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Generalised hyper-Ramsey spectroscopy of two-level atoms in an optically dense medium

K.A. Barantsev, T. Zanon-Willette, A.N. Litvinov

Abstract. **The peculiarities of Ramsey resonance and its sensitivity to the light shift from an optically dense medium of cold atoms are investigated. Different composite pulse protocols for clock spectroscopy, including hyper-Ramsey, modified and generalised hyper-Ramsey schemes, are compared. Error signals change significantly due to the processes of absorption and dispersion in the atomic medium. The dependence of the position of the central fringe resonance with a residual uncompensated light shift of the atomic transition is studied with the attenuation of the radiation intensity in the medium taken into account. It is shown that using a combination of generalised hyper-Ramsey error signals allows one to suppress the sensitivity to the light shift for any length of the medium.**

Keywords: *Ramsey resonance, light shift, cold atoms, optically dense medium, hyper-Ramsey scheme.*

1. Introduction

The use of pulsed laser radiation for interrogation of atoms compared to a continuous one allows one to reduce the width of the atomic resonance line. The method was proposed by N.F. Ramsey in 1949 [1]. The essence of this scheme is that two pulses are used, separated by a free evolution time, leading to narrow coherent interference fringes with much lower sensitivity to radiation inducing frequency shifts in comparison with the continuous radiation Rabi interrogation technique. Ramsey spectroscopy is nowadays widely used in the field of atomic frequency standards [2, 3].

Ramsey spectroscopy received a new impetus in 2010, when a so-called hyper-Ramsey method was proposed [4]. Its essence is to use a sequence of time-divided pulses, which can have different durations, frequencies and phases. It turns out that with such a polling scheme, the dependence of the residual frequency offset of the central hyper-Ramsey fringe with the light shift is similar in shape to an 'anti-symmetric' cubic parabola. This means that there is a region near the resonance, where its position depends very weakly on the light shift. The use of the original hyper-Ramsey interrogation scheme for optical frequency standards based on ultra narrow transitions was demonstrated to be a very successful and versatile technique, as evidenced by the recent experiment on a single $^{171}Yb^{+}$ ion clock with the electric octupole (E3) atomic transition [5] and extended theoretical investigations of other composite pulses interrogation schemes related to hyper-Ramsey spectroscopy [6–9].

The presence of absorption in a medium leads to the fact that the shape of the absorption line changes in comparison with an optically thin medium. Despite the fact that the effects of a dense medium are weakly manifested in the study of cold atoms, there are still a number of options where effects associated with an optically dense medium can occur. Thus, for example, one can expect the manifestation of the effects of an optically dense medium during the construction of atomic clocks based on ensembles of emitters such as three-dimensional optical-lattice clocks, atomic arrays on Sr atoms [10], in Coulomb crystals on $^{115}In^{+}$ and $^{40}Ca^{+}$ ions [11, 12]. The ensembles make use of about one thousand of atoms which can ensure a significant optical density. An increase in the number of atoms (or ions) in clocks is promising due to the suppression of the atomic shot noise.

Barantsev and Litvinov [13] investigated the properties of the Ramsey resonance and its sensitivity to the light shift of the frequency of the atomic transition in the case of the hyper-Ramsey interrogation scheme in an optically dense medium of cold atoms. This work is a continuation of paper [13] and aims to analyse the sensitivity of the light shift of atomic resonance using various protocols based on generalised hyper-Ramsey (GHR) spectroscopy [14, 15]. Thus, the problem to be solved is quite relevant from a fundamental point of view, since it contributes to the understanding of the physics of processes accompanying the interaction of an atom with sequences of pulses of various types. Understanding the new features caused by the presence of radiation absorption in the medium using GHR protocols as an example may help in the analysis and interpretation of error signals in more complex interrogation schemes, such as, for example, auto-balance Ramsey scheme applied with atomic vapour cells based on coherent population trapping effect [16, 17].

2. Analysis of the error signal of an atomic clock in an optically dense atomic ensemble

The physical part of an atomic clock consists of the atoms probed by the pump laser. In a Ramsey interrogation scheme, laser pulses rotate the Bloch vector of the atoms by some angle. After that atomic fluorescence is detected. The fluorescence signal is proportional to the value of population of the excited level after the action of the pulses. Usually in experiments, for example, for neutral Yb atoms, the lifetime γ^{-1} of

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an excited state of an atom is much longer than the duration T_m of the pulse sequence, thus we can put $\gamma^{-1} \gg T_m$ and neglect the spontaneous decay rate γ in the equations for density matrix during a pulse train.

The scheme of interaction of the radiation with a two level atom is shown in the Fig. 1a. The laser has a carrier frequency ν that is quasi-resonant to an atomic transition with a frequency ω_a. In this case, the detuning $\delta = \nu - \omega_a$ obeys the inequality $\delta \ll \omega_a$. Identical atoms are located in an ensemble of length L (Fig. 1b) along the direction of radiation propagation, so that the ensemble is optically dense ($n_a\lambda^2 L > 1$, where n_a is the atomic density, and λ is the wavelength of the atomic transition). At the same time, the ensemble is quite dilute that the wavelength of the incident radiation is less than the average inter-atomic distance ($n_a\lambda^3 < 1$). It allows one to neglect the effects of recurrent light scattering [18–21]. The mathematical description of interaction of the laser field with an optically dense atomic ensemble is given in work [13].

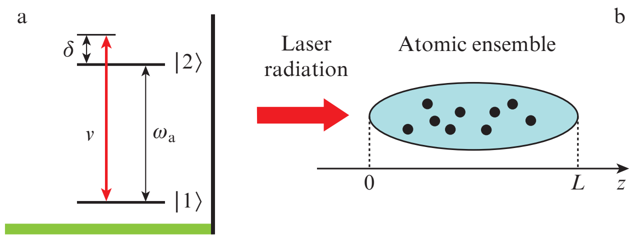

Figure 1. (a) Interaction of the electromagnetic field at a frequency ν with the atomic transition at a frequency ω_a and (935b) an atomic ensemble optically dense along the z axis.

It is possible to achieve a lower sensitivity of the atomic resonance shift depending on the light shift of the reference transition when detecting an error signal [6, 14, 15]. The error signal is generated upon double interrogation of atoms by a sequence of pulses with a phase-step. The fluorescence signal is detected after each interrogation with different phases and these signals are subtracted. Due to the symmetry of the scheme, equal and opposite shifts of the reference resonance appear for each of the two interrogations. During the formation of the error signal, some of them are completely subtracted, which makes the error signal much less sensitive to variations in the laser field.

There are several protocols which generate a phase-step error signal [14]. For formal designation, we divide the sequence of laser pulses into four parts. We will denote the area of the ith pulse in degrees by θ_i and the initial phase of the ith pulse in radians by φ_i (Fig. 2). The fluorescence signal is proportional to the population of the excited state; therefore, one can use the value $P(\varphi_1,\varphi_3,\varphi_4) = \rho_{22}|_{\rm ALP}$ (ALP stands for after the last pulse) for its numerical expression. Here $\rho_{22}|_{\rm ALP}$ is the population of the excited level after interaction of atoms with the sequence of pulses, shown in Fig. 2, the initial phases of which have the values φ_1, φ_3 and φ_4. The initial phase φ_2 in the second region, where the field is missing, is equal to zero. Thus the error signal is

$$\Delta E = P(\varphi_1,\varphi_3,\varphi_4) - P(\varphi_1',\varphi_3',\varphi_4'), \tag{1}$$

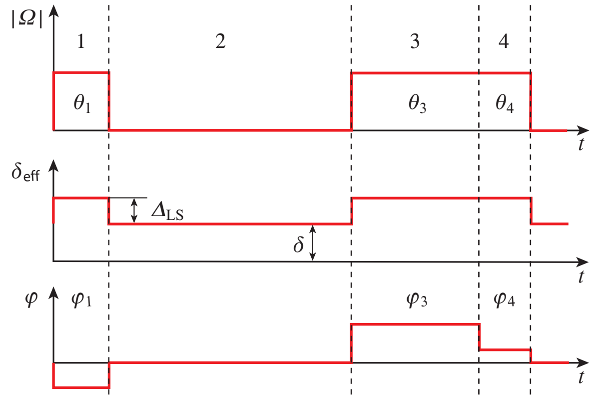

Figure 2. Pulse sequence with arbitrary areas θ_i and initial phases φ_i at the entrance to the medium and the corresponding frequency detuning; $\Delta_{\rm LS}$ is the light shift of the atomic transition frequency.

where φ_i' are the initial phases of the pulses in the secondary interrogation of atoms. The time between interrogations is assumed to be long enough for all atoms to go to the ground state.

Conventionally, the double pulse sequence is denoted as

$$\theta_{1(\varphi_1,\varphi_1')} - \delta T - \theta_{3(\varphi_3,\varphi_3')}\theta_{4(\varphi_4,\varphi_4')}, \tag{2}$$

where δT is the symbolic designation of the free evolution time. Various protocols listed in Table 1 are borrowed from Ref. [14]. The last three protocols make it possible to completely suppress the sensitivity of the position of the resonance to the light shift of the atomic transition in an optically thin medium.

Table 1. Double interrogation protocols of the composite pulses using a phase-step [14].

Protocol	Composite pulses
Hyper-Ramsey (π)	$90_{(\pi/2,-\pi/2)} - \delta T - 180_{(\pi,\pi)}90_{(0,0)}$
Modified hyper-Ramsey	$90_{(\pi/2,0)} - \delta T - 180_{(\pi,\pi)}90_{(0,-\pi/2)}$
Generalised hyper-Ramsey (π/4)	$90_{(0,0)} - \delta T - 180_{(\pi/4,-\pi/4)}90_{(0,0)}$
Generalised hyper-Ramsey (3π/4)	$90_{(0,0)} - \delta T - 180_{(3\pi/4,-3\pi/4)}90_{(0,0)}$

The presence of absorption in a medium leads to a significant distortion of the pulse shape. Moreover, this distortion occurs in different ways for the first pulse train with the phases φ_i and the second pulse train with the phases φ_i', which leads to substantially asymmetrical shifts that do not compensate for each other when forming the error signal. Thus, protocols that completely suppress the light shift in an optically thin medium require modification in an optically dense medium. Below we will consider each of them separately.

In work [13] one can see from the field propagation equation (7) that the field absorption depends on the coefficient $q = 4\pi n_a|d_{12}|^2 k/\hbar$ in the right-hand side of this equation (n_a is the atomic density, d_{12} is the matrix element of the transition, and k is the wave number). Hereinafter, we introduce a dimensionless length $Z = qz/\Omega_0$, where Ω_0 is the amplitude of the pulse at the input of the medium. In terms of this length, the calculations will take on a universal form for an arbitrary set of these parameters.

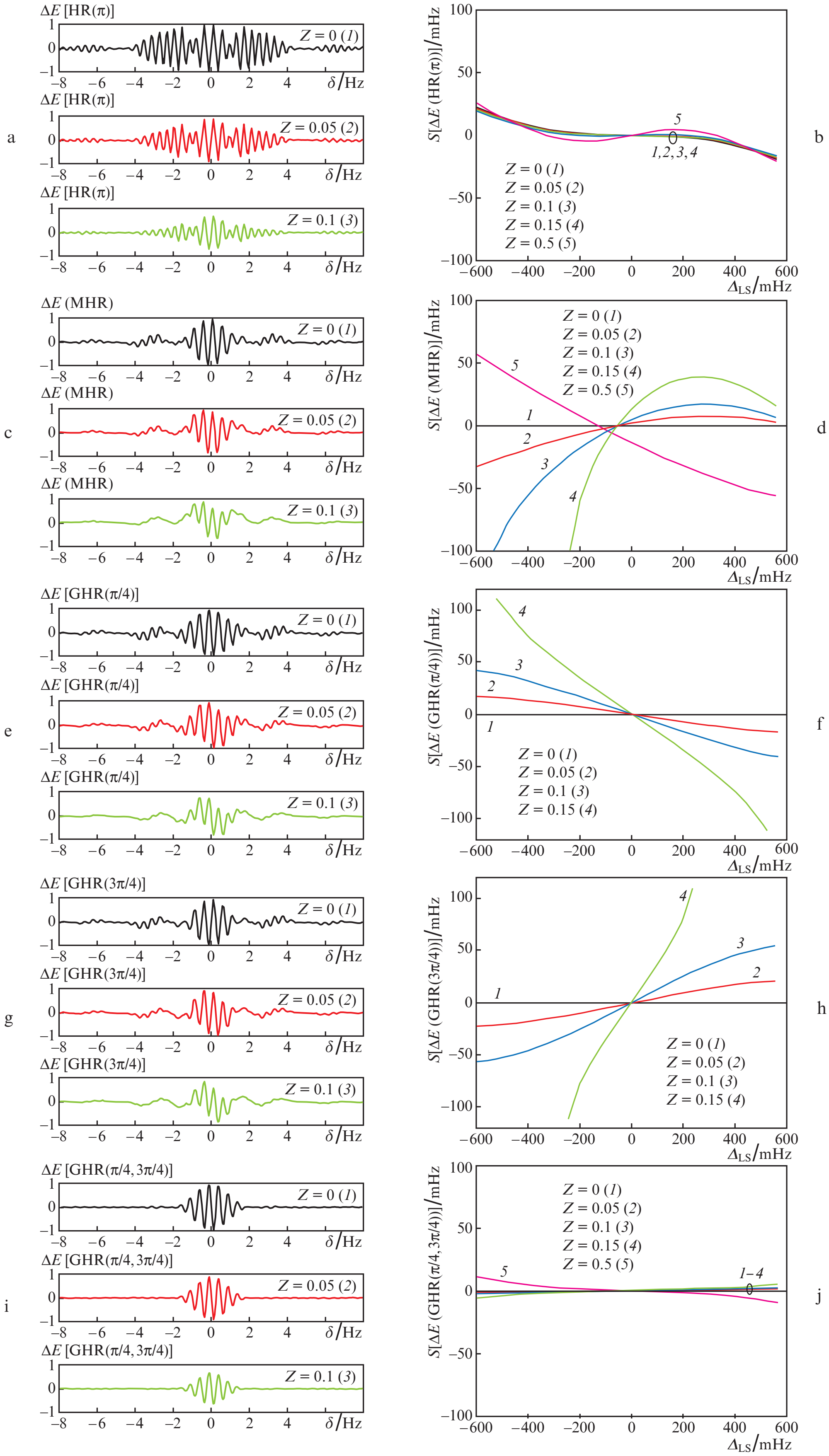

Figure 3. (a, c, e, g, i) Error signal ΔE and its change with increasing longitudinal coordinate Z for different schemes and (b, d, f, h, j) change in the dependence of the position S of zero point of the error signal on the light shift Δ_{LS} of the atomic transition with the longitudinal coordinate Z. Durations of the pulses at the entrance of the medium: $\tau_1 = \tau_3 = \tau$, $\tau_2 = 2\tau$, $T = 9\tau$; pulse amplitudes: $\Omega_0 = (\pi/2)\tau^{-1}$, $\tau = 0.2$ s.

Hyper-Ramsey scheme [HR(π)]. Let us consider the dependence of the error signal on the detuning of the laser field for the hyper-Ramsey sequence of pulses $90_{(\pi/2,-\pi/2)} - \delta T - 180_{(\pi,\pi)}90_{(0,0)}$. The error signal is

$$\Delta E_{\rm HR} = P(\pi/2,\pi,0) - P(-\pi/2,\pi,0). \quad (3)$$

Figure 3a shows several graphs of the error signal in various sections along the atomic cloud. When $Z = 0$, the signal in the zero offset region can be approximated by a straight line with a positive slope (curve *1*). The evolution of the error signal with the changes in the Z coordinate is shown by curves *2* and *3*.

Figure 3b presents the dependence of the position S of the zero point of the error signal on the light shift $\Delta_{\rm LS}$ of the atomic transition. At the entrance to the medium there is a horizontal plateau (curve *1*), but with increasing Z coordinate, the plateau rotates, similar to the fluorescence signal.

Modified hyper-Ramsey scheme (MHR). Hobson et al. [8] proposed some modifications of the hyper-Ramsey interrogation scheme, making it possible to completely suppress the sensitivity of the position of the zero point of the error signal to a light shift in an optically thin medium. Such schemes include a modified hyper-Ramsey scheme, described by a formula $90_{(\pi/2,0)} - \delta T - 180_{(\pi,\pi)}90_{(0,-\pi/2)}$. The error signal for this scheme is

$$\Delta E_{\rm MHR} = P(\pi/2,\pi,0) - P(0,\pi,-\pi/2). \quad (4)$$

Figures 3c and 3d show the change in the error signal and sensitivity of the atomic transition to the light shift as it moves along the Z coordinate for MHR scheme. At the entrance to the medium the sensitivity to light shift is completely absent (Fig. 3d, curve *1*). However, as the Z coordinate increases, the horizontal line rotates, and the effect is stronger than that for the HR scheme.

Generalized hyper-Ramsey scheme (GHR). The error signal for the GHR scheme of double interrogation of atoms with arbitrary phase is

$$\Delta E_{{\rm GHR}(\varphi_3)} = P(0,\varphi_3,0) - P(0,-\varphi_3,0). \quad (5)$$

It is possible to get different versions of this interrogation scheme by setting different values of the phase φ_3. Figures 3e–3h show the error signals and dependences of the position of the zero point on the light shift for the two cases: GHR(π/4) and GHR(3π/4). One can see that the horizontal straight lines in Figs 3f and 3h for $Z = 0$ begin to rotate in opposite directions with increasing the Z coordinate. The magnitude of the rotation angle is comparable to that for the MHR scheme.

The opposite directions of plateau rotation for these two schemes have certain symmetry and make it possible to build an additional error signal in which these effects cancel each other out:

$$\Delta E_{{\rm GHR}(\pi/4,3\pi/4)} = \frac{1}{2}(\Delta E_{{\rm GHR}(\pi/4)} - \Delta E_{{\rm GHR}(3\pi/4)}). \quad (6)$$

For this scheme GHR(π/4, 3π/4) it is necessary to interrogate the atoms by a sequence of pulses four times. As was shown in work [15], there are several variants of such hybrid interrogation schemes which use 4 or even 8 atomic population measurements. They consists of different combinations of direct and mirror-like GHR(π/4) and GHR(3π/4) protocols with different initial conditions for the atomic system (atoms are prepared in the ground or in the excited state). In this work, we will use only direct (not mirror-like) protocols with state initialisation in the ground state. The signal and the dependence of the zero point on the light shift for this hybrid scheme are shown in Figs 3i and 3j. One can see that the protocol is significantly more resistant to shifts in an optically dense medium. Moreover, in contrast to the method described in work [13] allowing one to achieve suppression of sensitivity to light shift in a certain point of the medium, this method allows us to suppress the sensitivity to light shifts along the whole medium.

Table 2 lists the angles of rotation of the horizontal part of the dependence of the zero point of the error signal on the light shift of the atomic transition for the value of the longitudinal coordinate Z= 0.15 for all considered protocols. The smallest rotation has the hybrid GHR(π/4, 3π/4) scheme, and comparable result is for HR(π) scheme. The advantages of GHR(π/4, 3π/4) scheme include the fact that the width of the flat area is wider than that of the HR(π) scheme. However GHR(π/4, 3π/4) is a four-pass scheme, which lengthens the interrogation time of atoms compared to a single-pass HR(π) scheme.

Table 2. Angles of plateau rotation of different protocols for the longitudinal coordinate $Z = 0.15$.

Protocol	Angle of plateau rotation/deg
HR (π)	0.36
MHR	14.35
GHR (π/4)	−9.81
GHR (3π/4)	19.79
GHR (π/4, 3π/4)	0.30

The rest of considered interrogation schemes [MHR, GHR(π/4), GHR(3π/4)] have much larger (two orders of magnitude) angles of rotation of horizontal section of the dependence of zero point of the error signal on the light shift. It makes these schemes unstable in the case of an optically dense medium, despite the fact that in an optically thin medium they completely suppress the dependence on the light shift.

3. Conclusions

In this theoretical work, we have investigated for the first time hyper-Ramsey interrogation schemes for ‘two-level’ atoms under conditions of finite optical thickness and with the presence of collective effects in a dilute medium. It is found that the sensitivity of the resonances to residual uncompensated light shift of the atomic transition may increase due to absorption in the atomic medium.

Different protocols with various composite interrogation pulse trains were analysed. Namely, the error signals for hyper-Ramsey, modified and generalised hyper-Ramsey protocols were investigated. Despite the fact that these methods make it possible to fully compensate for the sensitivity to the light shift, even a small absorption in the medium leads to a strong dependence of the resonance on the light shift. This is expressed as the rotation of the flat area on the graph of the dependence of the shift of the resonance on the light shift of the atomic transition. However, the use of a hybrid combination of the generalised hyper-Ramsey (GHR) scheme (atoms are interrogated 4 times) allows one to compensate for this additional rotation. Thus, a method of suppression of sensitivity of an atomic resonance to the light shift in any spatial position of a dense medium is found with a basic investigation of Maxwell – Bloch equations.

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