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Ultrashort video pulses of a transverse wave field in an effective spin system with an anisotropic spin–spin interaction

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Abstract. The propagation of the circularly polarized ultrashort video pulse in a spin system with an anisotropic spin–spin interaction has been studied. A few physical realizations of this model have been suggested. Steady-state soliton-like solutions without using a slowly varying envelope and rotating-wave approximations have been obtained. The conditions of two-component video pulse bound-state formation have been investigated.

1. Introduction

At present the developing laser technology seeks ways of creating more high-power lasers generating pulses having durations in the femtosecond regime (Auston *et al* 1984, Akhmanov *et al* 1988, Belenov and Nazarkin 1990, Maimistov and Elyutin 1991, Nakata 1991, Sazonov 1991). In connection with this the interaction of ultrashort pulses (USPs) containing one period of light oscillations with matter is of much interest. Employing radiophysical terminology, such pulses for which the slowly varying envelope cannot be applicable will be referred to as 'video pulses' (Akhmanov *et al* 1988). Not until recently under laboratory conditions has the generation of a one-wave electromagnetic video pulse of infrared range proved to be possible (Darrow *et al* 1990).

For USP the rotating-wave and slowly varying envelope approximations (Allen and Eberly 1975) cannot be applied. The consideration of any pair of atomic levels (the approximation of a two-level medium) is irrelevant as the pulse spectrum width $\Delta\omega \sim \tau^{-1}$ (τ is the temporal pulse duration) becomes large: $\Delta\omega \gtrsim \omega_0$ (ω_0 is the atomic transition frequency). So, if we are willing to keep to a two-level model, it is necessary to use a pair of fairly distant levels.

In the present paper the propagation of the circularly polarized USPs of the wave field in a spin system with an anisotropic interaction between effective spins in the 'molecular-field' approximation (Baxter 1982) has been studied. Here we shall call the dynamic quantum variables the operators of which obey the Pauli commutational relationships the effective spins. In the case of paramagnets interacting with a magnetic field, real electron spins as well as a density operator corresponding to the quantum transition between two Zeeman sublevels (Pake 1962) can be taken as the effective spin. In the case of the interaction between a two-level quantum system and an electric field, two transverse components of the effective spin correspond to the transition electric dipole moment, the third component of the effective spin corresponding to a two-level system inversion (Allen and Eberly 1975).

In further discussion in the framework of the effective spin–spin interaction, in the first case we shall deal with the interaction between real electron spins, in the second case

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with the interaction between the Zeeman sublevels, and in the third case with an electric dipole–dipole interaction between two-level atoms absorbing and reradiating a light field.

For convenience we employ the following models to represent possible physical realizations of the interaction of an effective spin system with a transverse wave field. *Model A* corresponds to the interaction of a magnetic USP with a paramagnet, *model B* to the interaction of an electric USP with a two-level medium at transitions between levels, when the corresponding dipole moment matrix element is the complex number (Allen and Eberly 1975) and *model C* to the interaction of an acoustic USP with paramagnetic impurities.

A unitary transformation of the spin Hamiltonian enabling one to diagonalize its part related to the spin–spin interaction is presented in section 2. This transformation simplifies substantially the analysis of the equations for the wave and spin variables. Steady-state soliton-like solutions without using the rotating-wave and slowly varying envelope approximations are obtained and analysed in section 3. In section 4, model B illustrates the physical meaning of the restrictions imposed on the pulse propagation velocity. Model C illustrates the possibility of controlling the steady-state strain USP formation due to the variation in the Zeeman splitting value.

2. Basic equations

Consider a system of effective spins $S = \frac{1}{2}$ with an anisotropic spin–spin interaction. Let the USP propagate along the z axis. In the 'molecular-field' approximation the spin Hamiltonian may be written in the form

$$H = \hbar \sum_i \left\{ \omega_0 S_z^i - \sum_{\alpha,\beta} (\mathcal{J}_{\alpha\beta} S_\alpha^i \langle S_\beta \rangle + \Omega_\alpha S_\alpha^i) \right\} \tag{1}$$

where $\hbar$ is the Planck constant, $\alpha, \beta = x, y, z$, ω_0 is the Zeeman splitting frequency produced by the external magnetic field $H_0 \| z$ for models A and C, or the atomic transition frequency between quantum levels for model B, $\mathcal{J}_{\alpha\beta}$ are the components of the symmetrical tensor of effective spin–spin interaction ($\mathcal{J}_{\alpha z} = 0$) and S_α^i is the αth component of the ith effective spin operator (in model B, S_x^i and S_y^i are the xth and yth dipole moment operator components, respectively, and S_z^i is the inversion operator). For model A, $\Omega_\alpha = \mu_0 g_{\alpha\alpha} H_\alpha \hbar^{-1}$, where μ_0 is the Bohr magneton, $g_{\alpha\alpha}$ are the components of the diagonal Landé tensor and H_α are the components of the magnetic intensity vector ($H_z = 0$); for model C, $\Omega_\alpha = \omega_0 F_{\alpha z} \epsilon_{\alpha z}$, where $\epsilon_{\alpha z}$ ($\alpha = x, y$) are the components of the strain tensor and $F_{\alpha z}$ ($\alpha = x, y$) is the tensor of spin–acoustic interaction (Tucker 1966); for model B, $\Omega_\alpha = d_\alpha E_\alpha \hbar^{-1}$ ($\alpha = x, y$), where E_α is the αth component of the electric intensity vector, and d_α is the real component ($\alpha = x$) and d_y the imaginary ($\alpha = y$) component of the matrix element of the dipole moment.

If we derive the Heisenberg equations for the spin operators from the Hamiltonian (1), then they will turn out to be sufficiently complicated for the analysis to be performed with a non-diagonal form of the tensor of effective spin–spin interaction. So, to simplify calculation further, we shall diagonalize the tensor $\mathcal{J}_{\alpha\beta}$ in the Hamiltonian (1). For this purpose we move from the axes l_α to the major axes l'_α of the tensor $\mathcal{J}_{\alpha\beta}$:

$$l'_\alpha = \sum_\beta B_{\alpha\beta} l_\beta .$$

The matrix $\hat{\mathbf{B}}$ describing a rotation in the (x, y) plane takes the form

$$\hat{\mathbf{B}} = \begin{pmatrix} l_1 & -l_2 & 0 \\ l_2 & l_1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad (2)$$
$$\hat{\mathbf{B}}^{\mathrm{T}} = \hat{\mathbf{B}}^{-1}$$
$$\det \hat{\mathbf{B}} = 1$$

where

$$\begin{aligned} &l_1 = \mathcal{J}_{xy}/(\mathcal{J}_{xx} - \mathcal{J}_{yy}) \qquad l_2 = -1/A \\ &A = (\mathcal{J}_{xx} - \mathcal{J}_{yy})(\mathcal{J}_{xx} - \mathcal{J}_{yy} - 2D)/2\mathcal{J}_{xy}^2 \\ &D^2 = (\mathcal{J}_{xx} - \mathcal{J}_{yy})^2/4 - \mathcal{J}_{xy}^2. \end{aligned} \qquad (3)$$

The operators S_α^i as well as their average values will be transformed according to the law

$$\tilde{S}_\alpha = \sum_\beta (\hat{\mathbf{B}}^{-1})_{\alpha\beta} S_\beta \qquad S_\alpha = \sum_\beta B_{\alpha\beta} \tilde{S}_\beta \qquad \alpha, \beta = x, y, z. \qquad (4)$$

The eigenvalues of the tensor $\mathcal{J}_{\alpha\beta}$ are equal to

$$\begin{aligned} &\mathcal{J}_x = 2D/A - \mathcal{J}_{xx} \\ &\mathcal{J}_y = -2D/A - \mathcal{J}_{yy} \\ &\mathcal{J}_z = 0 \end{aligned} \qquad (5)$$

where D and A are defined in (3).

For new variables the Hamiltonian (1) is written in the form

$$H = \hbar \sum_i \left(\omega_0 \tilde{S}_z^i - \sum_\alpha (\mathcal{J}_\alpha \tilde{S}_\alpha^i \langle \tilde{S}_\alpha \rangle + \Omega_\alpha \tilde{S}_\alpha^i) \right) \qquad (6)$$

where $\tilde{\Omega}_\alpha = \sum_\beta B_{\alpha\beta} \Omega_\beta$; $\alpha, \beta = x, y$.

As $\hat{\mathbf{B}}^{\mathrm{T}} = \hat{\mathbf{B}}^{-1}$, then commutational relations are retained:

$$[\tilde{S}_\alpha^k, \tilde{S}_\beta^j] = \mathrm{i}\delta_{jk}\epsilon_{\alpha\beta\gamma}\tilde{S}_\gamma \qquad \alpha, \beta, \gamma = x, y, z.$$

Here $\epsilon_{\alpha\beta\gamma}$ is the unit antisymmetric tensor.

Using the Heisenberg representation, upon quantum averaging we obtain the following matter equations:

$$\begin{aligned} &\partial R_x/\partial t = -\omega_0 R_y - (\mathcal{J}_y R_y + \tilde{\Omega}_y) R_z \\ &\partial R_y/\partial t = \omega_0 R_x + (\mathcal{J}_x R_x + \tilde{\Omega}_x) R_z \\ &\partial R_z/\partial t = (\mathcal{J}_y - \mathcal{J}_x) R_x R_y - \tilde{\Omega}_x R_y + \tilde{\Omega}_y R_x \end{aligned} \qquad (7)$$

where $R_\alpha \equiv \langle S_\alpha^i \rangle$, $\alpha = x, y, z$, and the brackets designate quantum averaging.

From the Maxwell equations for H_α (model A) or for E_α (model B) it follows that

$$\begin{aligned}\Box\tilde{\Omega}_x &= c^{-2}(\partial^2/\partial t^2)(l_1 a_x\langle S_x\rangle - l_2 a_y\langle S_y\rangle)\\ \Box\tilde{\Omega}_y &= c^{-2}(\partial^2/\partial t^2)(l_2 a_x\langle S_x\rangle + l_1 a_y\langle S_y\rangle)\end{aligned} \tag{8}$$

where

$$\Box = \partial^2/\partial z^2 - \partial^2/\partial(ct)^2 \qquad a_\alpha = 8\pi(\mu_0 g_{\alpha\alpha})^2 n\hbar^{-4}$$

and n is the concentration of the atoms interacting with the wave field.

For the acoustic field (model C) from the elasticity theory, one can obtain equations similar to set (8) with the substitution

$$\partial^2/\partial(ct)^2 \rightarrow \partial^2/\partial z^2$$

in the right-hand side of these equations and

$$\Box \rightarrow c_s^2\partial^2/\partial z^2 - \partial^2/\partial t^2$$

in the left-hand side, where c_s is the sound velocity for transverse components of the acoustic field (we suppose that the velocities of the two components are equal). The constants a_α are defined as follows: $a_\alpha = n(H_0 F_{\alpha z})^2/\rho\hbar$, where ρ is the medium density and n is the concentration of paramagnetic centres.

3. Steady-state pulses

Suppose that a medium possesses an axial symmetry with respect to the spin–wave interaction. This means that $a_x = a_y = a_\perp$. For model A this is identical with $g_{xx} = g_{yy} = g_\perp$. Consequently, for model B we have $F_{xz} = F_{yz} = F_\perp$. For both models the latter conditions are valid, for instance, in the case of rhombic and cubic symmetry crystals. For model C the equality $a_x = a_y$ is equivalent to the equality of real and imaginary parts of the matrix element of the transition dipole moment: $d_x = d_y$. An electromagnetic wave reradiated by such a system possesses a circular polarization (Allen and Eberly 1975). In this case, subject to (2) and (4), system (8) can be written in the following form:

$$\Box\tilde{\Omega}_\alpha = a_\alpha c^{-2}\partial^2 R_\alpha/\partial t^2 \qquad \alpha = x, y. \tag{9}$$

We shall seek solutions in the form of steady-state solitary pulses depending on the variable $\xi = t - z/v$, where v is the pulse velocity.

Let an initial state of the spin system be as follows:

$$\begin{aligned}&\Omega_x(\xi = -\infty) = \Omega_y(\xi = -\infty) = R_x(\xi = -\infty) = R_y(\xi = -\infty) = 0\\ &R_z(\xi = -\infty) = -|R_z^\infty| < 0.\end{aligned} \tag{10}$$

Such a state, when $\mathcal{J}_{x,y} < \omega_0$, corresponds to a thermodynamic equilibrium state at any temperature T, and in the case $\mathcal{J}_{x,y} > \omega_0$ to a thermodynamic equilibrium state at $T > T_k$, where T_k is the temperature of the spin phase transition to an ordered state (Baxter 1982). For models A and B this phase transition (at $T < T_k$) corresponds to weak ferromagnetism

($T_k \simeq 0.1$ K) caused by remote action spin–spin interactions (Harrison 1970). In the case of model C at $T < T_k$ we have a phase ferroelectric-like transition of the order–disorder type.

Using suggestions made for the case of steady-state pulses, the set of equations (9), subject to (10), is easily integrated:

$$\tilde{\Omega}_\alpha = \tilde{a}_\alpha R_\alpha \qquad \alpha = x, y \tag{11}$$

where, for models A and B, $\tilde{a}_x = \tilde{a}_y = a_\perp(c^2/v^2 - 1)^{-1}$ and, for model C, $\tilde{a}_x = \tilde{a}_y = a_\perp(c_s^2 - v^2)^{-1}$.

Substituting (11) into (7) we obtain

$$\begin{aligned} \mathrm{d}R_x/\mathrm{d}\xi &= -\omega_0 R_y - b_y R_z R_y \\ \mathrm{d}R_y/\mathrm{d}\xi &= \omega_0 R_x + b_x R_z R_x \\ \mathrm{d}R_z/\mathrm{d}\xi &= (b_x - b_y) R_x R_y \end{aligned} \tag{12}$$

where $b_\alpha = \mathcal{J}_\alpha + \tilde{a}_\alpha$, $\alpha = x, y$.

The set of equations (12) has first the two integrals

$$\begin{aligned} -2\omega_0 R_z - b_y R_z^2 - 2\omega_0 |R_z^\infty| + b_y |R_z^\infty|^2 &= (b_y - b_x) R_x^2 \\ 2\omega_0 R_z + b_x R_z^2 + 2\omega_0 |R_z^\infty| - b_x |R_z^\infty|^2 &= (b_y - b_x) R_y^2 \end{aligned} \tag{13}$$

which enables one to integrate equations (12) by quadrature. After substituting (13) into equations (12) we have

$$\mathrm{d}u/\mathrm{d}\xi = \sqrt{b_x b_y}\, u[-(u_1 + u)(u_2 + u)]^{1/2} \tag{14}$$

where $u = R_z + |R_z^\infty|$ and $u_{1,2} = -2|R_z^\infty| + 2\omega_0/b_{x,y}$. For the existence of a soliton-like solution to (14) it is necessary that $u_1 u_2 < 0$. Let $u_1 < 0$, $u_2 > 0$, i.e.

$$\omega_0/b_x < |R_z^\infty| < \omega_0/b_y \tag{15}$$

which is met at $\mathcal{J}_x > \mathcal{J}_y$. If the effective spin–spin interactions are absent, then the formation of steady-state solitary pulses in the system under consideration in the presence of axial symmetry with respect to the spin–wave interaction is not possible (Sazonov and Yakupova 1992).

Integrating (14), we find that

$$\begin{aligned} &R_z = |R_z^\infty| - 2\omega_0/b_x - 2\omega_0(b_y^{-1} - b_x^{-1}) \tanh^2(\xi/\tau)/[p^2 - \mathrm{sech}^2(\xi/\tau)] \\ &p^2 = \omega_0(b_y^{-1} - b_x^{-1})/(|R_z^\infty| - \omega_0 b_x^{-1}) \qquad \tau^{-2} = \omega_0(1 - b_y/b_x)(b_x|R_z^\infty| - \omega_0). \end{aligned} \tag{16}$$

Using (13), we express R_x and R_y as functions of R_z. Substituting the expressions obtained in (11), we find the functions $\tilde{\Omega}_\alpha(\xi)$, $\alpha = x, y$. Going from the functions $\tilde{\Omega}_\alpha$ to the functions Ω_α according to the relationships

$$\Omega_\alpha = \sum_\beta (\hat{\mathbf{B}}^{-1})_{\alpha\beta} \tilde{\Omega}_\beta \qquad \alpha, \beta = x, y \tag{17}$$

we obtain

$$\Omega_x = [l_1\tilde{\Omega}_x^0 + l_2\tilde{\Omega}_y^0 \tanh(\xi/\tau)]\,\mathrm{sech}(\xi/\tau)/[p^2 - \mathrm{sech}^2(\xi/\tau)]$$
$$\Omega_y = [-l_2\tilde{\Omega}_x^0 + l_1\tilde{\Omega}_y^0 \tanh(\xi/\tau)]\,\mathrm{sech}(\xi/\tau)/[p^2 - \mathrm{sech}^2(\xi/\tau)] \qquad (18)$$

where

$$\tilde{\Omega}_x^0 = 2\omega_0[(1 - b_y/b_x)(\omega_0 - b_y|R_z^\infty|)/(b_x|R_z^\infty| - \omega_0)]^{1/2}$$
$$\tilde{\Omega}_y^0 = 2[\omega_0(\omega_0 - b_y|R_z^\infty|)]^{1/2}. \qquad (19)$$

Fix the observation plane z = constant. Then the end of the intensity vector $\Omega = \Omega_x l_x + \Omega_y l_y$ passing through this plane will describe the following curve:

$$(l_1\Omega_x - l_2\Omega_y)^2/\tilde{\Omega}_x^{02} = [\tfrac{1}{4} + p^2(l_2\Omega_x + l_1\Omega_y)/\tilde{\Omega}_y^0]^{1/2} - (p^2-1)[(l_2\Omega_x + l_1\Omega_y)/\tilde{\Omega}_y^0] - \tfrac{1}{2}. \qquad (20)$$

In this sense the wave field pulse may be considered to be circularly polarized. The graph of curve (20) is presented in figure 1. An angle between the axis l_x and the symmetry axis MN is defined by the elements of matrix (4). An angular value is defined by the medium parameters

$$\varphi = \tan^{-1}[\mathcal{J}_{xy}/(\mathcal{J}_{xx} - \mathcal{J}_{yy})].$$

If $\mathcal{J}_{xy} = 0$, we find that $\varphi = 0$, i.e. the axis MN is directed along the l_x axis.

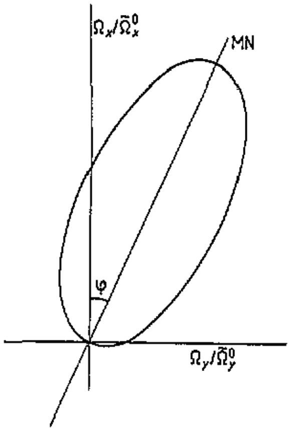

Figure 1. The curve (20) which describes the intensity vector $\Omega = \Omega_x l_x + \Omega_y l_y$ of a video pulse passing through the fixed plane z = constant; $\tilde{\Omega}_x^0/\tilde{\Omega}_y^0 = 4$; $\varphi = 25°$.

The double inequality (15) imposes restrictions on the propagation velocity of a circularly polarized solitary video pulse:

$$v_x < v < v_y \qquad (21)$$

where

$$v_\alpha = c[1 + a_\alpha|R_z^\infty|/(\omega_0 - \mathcal{J}_\alpha|R_z^\infty|)]^{1/2} \qquad \alpha = x, y \qquad (22)$$

for an electromagnetic pulse (models A and B), and

$$v_\alpha = [c_s^2 - a_\alpha|R_z^\infty|/(\omega_0 - \mathcal{J}_\alpha|R_z^\infty|)]^{1/2} \qquad \alpha = x, y \qquad (23)$$

for an acoustic pulse (model C).

Let us return to the condition $u_1 < 0$, $u_2 > 0$ ($\mathcal{J}_x > \mathcal{J}_y$). For the inverse relation $\mathcal{J}_x < \mathcal{J}_y$, corresponding solutions with $u_1 > 0$ and $u_2 < 0$ may be obtained from (5), (7), (9) and (21) with the help of the substitutions $\Omega_x \rightleftarrows \Omega_y$, $R_x \rightleftarrows -R_y$, $B_x \rightleftarrows B_y$ and $v_x \rightleftarrows v_y$.

4. Analysis of restrictions on the pulse and medium parameters

To understand the physical meaning of the values v_x and v_y in (22) we linearize the set of equations (7) near $R_z = -|R_z^\infty|$, $R_{x,y} = \Omega_{x,y} = 0$. Consider solutions of this system in the form $R_{x,y} \sim \Omega_{x,y} \sim \exp(i\omega t - ikz)$. We confine attention to model B. Model A may be considered in a similar way.

Using definitions of the dielectric permeability tensor $\epsilon_{\alpha\beta}$ and the dielectric susceptibility tensor $\chi_{\alpha\beta}$ given by

$$\epsilon_{\alpha\beta} = \delta_{\alpha\beta} + 4\pi\chi_{\alpha\beta} \qquad d_\alpha n R_\alpha = \sum_\beta \chi_{\alpha\beta} E_\beta \qquad \alpha, \beta = x, y$$

we find that the diagonal components $\epsilon_{\alpha\alpha}$ are given by

$$\epsilon_{\alpha\alpha} = 1 + \frac{8\pi d_\alpha^2 n |R_z^\infty|(\omega_0 - \mathcal{J}_\alpha |R_z^\infty|)}{[(\omega_0 - \mathcal{J}_y |R_z^\infty|)(\omega_0 - \mathcal{J}_x |R_z^\infty|) - \omega^2]} \qquad \alpha = x, y. \tag{24}$$

From (22) and (24) it is seen that at $\omega \to 0$ we have $v_x \to c/\sqrt{\epsilon_{xx}}$ and $v_y \to c/\sqrt{\epsilon_{yy}}$. Therefore, the upper and lower restrictions on the pulse velocity v correspond to the phase velocities of linear low-frequency electromagnetic waves along the main axes of the tensor $\mathcal{J}_{\alpha\beta}$. If $\omega_0 \gg \mathcal{J}_{xy}$, then the range of admissible values of velocities $v_x < v < v_y$ for electromagnetic video pulses is rather narrow (see (22)), and v is near the speed c of light.

For acoustic video pulses (model C) because the expressions for $v_{x,y}$ have another form (see (23)) the relative interval $(v_x/c_s, v_y/c_s)$ is wider.

The physical meaning of the restrictions (21) and (23) on the acoustic pulse velocity is rather clear. In the absence of spin–acoustic interaction ($F_{\alpha z} = 0$) a transverse acoustic wave propagates in a medium with the sound velocity c_s. In the presence of interaction between the spins and strain field the velocity of the acoustic pulse decreases (owing to the interaction delay). By controlling the external parameters (the Zeeman splitting frequency ω_0) one may act on the formation of the bound state of a two-component acoustic video pulse.

At $\omega_0 > c_s^2 m n_0 |R_z^\infty|/n\hbar F_{\alpha z}^2$ (see (23)), where m is the mass of an atom in the crystal lattice and n_0 is the concentration of atoms, a soliton-like strain pulse cannot be formed. Such values are possible in EPR spectroscopy. Taking $n_0/n \simeq 10$, $m \simeq 10^{-23}$ g, $F_{\alpha z} \simeq 10$ (Tucker 1966), $|R_z^\infty| \simeq \frac{1}{2}$, $c_s \simeq 3 \times 10^5$ cm s^{-1}, we obtain that, as the Zeeman splitting frequency is of the order of 10^{11} s^{-1}, corresponding to a magnetic field $H_0 \simeq 1$ T, we can qualitatively affect the character of strain video pulse propagation.

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