

NONLINEAR OPTICAL PHENOMENA

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Formation of optical pulses by modulating the resonant quantum transition frequency in a spectrally inhomogeneous medium

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***Abstract*. We consider the conversion of monochromatic radiation in the case of resonant interaction with a quantum system under the condition of harmonic modulation of the quantum transition frequency by the action of additional nonresonant radiation due to the Stark or Zeeman effect, taking into account the inhomogeneous broadening of the quantum transition line. It is shown analytically and numerically that resonant radiation can be converted in a train of ultrashort pulses with a peak intensity exceeding manifold the incident wave intensity. The possibility of the additional compression of the produced pulses is studied by compensating the inherent frequency modulation in a medium with a quadratic or programmable dispersion. The optimal values of the radiation – matter interaction parameters are found numerically. It is shown that generation of femtosecond optical pulses of radiation quasi-resonant to the δ transition of the atomic hydrogen Balmer series is possible.**

Keywords: *ultrashort pulses, coherent optics, resonant interaction, inhomogeneous broadening, Stark effect, Zeeman effect.*

1. Introduction

The technique of laser pulse generation by the mode-locking method has now acquired its final form [1]. The advance in the field of shorter durations and higher peak intensities, formation of far-IR and vacuum UV pulses, X-ray and gamma pulses is based on new physical methods. These methods involve compression of laser pulses in the soliton propagation regime in a two-level medium [2, 3], compression of optical solitons in plasma channels produced in a gas [4, 5], self-action of tightly focused laser pulses in a transparent condensed medium [6], generation of tunable femtosecond pulses in optical fibres with length-variable dispersion [7], formation of few-cycle far-IR and terahertz pulses during the interaction of radiation with a relativistic electron beam [8, 9], generation of a broad line spectrum and formation of sub-femtosecond optical pulses during the stimulated Raman scattering (SRS) on vibrational or/and rotational transitions of molecules [10 – 14]. When coherence in the SRS medium is excited by a driving laser pulse, a train of femtosecond pulses is obtained experimentally [15], and the possibility of producing a single 1-fs pulse is shown [16]. The decrease in the pulse repetition rate is feasible via SRS on a coherence induced between the superfine sublevels [17, 18]. Formation of attosecond pulses with a duration less than 100 as is achieved during generation and in-phase summation of high laser radiation harmonics in the case of tunnel ionisation and recombination of atoms in the field of a terawatt ultrashort laser pulse [19 – 24]. Generation of shorter pulses (with a duration less than 1 as) is predicted in the case of ionisation of a solid target in a superstrong optical field [25 – 29].

We proposed the method of ultrashort electromagnetic pulse formation [30, 31] based on the deep amplitude and frequency modulation of incident monochromatic radiation due to the resonant interaction with a quantum system under the condition of harmonic modulation of the resonant quantum transition frequency. It can be used for generating ultrashort pulses in different spectral regions, from the microwave to the gamma region. Unlike most of the above-mentioned approaches, efficient in the region of optical transparency of the medium, this method allows one to use strong resonant interaction for generating a broad spectrum and compensating for the phases of the emerging harmonics. Further comparison of the proposed method with that mentioned above, especially with respect to the generation efficiency and the limiting values of the pulse parameters, requires separate investigations. One such investigation is the present paper.

In this paper, we consider the application of the above method for producing ultrashot optical pulses in the case of resonant interaction with a quantum system under conditions of inhomogeneous broadening of the resonant transition line. The harmonic modulation of the quantum transition frequency is achieved due to oscillations of the position of atomic levels caused by additional nonrsonant radiation due to the Stark or Zeeman effect.

2. Formulation of the problem and analytic solution

Consider the conversion of a plane monochromatic electromagnetic wave in a plane dielectric layer of a medium under the condition of harmonic modulation of the resonant quantum transition frequency by a low-frequency electromagnetic field due to the Stark or Zeeman effect.

The electric field of the incident wave is

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$$\boldsymbol{E}_{\rm in} = \frac{1}{2}\,\boldsymbol{x}_0 E_0 \exp(\mathrm{i}k_0 z - \mathrm{i}\omega_0 t) + \mathrm{c.c.}, \qquad (1)$$

where $\boldsymbol{x}_0$ is the polarisation unit vector; ω_0 is the incident wave frequency; $k_0 = \omega_0/c$ is the wave number of the incident wave; c is the speed of light in vacuum. The field in the medium satisfies the wave equation

$$\frac{\partial^2 \boldsymbol{E}}{\partial z^2} - \frac{\varepsilon}{c^2}\frac{\partial^2 \boldsymbol{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \boldsymbol{P}}{\partial t^2}, \qquad (2)$$

where $\boldsymbol{P}$ is the resonant polarisation vector of the medium; ε is the nonresonant dielectric constant.

The frequencies ω_{21} of the resonant quantum transition of the particles in a spectrally inhomogeneous medium are characterised by the distribution $p(\omega_{21})$ and the average value $\omega_{21}^0 = \int \omega_{21} p(\omega_{21}) \mathrm{d}\omega_{21}$. The resonant polarisation vector of the isotropic medium is codirected with the field vector ($\boldsymbol{P} \| \boldsymbol{E}$) and is expressed via the density matrix elements:

$$\boldsymbol{P} = \boldsymbol{x}_0 N \int \rho_{21}(\omega_{21}) d_{12}\, p(\omega_{21}) \mathrm{d}\omega_{21} + \mathrm{c.c.}, \qquad (3)$$

where d_{12} is the average dipole moment of the quantum transition; ρ_{21} is the nondiagonal element of the density matrix; N is the concentration of the resonant particles. The quantity ρ_{21} depends on ω_{21} as on a parameter and satisfies the equation

$$\frac{\partial \rho_{21}}{\partial t} + (\mathrm{i}\omega_{21} + \gamma_{21})\rho_{21} = \frac{\mathrm{i}}{\hbar}\, n_{12} d_{21} E, \qquad (4)$$

where γ_{21} is the half-width of the homogeneous component of the spectral transition line; $n_{12} = \rho_{11} - \rho_{22}$ is the population difference between the lower and upper levels of the quantum transition. We will restrict our consideration to the approximation linear in the field, when the population difference is unperturbed: $n_{12} = n_{12}^0$. We will assume that $n_{12}^0 > 0$, which corresponds to the resonant field absorption.

In the presence of the additional nonresonant radiation at the frequency Ω, which we will call modulating, the atomic energy levels start oscillating at this frequency due to the Stark or Zeeman effect, which leads to modulation of the transition frequency $\omega_{21}(t)$. If the medium thickness h is small compared to the modulating radiation wavelength ($h \ll 2\pi c/\Omega$), the quantum transition frequencies of all the particles change in-phase:

$$\omega_{21} = \bar{\omega}_{21} + \varDelta \cos(\Omega t), \qquad (5)$$

where $\bar{\omega}_{21}$ is the transition frequency in the absence of the modulating field; $\varDelta$ is the modulation depth of the quantum transition frequency. Equation (4) for the nondiagonal element of the density matrix in the presence of the modulating field has the form

$$\frac{\partial \rho}{\partial t} + (\mathrm{i}\bar{\omega}_{21} + \gamma_{21})\rho_{21} + \mathrm{i}\varDelta\cos(\Omega t)\rho_{21} = \frac{\mathrm{i}}{\hbar}\, n_{12} d_{21} E. \qquad (6)$$

A convenient way to solve problems (2), (3), (6) is the change of the variable, $t \to t_{\rm a}$, which is determined from the condition of constancy of the resonant transition frequency:

$$\bar{\omega}_{21} t_{\rm a} = \int_0^t [\bar{\omega}_{21} + \varDelta\cos(\Omega t')]\mathrm{d}t'. \qquad (7)$$

It is easy to see that when the conditions $|\omega_0 - \bar{\omega}_{21}| \ll \bar{\omega}_{21}$ and $\varDelta \ll \bar{\omega}_{21}$ are fulfilled, we have the equality

$$\omega_0 t = \omega_0 t_{\rm a} - \frac{\varDelta}{\Omega}\sin(\Omega t_{\rm a}). \qquad (8)$$

The change $t \to t_{\rm a}$ transforms equation (6) to the form coinciding with (4). In this case, the expression for the incident wave takes the form

$$\boldsymbol{E}_{\rm in} = \frac{1}{2}\boldsymbol{x}_0 E_0 \exp[\mathrm{i}R\sin(\Omega t_{\rm a})]\exp(\mathrm{i}k_0 z - \mathrm{i}\omega_0 t_{\rm a}) + \mathrm{c.c.}$$

$$= \frac{1}{2}\,\boldsymbol{x}_0 E_0 \exp(\mathrm{i}k_0 z) \sum_{n=-\infty}^{+\infty} J_n(R) \exp[-\mathrm{i}(\omega_0 - n\Omega)t_{\rm a}] + \mathrm{c.c.}, \qquad (9)$$

where $R = \varDelta/\Omega$; $J_n(R)$ are the Bessel functions of the first kind.

Thus, the above change reduces the study of the monochromatic wave conversion (1) in the medium with the harmonically modulated transition frequency (5) to the investigation of the frequency-modulated wave conversion (9) in the medium with the fixed frequency of the quantum transition.

Below, we assume the dielectric constant of the medium to be close to unity ($\varepsilon \approx 1$) so that the boundary conditions on the sample faces would be reduced to the equality of the electric field strengths in the medium and outside its boundaries: $\boldsymbol{E}|_{z=0} = \boldsymbol{E}_{\rm in}|_{z=0}$ and $\boldsymbol{E}_{\rm tr}|_{z=h} = \boldsymbol{E}|_{z=h}$ ($E_{\rm tr}$ is the strength of the electric field transmitted through the medium).

We will seek the field satisfying equations (2)–(4) and boundary conditions depending on the variables $z, t_{\rm a}$ in the form

$$\boldsymbol{E}(z, t_{\rm a}) = \frac{1}{2}\,\boldsymbol{x}_0 E_0 \exp(\mathrm{i}kz) \sum_{n=-\infty}^{+\infty} J_{-n}(R)\exp(-g_n z) \times \exp(-\mathrm{i}\omega_n t_{\rm a}) + \mathrm{c.c.}, \qquad (10)$$

where $\omega_n = \omega_0 + n\Omega$; $g_n \equiv g(\omega_n)$ are the complex decrements of the field harmonics; $k = \sqrt{\varepsilon}\omega_0/c$ is the wave number in the medium. We will seek the elements of the density matrix $\rho_{21}(\bar{\omega}_{21})$ in the form

$$\rho_{21}(\bar{\omega}_{21}) = \exp(\mathrm{i}kz) \sum_{n=-\infty}^{+\infty} J_{-n}(R) b_n(\bar{\omega}_{21}) \times \exp(-g_n z)\exp(-\mathrm{i}\omega_n t_{\rm a}). \qquad (11)$$

By substituting expressions (10), (11) into equations (2)–(4), using the resonant approximation, $|\omega_0 - \bar{\omega}_{21}| \ll \bar{\omega}_{21}$, and slowly varying amplitude approximation, $\max|g_n|$, $\varDelta \ll \bar{\omega}_{21}$, we obtain

$$g_n = \frac{2\pi N n_{12}^0 \omega_0 |d_{12}|^2}{\sqrt{\varepsilon}\hbar c} \int \frac{p(\bar{\omega}_{21})\mathrm{d}\bar{\omega}_{21}}{\gamma_{21} + \mathrm{i}(\bar{\omega}_{21} - \omega_n)}. \qquad (12)$$

If the spectral inhomogeneity is caused by the thermal motion of particles or by statistically independent random perturbations of the local fields in the medium, the distribution of resonant frequencies of the particles in the medium will be close to Gaussian

$$p(\bar{\omega}_{21}) = \frac{1}{\sqrt{\pi}\sigma} \exp\left[-\frac{(\bar{\omega}_{21} - \omega_{21}^0)^2}{\sigma^2}\right]. \quad (13)$$

In the case $\sigma \gg \gamma_{21}$, taking into account that the sequence of Lorentz curves is reduced to a δ-function by decreasing the width, we obtain

$$g_n = \frac{2\pi^{3/2} N n_{12}^0 \omega_0 |d_{12}|^2}{\sqrt{\varepsilon}\hbar c \sigma} \times \left[\exp(-\delta_n^2) - \frac{\mathrm{i}}{\pi} \int_{-\infty}^{+\infty} \frac{\mathrm{e}^{-(\zeta+\delta_n)^2} - \mathrm{e}^{-\delta_n^2}}{\zeta} \mathrm{d}\zeta\right], \quad (14)$$

where $\delta_n = (\omega_n - \omega_{21}^0)/\sigma$.

Expressions (10), (14) completely determine the dependence of the field on the variables z, t_a, transition to the variables z, t being performed by the transformation inverse to transformation (8):

$$\omega_0 t_a = \omega_0 t + \frac{\Delta}{\Omega} \sin(\Omega t). \quad (15)$$

In this case, the expression for the field, transformed in the medium, takes the form

$$\boldsymbol{E} = \frac{1}{2} \boldsymbol{E}_0 \exp(\mathrm{i}kh) \sum_{m=-\infty}^{+\infty} \sum_{n=-\infty}^{+\infty} (-1)^m J_n(R) J_{n-m}(R) \times \exp(-g_n h) \exp[\mathrm{i}k_m(z-h) - \mathrm{i}\omega_m t] + \mathrm{c.c.} \quad (16)$$

where $\omega_m = \omega_0 + m\Omega$; $k_m = \omega_m/c$.

The resonant interaction of the monochromatic radiation with the medium under the condition of harmonic modulation of the quantum transition frequency can lead to a qualitative change in the emission spectrum. The time dependence of the field, which is responsible in the general case to its amplitude and frequency modulation, changes correspondingly.

3. Formation of pulses in a resonant frequency-modulated medium

Four dimensionless parameters

$$R = \frac{\Delta}{\Omega}, \quad \eta \equiv \frac{\Omega}{\sigma}, \quad \xi \equiv \frac{\omega_0 - \omega_{21}^0}{\sigma}, \quad G \equiv h \mathrm{Re} g_{\max}, \quad (17)$$

which denote the modulation depth of the quantum transition frequency with respect to the modulation frequency (R), the modulation frequency with respect to the inhomogeneous width of the quantum transition line (η), frequency detuning of the incident wave from the quantum transition frequency with respect to the inhomogeneous quantum transition linewidth (ξ), and the optical thickness (G) of the medium, determine the harmonic amplitudes and phases of field (16) and its time dependence. At certain values of the mentioned parameters, after transformation in the medium, the field takes the form of a pulse train. There appears the problem of determining the optimal values of parameters (17) at which the maximum peak intensity and the minimum pulse duration are achieved. This problem is solved numerically.

Consider the results of the numerical search for the optimal values of parameters (17) at which the pulses at the medium output have the maximum peak intensity with respect to the incident wave intensity:

$$I_{\max}/I_0 \to \max. \quad (18)$$

We found several sets of optimal values of the parameters whose final choice is determined by the possibilities of experimental realisation. We will consider the physical mechanism of the pulse formation in the medium with a variable frequency of the resonant quantum transition by the example of one of these sets ($R = 2.0$, $\eta = 10$, $\xi = -12.5$, $G = 10$). The spectrum of the incident monochromatic wave (1) assuming form (9) after the substitution $t \to t_a$ (8) is shown in Fig. 1 against the background of the curves of the resonant absorption and resonant dispersion. As the frequency-modulated wave (9) propagates in the medium with the fixed parameters with respect to the variables z, t_a, the amplitudes and phases of the field harmonics change due to the resonant absorption and resonant dispersion. According to the results of the numerical research, the range of the values of the parameters $R\eta \gg 1$ in which the width of the radiation spectrum (9) significantly exceeds the transition linewidth is optimal for producing intense pulses. In this case, the resonant dispersion plays a crucial role in the pulse formation, while the resonant absorption is insignificant. After radiation is transmitted through the medium, its spectrum (10) acquires the shape presented in Fig. 2.

Returning to the initial variable [$t_a \to t$ (15)], the radiation spectrum takes the shape (16) shown in Fig. 3.

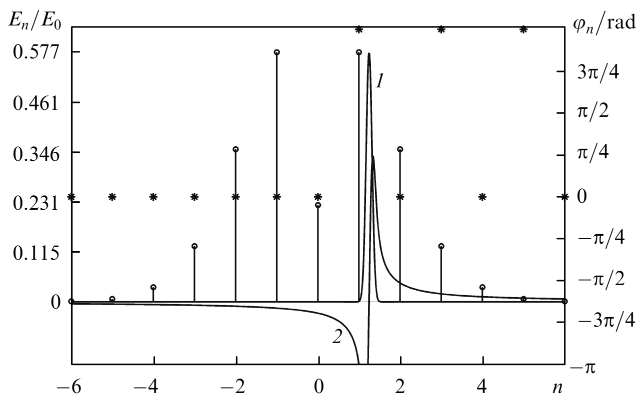

Figure 1. Spectrum of an incident monochromatic wave [amplitudes (○) and phases (∗) of the field harmonics] as a function of t_a at $R = 2.0$, $\eta = 10$, $\xi = -12.5$, $G = 1$ against the background of the profiles of the resonant absorption (*1*) and resonant dispersion (*2*).

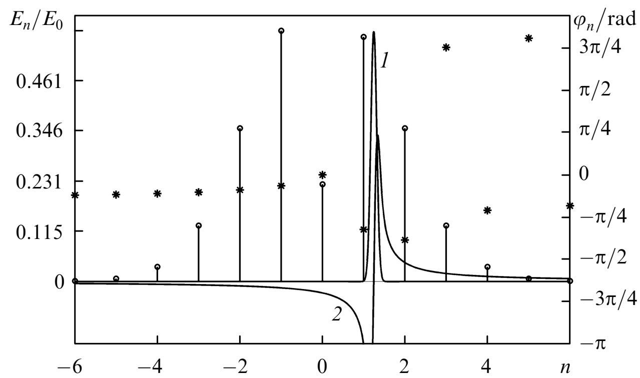

Figure 2. Spectrum of the wave transformed due to the resonant interaction with the quantum system as a function of t_a for the same parameters as in Fig. 1.

The corresponding time dependence of the field intensity represents a train of pulses shown in Fig. 4. Comparison of Figs 1, 2, and 3 shows that the selection of optimal values of the parameters is reduced to the minimisation of the energy losses due to resonant absorption as well as to synchronisation to the best advantage of spectral components due to the resonant phase incursion. As a result, the average field intensity at the output hardly decreases, while the redistribution of the intensity with respect to time due to synchronisation of spectral components leads to deep dips and high-power bursts, which significantly exceed the intensity at the medium input. For the selected values of parameters (17) the peak pulse intensity of the incident wave is more than four times greater than the incident radiation intensity. The pulse duration is 1/6 of the repetition period. The produced pulses are frequency-modulated; the intrinsic frequency modulation is shown in Fig. 5. The compensation of frequency modulation makes it possible to compress the produced pulses and to increase the peak intensity.

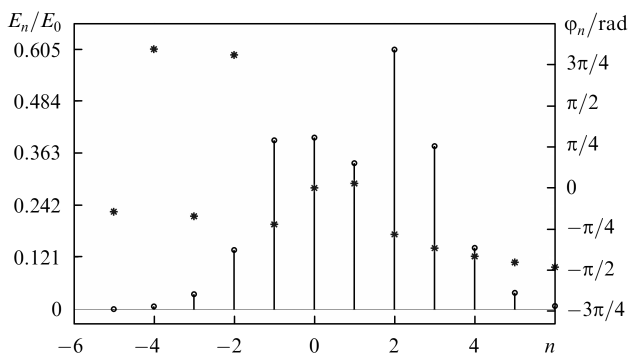

Figure 3. Spectrum corresponding to that presented in Fig. 2 in the laboratory reference frame.

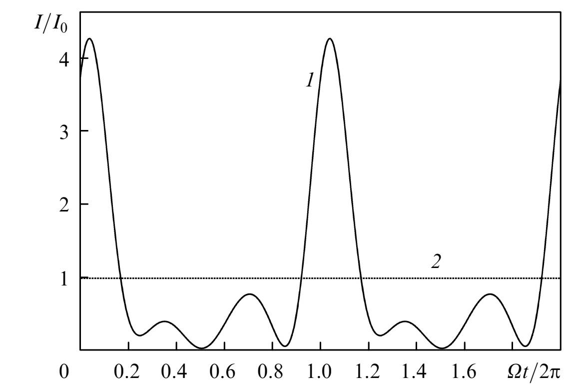

Figure 4. Time dependences of the instant (*1*) and average (*2*) intensities of produced pulses for the same parameters as in Fig. 1.

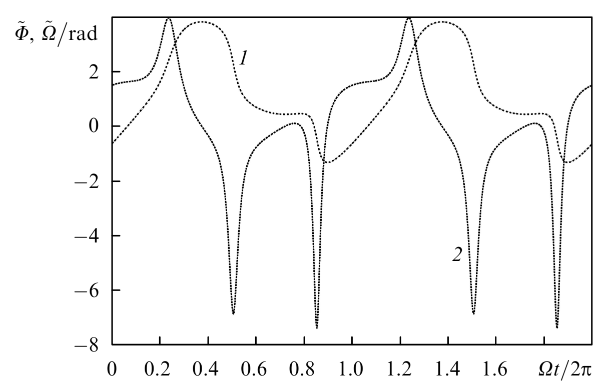

Figure 5. Time dependences of the instant phase $\tilde{\Phi}$ (*1*) and frequency $\tilde{\Omega}$ (*2*) of produced pulses for the same parameters as in Fig. 1.

4. Compression of produced pulses

Consider conversion of radiation experiencing resonant interaction with the frequency-modulated medium in a transparent dispersive medium. The relative phases of harmonics change as radiation propagates in the dispersive medium. Consider a medium with a quadratic dispersion:

$$k(\omega + n\Omega) \simeq k(\omega) + k'_{\omega} n\Omega + \frac{1}{2}\, k''_{\omega\omega} n^2 \Omega^2. \tag{19}$$

The field (16) transmitted through the dispersive medium layer of thickness L has the form

$$\begin{aligned} \boldsymbol{E} = \frac{1}{2}\, \boldsymbol{E}_0 \exp[\mathrm{i}\varphi_0(h, L)] \sum_{m=-\infty}^{+\infty} \sum_{n=-\infty}^{+\infty} (-1)^m J_n(R) J_{n-m}(R) \\ \times \exp\left(\mathrm{i}\, \frac{1}{2}\, k''_{\omega\omega} m^2 \Omega^2 L\right) \exp(-g_n h) \\ \times \exp[\mathrm{i}k_m z - \mathrm{i}\omega_m (t - t_0)] + \text{c.c.} \end{aligned} \tag{20}$$

The choice of the optimal value of the parameter $k''_{\omega\omega}\Omega^2 L$ characterising the medium dispersion allows one to compensate for the linear component of the frequency deviation of the field (20). The numerical optimisation of compression showed the possibility of a substantial decrease in the duration and an increase in the peak intensity of produced pulses. One of the optimal solutions corresponds to $R = 6.8$, $\eta = 1.4$, $\xi = -4.8$, $G = 7.8$, $k''_{\omega\omega}\Omega^2 L = -2.800$. Figure 6 presents the time dependence of the intensity. The peak intensity exceeds the incident wave intensity by more than seven times, the pulse duration being 1/15 of the repetition period.

Compensation for the nonlinear component of the frequency deviation achieved in the prism compressors and mirrors with a programmable dispersion allows further

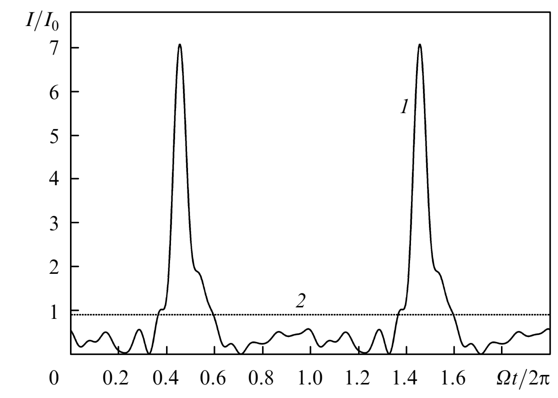

Figure 6. Instant (*1*) and average (*2*) intensities of pulses, produced in the case of the resonant interaction with the quantum system and compressed in a medium with the quadratic dispersion at $R = 6.8$, $\eta = 1.4$, $\xi = -4.8$, $G = 7.8$, $k''_{\omega\omega}\Omega^2 L = -2.800$.

pulse compression. In the case of the complete compensation for the phase difference of harmonics, the field (16) takes the form

$$E = \frac{1}{2} E_0 \exp(\mathrm{i}\varphi_0) \sum_{m=-\infty}^{+\infty} \sum_{n=-\infty}^{+\infty} |J_n(R) J_{n-m}(R)| \exp(-h \mathrm{Re}\, g_n)$$

$$\times \exp[\mathrm{i}k_m(z-h) - \mathrm{i}\omega_m t] + \mathrm{c.c.} \qquad (21)$$

According to the results of the numerical analysis, the compensation for both the linear and nonlinear components of the frequency deviation makes it possible to improve significantly the pulse parameters. Figure 7 demonstrates the time dependence of the intensity of the pulses produced at $R = 16$, $\eta = 22.4$, $\xi = -291.2$, $G = 80$ and at the complete compensation for the difference in the harmonic phases. The peak intensity exceeds the incident wave intensity by 14 times, the average intensity is equal to $0.94 I_0$, the pulse duration is 1/34 of the repetition period.

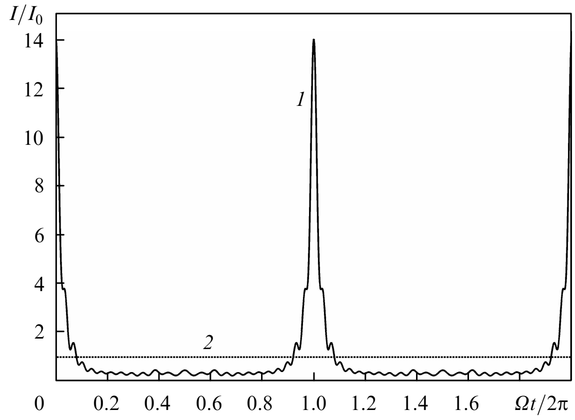

Figure 7. Instant (*1*) and average (*2*) intensities of pulses produced in the case of the resonant interaction with the quantum system and compressed in a medium with the programmable dispersion at $R = 16$, $\eta = 22.4$, $\xi = -291.2$, $G = 80$.

The presented approach to the formation of the electromagnetic pulses is not limited by a specific frequency range neither with respect to the initial resonant radiation nor with respect to the modulating radiation. It is obvious that the experimental realisation allows the use of radiation pulses if their duration exceeds the relaxation time of the resonant polarisation of the medium and the modulating field period. One can see from Figs 6 and 7 that the duration of the produced pulses can be less than 1/30 of the modulating field period and less than 1/600 of the relaxation time of the resonant polarisation. The selection of the optimal medium as well as of modulating and resonant radiation sources can provide the possibility for producing ultrashort pulses in differnet frequency ranges, from far-IR to UV.

Consider, for example, a 1-mm-long cell filled with atomic hydrogen at a pressure of 20 Torr and a temperature of 400 K under the conditions of a glow discharge. The second harmonic of a Ti:sapphire laser at 409.45 nm interacts with the 410.17-nm δ transition of the Balmer series, which has a Gaussian profile of the absorption line of width 5 GHz. The population difference n_{12} of the δ transition energy levels (with the principal quantum numbers equal to 2 and 6) is maintained equal to 0.02. Modulation of the δ-transition frequency due to the Stark effect is produced by the gyrotron radiation at a frequency of $\Omega/2\pi = 80$ GHz focused into a beam with the effective cross section $S = 1$ cm^2. At a 770-kW pulse power of the gyrotron, the optical pulses formed in a medium with a modulated resonant quantum transition frequency will have the shape presented in Fig. 7, after the compensation for the frequency modulation. The pulse duration is $\tau = 370$ fs and the pulse repetition rate is $T = 12.5$ ps. If a CO_2 laser is used as a modulating radiation source, it is possible to form pulses with duration of the order of a femtosecond.

5. Conclusions

Therefore, we have considered monochromatic radiation conversion in the case of the resonant interaction with a quantum system under the condition of harmonic frequency modulation and inhomogeneous broadening of the resonant quantum transition line. The harmonic modulation of the transition frequency is produced by additional nonresonant radiation due to the Stark and Zeeman effect. We have shown that incident resonant radiation under certain conditions is converted into a train of ultrashort pulses whose duration is inversely proportional to the modulation depth of the quantum transition frequency, the repetition period being equal to the modulating radiation period, and the peak intensity being able to exceed the incident wave intensity by many times. The possibility of the pulse compression by compensating the inherent frequency modulation has been demonstrated. We have studied numerically the pulse compression in media with a quadratic or programmable dispersion. It has been established that the compensation for the linear and nonlinear components of the frequency deviation allows one to significantly increase the peak intensity and decrease the pulse duration. We have optimised numerically the pulse formation and determined the optimal values of the radiation–matter interaction parameters. The possibility of producing femtosecond optical pulses of radiation quasi-resonant to the δ transition of the atomic hydrogen Balmer series has been shown.

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