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Analysis of the light shift in the hyper-Ramsey scheme of two-level atom interrogation in an optically dense medium

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Abstract. **The properties of the Ramsey resonance and its sensitivity to the light shift of the atomic transition frequency are investigated in the case of the hyper-Ramsey interrogation in an optically dense medium of cold atoms. The shape of the interrogating pulses varies significantly due to the processes of absorption and dispersion in the atomic medium, which leads to a distortion of the Ramsey resonance and a periodic change in the central minimum to the maximum and vice versa. The dependence of the position of the central resonance on the light shift of the atomic transition is found with the attenuation of radiation in the medium taken into account. It is shown that in a certain section of the medium this dependence becomes N-shaped.**

Keywords: *light shift, hyper-Ramsey interrogation scheme, optical frequency standard, two-level atom, optically dense medium.*

1. Introduction

The studies of magnetic resonance were started about 80 years ago [1]. Thanks to the improvement of the measuring technique, Rabi was able to increase the resolution of spectral lines and obtain much new information not only about atomic and molecular structures, but also about atomic properties [2]. Because a two-level atom is similar to a particle with a half-integer spin in a magnetic field, the basic dynamic equations describing the evolution of a two-level atom practically coincide with the equations describing spins. Therefore, the Bloch formalism for the spin vector, developed to describe magnetic resonance, can be transferred to optical resonance problems.

Optical resonance at the transition between two (ground and excited) quantum levels can be used as a reference frequency standard. The object on which the optical frequency standard can be implemented are single ions [3–5], neutral atoms in the optical lattice [6–8], and also the UV transition in the thorium-229 nucleus [9, 10]. The use of an optical transition as a reference made it possible to achieve atomic clock stability of 10^{-18} in one second. Such optical atomic clocks open up new possibilities for measuring the drift of fundamental constants [11], verifying the laws of quantum electrodynamics [11] and cosmological gravimetry [12], and detecting dark matter [13]. It is expected that active research in this area will make it possible in the near future to overcome the stability threshold of 10^{-18} in one second [14].

In 1949, Ramsey proposed using a sequence of pulses separated by a dark pause instead of continuous radiation to interrogate atoms [15]. This scheme makes it possible to reduce the width of the resonance line, determined by the finite time of interaction of the particle with the field. This method quickly found application in quantum frequency standards, primarily in microwave standards [16, 17]. Later it found application in optical frequency standards by using a sequence of three, four (or more) pulses.

When an atom interacts with laser radiation in a standard Ramsey interrogation scheme, due to the presence of non-resonant atomic transitions, a light shift of the resonance frequency (the Stark effect) occurs, which linearly depends on the intensity. In ultra-precise optical clocks, it is necessary to know the exact position of the resonance; however, the intensity of the laser implementing the interrogation fluctuates, which limits the accuracy of determining the position of the resonance line and, therefore, the stability of the optical clock. In 2010, a paper was published in which the hyper-Ramsey interrogation scheme was proposed [18]. The idea was to use a sequence of pulses separated in time, which can have different durations, frequencies and phases. For certain parameters of the pulse sequence, the dependence of the resonance line position on the light shift of the resonance transition frequency is similar in shape to a cubic parabola. Therefore, there is a region near the resonance, where its position does not depend on the light shift. Thus, the use of such an interrogation scheme can improve the stability of the optical clock. An experimental demonstration of the hyper-Ramsey method [19] gave a new impetus to research in this area. Thus, in a number of papers, the influence of various pulse parameters, frequencies, and phases on the position of the resonance was studied [20–22], and the role of the probe laser field fluctuations in the hyper-Ramsey spectroscopic scheme was considered [23].

Ramsey spectroscopy is also widely used in the development of microwave frequency standards in gas cells with alkaline atoms. Thus, in a recent paper [24], an auto-balanced interrogation scheme for such frequency standards was proposed, which was experimentally studied in Ref. [25], where the stability of 2.5×10^{-15} in 10^4 s was achieved in atomic clocks based on the effect of coherent population trapping. While this scheme is fundamentally different from the hyper-Ramsey one, since it is two-loop, both schemes are similar in that they use different pulse sequences.

When constructing the microwave frequency standard in a gas cell with alkaline atoms, in order to enhance the signal,

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one has to use higher concentrations of active atoms, increasing the temperature. With growing temperature, the concentration of atoms can reach such values that collective effects begin to appear, associated with the absorption of laser radiation as it passes through the atomic medium. In this case, the medium becomes optically dense. In such a medium, the shape of the absorption resonance line changes in comparison with the shape of this line in an optically thin medium [26, 27]. Barantsev et al. [28] showed that the presence of an optically dense medium significantly distorts the shape of the resonance line of coherent population trapping in the Ramsey interrogation scheme. The study of the auto-balance scheme in an optically dense medium seems to be a very important and urgent task. However, this scheme uses a more complex interrogation scheme. Based on the results of [28], one can expect that in the case of an optically dense medium, such an interrogation scheme will have a rather complicated picture of the propagation of pulses. In order to understand the physics of the processes with allowance for the laser radiation absorption, we solve in this paper the simpler problem of using the hyper-Ramsey interrogation scheme in an optically dense medium. The aim of our work is to study the physics of processes using a simpler example (hyper-Ramsey interrogation design), which would allow us to rely on known results in limiting cases [29]. Thus, the problem to be solved is rather urgent from a fundamental point of view, since it helps to understand the physics of the processes that accompany 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 the example of the hyper-Ramsey interrogation scheme, simplifies analysis and interpretation of the results of more complex interrogation schemes for atomic systems.

2. Mathematical model and basic approximations

Let us consider the interaction of pulsed laser radiation with an atomic ensemble consisting of identical stationary atoms, in which the frequency ω_{at} of one of the transitions ($|1\rangle \leftrightarrow |2\rangle$) is close to the carrier frequency of the external field ν (the detuning is $\delta = \nu - \omega_{at} \ll \omega_{at}$). The ensemble has a length L along the z axis of the electromagnetic radiation propagation (Fig. 1). In this case, the mean free path of the photon is much shorter than the ensemble length, which makes the atomic medium optically dense ($n_a \sigma L > 1$, where n_a is the concentration of atoms, and σ is the cross section for scattering of photons by an atom). The ensemble is supposed to be sufficiently sparse, so that less than one atom corresponds to the average wavelength λ of the incident radiation ($n_a \lambda^3 < 1$). This assumption allows us to neglect the effects of recurrent scattering of light [30–33] and to consider the interaction of each atom with radiation independently from the point of view of quantum correlations. However, the interaction of radiation with each atom of the ensemble is not completely independent due to its optical density. The radiation incident on the atoms of the far layers of the ensemble depends on the state of the atoms of the neighbouring layers, which is a manifestation of collective light scattering [34, 35].

Assuming that the electric field of the wave is a scalar, we define it by the expression

$$E(z,t) = E_0(z,t)\,e^{i(kz - \nu t)} + \text{c.c.}, \tag{1}$$

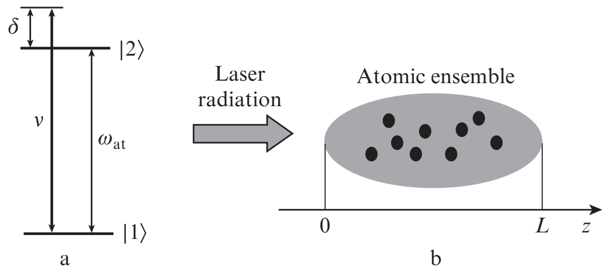

Figure 1. (a) Levels of the atomic transition with frequency ω_{at}, interacting with an electromagnetic field with frequency ν, and (b) scheme of interaction of the field with an atomic ensemble optically dense along the coordinate z.

where $E_0(z,t)$ is the complex amplitude and k is the wave number. For the laser radiation intensities under consideration, a semiclassical approach is applied, in which the radiation is described classically, and the atoms and their interaction with the field are described quantum mechanically. Then the Hamiltonian of the system has the form

$$\hat{H} = \hat{H}_0 + \hbar \hat{V}, \tag{2}$$

where $\hat{H}_0 = \sum_n \varepsilon_n |n\rangle\langle n|$ is the Hamiltonian of the atom in the absence of laser field; ε_n are the energies of atomic levels (n = 1, 2);

$$\hat{V} = -\frac{\Omega^*}{2} e^{i(\nu t - kz)} |1\rangle\langle 2| - \frac{\Omega}{2} e^{-i(\nu t - kz)} |2\rangle\langle 1| \tag{3}$$

is the interaction operator in the rotating wave approximation; $\Omega = (2/\hbar) d_{21} E_0$ is the Rabi frequency of the interaction of the atom with the field; and d_{21} is the transition matrix element of the dipole moment operator.

We will describe the atomic system using the Liouville equation for the single-atom density matrix $\hat{\rho}$:

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar}[\hat{H}, \hat{\rho}] + \hat{\hat{R}}\{\hat{\rho}\}, \tag{4}$$

where $\hat{\hat{R}}\{\hat{\rho}\}$ is the superoperator describing the spontaneous decay of the excited atomic level. The radiation propagation in the plane wave approximation is described by the one-dimensional wave equation for the complex amplitude:

$$\frac{\partial E_0(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E_0(z,t)}{\partial t} = 4\pi i P_0(z,t) k, \tag{5}$$

where c is the speed of light in vacuum and $P_0(z,t)$ is the slowly varying amplitude of the polarisation of the medium. The polarisation of the medium is expressed, in turn, through the atomic density matrix and the operator of the transition dipole moment $\hat{d}$:

$$P = n_a \mathrm{Sp}(\hat{\rho}\hat{d}) = n_a(\rho_{21} d_{12} + \rho_{12} d_{21})$$

$$= P_0 e^{-i(\nu t - kz)} + P_0^* e^{i(\nu t - kz)}. \tag{6}$$

Substituting expressions (2) and (3) into Eqn (4) and P_0 from expression (6) into Eqn (5), we select in the off-diagonal elements of the density matrix the rapidly oscillating factor $\rho_{12} = \tilde{\rho}_{12} e^{i(\nu t - kz)}$. Using the rotating wave approximation, we obtain a system of Maxwell–Bloch differential equations describing the dynamics of the density matrix and the propagation of the radiation field:

$$\dot{\rho}_{11}(z,t) = -\mathrm{i}\frac{\Omega(z,t)}{2}\tilde{\rho}_{12}(z,t) + \mathrm{i}\frac{\Omega^*(z,t)}{2}\tilde{\rho}_{21}(z,t) + \gamma\rho_{22}(z,t),$$

$$\dot{\rho}_{22}(z,t) = \mathrm{i}\frac{\Omega(z,t)}{2}\tilde{\rho}_{12}(z,t) - \mathrm{i}\frac{\Omega^*(z,t)}{2}\tilde{\rho}_{21}(z,t) - \gamma\rho_{22}(z,t),$$

$$\dot{\tilde{\rho}}_{12}(z,t) = [-\mathrm{i}\delta(z,t) - \gamma/2]\tilde{\rho}_{12}(z,t) - \mathrm{i}\frac{\Omega^*(z,t)}{2} \times [\rho_{11}(z,t) - \rho_{22}(z,t)], \tag{7}$$

$$\frac{\partial\Omega(z,t)}{\partial z} + \frac{\partial\Omega(z,t)}{c\partial t} = \frac{4\pi\mathrm{i}n_{\mathrm{a}}|d_{12}|^2k}{\hbar}\tilde{\rho}_{21}(z,t).$$

Here, the field propagation equation is written for the Rabi frequency and γ is the spontaneous decay rate of the excited level.

In the system of Eqns (7), the detuning $\delta(z,t)$ of the laser radiation frequency from the atomic transition frequency depends on the coordinate and time, since laser radiation, generally, interacts with other transitions of the atom, which causes a light shift of the resonance transition frequency $|1\rangle \leftrightarrow |2\rangle$. Suppose that under the action of a rectangular laser pulse with amplitude Ω_0, there occurs a shift in the transition frequency by Δ_{LS}. This shift takes place only under the influence of pulses, and during the dark pause, it is equal to zero. Thus, if the laser radiation frequency has a detuning δ from the frequency of the unperturbed transition in a dark pause, then during the pulse, the detuning will acquire an increment: $\delta + \Delta_{\mathrm{LS}}$. However, laser pulses when passing through an optically dense medium change their amplitude and cease to be rectangular. Since the light shift is proportional to the radiation intensity, the detuning of the laser field at time t at a point in space with coordinate z will have the form

$$\delta(z,t) = \delta + \Delta_{\mathrm{LS}}\frac{|\Omega(z,t)|^2}{|\Omega_0|^2}. \tag{8}$$

3. Discussion of results

In using the classical Ramsey interrogation scheme, a sequence of two identical pulses with an area of $\pi/2$ each affects atoms (Fig. 2a). The corresponding change in detuning according to Eqn (8) at $z = 0$ is shown in Fig. 2b.

In an atomic clock, the sensitivity of the Ramsey resonance to fluctuations in the light shift Δ_{LS} of the atomic transition directly affects their stability. We consider Δ_{LS} as a free parameter, which in a real experiment is determined by the degree of interaction of laser radiation with nonresonance atomic transitions and radiation intensity. By the position of the resonance, we mean the position of its extremum S on the frequency axis. For the classical Ramsey interrogation scheme, the dependence $S(\Delta_{\mathrm{LS}})$ is linear in the vicinity of point $\Delta_{\mathrm{LS}} = 0$. Yudin et al. [18] proposed to use a sophisticated sequence of pulses, which made it possible to reduce the sensitivity of the resonance position to the light shift (hyper-Ramsey method). The pulse sequence shown in Fig. 2c allows the reduction of the dependence $S(\Delta_{\mathrm{LS}})$ near zero to a cubic one, which significantly reduces the sensitivity of the resonance to fluctuations of the light shift. In this sequence, the first pulse has the area $\pi/2$, and the second pulse consists of two parts with the areas $-\pi$ and $\pi/2$. The change of the detuning in time for such a sequence, according to Eqn (8), is shown in Fig. 2d.

The Ramsey resonance for the coordinate $z = 0$ using the pulse sequence of Fig. 2c is shown in Fig. 3. The dependence of the position of its central minimum on the light shift is a cubic parabola (Fig. 4, $z = 0$). Let us analyse how the Ramsey resonance and the dependence $S(\Delta_{\mathrm{LS}})$ change in the course of propagation through an optically dense medium. Usually, in experiments, the lifetime of an excited state of an atom is much longer than the duration of a hyper-Ramsey sequence of pulses; therefore, in the calculations we set the spontaneous decay rate $\gamma = 0$.

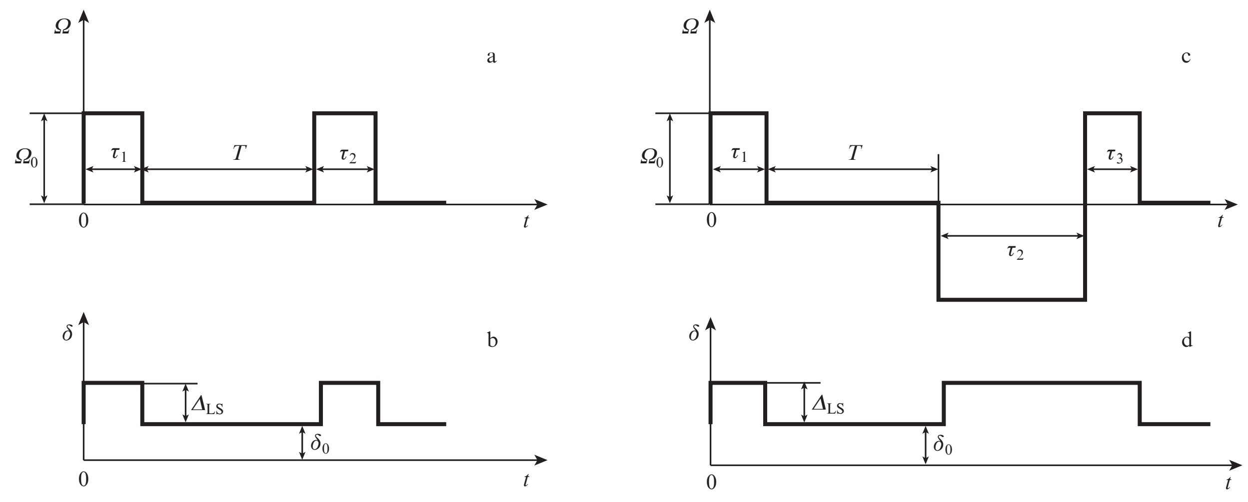

Figure 2. (a) Sequence of laser pulses of the Ramsey interrogation scheme at the input to the medium at the pulse areas $\Omega_0\tau_1 = \Omega_0\tau_2 = \pi/2$ and (c) sequence of laser pulses of the hyper-Ramsey interrogation scheme at the entrance to the medium for the pulse areas $\Omega_0\tau_1 = \Omega_0\tau_3 = \pi/2$, $\Omega_0\tau_2 = -\pi$, as well as (b, d) corresponding changes in the detunings of the frequencies of the laser field during the action of pulses by the value of the light shift Δ_{LS} for the detuning δ_0 from the unperturbed transition.

It can be seen from Fig. 3 that at the entrance to the medium the central resonance has the form of a minimum. Further, in the section with the coordinate z = 17 μm, it inverts and becomes a maximum. Then the process is repeated and in the section with the coordinate z = 98 μm, the minimum takes place again. Thus, in the case of a fixed detuning, when the coordinate z changes, there occur oscillations of the upper level population.

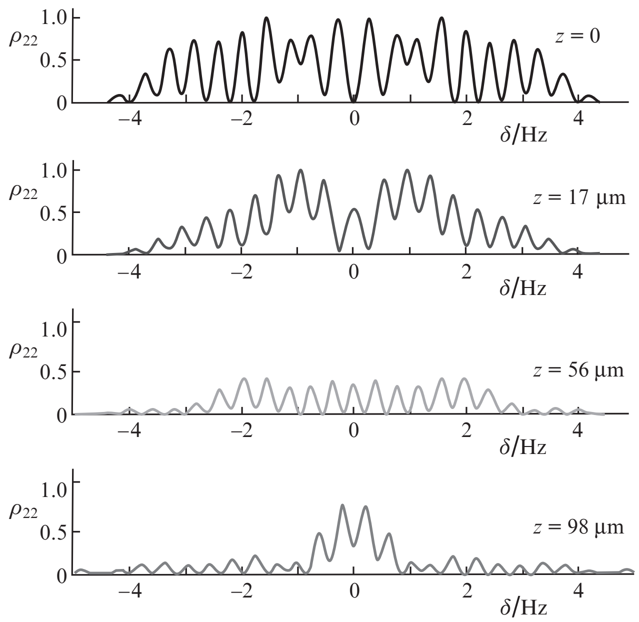

Figure 3. Ramsey resonance for various values of longitudinal coordinate z along the propagation of radiation in the case of the hyper-Ramsey interrogation scheme with parameters $\tau_1 = \tau_3 = \tau$, $\tau_2 = 2\tau$, $T = 9\tau$, $\Omega_0 = (\pi/2)\tau^{-1}$, $\tau = 0.2$ s, $\gamma = 0$, $n_a = 10^{11}$ cm^{-3}.

Figure 4 shows the dependences $S(\Delta_{LS})$ for the resonance at various z. For the maximum in the cross section with z = 17 μm the dependence represents an N-shaped curve. In this case, near zero, there is a significant sensitivity to light shift in the linear portion of the N-shaped curve. The smallest sensitivity to fluctuations in the light shift is achieved at its extremums at $\Delta_{LS} \approx \pm 200$ mHz. It should be taken into account that, as the coordinate z increases, the resonance amplitude, of course, decreases because of radiation absorption in the medium.

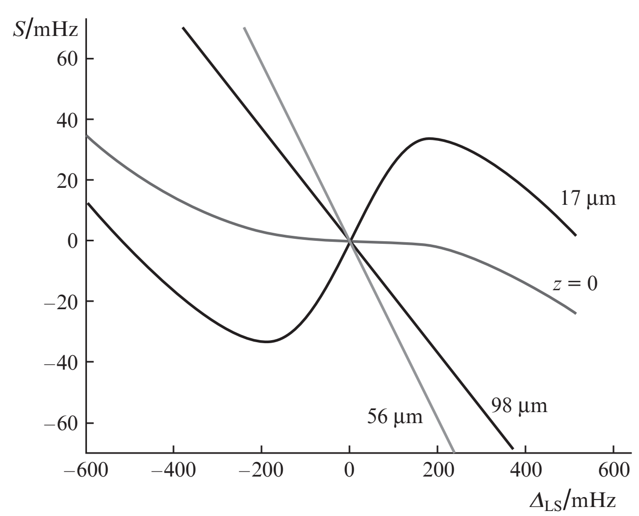

Figure 4. Dependence of the position of resonance on the light shift of the atomic transition frequency for various values of the longitudinal coordinate z along the propagation of radiation. The parameters are the same as in Fig. 3.

With a further increase in the z coordinate, the pulse sequence shown in Fig. 2c is already significantly distorted due to the processes of absorption and re-emission of photons by the atomic medium. The latter process leads, in particular, to the fact that in the dark pause, the field re-emitted by atoms of previous layers acts on the atoms. The dependence $S(\Delta_{LS})$ in this case becomes linear, as in the classical Ramsey interrogation scheme (Fig. 4, z = 56 and 98 μm).

4. Conclusions

The theory of the hyper-Ramsey scheme for interrogating a two-level atom under the conditions of the finite optical thickness of a rarefied medium and the presence of collective effects is developed. A cold atomic ensemble is considered. The mathematical model is a system of equations consisting of the dynamic part for the density matrix, which is solved together with the transport equations for the electromagnetic field and allows for collective effects. Within the framework of the developed model, it is found that the shape of the optical resonance in the case of the hyper-Ramsey interrogation scheme changes substantially in the course of propagation through the medium, namely, the central resonance at zero detuning periodically changes from minimum to maximum and vice versa, while its amplitude decays exponentially. Analysis of the light shift shows that its dependence on the detuning has the form of a cubic parabola only for a thin medium (z = 0). With propagation through the medium, this cubic parabola changes and becomes linear.

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