Stationary ultrashort pulses in resonant molecular media

M. V. Bogdanova, Yu. G. Khronopulo, and E. I. Yakubovich

Research Institute for Organic Intermediate Products and Dyes
(Submitted 9 July 1981)
Zh. Eksp. Teor. Fiz. **82**, 421–428 (February 1982)

A soliton-type solution is obtained for resonant ultrashort pulses (USP) whose propagation in a molecular gas medium is accompanied by a large number of multiphoton processes that lift the forbiddenness of vibrational-rotational transitions with $|\Delta j|>1$ (j is the rotational quantum number). An analytic solution is found for an arbitrary dependence of the susceptibility $\chi(|\mathscr{E}|^2)$ of the substance ($\mathscr{E}$ is the radiation amplitude), i.e., for any number of mulitphoton processes of various orders occurring in the resonance transition. The solution describes the waveform of the stationary USP with account of both the quadratic Stark effect and phase modulation of the radiation. It is shown that the presence of a small dipole moment of the resonance transitions, due for example to collisions, may ensure the existence of stationary USP. For ordinary multiphoton processes of orders higher than the second this would be impossible. The pulse waveform is investigated for $\chi=d_{12}+\varkappa^{(3)}|\mathscr{E}|^2$ which describes simultaneously two processes, one involving a single photon and the other three photons in the elementary absorption act.

PACS numbers: 33.80.Kn, 51.70. + f

Coherent interaction of ultrashort light pulses (USP) with a medium in the case of one-photon absorption (OPA), as well as in the case of second-order resonances such as two-photon absorption (TPA) and stimulated Raman scattering (SRS), has been investigated quite fully.[1-5] The possibilities of formation of stationary USP had been discussed, and analytic solutions were obtained for stationary amplitude profiles in the presence of one of the resonances, OPA or TPA (SRS). A substantially different situation is possible, however, when the USP propagates in a molecular medium, and its carrier frequency lies in the IR band and is at resonance with any one of the frequencies of the vibrational-rotational transitions in the molecule. At such a resonance, absorption of intense radiation is due as a rule not only to one-photon but also to three-, five-, and even seven-photon processes in the elementary act of which one photon is absorbed (OPA of third, fifth, etc. order[6,7]).

These "hindrance lifting" processes take place effectively at intensities 10–100 MW/cm^2, thus indicating that they can influence strongly the propagation of USP in OPA. Such a situation is perfectly feasible if, for example, the USP are nanosecond pulses from a CO_2 laser. In the investigation of coherent interaction of USP with molecular media, particularly when considering the possibility of self-induced transparency or the wave forms of the stationary USP, it is therefore necessary to take simultaneous account of both the OPA and the various multiphoton processes.

This calls in fact for the solution of a problem more general than that considered in Refs. 4 and 5, that of the existence of stationary pulses in a resonant medium with a susceptibility that is a more complicated function of the radiation amplitude than in the case of TPA or SRS. We shall show below that this problem can be solved. Its solution yields the waveform of the stationary pulse when simultaneous account is taken of OPA and of multiphoton processes of the hindrance-lifting type, and, as a particular case, the waveform of the stationary pulse in all the heretofore investigated processes—OPA, TPA, and SRS. We note that on the basis of the asymptotic solution for the front of a stationary USP in three-photon absorption, and also with the aid of an analog of the area theorem and the Manley-Rowe relations, it was concluded earlier that stationary pulses cannot exist in ordinary multiphoton processes of order higher than the second.[8,9] The procedure proposed here makes possible an analytic solution of this problem. It will be shown that a distinguishing feature of multiphoton processes of the hindrance-lifting type is the possible existence of stationary pulses when OPA of higher orders play a decisive role, if the dipole moment that causes the one-photon absorption of first order is arbitrarily small but different from zero.

1. BASIC EQUATIONS

We consider the propagation of pulsed laser radiation

$$E(z,t)=\sum_j \mathscr{E}_j(z,t)\exp[i(\omega_j t-k_j t)]$$

in a molecular absorbing medium. If the field E has a frequency $\omega_1=\omega_{21}$ that is at resonance with a certain transition 1–2, the resonance conditions for a multiphoton process of the hindrance-lifting type[6,7] are of the form

$$2\omega_1-\omega_1=\omega_{21},\quad 3\omega_1-2\omega_1=\omega_{21}, \tag{1}$$

etc. The conditions (1) mean that in a 1–2 transition there can take place, besides first-order OPA, also OPA of third, fifth, etc. order.[6]

For ordinary multiphoton resonance we have

$$\sum_j n_j\omega_j=\omega_{21}, \tag{2}$$

where n_j are positive integers that indicate the multiplicity of the degeneracy of the resonance with respect to the frequency ω_j, and the sum $\sum_j n_j=q$ yields the order of the resonance.

Assume that the stationary amplitude profiles $\mathscr{E}_j(z,t)$

$=\mathscr{E}_j(\tau=t-z/v)$, propagating with velocity $v<u_j$ (u_j is the speed of light in the given medium) are established at a certain distance in a one-dimensional absorbing medium. The equations that describe the stationary USP, neglecting relaxation processes and the Stark effect (the role of the Stark effect is considered in Sec. 4) take the following form. For a multiphoton process of the hindrance-lifting type (1)

$$d\mathscr{E}/d\tau=i\gamma_1^{(1)}\chi(|\mathscr{E}|)\sigma_{12}, \tag{3}$$
$$d\sigma_{12}/d\tau=-i\gamma_2\chi(|\mathscr{E}|)\eta_{12}\mathscr{E}, \tag{4}$$
$$d\eta_{12}/d\tau=\gamma_3\chi(|\mathscr{E}|)\,\mathrm{Im}\,(\sigma_{12}\mathscr{E}^*). \tag{5}$$

Here $\mathscr{E}$ is the field at the resonance frequency $\omega=\omega_{21}$, and $\eta_{12}=\sigma_{11}-\sigma_{22}$ is the difference between the populations of the working levels; σ_{11} and σ_{22} are the diagonal elements and σ_{12} is the off-diagonal element of the density matrix;

$$\gamma_1^{(1)}=2\pi\omega_1N_0/u_1\nu_1,\quad \gamma_2=1/\hbar,\quad \gamma_3=4/\hbar,$$

N_0 is the particle-number density, and $\nu_1=1/v-1/u_1$ is the velocity mismatch. Since resonances of third, fifth, etc. orders take place in the process (1) under consideration besides one-photon absorption, we have

$$\chi(|\mathscr{E}|)=d_{12}+\sum_{m=1}\varkappa^{(2m+1)}|\mathscr{E}|^{2m},$$

where d_{12} is the dipole moment and $\varkappa^{(2m+1)}$ are polarizabilities of order $q=2m+1$.

From (3) and (5) follows the first integral, which establishes the connection between the stationary amplitude and the population difference. Regardless of the order of the resonance we have

$$\eta_{12}(|\mathscr{E}|)=\eta_{12}^0-\frac{\gamma_3}{2\gamma_1^{(1)}}|\mathscr{E}|^2, \tag{6}$$

where η_{12}^0 is the initial population difference. In addition, if we introduce the quantity

$$S=2\hbar^{-1}\int_{-\infty}^{\tau}\chi(|\mathscr{E}|)|\mathscr{E}|d\tau,$$

it follows from Eqs. (4) and (5), in the absence of phase modulation, that $\eta_{12}=\eta_{12}^0\cos S$. Taking this into account, it is easy to find from (6) that the increment of S during the time of the pulse is $2\pi n$. Thus, if a stationary pulse is produced in the process under consideration, the quantity $S(\infty)$ is finite and is a multiple of 2π, regardless of the order of the resonance.

The change of variable

$$\zeta=\int_{-\infty}^{\tau}\chi(|\mathscr{E}|)d\tau$$

reduces the system (3)–(5) to a single second-order equation for the field $\mathscr{E}$:

$$d^2\mathscr{E}/d\zeta^2-\gamma_1^{(1)}\gamma_2\eta_{12}(|\mathscr{E}|)\mathscr{E}=0, \tag{7}$$

where $\eta_{12}(|\mathscr{E}|)$ is the known function (6). This equation will be investigated below, and at present we shall show that stationary USP are described by an equation similar to (7) also for the usual q-photon nondegenerate resonance (2).

The system of equations for the q-photon resonance (2) is of the form

$$\frac{d\mathscr{E}_j}{d\tau}=i\gamma_1^{(j)}\frac{\varkappa^{(q)}}{\mathscr{E}_j^*}\Pi_q^*\sigma_{12},\quad j=1,2,\ldots q, \tag{8}$$
$$d\sigma_{12}/d\tau=-i\gamma_2\eta_{12}\varkappa^{(q)}\Pi_q, \tag{9}$$
$$d\eta_{12}/d\tau=\gamma_3\varkappa^{(q)}\,\mathrm{Im}\,(\sigma_{12}\Pi_q^*), \tag{10}$$

where $\Pi_q=\mathscr{E}_1\mathscr{E}_2\ldots\mathscr{E}_q$. It is easy to show that the q equations for the amplitudes $\mathscr{E}_j$ in this system can be replaced by a single equation for the quantity Π_q, which coincides in form with Eq. (3) for the multiphoton resonance (1) of the hindrance-lifting type. Indeed, it follows from (8) that the amplitude product Π_q satisfies the equation

$$\frac{d\Pi_q}{d\tau}=i\varkappa^{(q)}|\Pi_q|^2\sigma_{12}\sum_{j=1}^{q}\frac{\gamma_1^{(j)}}{|\mathscr{E}_j|^2}. \tag{11}$$

The Manley-Rowe relations for Eqs. (8) yield the connection between the stationary amplitude profiles $|\mathscr{E}_k|^2=(\gamma_1^{(k)}/\gamma_1^{(j)})|\mathscr{E}_j|^2$. Therefore any amplitude $|\mathscr{E}_j|$ can be expressed in terms of the modulus of the product $|\Pi_q|$:

$$|\mathscr{E}_j|^2=(\gamma_1^{(j)}/\gamma_1)|\Pi_q|^{2/q}, \tag{12}$$

where $\gamma_1=[\gamma_1^{(1)}\gamma_1^{(2)}\cdots\gamma_1^{(q)}]^{1/q}$. Substituting (12) in (11) we obtain for Π_q an equation similar to (3):

$$d\Pi_q/d\tau=i\gamma_1\chi(|\Pi_q|)\sigma_{12}, \tag{13}$$

where

$$\chi(|\Pi_q|)=q\varkappa^{(q)}|\Pi_q|^{2(q-1)/q}.$$

Just as in the preceding case, the change of variable

$$\zeta=\int_{-\infty}^{\tau}\chi(|\Pi_q|)d\tau$$

reduces the system (13), (9), and (10) to a single second-order equation for Π_q:

$$\frac{d^2\Pi_q}{d\zeta^2}-\gamma_1\gamma_2\eta_{12}(|\Pi_q|)\frac{\varkappa^{(q)}}{\chi(|\Pi_q|)}\Pi_q=0, \tag{14}$$

where $\eta_{12}(|\Pi_q|)=\eta_{12}^0-(\gamma_3/2\gamma_1)|\Pi_q|^{2/q}$ is the first integral of the system (8)–(10). We note that in the case of q-photon resonance the quantity

$$S(\infty)=2\varkappa^{(q)}\hbar^{-1}\int_{-\infty}^{\infty}|\Pi_q|d\tau$$

should also be a multiple of 2π for stationary pulses.

Equation (7) for the field, in the case of a resonance of the hindrance-lifting type (1), and Eq. (14) for the product of the fields in q-photon resonance (2), are of the same type and are solved by the same method.

In the absence of phase modulation, when the initial conditions $\mathscr{E}(\tau\to-\infty)$, and $\sigma_{12}(\tau\to-\infty)=0$ are satisfied, the solution takes the form

$$\tau=\pm\int_{F_0}^{F}\left[\chi(F)\left(2\gamma_1\gamma_2\int_0^F\theta(F)FdF\right)^{1/2}\right]^{-1}dF, \tag{15}$$

where

$$F=\mathscr{E},\quad \theta(F)=\eta_{12}(\mathscr{E}),\quad \gamma_1=\gamma_1^{(1)}$$

for the resonance of the hindrance-lifting type (1);

$$F=\Pi_q,\quad \theta(F)=[\varkappa^{(q)}/\chi(\Pi_q)]\eta_{12}(\Pi_q),\quad \gamma_1=[\gamma_1^{(1)}\gamma_1^{(2)}\ldots\gamma_1^{(q)}]^{1/q}$$

for ordinary q-photon resonance (2). The solution (15) describes pulses of symmetrical shape with a vertex at $\tau=0$ and with a maximum value $F=F_0$. The plus sign corresponds to the rising part of the pulse ($\tau < 0$) and the minus sign to the decreasing part ($\tau > 0$). Let us now investigate the pulses (15) in specific cases.

2. STATIONARY PULSES IN q-PHOTON RESONANCE

Integration in (15) yields the waveforms of the stationary pulses in implicit form. For resonances of odd order $q = 2m-1, m = 1, 2, \ldots$ we have

$$\frac{\tau}{\tau_q} = \mp\left\{\frac{[1-\mathscr{E}_j^2/\mathscr{E}_{j0}^2]^{1/2}}{2m-1}\sum_{k=1}^{m-1}\frac{(2m-1)(2m-3)\ldots(2m-2k-1)}{2^k(m-1)(m-2)\ldots(m-k)} \times \frac{1}{(\mathscr{E}_j/\mathscr{E}_{j0})^{2(m-k)}} + \frac{(2m-3)!!}{2^m(m-1)!}\ln\frac{1+[1-\mathscr{E}_j^2/\mathscr{E}_{j0}^2]^{1/2}}{1-[1-\mathscr{E}_j^2/\mathscr{E}_{j0}^2]^{1/2}}\right\}, \qquad (16)$$

for resonances of even order $q = 2m, m = 1, 2, \ldots$

$$\frac{\tau}{\tau_q} = \mp\sum_{k=0}^{m-1}\frac{1}{2k+1}\binom{m-1}{k}\frac{[1-\mathscr{E}_j^2/\mathscr{E}_{j0}^2]^{(2k+1)/2}}{(\mathscr{E}_j/\mathscr{E}_{j0})^{2k+1}}, \qquad (17)$$

where

$$\mathscr{E}_{j0}^2 = 4\eta_{12}^0\gamma_1^{(j)}/\gamma_3, \quad \tau_q = \hbar/\varkappa^{(q)}\mathscr{E}_{10}\mathscr{E}_{20}\ldots\mathscr{E}_{q0}.$$

Equations (16) and (17) correspond in the particular cases of OPA and TPA to stationary amplitude profiles of USP in self-induced transparency[1-4]:

$$\mathscr{E}_1 = \mathscr{E}_{10}/\mathrm{ch}(\tau/\tau_1), \quad q=1, \qquad (18)$$

$$\mathscr{E}_{1,2} = \mathscr{E}_{10,20}/[1+\tau^2/\tau_2^2]^{1/2}, \quad q=2. \qquad (19)$$

We note that the solution (17) describes also stationary pulses in the case of SRS in an absorbing medium with anomalous dispersion.[5] By the parameter $\gamma_1^{(2)}$, which corresponds to a Stokes pulse, is meant in this case its modulus $|\gamma_1^{(2)}|$. The stationary amplitude profiles of the pump and of the Stokes component of the SRS are given by (19).

For q-photon resonance of order higher than $q = 2$, the expressions (16) and (17) contain terms $\sim 1/\mathscr{E}^{q-1}$. As $\tau \to \pm\infty$ the amplitudes $\mathscr{E}_j \to 0$, and these terms become dominant. The attenuation of the pulse fronts in accordance with the law $\mathscr{E}_1 \sim 1/\tau^{1/(q-1)}$ is too slow to ensure a finite energy. The divergence of the energy indicates that in principle no stationary USP are possible in q-photon resonance if $q > 2$. This conclusion agrees with the results of investigations of the asymptotic behavior of the USP (Refs. 8 and 9).

3. STATIONARY PULSES IN A MULTIPHOTON PROCESS OF THE HINDRANCE-LIFTING TYPE

We consider the resonant process (1) when, besides one-photon absorption resonances of third, fifth, etc. order also take place in the transition 1–2, i.e.,

$$\chi(|\mathscr{E}|) = d_{12} + \sum_{m=1}\varkappa^{(2m+1)}|\mathscr{E}|^{2m}.$$

We note immediately that if the dipole moment $d_{12} = 0$, the multiphoton process under consideration is described in the absence of phase modulation by the same equations (16) as the ordinary q-photon absorption (2).

It has shown in the preceding section that there are no stationary pulses at a resonance of order $q > 2$. We shall assume that $d_{12} \neq 0$ and investigate the role of dipole absorption in the multiphoton process (1). We confine ourselves to OPA of third order, i.e., we assume that

$$\chi(|\mathscr{E}|) = d_{12} + \varkappa^{(3)}|\mathscr{E}|^2.$$

Integrating in (15) for the indicated case, we obtain

$$\tau = \pm\frac{\hbar}{2d_{12}\mathscr{E}_0}\left\{\frac{1}{[1+d_{12}/\varkappa^{(3)}\mathscr{E}_0^2]^{1/2}} \times \ln\frac{[1+d_{12}/\varkappa^{(3)}\mathscr{E}_0^2]^{1/2}+[1-\mathscr{E}^2/\mathscr{E}_0^2]^{1/2}}{[1+d_{12}/\varkappa^{(3)}\mathscr{E}_0^2]^{1/2}-[1-\mathscr{E}^2/\mathscr{E}_0^2]^{1/2}} - \ln\frac{1+[1-\mathscr{E}^2/\mathscr{E}_0^2]^{1/2}}{1-[1-\mathscr{E}^2/\mathscr{E}_0^2]^{1/2}}\right\}. \qquad (20)$$

An investigation of (20) shows that $\mathscr{E}(\tau)$ is a stationary pulse of symmetrical shape with a peak at $\tau = 0$ and with a maximum value $\mathscr{E}_0 = [4\gamma_1^{(1)}\eta_{12}^0/\gamma_3]^{1/2}$. At $\mathscr{E}^2 \ll \mathscr{E}_0^2$ the influence of the nonlinear absorption becomes vanishingly small and the edges of the pulse fall off exponentially $\mathscr{E}(\tau) \sim e^{-|\tau|/\tau_d}$, $\tau_d = \hbar/d_{12}\mathscr{E}_0$, thereby ensuring a finite pulse energy.

Thus, the presence of dipole absorption in the transition 1–2 leads to formation of stationary USP, despite the presence of third-order OPA.

At small nonlinearity, $\varepsilon = \varkappa^{(3)}\mathscr{E}_0^2/d_{12} \ll 1$, Eq. (20) leads to an explicit expression for the wave form of the stationary pulse:

$$\mathscr{E}(\tau) = \mathscr{E}_0(1-\varepsilon\,\mathrm{th}^2(\tau/\tau_d))/\mathrm{ch}(\tau/\tau_d). \qquad (21)$$

Figure 1 shows the wave form of the pulse for $d_{12} = 10^{-20}$ cgs esu, $\varkappa^{(3)} = 10^{-26}$ cgs esu, $\mathscr{E}_0 = 3\cdot 10^3$ cgs esu. We note that in a molecular medium with a dipole moment $d_{12} = 10^{-20}$ cgs esu, which is typical of laser transitions, satisfaction of the condition $S(\infty) \geq \pi$ for an input pulse of picosecond duration, in ordinary one-photon resonance, calls for a peak power $\sim 10^{12}$ W/cm^2. The presence of third-order polarizability makes it possible to observe stationary pulses on laser transitions at a power smaller by four orders of magnitude. Estimates show that the power needed to form a stationary pulse with a duration $\sim 10^{-12}$ sec on account of third-order OPA at $\varkappa^{(3)} = 10^{-26}$ cgs esu becomes equal to the corresponding power for ordinary one-photon absorption when $d_{12} = 5\cdot 10^{-10}$ cgs esu.

It must be emphasized that a solution in the form of a solitary stationary pulse (15) was obtained at zero initial conditions. If $\sigma_{12} = \sigma_0 \neq 0$ as $\tau \to \pm\infty$, then one cannot exclude the possibility of a periodic solution similar to the soliton solutions in Ref. 5. Thus, in the case of hindrance lifting we have from (15) $\tau = \pm\pi(h, \varphi, k)$, where $\pi(h, \varphi, k)$ is an elliptic integral of the third kind,

$$h = \varkappa\mathscr{E}_1/(d+\varkappa\mathscr{E}_1^2), \quad k = \mathscr{E}_1/(\mathscr{E}_1^2+|\mathscr{E}_2|^2)^{1/2}, \quad \varphi = \arcsin(1-\mathscr{E}^2/\mathscr{E}_1^2)^{1/2},$$

$$\mathscr{E}_{1,2} = 2\gamma_1\eta_{12}^0/\gamma_3 \pm [(2\gamma_1\eta_{12}^0/\gamma_3)^2 + (\sigma_0 2\pi\omega/uv)^2]^{1/2}.$$

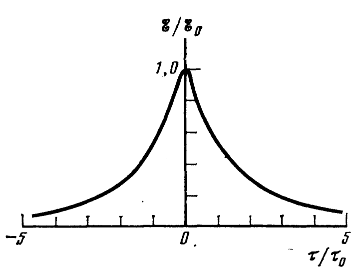

FIG. 1. Waveform of stationary pulse in third-order OPA, $\tau_0 = 8\cdot 10^{-12}$ sec.

In particular, if $h=k$, then the amplitude $\mathscr{E}(\tau)$ is determined from the relation

$$(1-\mathscr{E}^2/\mathscr{E}_1^2)^{1/2}=\mathrm{sn}\left\{2\tau-\frac{1}{1-k}\mathrm{arctg}\left[1-k\,\mathrm{tg}\frac{\arcsin(1-\mathscr{E}^2/\mathscr{E}_1^2)^{1/2}}{[1-k^2(1-\mathscr{E}^2/\mathscr{E}_1^2)]^{1/2}}\right]\right\},$$

in which $\mathrm{sn}(z)$ is an elliptic function. A more detailed investigation of the properties of the solution, such as determination of the period etc., calls for numerical calculations.

4. ALLOWANCE FOR THE STARK EFFECT

Stationary pulses in multiphoton interactions were considered in the preceding section without allowance for the Stark effect. In strong fields, and in the presence of linear polarizabilities of the resonance equations, it is necessary to take into account the additional nonlinearity due to the Stark shift of the levels. The problem of finding the waveform of the stationary pulse in this case is much more complicated, but the nonlinear equations can be analytically solved with the aid of the Hamilton-Jacobi method.[10]

We consider a multiphoton process of the hindrance-lifting type (1), in which

$$\chi(|\mathscr{E}|)=d_{12}+\sum_m \varkappa^{(2m+1)}|\mathscr{E}|^{2m},$$

and take into account the Stark level shift $\Omega(|\mathscr{E}|)=\Omega_0|\mathscr{E}|^2$. The complex amplitude $\mathscr{E}(\tau)$ of the stationary pulse is described by the system of equations

$$d\mathscr{E}/d\tau=i\gamma_1\chi(|\mathscr{E}|)\sigma_{12},\tag{22}$$

$$d\sigma_{12}/d\tau-i\Omega(|\mathscr{E}|)\sigma_{12}=-i\gamma_2\chi(|\mathscr{E}|)\eta_{12}(|\mathscr{E}|)\mathscr{E},$$

where

$$\eta_{12}(|\mathscr{E}|)=\eta_{12}^0-(\gamma_3/2\gamma_1)|\mathscr{E}|^2.$$

The change of variable

$$\zeta=\int_{-\infty}^{\tau}\chi(|\mathscr{E}|)\,d\tau$$

reduces the system (22) to a single equation for the amplitude $\mathscr{E}$:

$$\frac{d^2\mathscr{E}}{d\zeta^2}-\frac{i\Omega(|\mathscr{E}|)}{\chi(|\mathscr{E}|)}-\gamma_1\gamma_2\eta_{12}(|\mathscr{E}|)\mathscr{E}=0.\tag{23}$$

We separate the real and imaginary parts of the complex amplitude $\mathscr{E}=x+iy$. Then the system of equations

$$\frac{d^2x}{d\zeta^2}+\frac{\Omega}{\chi}\frac{dy}{d\zeta}=\gamma_1\gamma_2\eta_{12}x,\quad \frac{dy}{d\zeta^2}-\frac{\Omega}{\chi}\frac{dx}{d\zeta}=\gamma_1\gamma_2\eta_{12}y,\tag{24}$$

obtained from (23) coincides formally with the system describing the motion of a particle with unity mass and unity charge in an electric field and a magnetic field which are constant but nonuniform. Using this formal analogy, we solve the system (24) by the Hamilton-Jacobi method[10] developed for the solution of problems in classical mechanics. This method was applied to the propagation of nonlinear wave in Refs. 11 and 12.

The Hamilton-Jacobi equation for the action function S takes in cylindrical coordinates of the form

$$\frac{dS}{d\zeta}+\frac{1}{2}\left(\frac{\partial S}{\partial r}\right)^2+\frac{1}{2}\left(\frac{1}{r}\frac{\partial S}{\partial\varphi}-A\right)^2+V=0,\tag{25}$$

in our case

$$V=-\gamma_1\gamma_2\frac{r^2}{2}\left(\eta_{12}^0-\frac{\gamma_3}{4\gamma_1}\cdot r^2\right),\quad A=-\frac{1}{r}\int_0^r\frac{r\Omega(r^2)}{\chi(r^2)}dr,\quad r^2=x^2+y^2.$$

Separating the variables $S=-\lambda_1\zeta+\lambda_2\varphi+S_0(r)$, we obtain

$$S_0(r)=\pm\int\left[2\lambda_1-2V-\left(\frac{\lambda_2}{r}-A\right)^2\right]^{1/2}dr;\quad \lambda_{1,2}=\mathrm{const.}\tag{26}$$

Using the fact that in the Hamilton-Jacobi method

$$\frac{dS}{\partial\lambda_1}=-\zeta+\frac{\partial S_0}{\partial\lambda_1}=C_1=\mathrm{const},\quad \frac{\partial S}{\partial\lambda_2}=\varphi+\frac{\partial S_0}{\partial\lambda_1}=C_2=\mathrm{const},\tag{27}$$

we obtain the solution in the form

$$C_1+\zeta=\pm\int\frac{dr}{[2\lambda_1-2V-(\lambda_2/r-A)^2]^{1/2}},\tag{28}$$

$$\varphi-C_2=\mp\int\frac{(A-\lambda_2/r)\,dr}{r[2\lambda_1-2V-(\lambda_2/r-A)^2]^{1/2}}.\tag{29}$$

Equation (28) yields implicitly the dependence of the field amplitude r on the temporal variable ζ, while Eq. (29) establishes the connection between the phase φ and the amplitude r. We change over in (28) to a running time τ, using the fact that $d\zeta/d\tau=\chi(|\mathscr{E}|)$:

$$\tau=\pm\int_{|\mathscr{E}_0|}^{|\mathscr{E}|}\left\{\chi(r^2)\left[2\lambda_1-2V-\left(\frac{\lambda_2}{r}-A\right)^2\right]^{1/2}\right\}^{-1}dr.\tag{30}$$

Here $\mathscr{E}_0$ is the maximum value of the amplitude $r=(x^2+y^2)^{1/2}=|\mathscr{E}|$. If the dipole moment in the considered process is $d_{12}\neq0$, then the function $A(r=0)=0$. The initial conditions $\mathscr{E}(\tau\to-\infty)=0$ and $\sigma_{12}(\tau\to-\infty)=0$ are satisfied if the constants $\lambda_1=\lambda_2=0$. We present the form of the solution for the particular case of the resonance (1) for third-order OPA, i.e., for $\chi(r^2)=d_{12}+\varkappa^{(3)}r^2$:

$$\tau=\pm\int_{\mathscr{E}_0}^{|\mathscr{E}|}dr(d_{12}+\varkappa^{(3)}r^2)^{-1}\left\{\gamma_1\gamma_2r^2\left(\eta_{12}^0-\frac{\gamma_3}{4\gamma_1}r^2\right)-\frac{\Omega_0^2}{4r^2}\left[\frac{r^2}{\varkappa^{(3)}}-\frac{d_{12}}{(\varkappa^{(3)})^2}\ln\left(1+\frac{\varkappa^{(3)}r^2}{d_{12}}\right)\right]^2\right\}^{-1/2}.\tag{31}$$

We investigate the influence of the Stark effect on the stationary wave form of the pulse in the case of small nonlinearity $\varkappa^{(3)}\mathscr{E}_0^2/d_{12}\ll1$. In this case it follows from (31) that the Stark shift of the levels decreases the maximum amplitude of the stationary pulse:

$$\mathscr{E}_0^2=\frac{4\gamma_1\eta_{12}^0}{\gamma_3}-\frac{\Omega_0^2}{2d_{12}\gamma_2\gamma_3}.\tag{32}$$

The pulse waveform remains the same and is described by expression (21), which was obtained with the Stark effect neglected. It must be emphasized that the stationary pulse determined from (31) is meaningful only at $d_{12}\neq0$. If $d_{12}=0$, then just as in the absence of the Stark effect, pulses of infinite energy are obtained.

We note in conclusion that expressions (29) and (30) yield the wave form of a stationary USP for arbitrary multiphoton processes of the hindrance-lifting type with allowance for both Stark effect and phase modulation. A distinguishing feature of these processes is the dependence of the function χ on $|\mathscr{E}|^2$. For ordinary q-photon resonance we have $\chi=\chi[(\mathscr{E}^*)^{q-1}]$. In this case the procedure presented for solving equations with Stark nonlinearity is inapplicable.

In molecular media, multiphoton processes of the hindrance-lifting type can be used to shape much shorter pulses than in ordinary one-photon absorption, by using resonance with vibrational-rotational transitions $|\Delta j| > 1$, j is the rotational quantum number. The possible existence of stationary USP is ensured in this case by the presence of small dipole moment of the resonant transition, due, for example, to collisions.

[1]S. L. McCall and E. L Hahn, Phys. Rev. Lett. **18**, 908 (1967).
[2]S. L. McCall and E. L. Hahn, Phys. Rev. **183**, 457 (1969).
[3]I. A. Poluéktov, Yu. M. Popov, and V. S. Roĭtberg, Kvant. Élektron. (Moscow) **1**, 757 (1974) [Sov. J. Quantum Electron. **4**, 423 (1974)].
[4]I. A. Poluéktov, Yu. M. Popov, and V. S. Roĭtberg, Kvant. Élektron. (Moscow) **2**, 1147 (1975) [Sov. J. Quantum Electron. **5**, 620 (1975)].
[5]T. M. Makhviladze and M. E. Sarychev, Zh. Eksp. Teor. Fiz. **71**, 896 (1976) [Sov. Phys. JETP **44**, 471 (1976)].
[6]V. R. Block, G. M. Krochik, and Yu. G. Khronopulo, Zh. Eksp. Teor. Fiz. **76**, 46 (1979) [Sov. Phys. JETP **49**, 23 (1979)].
[7]G. M. Krochik and Yu. G. Khronopulo, Zh. Eksp. Teor. Fiz. **78**, 485 (1980) [Sov. Phys. JETP **51**, 245 (1980)].
[8]B. V. Thompson, Opt. Comm. **34**, 488 (1980).
[9]V. S. Butylkov, V. S. Grigor'yan, and M. E. Zhabotinskiĭ, Zh. Eksp. Teor. Fiz. **80**, 933 (1981) [Sov. Phys. JETP **53**, 475 (1981)].
[10]L. D. Landau and E. M. Lifshitz, Mechanika (Mechanics), Nauka, 1973, p. 184 [Pergamon, 1978].
[11]E. I. Yakubovich, Candidate's dissertation, Gor'kiĭ, 1971.
[12]E. I. Yakubovich, in: Nelineĭnye volny (Nonlinear Waves), A. V. Gaponov-Grekhov, ed., Nauka, 1981, p. 103.

Translated by J. G. Adashko