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Precursor pulse and frequency modulation of quasi-resonance self-induced transparency pulses in the presence of irreversible relaxation

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***Abstract*.** **The quasi-resonance interaction of a laser pulse with an inhomogeneously broadened quantum transition is numerically simulated under conditions corresponding to the experiment on the study of self-induced transparency in rubidium vapours. It is shown that the experimental shift of the pulse spectrum is caused by the initial stage of the formation of a precursor pulse and the influence of irreversible relaxation. It is found that the precursor pulse is more stable to the destructive action of relaxation than the 2π pulse and appears even in the case when the input pulse area is insufficient for the formation of the 2π pulse.**

Keywords: *self-induced transparency, quasi-resonance, frequency modulation, relaxation processes.*

1. Introduction

The discovery of the effect of self-induced transparency (SIT) [1] stimulated theoretical and experimental investigations of the resonance coherent interaction of light with an ensemble of two-level quantum objects [2 – 4]. The study of SIT in a quasi-resonance case, when the central frequency of the input pulse differs from that of an inhomogeneously broadened quantum transition, has required the consideration of the possible frequency modulation of a light signal. The theory developed in papers [5 – 10] has shown that, if the input pulse area exceeds π, then, as in the case of an exact resonance, one or several solitons, called 2π pulses, are formed in the large-distance limit. Optical solitons have no frequency modulation. However, during their formation, the central radiation frequency shifts from the central frequency of a quantum transition. In addition, the numerical experiment [8] showed that during the formation of a single 2π pulse from the input pulse with an area lying in the interval π – 2π, an additional pulse appears, which was called by the authors of Ref. [8] a precursor pulse. The precursor pulse propagates faster than the 2π pulse, it has a strong frequency modulation, and disappears in the infinite-distance limit. The precursor pulse was observed experimentally [11], however, its frequency modulation was not measured.

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In experiment [12], which is recognised as the most adequate confirmation of the SIT theory, the shift of the central frequency of the pulse spectrum from the central frequency of the quantum transition was measured. The aim of our paper is to show that this frequency shift is caused, on the one hand, by the formation of a frequency-modulated precursor pulse and, on the other, by the SIT destruction due to irreversible relaxation.

2. Basic equations

In experiment [12], radiation from a mercury laser excited ^{87}Rb vapours in a constant magnetic field whose strength vector was directed along the propagation of the light pulse. The Zeeman effect produced a quantum transition with nondegenerate energy levels, which was resonant with laser radiation. The lower level of this transition was the ground level and corresponded to the $5s^2 S_{1/2}$ $(M_J = -1/2, M_I = 3/2)$ state, while the upper level corresponded to $5p^2 P_{1/2}$ $(M_J = 1/2, M_I = 3/2)$ state, where M_J and M_I are the quantum numbers of the projections of the total electronic and nuclear angular momenta on the direction of the constant magnetic field. Let us denote these levels by numbers 1 and 2, respectively. Between these levels, the $5s^2 S_{1/2}$ $(M_J = 1/2, M_I = 3/2)$ level was located, which we denoted by number 3. Under experimental conditions [12], irreversible relaxation was manifested only as spontaneous radiative transitions from level 2 to lower-lying levels 3 and 1.

We will choose the right orthonormalised basis ***ijk*** of the laboratory coordinate system xyz so that the vector $\boldsymbol{k}$ is directed along the direction of laser pulse propagation. Let us introduce the notation

$$p = \frac{1}{\sqrt{2}}(p_{x12} - \mathrm{i}p_{y12}),$$

where p_{x12} and p_{y12} are the x and y components of the vector of the 1 – 2 transition electric dipole moment; ω_{21}^0 is the central frequency of the inhomogeneously broadened line of this transition; T is the characteristic time determining the density of distribution of frequencies ω_{21} of the 1 – 2 transitions in atoms according to the expression

$$g(\omega_{21}) = \frac{T}{\sqrt{\pi}} \exp\left[-T^2(\omega_{21} - \omega_{21}^0)^2\right];$$

T_{21} and T_{23} are the lifetimes of the level 2 for spontaneous transitions to levels 1 and 3, respectively; and T_{21}' is the transverse relaxation time for the 1 – 2 transition.

Let us represent the strength of a circularly polarised electric field used in experiment [11] in the form

$$\boldsymbol{E} = \mu(\boldsymbol{i} + \mathrm{i}\boldsymbol{j})a(z,t)\exp\left[\mathrm{i}\omega\left(\frac{z}{c} - t\right)\right] + \text{c.c.}, \quad (1)$$

where $\mu = \hbar/(2^{3/2}T|p|)$; $a(z,t)$ and ω are the complex envelope and carrier frequency of the laser pulse, respectively. Because the choice of the frequency ω is somewhat arbitrary, we assume for definiteness that this frequency is equal to the carrier frequency of the incident laser radiation.

Let us introduce dimensionless independent variables

$$s = \alpha T z, \quad w = \frac{t}{T} - \frac{s}{\beta}, \quad (2)$$

where

$$\alpha = \frac{2\pi\omega|p|^2 N}{c\hbar}; \quad \beta = \alpha T^2 c;$$

N is the total concentration of atoms on levels 1 and 2 before the laser pulse arrival. Let us also introduce the quantities σ_{ik} $(i, k = 1, 2)$

$$\sigma_{21} = \sigma_{12}^* = \frac{2p}{|p|}\rho_{21}\exp\left[\mathrm{i}\omega\left(t - \frac{z}{c}\right)\right], \quad \sigma_{ii} = \rho_{ii},$$

where ρ_{ik} are the elements of the density matrix in the Schrödinger representation for a two-level quantum system simulating a ^{87}Rb atom. By using the first approximation of the averaging procedure for the density matrix [13] and truncated Maxwell's equations, we obtain in the first approximation [14] of the dispersion theory the system of equations describing the interaction of a laser pulse with a medium

$$\begin{aligned}
&\frac{\partial a}{\partial s} = \frac{\mathrm{i}}{\sqrt{\pi}}\int_{-\infty}^{\infty}\sigma_{21}\exp\left[-(\varepsilon - \varepsilon_0)^2\right]\mathrm{d}\varepsilon,\\
&\frac{\partial \sigma_{21}}{\partial w} + \mathrm{i}\varepsilon\sigma_{21} = \mathrm{i}a(\sigma_{11} - \sigma_{22}) - \gamma\sigma_{21},\\
&\frac{\partial \sigma_{22}}{\partial w} = -\frac{1}{2}\mathrm{Im}(a\sigma_{21}^*) - \delta\sigma_{22},\\
&\frac{\partial \sigma_{11}}{\partial w} = \frac{1}{2}\mathrm{Im}(a\sigma_{21}^*).
\end{aligned} \quad (3)$$

Here, the notation

$$\varepsilon_0 = T(\omega_{21}^0 - \omega), \quad \varepsilon = T(\omega_{21} - \omega),$$
$$\gamma = \frac{T}{T_{21}'}, \quad \delta = T(T_{23}^{-1} + T_{21}^{-1}) \quad (4)$$

is used, the parameter ε_0 characterising the degree of deviation of the carrier frequency of the incident laser radiation from the central frequency of the quantum transition. The terms $-\gamma\sigma_{21}$ and $-\delta\sigma_{22}$ in the second and third equations of system (3) are introduced to take into account spontaneous transitions in atoms from the level 2 to levels 1 and 3. Due to the presence of these terms, the system (3) differs from the system of equations, whose numerical solution predicted the formation of a precursor pulse [8]. On the other hand, the theory based on system (3) and the representation of the field in the form (1) allows us to take into account the frequency modulation of the laser pulse induced by the medium.

The system (3) was supplemented with the boundary condition $(s = 0)$

$$a(s = 0, w) = a_0(w) \quad (w \geqslant 0), \quad (5)$$

where $a_0(w)$ is the envelope of the incident laser pulse. As the initial conditions $(w = 0)$, the relations

$$\sigma_{12}(s, w = 0) = 0, \quad \sigma_{11}(s, w = 0) = 1,$$
$$\sigma_{22}(s, w = 0) = 0 \quad (s \geqslant 0), \quad (6)$$

were used, which correspond to the excited state of the medium before the laser pulse arrival.

Boundary value problem (3), (5), (6) was solved numerically using the program presented in paper [15]. The results of calculations performed with the help of this program well agree with the analytic results of the SIT theory [1] and the theory of nonstationary double resonance [16]. The problem under study at $\gamma = \delta = 0$ is equivalent to the well-studied problem describing SIT. By accompanying each calculation in which γ and δ are nonzero be the calculation for $\gamma = \delta = 0$, we tested the results obtained by comparing them with the analytic results of the SIT theory.

3. Parameters of the medium and characteristics of the input pulse

Using the data presented in paper [12], we employed in our calculations the following values of the parameters of the resonance quantum transition: $|p| = 6.16$ electrostatic system units, $\omega_{21}^0 = 2.37\times10^{15}$ s^{-1}, $T_{21} = 42$ ns, $T_{23} = 84$ ns, and $T_{21}' = 56$ ns. The FWHM of the Doppler profile of the 1–2 transition equal to 550 MHz gives $T = 0.48$ ns. We find from (4) that $\gamma = 8.6\times10^{-3}$ and $\delta = 1.7\times10^{-2}$.

For boundary conditions (5), we assume that

$$a_0(w) = h\left[\exp\left(\frac{w - w_0}{\tau}\right) + \exp\left(-3\,\frac{w - w_0}{\tau}\right)\right]^{-1}. \quad (7)$$

The dimensionless parameters h, τ, and w_0 in (7) determine the height, duration, and position of the input laser pulse on the axis w. The pulse (7) has an asymmetric shape (close to the shape of experimental pulses [12]), with a greater steepness of the leading edge compared to that of the trailing edge. The required value of the input pulse area is provided by fitting the parameter h. As the parameter τ is varied from 10 to 20, the FWHM of the function $|a_0(w = t/T)|^2$ describing the laser pulse intensity lies within 5–10 ns. Input pulses of such durations were used in the experiment [12].

The values of some parameters were taken from section IV.8.2 in paper [12], where the results of experiments performed in the quasi-resonance case are reported. For example, for $(\omega - \omega_{21}^0)/2\pi = 0.7$ GHz, we obtain from (4) $\varepsilon_0 = -2.1$. According to the data presented in this section, the value of $\alpha' L$ (where α' is the linear absorption coefficient at the laser frequency, and L is the length of a cell containing rubidium vapours) was estimated as ~ 7. At the same time, the maximum concentration of ^{87}Rb atoms was approximately 10^{13} cm^{-3}, whereas the maximum value of L was 10 mm. By using the first of the expressions in (2), we can conclude that the dimensionless distance s corresponding to the experiment is approximately 100.

4. Method for representing the calculation results

The results of calculations are represented as functions $A_s(w)$, $\varphi_s(w)$, $w_{\rm m}(s)$, and $\Theta(s)$, where

$$A_s(w) = |a(w, s = {\rm const})|, \quad \varphi_s(w) = \arg a(w, s = {\rm const})$$

is the real envelope of a pulse and the phase addition to it when s is fixed; $w_{\rm m}(s)$ is the dependence on s of the value of w at which the envelope $A_s(w)$ achieves the maximum value $A_{s{\rm m}}$; and

$$\Theta(s) = \int_{-\infty}^{\infty} A_s(w){\rm d}w$$

is the dependence of the area under the real envelope on s. In this case, $\Theta(0)$ is the area under the real envelope of the input laser pulse. The pulse duration is characterised by the dimensionless quantities τ_1 and τ_2, which are defined as the distances alone the axis w between the points at which the function $A_s(w) = 0.5A_{s{\rm m}}$ and ${\rm sech}(1)A_{s{\rm m}}$, respectively. When the pulse deformation during its propagation is insignificant, we consider its velocity υ in the coordinate system ws, which is defined by the expression $\upsilon = ({\rm d}w_{\rm m}/{\rm d}s)^{-1}$. For a 2π pulse, the quantities τ_2, $A_{s{\rm m}}$, and υ are related by the expressions

$$A_{s{\rm m}} = \frac{4}{\tau_2}, \quad \upsilon = \sqrt{\pi}\left\{\tau_2^2 \int_{-\infty}^{\infty} \frac{\exp\left[-(\varepsilon-\varepsilon_0)^2\right]}{4+\tau_2^2\varepsilon^2}{\rm d}\varepsilon\right\}^{-1}. \quad (8)$$

These expressions are used then to detect the 2π pulse in numerical experiments. The symbol Δt denotes below the dimensional duration of the pulse, which is used in paper [12], and is defined as the time interval between two points at which the function $A_s^2(w = t/T)$ achieves half its maximum value.

Depending on s the shift $\Delta\nu$ of the centre of the pulse spectrum with respect to the carrier frequency $\nu = \omega/2\pi$ of the input radiation is determined by the expression

$$\Delta\nu = \frac{1}{2\pi T}\int_{-\infty}^{\infty} \varDelta F_s(\varDelta){\rm d}\varDelta \bigg/ \int_{-\infty}^{\infty} F_s(\varDelta){\rm d}\varDelta. \quad (9)$$

Here, $F_s(\varDelta)$ is the square of the modulus of the Fourier transform of the function $\boldsymbol{E}/(\mu T)$ at the point $\omega - \varDelta/T$ for s fixed. If $\varepsilon_0\Delta\nu > 0$ ($\varepsilon_0\Delta\nu < 0$), then the central frequency of the pulse spectrum is shifted away from (toward) the resonance with respect to the carrier frequency of the input laser pulse.

5. Results of calculations

Let us set the parameters $h = 0.233$, $\tau = 14$, and $w_0 = 40$ in (7), which corresponds to the input pulse with $\Theta(0) = 1.1\pi$ and $\Delta t = 7$ ns. The choice of the value of $\Theta(0)$ close to π is determined by experimental conditions. Figures 1a–c show the functions $A_s(w)$ and $\varphi_s(w)$ for $s = 100$, 240, and 360. The value $s = 100$ (see Fig. 1a), as mentioned above, corresponds to experimental conditions. For $s = 100$, the calculation gives $\Delta t = 19.3$ ns, which is almost three times longer than the input pulse duration, whereas $\Delta\nu = -12$ MHz (the negative sign of $\Delta\nu$ means that the

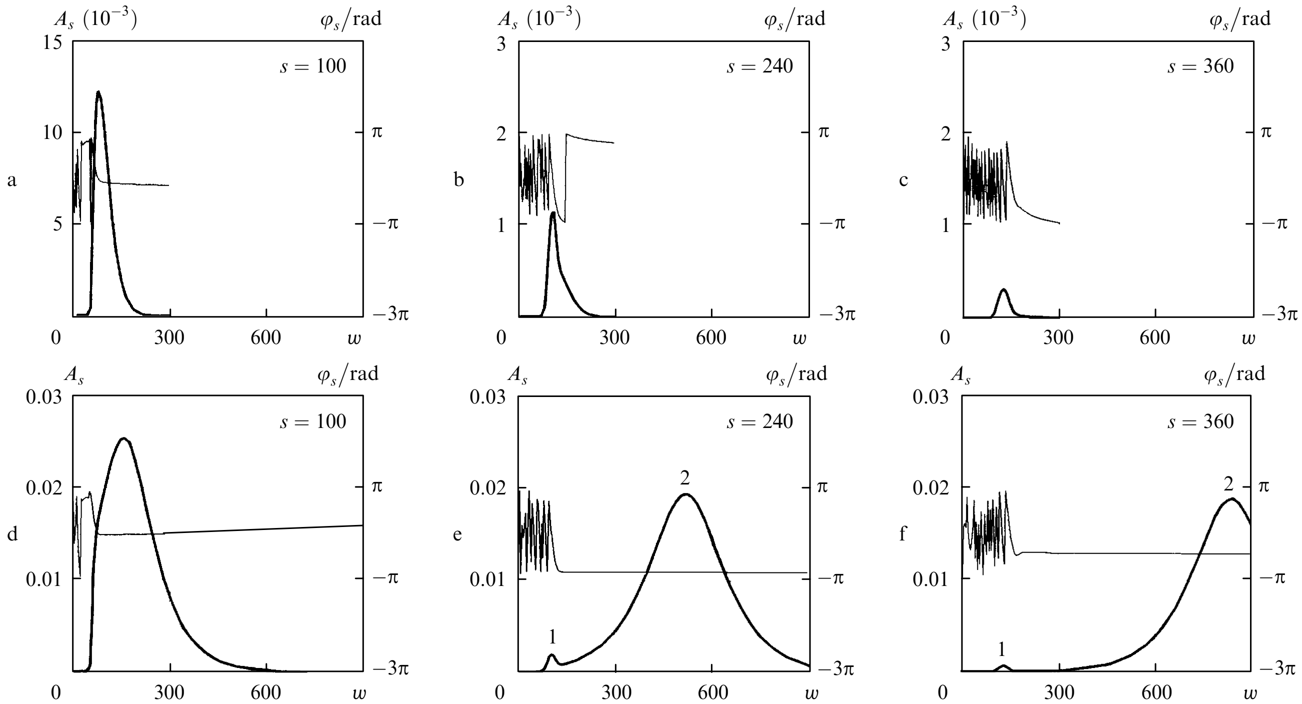

Figure 1. Real envelopes A_s (thick curves) and phase additions φ_s (thin curves) for different distances s and the input pulse area equal to 1.1π in the presence (a–c) and absence (d–f) of relaxation processes.

centre of the pulse spectrum is repulsed from the resonance). These results are in good agreement with the experimental data reported in section IV.8.2 of paper [12]. Figures 1b, c present the results of calculations performed for distances that greatly exceed experimental distances.

Figures 1d–f show the functions $A_s(w)$ and $\varphi_s(w)$ for the same values of s as above, but calculated neglecting relaxation ($\gamma = \delta = 0$). One can see that the pulse decomposes during its propagation into two separate pulses, which are denoted by numbers 1 and 2 according to the order of their propagation. The first pulse is substantially weaker than the second one and propagates at a greater velocity. The dependences $\varphi_s(w)$ show that the spectrum of the first pulse has the frequency modulation ($\mathrm{d}\varphi_s/\mathrm{d}w \neq 0$), whereas the spectrum of the second pulse has no modulation. As s increases, the shape and height of the second pulse are virtually invariable. For $s = 360$, $\tau_2 = 212$ and $A_{sm} = 0.0187$ for the second pulse. Figures 2 and 3 show the dependences $\Theta(s)$ and $w_{\mathrm{m}}(s)$. One can see that the area $\Theta(s)$ approaches 2π at large s. A linear dependence $w_{\mathrm{m}}(s)$ observed for $s > 100$ means that the velocity of the second pulse is constant, and according to this dependence, $v = 0.382$. By substituting the above-presented values of τ_2, A_{sm}, and v into (8), we obtain approximate inequalities, which are fulfilled with the relative error of 1 %. All this suggests that the second pulse is the 2π pulse. Then, according to Ref. [8], the first pulse is the precursor pulse. For the precursor pulse, as follows from the calculation, $\Delta\nu \simeq -100$ MHz; however, because of the presence of the 2π pulse without frequency modulation, the train of two pulses has the frequency shift with a significantly smaller modulus ($\Delta\nu \simeq -90$ kHz for $s = 240$). The precursor pulse, as the 2π pulse, has the symmetric bell-shaped envelope, however, unlike the 2π pulse, it decays during propagation even in the absence of relaxation processes.

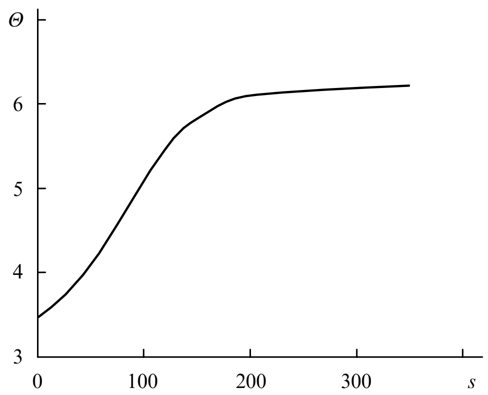

Figure 2. Dependence of the area Θ under the real envelope on the distance s for the input pulse area equal to 1.1π.

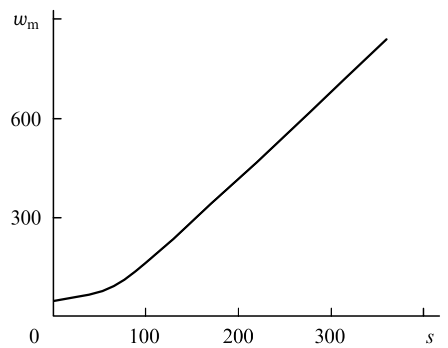

Figure 3. Dependence of the time position w_{m} of the maximum of the real envelope on the distance s for the input pulse area equal to 1.1π.

By comparing Figs 1 b, c with Figs 1 e, f, one can easily see that, taking into account relaxation for each fixed value s, the pulse is located on the axis w at the same place where the precursor pulse is located for the given s in the absence of relaxation. For example, for $s = 360$ (Figs 1c, f), the abscissas of the tops of these pulses and their durations τ_1 are equal to 130 and 40, respectively. For both pulses, $\Delta\nu \simeq -100$ MHz. All this suggests that relaxation processes at large distances suppress the formation of the 2π pulse but allow the formation of the precursor pulse.

At the distance $s \simeq 100$, corresponding to the experimental conditions [12], the process of transformation of the laser pulse to the precursor pulse is still in the initial stage. This follows from a small shift ($|\Delta\nu| = 12$ MHz) of the central frequency of the pulse spectrum compared to a similar shift ($|\Delta\nu| \simeq 100$ MHz) for the precursor. A comparison shows that, for s being fixed, the pulse intensity in the presence of relaxation is approximately half as much as for the precursor pulse in the absence of relaxation. This means that relaxation processes result in an increase in the decay rate of the precursor pulse during its propagation.

Let us present the results of calculations for the parameters in (7) $h = 0.181$, $\tau = 14$, and $w_0 = 40$, corresponding to $\Theta(0) = 0.9\pi$ and $\Delta t = 7$ ns. It is known [1, 8] that for such a value of $\Theta(0)$ the 2π pulse cannot be formed even in the absence of relaxation. The calculation in the presence of relaxation and $s = 100$ gave the value $\Delta\nu = -17$ MHz, which lies in the region of experimentally observed values [12]. Figure 4 presents the functions $A_s(w)$ for $s = 360$ in the presence and absence of relaxation. The values of $w_{\mathrm{m}}(s)$, Δt, and $\Delta\nu$ for both pulses are virtually the same, and $\Delta\nu \simeq -100$ MHz. The position of these pulses on the axis w is the same as for the precursor pulse in Fig. 1f. This means that the precursor pulse also appears for $\Theta(0) < \pi$, and this pulse is responsible for the frequency shift observed in experiments. Note that the calculations performed in paper [8] predicted the appearance of the precursor pulse only when the condition $\pi < \Theta(0) < 2\pi$ was fulfilled.

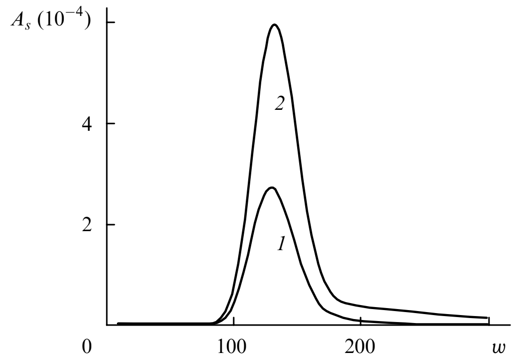

Figure 4. Dependences of the real envelope A_s at the distance $s = 360$ on w for the input pulse area equal to 0.9π in the presence (*1*) and absence (*2*) of relaxation processes.

Our calculations showed that the qualitative features of the evolution of the input pulse preserved when its duration Δt was further decreased. However, the frequency-shift magnitude increases. Thus, for $\Theta(0) = 1.1\pi$ and $\Delta t = 5$ ns, we have $\Delta\nu = -20$ MHz at the distance $s = 100$. An increase in the input pulse area results in a decrease in $|\Delta\nu|$. For example, for $\Theta(0) = 1.4\pi$ and $\Delta t = 5$ ns, the calculation gives $\Delta\nu = -5.5$ MHz for $s = 100$. The decrease in $|\Delta\nu|$ is qualitatively explained by the fact that self-induced transparency is not efficiently enough suppressed by relaxation processes with increasing $\Theta(0)$.

Note that the precursor appears within a rather broad range of variations of $\Theta(0)$. Figure 5 shows the dependences $A_s(w)$ for $s = 360$ for the input pulse with $\Theta(0) = 3.5\pi$ and $\Delta t = 5$ ns ($h = 1.0$ and $\tau = 10$), which were calculated taking relaxation into account or neglecting it. The first pulse from the origin in Figs 5a, b is the precursor pulse, for which $\Delta\nu \simeq -100$ MHz. The second and third pulses in Fig. 5a are 2π pulses, the top of the second pulse lying beyond the figure. The second pulse in Fig. 5b is not a soliton and decays with distance faster than the precursor pulse.

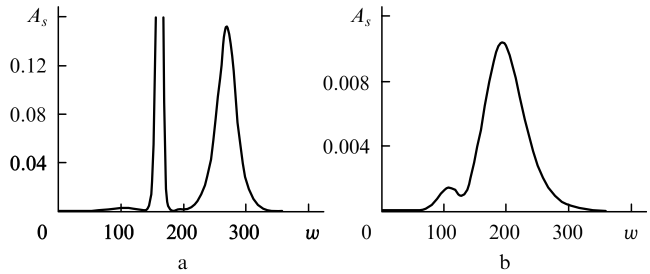

Figure 5. Dependences of the real envelope A_s at the distance $s = 360$ on w for the input pulse area equal to 3.5π in the absence (a) and presence (b) of relaxation processes.

6. Conclusions

Our calculations have shown that upon quasi-equilibrium interaction of radiation with an inhomogeneously broadened quantum transition, a precursor pulse appears when the area of $\Theta(0)$ of the input laser pulse can be both smaller and larger than π. A distinct feature of the precursor pulse is its strong frequency modulation, which is manifested as the repulsion of the central frequency of its spectrum from the resonance ($\Delta\nu \simeq -100$ MHz). In the absence of relaxation, the precursor pulse decays because of the Doppler spread of resonance frequencies. When $\Theta(0) < \pi$ almost total radiation energy is contained at large distances in the precursor pulse.

When $\Theta(0) > \pi$, the precursor pulse is accompanied by one or several 2π pulses. Because the 2π pulse in our case is somewhat more intense than the precursor pulse and has no frequency modulation, the value of $|\Delta\nu|$ for a train of pulses is considerably lower than that for the precursor pulse. The presence of even weak influence of relaxation processes (δ, $\gamma \ll 1$) results first of all in the suppression of SIT. The precursor pulse, despite the increase in its decay, contains the total energy at large distances. At the distance corresponding to the experiment described in section IV.8.2 of paper [12], the process of transformation of the pulse to the precursor pulse is in the initial stage. Therefore, $\Delta\nu$ changes from -20 to -10 MHz if $\Theta(0)$ is close to π. As $\Theta(0)$ increases, the value of $|\Delta\nu|$ decreases because the distance at which relaxation can suppress SIT increases. Note that precursor pulses obtained in our calculations have a symmetric bell-shaped profile, which is similar to that observed in experiment [11], whereas calculations performed in paper [8] gave a complicated asymmetric structure of the precursor envelope containing many peaks.

The results of numerical experiments presented in the paper are restricted by the only set of numerical values of the parameters of the resonance medium and a limited range of the input laser pulse parameters. Therefore, it is interesting to study the dependence of the laser pulse evolution on the degree of deviation of this pulse from the resonance and its possible modulation. We plan to continue studies in this direction.

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