Ultrahigh-Contrast Saturated-Absorption Resonance to Enhance Stability of CPT Atom Clocks

Moustafa Abdel Hafiz¹, Denis Brazhnikov^{2,3}, Grégoire Coget¹, Alexei Taichenachev^{2,3}, Valeriy Yudin^{2,3,4}, Emeric de Clercq⁵ and Rodolphe Boudot¹

¹ Department of Time and Frequency Standards, FEMTO-ST, CNRS, UBFC, Besancon, France

²Institute of Laser Physics SB RAS, Novosibirsk, Russia

³ Novosibirsk State University, Novosibirsk, Russia

⁴Novosibirsk State Technical University, Novosibirsk, Russia

⁵ LNE-SYRTE, Observatoire de Paris, PSL Research University, CNRS,

Sorbonne Universités, UPMC Univ. Paris, France

Emails: brazhnikov@laser.nsc.ru, rodolphe.boudot@femto-st.fr, emeric.declercq@obspm.fr

Abstract—The results of detailed experimental and theoretical investigations of new nonlinear effect in the field of saturatedabsorption spectroscopy of atom vapours are presented. The effect consists in observation of a high-contrast natural-linewidth absorption spike when ¹³³Cs atoms are being irradiated by the bichromatic counterpropagating laser beams. The results obtained can be useful in various fields of modern laser physics and applications where saturated-absorption resonances are used. For instance, the effect has been already implemented in the coherent-population-trapping atom clocks to enhance stabilization of optical frequencies and to improve stability of the clocks on the whole.

Keywords—saturated-absorption resonance; microwave atom clocks; coherent population trapping; quantum metrology

I. INTRODUCTION

Saturated-absorption resonance (SAR) is a widely used tool in fundamental laser spectroscopy of atoms and molecules [1], and in many experiments with laser radiation. It plays an important role in quantum metrology for producing optical frequency standards including compact and mobile versions for ground and space-borne applications. For instance, many laboratories develop He-Ne, Nd:YAG, Yb:YAG, diode and other lasers frequency locked to optical transitions in iodine or methane molecules by means of SAR, including stabilization of optical frequency combs (for instance, see [2-4]). Besides the molecular vapours, the alkali-metal atoms are also used for the same purposes [5,6].

New bright feature of SAR has been discovered in the recent experiments [7,8]. It consists in observation of a high-contrast natural-linewidth absorption spike under linearly polarized counterpropagating laser beams (see an example of the signal on Fig. 1 and a part of experimental setup on Fig. 2). Each of the beams has two optical frequency components to simultaneously excite both hyperfine levels of the Cs ground state (Fig. 3). Such kind of the natural-linewidth absorption peaks were observed and studied before. Indeed, it is well-known that several physical reasons may cause a nonlinear

absorption peak creation at the center of absorption profile. For instance, among these effects are the recoil effect [9,10], highorder spatial harmonics of atomic polarization [11], optical pumping effects [12–14] and magnetic-field-caused splitting [15]. Besides, Doppler-effect peculiarities for moving atom in a gas, whose open optical transition is being excited by two counterpropagating light waves, can also lead to the absorption peak observation [16,17].

Regularly, the contrast of SAR with respect to a wide background signal is not high in many studied configurations. However, the recent experiments [7,8] have revealed a saturated-absorption peak with ultrahigh contrast. In particular, under the appropriate conditions amplitude of the central resonance could be twice bigger than amplitude of the Doppler-broadened background. Moreover, the SAR in the considered light-field configuration has been studied at first time.



Fig. 1. Experimental resonance curves. The total laser power is 500 μ W, polarizations of the counterpropagating light beams are perpendicular. Fg, Fe are the total angular momenta of the ground and excited energy levels of Cs.



Fig. 2. Experimental setup: DFB – distributed feedback diode laser, OI – optical isolator, EOM – electro-optic modulator, LO – microwave frequency synthesizer, PBS – polarizing beam splitter, M – mirror, PD – photodiode.



Fig. 3. A-scheme of atomic energy levels in ^{133}Cs : $|F_g = 3\rangle \equiv |1\rangle$, $|F_g = 4\rangle \equiv |2\rangle$, $|F_e = 4\rangle \equiv |3\rangle$. Spontaneous relaxation rate of the excited state equals to 2γ . Branching ratio β governs the openness of the system.

The new effect has been already successfully implemented in the microwave coherent-population-trapping atom clocks in two laboratories (FEMTO-ST and LNE-SYRTE [18, 19]). It has appeared to be efficient for the optical frequency stabilization that improved stability of the clocks. At the same time, in spite of such the interest, physical reasons of the effect have not been understood and studied well enough yet. Therefore, our current research is devoted to comprehensive experimental and theoretical study of SAR under the light field composed of the bichromatic counterpropagating laser beams. The results help one to understand physical reasons for observation of the new effect as well as demonstrate influence of external magnetic field, Raman frequency detuning, laser beam polarizations and intensities on the properties of the nonlinear resonance. In this paper we just briefly discuss physical nature of the effect, while a full version of the study is going to be published soon [8].

II. PHYSICAL REASONS OF THE HIGH-CONTRAST SPIKE

The high-contrast-spike effect can be understood on the basis of the well-known Λ -scheme of atomic energy levels (see Fig. 3). In spite of the spectroscopic signal in our case is not the sub-natural linewidth resonance (as the "dark" resonance for instance), the effect is closely connected with the CPT

phenomenon [20,21]. The effect can be also caused just by the population redistribution over the ground-state sublevels of an atom due to optical pumping. All these factors should be considered carefully.

A. HFS-CPT effect

If the one-photon frequency detuning Δ is much larger than the natural linewidth of an optical transition ($\Delta >> \gamma$), the counterpropagating beams act on the different velocity groups of atoms in a gas due to the Doppler effect (here the onephoton detuning $\Delta = \omega_1 - \omega_{31} = \omega_2 - \omega_{32}$ is assumed to be the same for both frequency components of the light field, and also the condition $\Delta = 0$ means that two counterpropagating waves are both in resonance with the atoms at rest). In this case CPT ("dark" or non-coupled, NC) state can be created in each velocity group separately, embracing magnetic sublevels of the *different* hyperfine levels of the ground state (F_g=3 and F_g=4 for Cs). Therefore, level of absorption under $\Delta >> \gamma$ is low.

This effect can be explained on the basis of the Λ scheme depicted on Fig. 3 with $\Delta_g \approx 9.2$ GHz. The expression for the light field in a vapor cell is:

$$E(z,t) = E_1 \left[e^{-i(\omega_1 t - k_1 z)} + e^{-i(\omega_2 t - k_2 z)} \right] + E_2 \left[e^{-i(\omega_1 t + k_1 z - \phi + \phi_1)} + e^{-i(\omega_2 t + k_2 z + \phi + \phi_2)} \right] + c.c.,$$
(1)

where the amplitudes $E_{1,2}$ are considered to be real values, while the relative phases of two frequency components of the counterpropagating beam are denoted as $\phi_{1,2}$. The angle ϕ corresponds to the mutual angle between linear polarizations of the counterpropagating waves in a real experiment.

So, each of two two-frequency waves from (1) pumps the atoms into the following non-coupled (CPT) states:

$$\left| \operatorname{NC}_{1} \right\rangle = \left\{ \left| 1 \right\rangle - e^{ik_{12}z} \left| 2 \right\rangle \right\} / \sqrt{2} \quad , \tag{2}$$

$$|\mathrm{NC}_{2}\rangle = \left\{ |1\rangle - e^{-i(k_{12}z + \phi_{12} + 2\phi)} |2\rangle \right\} / \sqrt{2} , \qquad (3)$$

with $\phi_{12} = \phi_1 - \phi_2$ and $k_{12} = k_1 - k_2$. If the frequency detuning is not so large $(\Delta \sim \gamma)$, the counterpropagating beams act on the same atoms in a gas and the result depends on the fact if two NC states (2) and (3) are the same or orthogonal to each other. In the latter case one can observe the increased absorption in vicinity of $\Delta \approx \gamma$ (see an example on Fig. 4). Also, as it is seen from (2) and (3), the HFS-CPT effect depends on position of the mirror "M" or position of the cell (see Fig. 2), polarizations of the beams (the angle ϕ in particular) as well as the cell length. Moreover, different points on the *z*-axis (along the cell) can exhibit the constructive (low level of absorption) or destructive (high level of absorption) action of the counterpropagating waves via the NC₁ and NC₂ states. Apparently, the peak's effect does not depend essentially on the cell length when the length satisfies the condition $L > \pi/|k_{12}| \approx 1.6$ cm (in the case of Cs).

It should be noted that the qualitative analysis presented in this subsection has many in common with the one previously reported in [22], but for the counterpropagating light waves with σ^+ and σ^- polarizations.



Fig. 4. High-contrast absorption spike calculated on the basis of the optical Bloch equations. Solid line for the orthogonal polarizations of the counterpropagating beams, while dashed one is for parallel polarizations. Cell length equals to 2 cm, Rabi frequencies are $\sim \gamma$. In this paper we consider the total population of the excited state $|3\rangle$ multiplied by the factor 10^3 (for convinience of presentation) as the theoretical spectroscopic signal.

B. Zeeman CPT effect

This effect has many in common with HFS-CPT and it can be treated with the help of Λ scheme too, taking $\Delta_g = 0$, $k_2=-k_1$, $\omega_1=\omega_2$. In this case the non-coupled states consist of the coherent superpositions of magnetic sublevels of *the same* hyperfine level (namely F_g=4 for the resonance on Fig. 1, right). In contrast to the HFS-CPT, the Zeeman CPT effect is insensitive to position of the mirror or the cell length, but its sign and magnitude strongly depend on the polarizations of the counterpropagating beams.



Fig. 5. Absorption curves for parallel (ϕ =0, dashed black line) and orthogonal (ϕ = $\pi/2$, solid red line) light wave polarizations. The static magnetic field is zero. The system of levels is assumed to be closed (i.e. the branching ratio β =1, see Fig. 3).

The similar theory was considered in [23] for the subnatural-linewidth resonance in the Hanle configuration under the two counterpropagating waves of equal frequency. The analysis was done for the general case of light wave polarizations. For reasons of a concise style, here we do not extend the theory for our case of the natural-linewidth saturated-absorption resonances (for instance, see [8]). Let us just provide a reader with an example of the absorption profile demonstrating the Zeeman CPT effect (Fig. 5). As one can see, the result of absorption at the center of the curve strongly depends on mutual orientations of the linear polarizations of the counterpropagating waves.

It is also obvious that the Zeeman CPT effect depends on an external magnetic field applied to the cell. For example, if the field is strong enough, it destroys the CPT states composed of magnetic sublevels of the same level (F_g =4 in the case of Cs) and the influence of the Zeeman CPT effect on a possibility of the peak's creation is significantly reduced. In this situation only the HFS-CPT or the optical pumping effect, which is mentioned in the next paragraph, can cause the absorption peak creation. And it is apparent that the magnitude of the peak effect is higher when all three reasons take place. Therefore, a magnetic shield is required to defend the spectroscopic cell from the external stray field and to activate the Zeeman CPT reason.

C. Optical pumping effect

It appears that absorption-peak effect can also result from just the optical pumping process, which redistributes populations of the ground sublevels $|1\rangle$ and $|2\rangle$. This effect is pretty simple and can be treated on the basis of rate equations, even without taking into account the low-frequency coherence between the sublevels.

To demonstrate a sense of the optical-pumping reason, let us briefly analyze the scheme on Fig. 3, but in the simplest case of two counterpropagating waves with the same frequencies ($\omega_1=\omega_2$). It means that we consider the A-scheme, which has degenerate ground state under the null magnetic field as in the subsection devoted to the Zeeman CPT effect (see paragraph B). However, now we take the splitting Δ_g is large enough to destroy the coherence between the $|1\rangle$ and $|2\rangle$ states. So, we can consider only the populations of the states.

If the detuning is large $(\Delta \gg \gamma)$ the counterpropagating light waves act on the separate resonance groups of atoms in a gas. Moreover, each group of atoms can be effectively described by the open two-level system ($\beta < 1$), because one of two groundstate sublevels ($|1\rangle$ or $|2\rangle$) is not in resonance with the light wave and it accumulates the atomic population. The level of absorption in this situation is not so high due to the openness.

On the other hand, if the detuning Δ is swept near the center of the resonance curve ($\Delta \sim \gamma$), there are two symmetrical group of atoms with velocities $\upsilon_{1,2}=\pm \Delta_g/k$, which are resonant to both of the light waves (here we imply also $\Delta <<\Delta_g$). In this situation the Λ -scheme is closed (β =1), because no one ground-state sublevel accumulates the population. The level of light field absorption in the closed system is significantly higher than in the open system. It leads to the peak effect at the center of the resonance profile ($\Delta \sim \gamma$).

Note that the similar optical-pumping effect was also mentioned in [24] (the potassium atoms were studied).

III. CONCLUSIONS

To conclude we can state that the high contrast of the nonlinear resonance observed is due to the simultaneous constructive action of several physical phenomena. Qualitative physical explanation has been provided with the help of a simple Λ scheme of atomic energy levels. However, to explain quantitatively the peak's amplitude and width observed in the real experiments one should consider a real structure of atomic energy levels when all peak-caused physical reasons considered here work together. We have also carried out such kind of calculations and the results are in agreement with the experimental data (to be published elsewhere).

Besides the new theoretical knowledge about the saturatedabsorption resonances in a gas of atoms obtained during our investigations, the effect can be interesting and useful for modern quantum metrology, laser physics and high-resolution spectroscopy.

ACKNOWLEDGMENT

Russian part of the team thanks for the support RFBR (grant no. 15-02-08377), RSF (project no. 16-12-10147), and MESRF (project no. 3.1326.2017). French authors thank LabeX FIRST-TF, Région de Franche-Comté, EURAMET (EMRP programm, MClocks project) and DGA. The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union.

REFERENCES

- V. S. Letokhov and V. P. Chebotaev, Nonlinear Laser Spectroscopy. Heidelberg, Germany: Springer-Verlag, 1977.
- [2] N. Castagna et al., "Iodine stabilized IR laser sources", IEEE Xplore, January 2014 [Digest of the Joint UFFC, EFTF and PFM Symposium, Czech Republic, 21-25 July 2013, p. 405].
- [3] S. M. Ignatovich et al., "Yb:YAG/I₂ optical frequency standard at 515 nm with instability at the level 10⁻¹⁵", Journal of Physics: Conf. Series, vol. 793, 012010, February 2017.
- [4] N. A. Koliada et al., "Stabilisation of a fibre frequency synthesiser using acousto-optical and electro-optical modulators", Quantum Electron., vol. 46, pp. 1110–1112, December 2016.
- [5] D. C. Heinecke et al., "Optical frequency stabilization of a 10 GHz Ti:sapphire frequency comb by saturated absorption spectroscopy in ⁸⁷rubidium", Phys. Rev. A, vol. 80, 053806, November 2009.
- [6] M. Lezius et al., "Space-borne frequency comb metrology", Optica, vol. 3, pp. 1381–1387, December 2016.
- [7] M. Abdel Hafiz et al., "Doppler-free spectroscopy on the Cs D₁ line with a dual-frequency laser", Opt. Lett., vol. 41, pp. 2982–2985, July 2016.

- [8] M. Abdel Hafiz et al., "High-contrast sub-Doppler absorption spikes in a hot atomic vapor cell exposed to a dual-frequency laser field", New J. Phys., in press, 2017.
- [9] A.P. Kol'chenko, S.G. Rautian, and R.I. Sokolovskii, "Interaction of an atom with a strong electromagnetic field with the recoil effect taken into consideration", Sov. Phys. JETP, vol. 28, pp. 986–990, May 1969.
- [10] J.L. Hall, C.J. Borde, and K. Uehara, "Direct optical resolution of the recoil effect using saturated absorption spectroscopy", Phys. Rev. Lett., vol. 37, pp. 1339–1342, 15 November 1976.
- [11] J. Ishikawa et al., "Strong-field effects in coherent saturation spectroscopy of atomic beams", Phys. Rev. A, vol. 49, pp. 4794–4825, June 1994.
- [12] P.G. Pappas et al., "Saturation spectroscopy with laser optical pumping in atomic barium", Phys. Rev. A, vol. 21, pp. 1955–1968, June 1980.
- [13] E.G. Saprykin, A.A. Chernenko, and A.M. Shalagin, "Polarization phenomena in the transparency and adsorption effects induced by the field of counterpropagating waves", J. Exp. Theor. Phys., vol. 119, pp. 196–205, August 2014.
- [14] D.B. Lazebnyi et al., "Electromagnetically induced absorption and electromagnetically induced transparency for optical transitions Fg→Fe in the field of elliptically polarized waves", J. Exp. Theor. Phys., vol. 121, pp. 934–949, December 2015.
- [15] H.S. Lee et al., "Zeeman effect in the saturation spectroscopy of the ⁸⁷Rb D₂ line", J. Opt. Soc. Am. B, vol. 11, pp. 558–563, April 1994.
- [16] V.V. Vasil'ev et al., "Dual structure of saturated absorption resonance at an open atomic transition", J. Exp. Theor. Phys., vol. 112, pp. 770–779, May 2011.
- [17] D.V. Brazhnikov et al., "New polarisation effects in saturated absorption spectroscopy in the field of counterpropagating light waves", Quant. Electron., vol. 46, pp. 453–463, May 2016.
- [18] M. Abdel Hafiz et al., "A CPT-based Cs vapor cell atomic clock with a short-term fractional frequency stability of $3 \times 10^{-13} \tau^{-1/2}$ ", Journal of Physics: Conf. Series, vol. 723, 012013, July 2016.
- [19] P. Yun et al., "High-performance coherent population trapping clock with polarization modulation", Phys. Rev. Appl., vol. 7, 014018, January 2017.
- [20] G. Alzetta et al., "An experimental method for the observation of r.f. transitions and laser beat resonances in oriented Na vapour", Nuovo Cimento B, vol. 36, pp. 5–20, 11 November 1976.
- [21] E. Arimondo, "Coherent population trapping in laser spectroscopy", Progress in Optics, vol. 35, pp. 257–354, 1996.
- [22] S.V. Kargapoltsev et al., "High-contrast dark resonance in $\sigma_+ \sigma_-$ optical field", Laser Phys. Lett., vol. 1, pp. 495–499, 16 September 2004.
- [23] D.V. Brazhnikov, A.V. Taichenachev, and V.I. Yudin, "Polarization method for controlling a sign of electromagnetically-induced transparency/absorption resonances", Eur. Phys. J.D, vol. 63, pp. 315– 325, 23 June 2011.
- [24] D. Bloch et al., "Doppler-free spectroscopy of the D₁ line of potassium", Laser Phys., vol. 6, pp. 670–678, August 1996.