Zero ac Stark frequency shift of an atom trapped in pulsed laser light*

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Abstract. **We report a theoretical analysis of the spectral properties of atoms localised in an optical pulsed trap. It is shown that this analysis can be carried out using the correlation coefficient calculated from the averaged dynamics of atomic excitation by resonant probe laser light in the presence of a nonresonant pulsed field. It follows from calculations that at a laser pulse duration of 8 ps, atoms can be trapped at a zero shift in the D_2 line transition frequency of the Rb atom, caused by the ac Stark effect. This configuration can be used to build optical frequency standards without the need for localising laser radiation at the 'magic' wavelength.**

Keywords: optical pulsed trap, ac Stark effect, resonant probe laser light, rubidium.

1. Introduction

The importance of such an instrument as a clock in modern life and scientific experiment can hardly be overestimated. The precise measurement of time and frequency allows many fundamental experiments to be performed and also underlies many industrial technologies, such as satellite navigation. In recent years, the scientific application of clocks has led to a number of advances, which include tests of the special theory of relativity [1] and Einstein's principle of equivalence [2], the search for the drift of fundamental constants [3, 4], the search for dark matter [5], capture of the first image of a black hole with the Event Horizon Telescope [6] and many others.

In modern experiments, use is often made of optical frequency standards, which are more accurate and stable than microwave standards [7, 8]. Optical frequency standards are based on the measurement of absorption at a narrow line corresponding, as a rule, to a forbidden transition in an atom. To construct an optical frequency standard, atoms are localised in optical traps [9, 10], which makes it possible to increase the time of interaction of atoms with probe laser light. In addition, localisation of atoms in the region shorter than the laser light wavelength [11] suppresses the Doppler effect.

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Atoms are trapped as a result of the action of a dipole force, which is formed by the interaction of an atom with laser light detuned from the atomic transition frequency. The interaction of an atom with localising laser light leads to a shift in the absorption spectral line caused by the Stark effect in an ac field [12, 13]. This shift depends on the differential polarisability of the atomic levels of the clock transition and on the intensity of the localising laser light [14]. Because of this, apart from the shift of the line corresponding to the clock transition, there arises its additional inhomogeneous broadening caused by the dependence of the shift of the line on the position of the atom in the optical potential [15].

There are several approaches to suppressing the line shift caused by this ac Stark effect. The main one is the use of the 'magic' wavelength of the trapping laser light [16, 17]. In this approach, the wavelength of the trapping laser light is chosen such that the optical shifts of the ground and excited levels are the same. This corresponds to the zero value of the differential polarisability of the two levels and, as a consequence, leads to compensation for the spectral line shift. A necessary condition for this approach is the presence of a 'magic' wavelength of the trapping light [18–20].

Other approaches to compensating for the optical shift involve the interaction of an atom with additional laser light, which compensates for the shift [21], or switching off the trapping potential during the measurement [22, 23]. Switching off the trapping potential was originally proposed in [24, 25] to suppress heating of atoms during their localisation in an optical dipole trap. Now this approach is actively used for the trapping of single atoms [23, 26]. Switching off the trapping potential makes it possible to completely suppress the shift of the spectral line caused by the trapping field [22, 27], since the atom interacts only with probe laser light. This regime is achieved by amplitude modulation of the trapping laser light using, as a rule, acousto-optic modulators and corresponds to the quasi-pulse regime of interaction of atoms with laser fields. Trapping of atoms by pulsed laser light of picosecond [28, 29] and femtosecond [30–33] durations is an extremely short case of the trapping of an atom in a pulsed field.

There exits two atom-trapping regimes, i.e. pulsed and quasi-pulsed one. The main difference between the trapping of atoms by pulsed fields and their trapping in the quasi-pulsed regime is the duration of the stay of the atoms in the trapping potential and free state. In the quasi-pulsed regime, the times of interaction of atoms with the trapping and probe fields exceed the characteristic times inherent in the system in question: the period of oscillations of an atom in an optical potential and the lifetime of an atom in an excited state. This makes it possible to consider the processes of trapping and probing independently of each other under the assumption of

the presence of a stationary state of the system at each of the stages of trapping and probing.

The parameters of picosecond and femtosecond pulsed light do not allow one to analyse the trapping of atoms and their spectral properties using the same approaches as in the quasi-pulsed case. Indeed, the time of interaction of an atom with a single pulse of the trapping field (50 fs–10 ps) is much shorter than the period of atomic oscillations in the averaged optical potential (2–100 μs). The periodicity of the pulsed light used to trap atoms corresponds to a frequency of about 80 MHz. Because of this, the time interval when the atom does not interact with the trapping field is 12.5 ns, which is shorter than the lifetime of the excited state (for example, for $5P_{3/2}$ rubidium atoms it is 27 ns). For these reasons, the probing and trapping of atoms cannot be considered independently.

In [34], we used the spectrally selective heating of atoms [35] to measure the absorption spectrum of rubidium-85 atoms at the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition, localised in a pulsed optical trap formed by femtosecond light. Trapping was carried out by focused radiation of a Ti: sapphire laser with a pulse duration of 420 fs. This duration was chosen to suppress the heating of atoms due to the momentum diffusion at a high peak intensity of the pulsed light [33]. The choice of the pulse duration and the use of additional spectral filtering of the light made it possible to trap atoms by a pulsed field, the lifetime of which is comparable to the lifetime of atoms in a trap formed by cw laser light [36]. The results of the study showed that at low average intensities of pulsed light, the spectral properties of atoms trapped by pulsed light do not differ from the properties of atoms trapped by cw laser light. In both cases, there is a shift of the atomic absorption line caused by the ac Stark effect.

Theoretical calculations from Ref. [34] show that the interaction of atoms with pulsed trapping light is possible in the absence of an optical shift. This regime corresponds to the interaction of atoms with a laser 2π pulse and Choi et al. [37] were the first to propose to use it in a pulsed optical trap. Experimental work with pulsed trapping of atoms in a magneto-optical trap with amplitude modulation of the magnetic field [38] confirmed the validity of the assumptions made in [37] about the spectral properties of atoms in the interaction with a 2π pulse. However, the trapping of atoms in pulsed optical traps has not been analysed. The objective of this work is to carry out such an analysis in order to determine the optimal characteristics of pulsed laser light for optical trapping of atoms, at which there will be no optical shift of the spectral line caused by the ac Stark effect.

2. Theoretical model

Let us consider a two-level atom (Fig. 1) with a dipole moment d of the transition from the ground state $|g\rangle$ to the excited state $|e\rangle$, interacting with two laser fields, i.e. trapping and probe. The trapping field depends periodically on time with an electric field envelope $E_d(t)$, a Rabi frequency $\Omega_d(t) = dE_d(t)/\hbar$, and a frequency detuning δ_d from the precise frequency of the atomic transition. In subsequent calculations, the pulse repetition rate was taken equal to 80 MHz; the detuning δ_d to the red region with respect to the D_2 line of the rubidium atom (780 nm) excitation was 45 nm, which corresponds to the conditions of the experiment on the localisation of rubidium atoms in a pulsed optical trap [33, 36]. The envelope of a single field pulse was described by a Gaussian func-

tion: $E_d^s(t) = E_0\exp[-1.385(t/\tau)^2]$, where E_0 is the peak amplitude of the pulsed field, and τ is the pulse duration (in the calculations, it was taken equal to 420 fs). In the problem under consideration, the field pulses, with which the atom interacts, are phased. This means that the Fourier transform of the field $E_d(t)$ is the spectrum of the optical comb. It is important to note that the effect of a change in frequency f_{CEO} (corresponding to a phase shift of the carrier envelope of the pulsed field) on the interaction of the atom with the pulsed field is small, since this frequency and its possible deviations are much less than the frequency detuning of the laser field from the atomic resonance frequency. The probe field E_p is continuous with the Rabi frequency $\Omega_p = dE_p(t)/\hbar$ and the detuning δ_p, which is small compared to the detuning for the trapping field. The absorption line width Γ from the ground state to the excited one was chosen to be equal to the width of the D_2 line of rubidium ($2\pi \times 6$ MHz). The intensity of the probe field in the calculations was chosen such that the saturation parameter was equal to 0.43 ($I_p \approx 1.1$ mW cm^{-2}) and corresponded to the experimental conditions [34]. The interaction of such an atom with the considered configuration of laser fields is described by the equations for the density matrix:

$$\rho_{gg}(I_d, t) + \rho_{ee}(I_d, t) = 1,$$

$$\frac{d\rho_{ee}(I_d, t)}{dt} = i\frac{\Omega_p}{2}[\rho_{ge}(I_d, t)\exp(-i\delta_p t) - \rho_{eg}(I_d, t)\exp(i\delta_p t)]$$

$$+ i\frac{\Omega_d(t)}{2}[\rho_{ge}(I_d, t)\exp(-i\delta_d t) - \rho_{eg}(I_d, t)\exp(i\delta_d t)]$$

$$- \rho_{ee}(I_d, t)\Gamma, \qquad (1)$$

$$\frac{d\rho_{ge}(I_d, t)}{dt} = i\frac{1}{2}[\rho_{ee}(I_d, t) - \rho_{gg}(I_d, t)]$$

$$\times[\Omega_p\exp(i\delta_p t) + \Omega_d(t)\exp(i\delta_d t)] - \rho_{ge}(I_d, t)\frac{\Gamma}{2},$$

$$\rho_{eg} = \rho_{ge}^*,$$

where $\rho_{gg}(I_d, t)$ and $\rho_{ee}(I_d, t)$ are the diagonal elements of the density matrix, which determine the populations of the
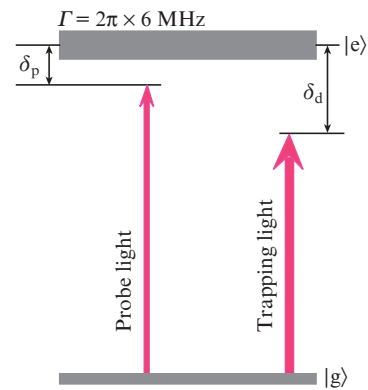

Figure 1. Energy diagram of a two-level atom, consisting of the ground and excited states, interacting with trapping nonresonant pulsed and quasi-resonant cw probe laser fields.

ground and excited states of a two-level atom, respectively; and $\rho_{ge}(I_d, t)$ are off-diagonal elements of the density matrix. The elements of the density matrix depend not only on time, but also on the average intensity I_d of pulsed laser light, which is used in the experiment to trap atoms. The average intensity of the laser light is determined at the laser beam centre, where the trapping optical potential is formed, and sets its depth. This intensity is related to the envelope of the Rabi frequency of a single field pulse by the expression (for a Gaussian pulse)

$$\Omega_d^s(t) \approx \frac{d\exp[-1.385(t/\tau)^2]}{\hbar}\sqrt{\frac{I_d}{\tau\nu}},$$

where τ is the pulse duration measured at the half maximum of the intensity distribution in time, and ν is the pulse repetition rate.

It is important to note that there is no analytical solution for this system of equations, and in the general case it does not have a stationary solution due to the time dependence of the Hamiltonian of the system [39]. As a result, the solution to the system of equations (1) will be found by numerical integration.

3. Analysis of the data obtained by numerical calculation

By solving the system of equations (1), Afanasiev et al. [34] determined the spectrum of the atom in the above configuration shown in Fig. 2a, i.e. the dependence of the averaged population of the excited state on the frequency of the probe laser light and the average intensity of the nonresonant pulsed trapping field. For convenience, the calculated depth (in temperature units) of the optical potential formed by the trapping field is also presented. Since we are interested in the case when there is no shift of the spectral line of the atom in the trap, it is sufficient to perform an analysis only at $\delta_p = 0$ (corresponds to the gray line in Fig. 2a). Three limiting cases can be distinguished: (i) there is no pulsed field ($I_d = 0$) (point d in Fig. 2a);

(ii) the parameters of the average intensity of the nonresonant pulsed field are such that there occurs a maximum frequency shift of the absorption spectral line and its splitting (point c in Fig. 2a); and (iii) the splitting and shift of the absorption line are absent in the presence of a pulsed probe field (point b in Fig. 2a). Let us consider the dynamics of the excitation of atoms by a probe resonant field at these points. The moment when the probe field is switched on is the zero moment of time, which coincides with the maximum intensity of the first pulse of the trapping field.

Figure 2d shows the dynamics of atomic excitation by a resonant probe field in the absence of a pulsed trapping field. The presented curve corresponds to the dynamics at point d in Fig. 2a. The dynamics of excitation in this case completely coincides with the solution of the equations for the density matrix of a two-level atom interacting with the resonant light, and can be described analytically:

$$\rho_{ee}(0, t) = \frac{8\Omega_p^2}{16\Omega_p^2 + 8\Gamma^2}\left\{1 - \exp\left(-\frac{3}{4}\Gamma t\right)\right. \tag{2}$$

$$\times\left[\cos\frac{1}{4}t\sqrt{16\Omega_p^2 - \Gamma^2} + \frac{3\Gamma}{\sqrt{16\Omega_p^2 - \Gamma^2}}\sin\frac{1}{4}t\sqrt{16\Omega_p^2 - \Gamma^2}\right]\right\}.$$

In the presence of a pulsed nonresonant periodic field and with an increase in its intensity, the dynamics of the population of the excited state $\rho_{ee}(I_d, t)$ exhibit time-periodic perturbations associated with the interaction of an atom with a pulsed field; their frequency coincides with the pulse repetition rate. These perturbations distort the dynamics of the excitation of the atom by the probe field due to the perturbation of the amplitude and phase of the internal degrees of freedom of the atom. The greatest perturbation is achieved at an average intensity of the pulsed field, $I_d = 25 \times 10^4$ W cm^{-2}. The dynamics of excitation in this case is shown in Fig. 2c. It can be seen that it dramatically differs from the dynamics in the case of the interaction of a free atom with a probe field
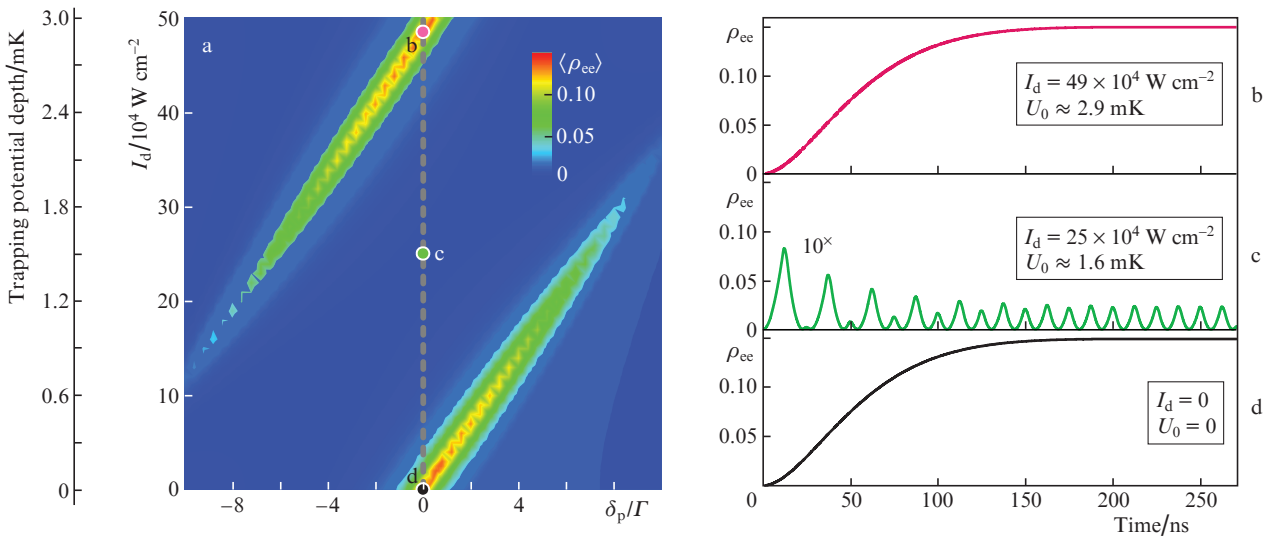

Figure 2. (Colour online) (a) Two-dimensional diagram showing the dependence of the average population of the excited state of a two-level atom on the frequency of the probe laser light and the average intensity of the nonresonant trapping pulsed field, as well as (b–d) the dynamics of excitation of the atom in the presence of a pulsed trapping field at $\delta_p = 0$.

(Fig. 2d). Firstly, the stationary value of the population has not been achieved and, secondly, its value averaged over time is small (the vertical scale in Fig. 2d is increased 10 times). It is seen that the interaction with the pulsed field almost completely suppresses the interaction with the probe resonant laser light, and only a periodic stationary value of the population is realised, similar to that considered in [39]. This intensity corresponds to point c in Fig. 2a. At this value of the pulsed field intensity, there occurs a significant optical shift and splitting of the atomic absorption line. As a result, a small population of the excited state of the atom by the probe field is realised.

A further increase in the intensity of the pulsed field transforms the excitation dynamics into the dynamics similar to that of excitation in the absence of a pulsed field. This is achieved at $I_d = 49 \times 10^4$ W cm^{-2} (Fig. 2b). It can be seen that the curve in this case almost completely repeats the curve shown in Fig. 2d. It leads us to conclude that the pulsed field exerts no influence on the internal degrees of freedom of the atom. This is possible only if the interaction of the atom with each individual pulse of the laser field changes the phases of the internal degrees of freedom by a value multiple of 2π. Then, the internal degrees of freedom of the atom after interaction with the laser field pulse will be identical to those before it arrives (under the assumption that the pulse duration is short in comparison with the relaxation times of the internal degrees of freedom of the atom). For this reason, the interaction of an atom with a probe laser field will be perturbed by a strong pulsed field, which is used to trap the atom.

4. Correlation function

The problem of interaction of an atom with a 2π laser field pulse has an analytical solution only at a zero frequency detuning of the pulsed laser field. In this case, the intensity of the 2π pulse will be determined from the condition

$$\int_{-\infty}^{\infty} \Omega(t)\,\mathrm{d}t = 2\pi,$$

where $\Omega(t)$ is the Rabi frequency envelope of a single pulse. In the general case, with an arbitrary time dependence of the pulse envelope function and an arbitrary detuning of the laser field from the atomic transition frequency, the problem is analytically difficult to solve. For rectangular pulses of duration τ, when the laser radiation frequency is detuned from the exact frequency of the atomic transition, the condition for a 2π pulse has the form

$$\int_{0}^{\tau} \tilde{\Omega}\,\mathrm{d}t = 2\pi,$$

where $\tilde{\Omega} = \sqrt{\delta^2 + \Omega^2}$ is the generalised Rabi frequency. This expression can be used only if the leading and trailing edges of the laser pulse are small in time. This condition is not met in the case of pulsed femtosecond or picosecond radiation, the shape of the envelope of which can be approximated by a Gaussian function.

A number of works [40–42] are devoted to the problem of interaction of a two-level atom with pulsed light in the case of different pulse envelopes and frequency detunings, but there is no exact analytical solution of the problem in general form. Perturbation theory can be used to assess the effect of a single

pulse on the dynamics of the internal degrees of freedom of an atom. But in a real experiment, the absorption spectrum of an atom is measured in the regime of interaction of an atom not with one pulse, but with their sequence. The need to take into account the initial amplitudes and phases for each individual pulse further complicates the analytical calculation, and the error caused by the approximations will accumulate.

At an arbitrary frequency detuning of the pulsed light, an analytical solution for a 2π pulse exists only for pulses whose envelope can be described by a hyperbolic secant [43, 44]. This makes it easy to determine the radiation parameters for spectroscopic applications. For example, when such radiation propagates in a medium, there is no absorption and its bleaching occurs [45, 46]. At present, this approach is used, among other things, to control the phase of a two-level system in the microwave region of the spectrum in order to control a superconducting qubit [47]. However, pulses of this shape are not always realisable experimentally, and in addition, the interaction of an atom with pulsed light strongly depends on the shape of its envelope [48]. For this reason, it is necessary to develop a simple and convenient method for numerically calculating the effect of a pulsed nonresonant field with an arbitrary envelope on the absorption of a probe resonant field by an atom.

Let us consider the differences in the dynamics of the population of atoms in the excited state $\rho_{ee}(I_d, t)$ in the interaction with a resonant probe field for different average intensities of a pulsed nonresonant field (Figs 2b–2d). To this end, the correlation coefficient C should be introduced, which will allow comparing the averaged populations of the excited state in the presence of a pulsed field with an average intensity I_d and in its absence ($I_d = 0$):

$$C(I_d) = \frac{\langle \rho_{ee}(I_d, t)\rho_{ee}(0, t)\rangle}{\langle \rho_{ee}(0, t)\rho_{ee}(0, t)\rangle}. \tag{3}$$

Expression (3) provides for averaging over a time interval sufficient to establish a stationary value of the population in an excited state in the absence of a pulsed laser field. As can be seen from Fig. 2d, this time for the selected parameters of the interaction of the atom with the probe field is ∼250 ns. With the same interaction parameters, the establishment of a periodic stationary value is observed in the presence of a pulsed laser field (Fig. 2c). The correlation function is normalised to the average value of the squared population of the excited state in the absence of a pulsed field. Note that the results presented in Fig. 2a demonstrate that $\langle \rho_{ee}(I_d, t)\rangle \leqslant \langle \rho_{ee}(0, t)\rangle$ for any values of the average intensity I_d of the trapping pulsed field. Because of this, for the coinciding population dynamics in the presence of a pulsed field and in its absence, the correlation coefficient is $C = 1$. The population dynamics of the upper level $\rho_{ee}(0, t)$ is determined by the analytical expression (2).

Figure 3 shows the result of a numerical calculation of the correlation coefficient of function (3) depending on the average intensity of pulsed laser radiation. For convenience, the average intensity value is recalculated into the depth of the optical potential, expressed in temperature units. It can be seen that for a pulse duration of 420 fs, the correlation coefficient is $C = 1$ at an average pulsed-light intensity $I_d = 49 \times 10^4$ W cm^{-2}, which corresponds to a zero optical shift of the spectral line in Fig. 2a. This means that at such an intensity, the dynamics of the excitation of atoms in the presence of pulsed radiation (Fig. 2b) coincides with the dynamics of exci-
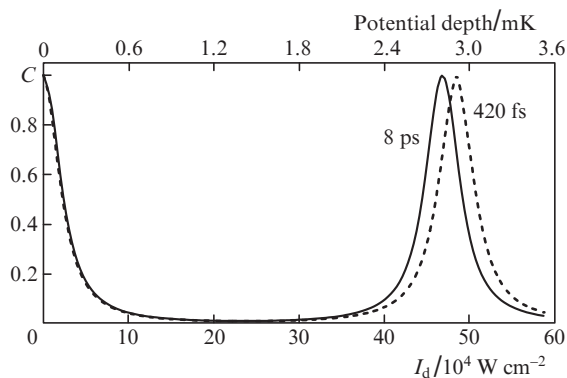
Figure 3. Dependences of the correlation coefficient C on the average intensity I_d of a pulsed laser field at pulse durations of 420 fs and 8 ps.

tation of atoms in its absence (Fig. 2d), i.e., the trapping field does not affect the spectral properties of a trapped atom.

The achievement of such a regime of nonperturbing interaction is impossible in the experiment. This is due to the short lifetime of atoms localised in the field of an optical dipole trap formed by pulsed light of femtosecond duration. Afanasiev et al. [34] and Mashko et al. [36] used radiation with a duration of 420 fs, and the lifetime of atoms in an optical trap was ~ 1.2 s at $I_d = 2.5 \times 10^4$ W cm^{-2}. The main problem of trapping by pulsed fields is the momentum diffusion of atoms caused by fluctuations of the dipole force at high peak intensities of pulsed light [33]. The peak intensity I_d^{peak}, which corresponds to the atom trapping demonstrated in experimental studies, is 7.5×10^8 W cm^{-2}. An increase in the average intensity of laser light to 49×10^4 W cm^{-2}, at which there should be no optical shift of the spectral line, will lead to an increase in the peak intensity and, consequently, to an increase in the momentum diffusion of trapped atoms. This will prevent their spectral study due to the short lifetime in the optical trap. The way out of this situation is to increase the pulse duration to maintain the peak radiation intensity at the same level. Calculations show that at $I_d = 49 \times 10^4$ W cm^{-2} and $I_d^{peak} = 7.5 \times 10^8$ W cm^{-2}, the pulse duration should be ~ 8 ps. Figure 3 shows the calculated correlation coefficient for a trapping laser pulse duration of 8 ps. The peak value of the correlation function reaches ~ 0.998 at $I_d = 47 \times 10^4$ W cm^{-2}. This means that the interaction of a resonant probe field with an atom in the presence of a nonresonant pulsed field with the indicated duration and average intensity will be the same as in the case of interaction with a free atom. The deviation of this value from unity may be due to numerical errors. Note that under the experimental conditions this value may differ from unity due to fluctuations in the intensity of the pulsed trapping field. Our estimates show that to ensure the measurement of the line of the rubidium atom $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ with an accuracy of no worse than 10%, it is necessary to stabilise the average intensity of the trapping pulsed light with an accuracy of $\sim 1\%$. Fluctuations in the pulse repetition rate affect the line shift in the second order of smallness.

5. Conclusions

Thus, the performed analysis shows that the experiment makes it possible to achieve the parameters of localisation of atoms in a pulsed optical trap, at which there will be no shift of the absorption spectral line caused by the ac Stark effect. The developed method of analysis, based on calculating the correlation coefficient of the dynamics of atomic excitation by a probe field in the presence of a localising pulsed field detuned from resonance, allows one to calculate the parameters required for the realisation of the conditions for the absence of an optical shift. It should be noted that the use of the described approach permits analysing not only pulsed light with an arbitrary pulse envelope, but also more complex temporal profiles of light fields.

Note also that the widespread use of 2π pulses in spectroscopy is limited by the specific features of the excitation of atoms by such radiation [49–51]. In this work, pulsed light is necessary for the trapping of atoms, while spectroscopic studies are carried out with an additional probe field. Pulsed interaction of the atom and the trapping field with the required parameters [average intensity $I_d = 47 \times 10^4$ W cm^{-2}, pulse duration $\tau = 8$ ps, detuning $\delta_d = -45$ nm relative to the frequency of the atomic transition $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ in the rubidium-85 atom], determined by the 2π pulse condition, will make it possible to develop atomic systems for constructing frequency standards in the optical range of the spectrum without the need to use 'magic' wavelengths for optical trapping of atoms.

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