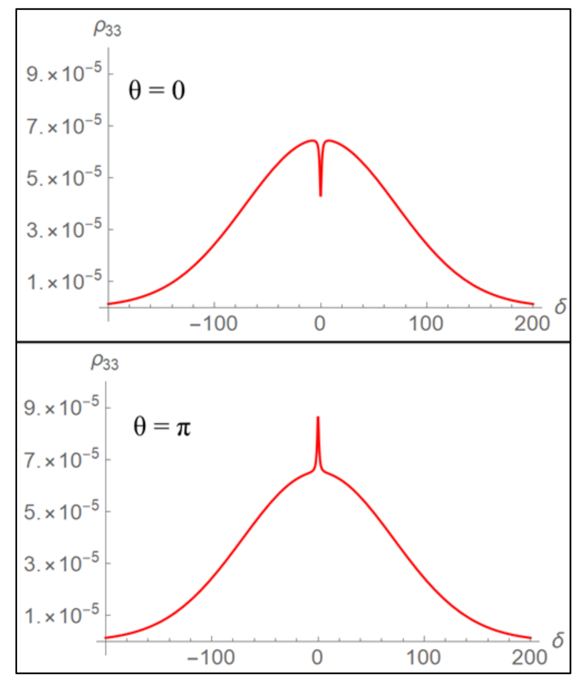

Fig. 4 Dependence of the population of the excited level on the frequency detuning δ. R = 0.05 γ, β = 1γ, Γ = $10^{-2}\gamma$, x_0 = 100γ