Effects of the nonlinear interaction of extremely short pulses in a dielectric paramagnet

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The propagation and parametric effects in the interaction of extremely short magnetic pulses (video pulses) with a dielectric paramagnet are analyzed. The paramagnet has a spin $s = 1/2$ and is in an external magnetic field H_0. In contrast with optical video pulses, steady-state, circularly polarized pulses may form in an inverted paramagnet under the condition $H_0 < H_0^*$, where H_0^* is determined by the parameters of the medium. If $H_0 > H_0^*$, the results predict amplification of the transverse component of the pulse as the longitudinal component reaches saturation. There may be a continuous parametric increase in the signal frequency.

1. INTRODUCTION

Interest has recently been drawn to the nonresonant interaction of extremely short electromagnetic pulses, consisting of about one oscillation period (video pulses), with matter.[1-7] This interest has been stimulated by progress in laser technology, which is now capable of generating femtosecond-range pulses.[8] Studies in this direction can significantly improve dynamic methods of nonresonant laser spectroscopy. The typical frequencies of the optical quantum transitions are $\omega_0 \approx 10^{15}\ \mathrm{s}^{-1}$. For rf-spectroscopy transitions, on the other hand, they are $\omega_0 \sim 10^9$–$10^{11}\ \mathrm{s}^{-1}$. These may be transitions between hyperfine levels or between Zeeman sublevels of paramagnetic atoms. By analogy with the spectroscopy of optical transitions, dynamic methods of nonresonant magnetic rf and IR spectroscopy may be developed. Since the values of ω_0 are small in the latter case (small in comparison with the optical values), it is sufficient to use video pulses with a length τ_p on the order of a picosecond. Infrared pulses in "one oscillation period" of length $\tau_p \sim 10^{-12}$–10^{-13} s have recently been generated under experimental conditions.[9]

Several theoretical studies[10-12] have also been carried out. They have dealt with the nonlinear propagation of electromagnetic pulses in a paramagnet. In particular, steady-state video pulses traveling at a constant velocity at a certain angle with respect to the external magnetic field $\mathbf{H}_0$ were studied in Refs. 10 and 11. Circularly polarized pulses propagating along $\mathbf{H}_0$ and satisfying the condition $\omega_0 \tau \gg 1$ were studied in Ref. 12. It was shown in that paper that these pulses can be described by a "differential nonlinear Schrödinger equation." It was shown in Ref. 13 that the latter equation can be integrated by the inverse-scattering method; an N-soliton solution of this equation was derived.[1)]

Let us consider the interaction of an electromagnetic video pulse with a dielectric paramagnet. We assume that the paramagnetism stems from the spin of an electron in the atomic S state. An external magnetic field causes Zeeman splitting of the state into two sublevels with a frequency separation $\omega_0 = g_\parallel \beta_0 H_0/\hbar$ ($g_\parallel$ is a component of the Landé tensor, β_0 is the Bohr magneton, and $\hbar$ is Planck's constant). The magnetic field $\mathbf{H}$ of the pulse propagating through such a medium can then cause magnetic-dipole transitions between the given sublevels. If the magnetic-dipole transitions are to effectively influence the nature of the propagation of the video pulse, there must be an appreciable difference between the populations of the corresponding sublevels. Such a difference is possible at temperatures $T < \hbar\omega_0/k_B$ (k_B is the Boltzmann constant). Taking $\omega_0 \sim 10^{11}\ \mathrm{s}^{-1}$, we find $T < 1$ K. The two-level approximation is legitimate for a paramagnetic medium if[14] $(d_j E/\hbar\omega_j)^2 \ll 1$, where d_j is the electric moment of a transition from one of the sublevels of interest to the nearest of the remote quantum levels, $\hbar\omega_j$ is the corresponding distance along the energy scale, and E is the electric field of the video pulse.

2. BASIC EQUATIONS

We assume that the paramagnet is a uniaxial crystal. An external magnetic field is directed along the cylindrical-symmetry axis of the crystal (the z axis). The Hamiltonian of this paramagnet, incorporating the interaction with the video pulse, is

$$\hat{H} = -\hbar(\omega_0 + \Omega_\parallel)\hat{S}_z - \hbar\Omega_x\hat{S}_x - \hbar\Omega_y\hat{S}_y . \tag{1}$$

Here $\hat{S}_x, \hat{S}_y, \hat{S}_z$ are electron spin operators; $\Omega_\parallel = g_\parallel\beta_0 H_z/\hbar$; $\Omega_{x,y} = g_\perp\beta_0 H_{x,y}/\hbar$; and H_x, H_y, H_z are components of the magnetic field of the video pulse. In the Heisenberg picture, we easily find from (1) the equations of motion for the electron spin:

$$\frac{\partial U}{\partial t} = (\omega_0 + \Omega_\parallel)V - \Omega_y W , \tag{2}$$

$$\frac{\partial V}{\partial t} = -(\omega_0 + \Omega_\parallel)U + \Omega_x W , \tag{3}$$

$$\frac{\partial W}{\partial t} = \Omega_y U - \Omega_x V , \tag{4}$$

where $U = \langle\hat{S}_x\rangle$, $V = \langle\hat{S}_y\rangle$, $W = \langle\hat{S}_z\rangle$, and $\langle\ldots\rangle$ means the operation of taking the expectation value.

We supplement Eqs. (2)–(4) with Maxwell's equations:

$$\boldsymbol{\Omega} - \mathrm{grad}\,\mathrm{div}\,\boldsymbol{\Omega} - \frac{1}{c^2}\frac{\partial^2\boldsymbol{\Omega}}{\partial t^2} = \frac{4\pi\beta_0^2}{\hbar c^2}\hat{g}n\frac{\partial^2}{\partial t^2}\langle\mathbf{S}\rangle\hat{g} . \tag{5}$$

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Here n is the concentration of paramagnetic atoms, and c is the velocity of light. We assume that the video pulse is propagating along the external magnetic field, which is directed along the z axis. The z component of Eq. (5) can then be integrated easily:

$$\Omega_{\parallel} = -\frac{4\pi^2\beta_0^2}{\hbar} g_{\parallel}^2 n(W - W_{\infty}), \tag{6}$$

where W_{∞} is the initial spin inversion $[(W_{\infty} = W(t = -\infty)]$. In integrating we note that in the absence of a video pulse the spins are oriented parallel to $(W_{\infty} > 0)$ or antiparallel to $(W_{\infty} < 0)$ $\mathbf{H}_0$.

The system (2)–(4) has the integral of motion $U^2 + V^2 + W^2$. Using the initial conditions at $t = -\infty$ $(U = V = 0, W = W_{\infty})$, we can transform to coordinates on a sphere $U^2 + V^2 + W^2 = |W_{\infty}|^2$:

$$U = W_{\infty}\cos\psi\sin\vartheta, \quad V = W_{\infty}\sin\psi\sin\vartheta, \quad W = W_{\infty}\cos\vartheta. \tag{7}$$

We also assume $\Omega_x = \Omega_{\perp}\cos\varphi$, $\Omega_y = \Omega_{\perp}\sin\varphi$. In this notation, Eqs. (2)–(4) become

$$\begin{aligned} \partial\vartheta/\partial t &= \Omega_{\perp}\sin(\psi - \varphi), \\ \partial\psi/\partial t &= -(\omega_0 + \Omega_{\parallel}) + \Omega_{\perp}\cos(\psi - \varphi)\operatorname{ctg}\vartheta. \end{aligned} \tag{8}$$

Alternatively, introducing the variable $\chi = \psi - \varphi$, we can write

$$\begin{aligned} \partial\vartheta/\partial t &= \Omega_{\perp}\sin\chi, \\ \partial\chi/\partial t &= \partial\varphi/\partial t - \omega_0 - \Omega_{\parallel} + \Omega_{\perp}\cos\chi\operatorname{ctg}\vartheta. \end{aligned} \tag{9}$$

We further assume

$$|\Omega_{\perp}| \gg |\partial\varphi/\partial t|, |\omega_0|, |\Omega_{\parallel}|. \tag{10}$$

Inequalities (10) constitute a generalization of the strong-field approximation[1-3,7] to the case of circular polarization. The condition $|\Omega_{\perp}| \gg |\Omega_{\parallel}|$ holds well for pulses which are travelling at velocities close to the velocity of light. Under these conditions, Eqs. (9) have a solution

$$\chi = \frac{\pi}{2}, \quad \vartheta = \int_{-\infty}^{t} \Omega_{\perp}(\varphi, \xi)d\xi, \tag{11}$$

which can be put in the form

$$U = -W_{\infty}\sin\varphi\sin\vartheta, \tag{12}$$

$$V = W_{\infty}\cos\varphi\sin\vartheta, \tag{13}$$

$$W = W_{\infty}\cos\vartheta. \tag{14}$$

If $\Omega_y = \Omega_{\parallel} = 0$, the solution in (12)–(14) is the same as the corresponding solutions derived in Refs. 1–3 and 7, by a different method, for the case of linear polarization.

Substituting (12) into (13) into the right sides of the (2) and (3), and using (6), we find

$$\begin{aligned} \frac{\partial S_{\perp}}{\partial t} &= \frac{\partial}{\partial t}(U + iV) \\ &= W_{\infty}\exp(i\varphi)[\tilde{\omega}_0\sin\vartheta - i\Omega_{\perp}\cos\vartheta - \frac{1}{2}\omega_0\lambda_{\parallel}\sin 2\vartheta], \end{aligned} \tag{15}$$

where

$$\tilde{\omega}_0 = \omega_0(1 + \lambda_{\parallel}), \quad \lambda_{\parallel(\perp)} = 4\pi\beta_0 g_{\parallel(\perp)} n W_{\infty}/h\omega_0.$$

From (5) and (15) we then find

$$\begin{aligned} \left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\Omega &= \omega_0\lambda_{\perp}W_{\infty}c^{-2}\frac{\partial}{\partial t}[e^{i\varphi}(\tilde{\omega}_0\sin\vartheta \\ &\quad - i\Omega_{\perp}\cos\vartheta - \frac{1}{2}\lambda_{\parallel}\omega_0\sin 2\vartheta)], \end{aligned} \tag{16}$$

where $\Omega = \Omega_x + i\Omega_y$.

Let us assume that the velocity of the pulse is close to the velocity of light, c. We can then transform from Eq. (16) to an equation which describes the motion of a pulse in one direction:

$$\frac{\partial\Omega}{\partial z} = -\frac{\lambda_{\perp}\omega_0 W_{\infty}}{2c} e^{i\varphi}(\tilde{\omega}_0\sin\vartheta - \frac{1}{2}\lambda_{\parallel}\omega_0\sin 2\vartheta - i\Omega\cos\vartheta), \tag{17}$$

$$\frac{\partial\vartheta}{\partial\tau} = \Omega_{\perp}, \tag{18}$$

where $\tau = t - z/c$.
Separating the imaginary and real parts, we find

$$\frac{\partial^2\vartheta}{\partial z\partial\tau} = -\frac{\lambda_{\perp}\omega_0 W_{\infty}}{2c}(\tilde{\omega}_0\sin\vartheta - \frac{1}{2}\lambda_{\parallel}\omega_0\sin 2\vartheta), \tag{19}$$

$$\frac{\partial\varphi}{\partial z} = \frac{\lambda_{\perp}\omega_0 W_{\infty}}{2c}\cos\vartheta. \tag{20}$$

3. STEADY-STATE, CIRCULARLY POLARIZED PULSES

The magnitude of the transverse component of the pulse field and the phase of its rotation can thus be expressed in terms of the solution of the double sine-Gordon equation. According to (6), the longitudinal component of the pulse field is given by

$$\Omega_{\parallel} = 2\omega_0\lambda_{\parallel}\sin^2\frac{\vartheta}{2}. \tag{21}$$

It is because of the longitudinal component of the vector Ω that a second term arises on the right side of Eq. (19). We know that solutions of the double sine-Gordon equation do not have soliton properties.[15] If the external magnetic field is so strong that the condition $\omega_0 \gg 4\pi\beta_0^2 g_{\parallel}^2 n|W_{\infty}|/\hbar$ holds, we can ignore the second term in (19). The propagating video pulses are then solitons of the sine-Gordon equation. The law describing the rotation of their polarization plane is given by (20).

For equation (19) we can write the "Hamiltonian density"

$$\mathcal{H} = \frac{1}{2}\frac{\partial\vartheta}{\partial z}\frac{\partial\vartheta}{\partial\tau} + \frac{\lambda_{\perp}\omega_0 W_{\infty}}{c}(\tilde{\omega}_0\sin^2\frac{\vartheta}{2} - \frac{1}{2}\omega_0\lambda_{\parallel}\sin^2\vartheta). \tag{22}$$

Following Refs. 16 and 17, we define the "vacuum state" as the state with a constant $\vartheta_{\rm vac}$ which minimizes $\mathscr{H}$. Corresponding studies show that under the condition $W_\infty > 0$ we have $\vartheta_{\rm vac} = 2\pi n$ ($n = 0,1,2,...$). A transition between the vacuum states with $n = 0$ and $n = 1$ corresponds to a solution in the form of a solitary pulse which is traveling parallel to $\mathbf{H}_0$ at a velocity $\mathbf{v}$ and which is rotating in the transverse plane (we call this a "stationary" pulse):

$$\Omega_\perp = \omega_0 \left(\frac{2\lambda_\perp}{1+\lambda_\parallel} \frac{v}{c-v} \right)^{1/2} \frac{\operatorname{sech}\zeta}{1+q^2\operatorname{sech}^2\zeta}, \tag{23}$$

$$W = W_\infty \left(1 - \frac{2}{1+(1+\lambda_\parallel)\operatorname{sh}^2\zeta} \right), \tag{24}$$

$$\varphi = f(z-ct) + \frac{\omega_0\lambda_\perp}{2c} z - \frac{g_\perp}{g_\parallel} \left(\frac{2v}{c-v} \right)^{1/2} \operatorname{arctg}(\lambda_\parallel^{1/2} \operatorname{th}\zeta). \tag{25}$$

Here

$$\zeta = (z-vt)/\Delta, \quad \Delta = \frac{1}{\omega_0} \left(\frac{2v(c-v)}{\lambda_\perp} \right)^{1/2}, \quad q^2 = \frac{\lambda_\parallel}{\lambda_\parallel + 1},$$

and $f(z-ct)$ is an arbitrary function determined by the initial (or boundary) conditions. If $f(z-ct) = 0$, inequality (10) is violated. Assuming $v \lesssim c$, we write

$$f(z-ct) \approx f(z-vt) \approx -(1-v/c) z f'(z-vt).$$

We assume that $f(z-vt)$ is equal to the third term in (25). We then have

$$\varphi \approx \frac{\omega_0\lambda_\perp}{2c} z \left(1 - \frac{2}{1+(1+\lambda_\parallel)\operatorname{sh}^2\zeta} \right). \tag{26}$$

Since we still have $v \neq c$, at a certain distance z_k inequality (10) becomes violated after the pulse in (25), (26) is formed, and the solutions found above are no longer valid. This distance can be estimated easily from the condition for the convergence of the series obtained by expanding f in powers of $(1-v/c)z/\Delta$. We thus have

$$z_k \approx \Delta(1-v/c)^{-1}. \tag{27}$$

Since $v \lesssim c$, this distance can be substantial in units of Δ.

The case considered here corresponds to, for example, a paramagnet at thermodynamic equilibrium:[2)]

$$W_\infty = \operatorname{th}(\hbar\omega_0/2k_BT). \tag{28}$$

The pulse corresponding to (23) is shown in Fig. 1. It follows from (24) that the electron spins "undergo a complete rotation" through an angle of 2π over the time taken for the pulse to pass.

If the initial state of the paramagnet is not an equilibrium state ($W_\infty < 0$), on the other hand, and if the condition

$$\lambda_\parallel > 1, \tag{29}$$

also holds, then there exists a $\vartheta^*_{\rm vac}$, which is found from

$$\sin^2(\vartheta^*_{vac}/2) = 1/2\lambda_\parallel. \tag{30}$$

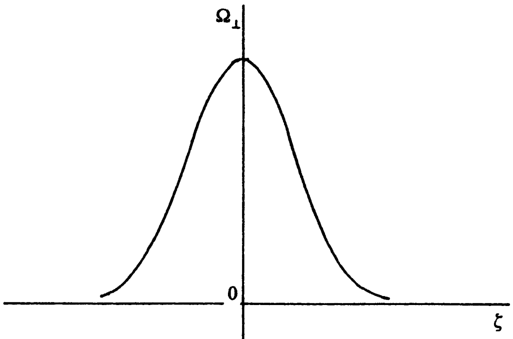

FIG. 1. Profile of the magnetic field of the pulse in the case of thermodynamic equilibrium.

The quantity $\vartheta^*_{\rm vac}$ corresponds to a cancellation of the static Zeeman splitting ω_0 by a dynamic Zeeman effect resulting from the longitudinal component of the magnetic field of the pulse [see (2), (3), (21)]. Thus the resultant Zeeman splitting vanishes at $\vartheta = \vartheta^*_{\rm vac}$. Equation (30) has a mechanical analogy in a pendulum whose suspension point is vibrating at a frequency ν.

Here we have[18] $\lambda_\parallel \sim \nu^2$. In this case the pendulum acquires a stable equilibrium position, which corresponds to $\vartheta^*_{\rm vac}$ in (30). The role of the source which causes the suspension point to vibrate is played by the longitudinal component of the pulse field.

The corresponding solitary pulse is described by the following expressions, where we are using $v \lesssim c$:

$$\Omega_\perp = 2(\lambda_\parallel - 1)^{1/2} \frac{v}{\Delta_0} \frac{\operatorname{sh}\zeta'}{\lambda_\parallel + (\lambda_\parallel - 1)\operatorname{sh}^2\zeta'}, \tag{31}$$

$$W = -|W_\infty| \left(1 - \frac{2}{\lambda_\parallel + (\lambda_\parallel - 1)\operatorname{sh}^2\zeta'} \right), \tag{32}$$

$$\varphi = -\frac{\omega_0\lambda_\perp}{2c} z \left(1 - \frac{\lambda_\parallel}{\lambda_\parallel + (\lambda_\parallel - 1)\operatorname{sh}^2\zeta'} \right). \tag{33}$$

Here

$$\zeta' = (z-vt)/\Delta_0, \quad \Delta_0 = \frac{1}{\omega_0} \left(\frac{2v(c-v)}{\lambda_\perp} \right)^{1/2}.$$

The pulse in (31) is shown in Fig. 2. The pulse is bipolar in this case, in contrast with the preceding case, and its area is zero.

Solution (31)–(33) corresponds to a single vibration of the spin around its equilibrium position $\vartheta = \vartheta^*_{\rm vac}$. The maximum azimuthal angle ϑ on the Bloch sphere is determined by

$$\sin^2 \frac{\vartheta_{max}}{2} = \frac{1}{\lambda_\parallel}, \tag{34}$$

which corresponds to $\Omega_\parallel = -2\omega_0$ [see (21)]. The resultant Zeeman splitting is $\omega_0 + \Omega_\parallel = -\omega_0 < 0$. A dynamic inversion of Zeeman sublevels has occurred: The excited sublevel has become the ground sublevel, and the ground sublevel the excited one. It is this effect which is responsible

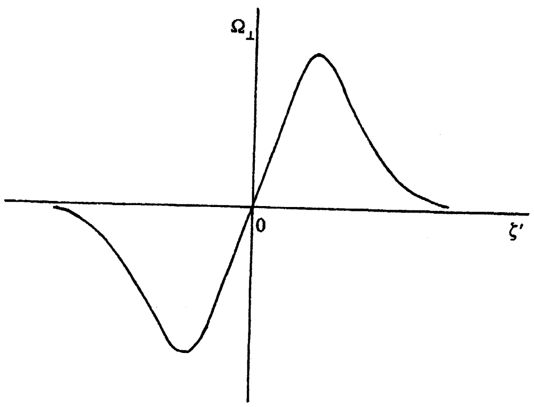

FIG. 2. Profile of the magnetic field of the pulse in the nonequilibrium case with $H_0 < H_k$.

for the stability of the pulse (31) as it propagates through an initially inverted system of spins. We mentioned above that if the longitudinal component of the pulse field is ignored we obtain the ordinary sine-Gordon equation. This equation does not allow the existence of stable stationary pulses in an inverted system ($W_\infty < 0$), while formal solutions in the form of superluminous pulses are unstable.

Condition (29) imposes a limitation on the external magnetic field H_0 which causes the static Zeeman splitting of the electronic S state. If we have $H_0 < H_k = 4\pi\beta_0 g_\parallel n|W_\infty|$, circularly polarized pulses of the type in (31), (33), (21) can propagate in a paramagnet. Otherwise, they cannot exist. We can thus say that a nonequilibrium phase transition occurs in the system as the control parameter H_0 is varied. Taking $|W_\infty| \sim 1$, $g_\parallel \approx 2$, and $n \sim 10^{17} - 10^{23}\ \mathrm{cm}^{-3}$, we find $H_k \sim 10^{-2}$–10^4 G. Such fields correspond to a Zeeman splitting $\omega_0 \sim 10^5$–$10^{11}\ \mathrm{s}^{-1}$. Comparing (31) with (21), we see the validity of the assertion that we have $|\Omega_\perp| \gg |\Omega_\parallel|$ for values of v close to the velocity of light. The latter inequality is important, since its use led to the approximate solutions of system (2)–(4).

4. AMPLIFICATION

We now assume that inequality (29) does not hold. In this case there can be no dynamic inversion of the Zeeman sublevels. There can thus be no formation of video pulses of the form (31), (33), which drive the paramagnet from its original excited state and then return it to that state. In this case, a pulse removes energy stored in the sample as it passes through the sample. As a result, the paramagnet goes to the ground state, amplifying the signal by virtue of an energy transfer to it. Under the condition $\lambda_\parallel \ll 1$, we have the sine-Gordon equation for ϑ. In this case the field and frequency of the pulse increase in proportion to the distance traversed.[1–3,7,15] Since the longitudinal component of the magnetic field of the pulse promotes the formation of circularly polarized video pulses, we would expect that this component would oppose amplification under the condition $\lambda_\parallel < 1$. In the nonequilibrium case we have a self-similar solution of the sine-Gordon equation for the field which depends on the variable $\eta = \xi\tau$ (Refs. 1–3, 7, 15). This solution is a function that changes sign and is nonzero primarily near $\tau = 0$. Its resultant area is π.

Following the papers just cited, we introduce this variable in our own analysis. Equation (19) then becomes

$$\eta\vartheta'' + \vartheta' = \frac{\lambda_\perp\omega_0}{2c}|W_\infty|(\tilde{\omega}_0 \sin\vartheta - \frac{1}{2}\lambda_\parallel\omega_0 \sin 2\vartheta). \quad (35)$$

The function ϑ and its derivatives are smooth everywhere. We can thus ignore the first term on the right side of (35) near $\eta = 0$. Near $\eta = 0$ we then find the approximate solution

$$\Omega_\perp = 2\omega\left(\frac{2-k}{2\exp(2\omega\tau) - k}\right)^{1/2} \frac{\exp(2\omega\tau)}{\exp(2\omega\tau) + 1 - k}, \quad (36)$$

$$\omega_r = \frac{\partial\varphi}{\partial\tau} = -\omega_0\frac{z^2}{l_0 l}. \quad (37)$$

Here

$$\omega = \omega_0 z/l, \quad l = l_0/(1-\lambda_\parallel), \quad l_0 = (\lambda_\perp\omega_0)^{-1}c,$$

$$k = \lambda_\parallel^2/(2-\lambda_\parallel) < 1.$$

Far from $\tau = 0$, the equation can be linearized near the point $\vartheta = \pi$. We introduce the variable $\sigma = \vartheta - \pi$ ($|\sigma| \ll \pi$). From the linearized versions of Eqs. (35) and (20) we then have

$$\Omega_\perp = -\omega J_1(\sqrt{2\omega\tau})/\sqrt{2\omega\tau}, \quad (38)$$

$$\omega_r \approx 0, \quad (39)$$

where $J_1(a)$ is the Bessel function of order unity. This solution is valid for $\omega\tau > 0$. For $\omega\tau < 0$ we find a solution in the form of a modified Bessel function of order unity, whose asymptotic form (for $|\omega\tau| \gg 1$) is

$$\Omega_\perp \approx \frac{|\omega|}{2\pi}\frac{1}{|2\omega\tau|^{3/4}}\exp(-\sqrt{2|\omega\tau|}). \quad (40)$$

If we ignore the longitudinal component of the pulse field, we have $\lambda_\parallel = 0$ and $\Omega_\perp = \Omega_{10} = \omega\,\mathrm{sech}(\omega\tau)$. Following Refs. 1–3 and 7, we interpret ω as the frequency of the pulse, which increases in proportional to the distance traversed by the pulse. The amplitude $\Omega_\perp^{\max}$, of the transverse component of the magnetic field, increases in the same fashion. The longitudinal field component of the pulse, on the other hand, cannot exceed a maximum value $2\lambda_\parallel\omega_0$ [see (21)]. From (36) we have

$$\Omega_\perp^{max} = \Omega_\perp(z, 0) = 2(1-\lambda_\parallel)/(2-k).$$

Since we have $2(1-\lambda_\parallel)/(2-k) < 1$ under the condition $\lambda_\parallel < 1$, we conclude that the effect of the longitudinal component of the pulse amounts to reducing the amplification effect ($\Omega_\perp^{\max} < \Omega_{10}^{\max}$) and slowing this effect ($l > l_0$). Adopting $n \sim 10^{21}\ \mathrm{cm}^{-3}$, we find $l_0 \sim 10$ m. With $\lambda_\parallel = 0.5$ we have $q = 0.17$, $l = 2l_0 \sim 20$ m, and $\Omega_\perp^{\max} \approx 0.27\Omega_{10}^{\max}$. According to (36) and (37), the inequality $|\Omega_\perp| > |\partial\varphi/\partial\tau|$ holds if the distance traversed by the pulse satisfies $z < 2l_0/(2-k) < l$. The analysis above is thus valid only at distances small in comparison with those over which the pulse frequency increases by ω_0.

5. PARAMETRIC FREQUENCY CONVERSION

Along with the effects which stem from the nonlinear propagation of the video pulses, it is interesting to look at certain parametric processes. An example is the generation of higher harmonics, which was studied in Refs. 1, 2, and 7 for the case of linearly polarized pulses. In our own case, the rotation of the spin in real space means that the transverse component of the pulse is circularly polarized. Furthermore, a longitudinal component is created. We introduce the complex dipole moment $S_\perp = U + iV$, and by analogy with Refs. 2 and 7 we introduce the "Josephson magnetic current," $J = \partial S_\perp / \partial t$ [see (15)]. This current generates a circular-polarization wave. The magnetic field vector rotates in a circle with a frequency $\omega = \partial\varphi / \partial t$. However, the spectrum of the reradiated signal acquires components with frequencies

$$\omega_{1,2} = \Omega_\perp \pm \omega\,, \qquad \omega_{3,4} = 2\Omega_\perp \pm \omega\,.$$

Solutions (12)–(14) describe nutation in an ultrastrong field. Superimposed on the rapid rotations of the spin in the meridional plane of the Bloch sphere, at a frequency $\Omega_\perp$, is a slow motion of the spin in the equatorial plane, at a frequency ω. The role of the longitudinal component of the spin is to generate the frequencies $\omega_{3,4}$. The ratio of the intensities of the spectral lines at the frequencies $\omega_{1,2}$ and $\omega_{3,4}$ is estimated to be $\sim(\omega_0\lambda_\parallel/4|\Omega_\perp|)^2$. Under the condition $|\Omega_\perp| \gg \omega_0$ we have $\omega_{3,4} \approx 2\omega_{1,2} \sim \sqrt{I}$, where I is the pulse intensity. By varying I smoothly, we can tune the frequency at the exit from the paramagnet continuously. In the case of a linearly polarized pulse with an electric dipole interaction, a discrete set of harmonics of the frequency ω is generated at the exit.[1,2,7] As the power is raised, the spectral intensities of progressively higher harmonics increase. In our own case, there are no harmonics at all. In this case, an increase in the pulse power has the result that the four spectral components which are generated ($\omega_{1,2}, \omega_{3,4}$) withdraw continuously toward progressively higher frequencies. Corresponding to the frequency $\omega_{1,2} \sim |\Omega_\perp| \sim 10^{12}$ s^{-1} is an intensity $I = cH^2/4\pi \sim 10^{12}$ W/cm^2.

The efficiency of the frequency conversion can be estimated by calculating an effective cross section σ_0 for the process. Since the direction of the reradiated signal is the same as the direction of the incident signal, we can restrict the discussion to an estimate of the integral cross section. From the definition of σ_0 we have $\sigma_0 = \hbar\omega|\partial W/\partial t|/I$. Using (15) and the expression for I, and taking a time average, we find ($|\Omega_\perp| = \mathrm{const}$)

$$\sigma_0 = 16 g_\perp^2 \frac{\beta_0^2 \omega}{\hbar c \Omega_\perp} \,\mathrm{th}\, \frac{\hbar\omega_0}{2k_B T}\,. \tag{41}$$

Taking $\omega/\Omega_\perp \sim 0.1$, we find $\sigma_0 \sim 10^{-23}$ cm. The mean free path of a photon in the medium in the case of a tenfold frequency increase is thus $l \sim (\sigma_0 n)^{-1} \sim 1$ cm, where $n \sim 10^{23}$ cm^{-3}. To achieve efficient conversion, we must keep the thickness of the paramagnetic sample above this value.

6. CONCLUSION

Experiments on the generation of extremely short electromagnetic pulses in the microwave and IR ranges[9] are stimulating theoretical work on the nonresonant interactions of such pulses with various media, which have previously been studied by methods of resonant coherent spectroscopy. There is interest in studies of pulse propagation processes and also various parametric effects.

The analysis above shows that the magnetic dipole interaction of pulses with a paramagnet is quite different from the electric dipole interaction of such pulses with a nonresonant two-level system. The pseudospin describing the two-level system "rotates" in isotopic space. The z component of the pseudospin (the inversion) is unrelated to the longitudinal component of the electric field. The spin of a paramagnet, on the other hand, "rotates" in real space. The z component of the spin thus creates a longitudinal component of the magnetic field of the pulse. This component can radically change propagation processes.

There is the possibility that circularly polarized stationary pulses can propagate in a nonequilibrium paramagnet by dynamically inverting the Zeeman sublevels of the S state caused by the longitudinal pulsed component of the magnetic field.

The propagation of a pulse in a system of inverted spins leads to continuous amplification of the transverse field component. The longitudinal component, on the other hand, is amplified under saturation conditions. Furthermore, its presence results in a slowing of the amplification.

The "rotation" of the spin in real space also makes possible continuous parametric frequency up-conversion of a signal.

1) Nakata *et al.*[10-12] correctly associate their results with low-temperature ferromagnets ($T \ll T_C$, where T_C is the Curie temperature). The exchange-interaction field can be ignored since the velocity of spin waves is much smaller than the velocity of light. The corresponding formal criterion can be derived by comparing the field of the pulse with the exchange-interaction field: $\varkappa \equiv k_B T_C a^2/(\hbar \tau_p c^2) \ll 1$, where a is the distance between the nearest spins in the crystal lattice. Substituting in the values $T_C \sim 10^2$ K, $a \sim 10^{-8}$ cm, and $\tau_p \sim 10^{-12}$ s, we find $\varkappa \sim 10^{-12}$. The results of this paper can thus also be extended to low-temperature ferromagnets.

2) For the case of a ferromagnet, the substitution $\tanh(\hbar\omega_0/2k_B T) \to 1$ should be made in expressions (28) and (41).

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