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Strain video pulses in solids with paramagnetic impurities

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Abstract. Non-linear propagation of a steady-state lateral strain video pulse in a crystal with rhombic symmetry in the presence of paramagnetic centres has been studied. On the basis of the solutions obtained the restrictions on the velocity of video pulse propagation have been analysed. The maximum value of the Zeeman splitting frequency at which formation of the steady-state strain video pulse is possible has been obtained.

1. Introduction

In recent years a number of experimental papers concerning the generation of femtosecond electromagnetic pulses have been published (Auston *et al* 1984, Akhmanov *et al* 1988). It is a characteristic property of these signals that they are video pulses. The term 'video pulse' is commonly used in radiophysics for pulses for which the term 'envelope' cannot be applied, i.e. video pulses contain only one period of oscillations (Akhmanov *et al* 1988). Such experiments stimulated a large body of theoretical work on the interaction of electromagnetic video pulses with matter (Belenov *et al* 1988, Belenov and Nazarkin 1990, Maimistov and Yelyutin 1991, Nakata 1991, Sazonov 1991). Obviously, within the theoretical framework that is used to describe the propagation of video pulses in a medium the slowly varying envelope approximation is not applicable. It was shown by Belenov *et al* (1988) and Belenov and Nazarkin (1990) that the propagation of high-power light video pulses in a two-level medium is described by the sine–Gordon equation. Consequently, these video pulses must possess soliton properties. The latter means that femtosecond electromagnetic signals may find wide application in information computer systems, optical spectroscopy of different media, etc.

The existence of different mechanisms for opto-acoustic interaction enables the generation of acoustic video pulses of nanosecond and picosecond duration (Leung *et al* 1985, Vodopyanov *et al* 1986).

In the early 1970s similar work with the electromagnetic-pulse acoustic self-induced transparency (SIT) on paramagnetic impurities has been produced (Shiren 1970, Denisenko 1971).

In connection with the above, the study of the propagation of acoustic video pulses in solids containing paramagnetic centres seems to be timely. Largely this results from the development of picosecond acoustic spectroscopy of paramagnetic media.

Let us consider the case when $l_p \gg l$, where l_p is the spatial scale length of the strain video pulse and l is the constant of a crystal lattice. This enables one to

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describe strain waves in the approximation of a continuous anisotropic medium. If $l \simeq 5 \times 10^{-8}$ cm and the velocity of a lateral strain is $a \simeq 5 \times 10^{5}$ cm s^{-1}, then the duration τ of the video pulse must satisfy the condition $\tau \gg 10$ ps.

We shall consider a crystal that is at the temperature of liquid helium. Later we shall neglect the attenuation of the strain pulse. Since the pulse is very short, we shall also ignore the relaxation terms in the equations for the spin movement.

2. Basic equations

Let a strain pulse propagate along the z axis parallel to an external magnetic field B_0. Also, let every dynamical variable depend only upon z and t, where t is the time. For simplicity consider only pulses of lateral strain in a crystal which has a symmetry that belongs to the rhombic system (Tucker and Rampton 1972). The Hamiltonian of the system 'strain plus paramagnetic centres' can then be written in the form

$$H = \int \left\{ n\beta_0 \boldsymbol{B}_0 \cdot \hat{\mathbf{g}}\boldsymbol{S} + \frac{1}{2\rho}(P_x^2 + P_y^2) + \frac{\rho}{2}\left[a_x^2\left(\frac{\partial u_x}{\partial z}\right)^2 + a_y^2\left(\frac{\partial u_y}{\partial z}\right)^2\right]\right\} \mathrm{d}\boldsymbol{r} \quad (1)$$

where ρ is the average medium density, u_x and u_y are the components of lateral displacement in solids, $P_{x\ (y)} = \rho\ \partial u_{x\ (y)}/\partial t$, n is the concentration of paramagnetic centres. $a_{x\ (y)}$ is the velocity of the lateral strain wave at the displacement in the x (y) direction in the absence of paramagnetic centres, β_0 is the Bohr magneton, $\boldsymbol{S} = (S_x, S_y, S_z)$ is the effective spin of a paramagnetic centre and $\hat{\mathbf{g}}$ is the Landé tensor. We assume that $S = \frac{1}{2}$. Then S_x, S_y, S_z are the Pauli operators. This is valid if two quantum levels described by the effective spin $S = \frac{1}{2}$ are at a sufficient distance from any other levels. Note that the constants a_x^2 and a_y^2 are related to the elastic moduli of a crystal (Tucker and Rampton 1972): $a_x^2 = c_{55}/\rho$; $a_y^2 = c_{44}/\rho$. The axes of x, y and z are perpendicular to the planes of crystal symmetry. The Landé tensor can be written in the form

$$\hat{\mathbf{g}} = \hat{\mathbf{g}}_0 + \delta\hat{\mathbf{g}} \quad (2)$$

where $\hat{\mathbf{g}}_0$ is the Landé tensor in the absence of crystal strain and $\delta\hat{\mathbf{g}}$ is its disturbance due to the strain pulse. Here the $\hat{\mathbf{g}}_0$ tensor is such that

$$\boldsymbol{B}_0 \cdot \hat{\mathbf{g}}_0\boldsymbol{S} = B_0 g_{zz} S_z. \quad (3)$$

For lateral strain waves we have (Denisenko 1971)

$$\boldsymbol{B}_0 \cdot \delta\hat{\mathbf{g}}\ \boldsymbol{S} = B_0 g_{zz}(F_x S_x\ \partial u_x/\partial z + F_y S_y\ \partial u_y/\partial z) \quad (4)$$

where $F_x \equiv F_{xzxz}$, $F_y \equiv F_{yzyz}$ are the components of the tensor of spin–strain interaction. Using the Hamiltonian formalism with respect to (1)–(4) gives

$$\partial u_{x\ (y)}/\partial t = \delta H/\delta P_{x\ (y)}$$
$$\partial P_{x\ (y)}/\partial t = \rho\ \partial^2 u_{x\ (y)}/\partial t^2 = -\delta H/\delta u_{x\ (y)}. \quad (5)$$

After quantum averaging, we find that

$$\partial^2\epsilon_{xz}/\partial t^2 - a_x^2\ \partial^2\epsilon_{xz}/\partial z^2 = q_x\ \partial^2 U/\partial z^2 \tag{6}$$

$$\partial^2\epsilon_{yz}/\partial t^2 - a_y^2\ \partial^2\epsilon_{yz}/\partial z^2 = q_y\ \partial^2 V/\partial z^2 \tag{7}$$

where $\omega_0 = g_{zz}\beta_0 B_0/\hbar$, $q_{x(y)} = n\hbar\omega_0 F_{x(y)}/\rho$; $\epsilon_{xz} = \partial u_x/\partial z$ and $\epsilon_{yz} = \partial u_y/\partial z$ are the components of the tensor of lateral strain, $U \equiv \langle S_x\rangle$, $V \equiv \langle S_y\rangle$ and $\langle\ldots\rangle$ is the operation of quantum averaging. For the spin operators we have the Heisenberg equations

$$\partial S_k/\partial t = (\mathrm{i}/\hbar)[H, S_k] \qquad k = x, y, z$$

where $\hbar$ is the Planck constant. After the quantum averaging we obtain

$$\partial U/\partial t = -\omega_0 V + \omega_0 F_y \epsilon_{yz} R \tag{8}$$

$$\partial V/\partial t = \omega_0 U - \omega_0 F_x \epsilon_{xz} R \tag{9}$$

$$\partial R/\partial t = \omega_0(F_x\epsilon_{xz} V - F_y\epsilon_{yz} U) \tag{10}$$

where $R \equiv \langle S_z\rangle$.

3. Steady-state solutions

We shall seek the solutions to the set of equations (6)–(10) in the form of steady-state pulses running along the z axis at a velocity v, i.e. assume that the functions R, U, V, ϵ_{xz} and ϵ_{yz} depend on one variable $\xi \equiv t - z/v$. We assume that before the interaction with an acoustic pulse all spins are in a state of thermodynamic equlibrium, i.e.

$$U_\infty = V_\infty = 0$$
$$R_\infty = -\tanh(\hbar\omega_0/k_{\mathrm{B}}T) = -|R_\infty| \tag{11}$$

where k_{B} is the Boltzmann constant and T is the absolute temperature. Using the variable ξ, after integrating equations (6) and (7) in light of (11), we obtain

$$\epsilon_{xz} = [q_x/(v^2 - a_x^2)]U \qquad \epsilon_{yz} = [q_y/(v^2 - a_y^2)]V. \tag{12}$$

Substituting (12) into (8)–(10) we find that

$$\dot V = (\omega_0 + \beta_x R)U \tag{13}$$

$$\dot U = -(\omega_0 + \beta_y R)V \tag{14}$$

$$\dot R = (\beta_y - \beta_x)UV \tag{15}$$

where $\beta_{x(y)} = n\hbar\omega_0^2 F_{x(y)}^2/\rho(a_{x(y)}^2 - v^2)$, and the dot denotes the derivative with respect to ξ. From (13) we find U through R and $\dot V$. Then we substitute it into (15). Integrating the expression obtained, subject to (11), we find that

$$V^2 = [2/(\beta_y - \beta_x)]\left[\omega_0(R - R_\infty) + \tfrac{1}{2}\beta_x(R^2 - R_\infty^2)\right]. \tag{16}$$

Similarly from (14) and (15) we obtain

$$U^2 = [2/(\beta_y - \beta_x)]\left[-\omega_0(R - R_\infty) - \tfrac{1}{2}\beta_y(R^2 - R_\infty^2)\right]. \tag{17}$$

Employing (15)–(17) we have

$$\dot{u} = \sqrt{\beta_x\beta_y}\,u\,[(u+u_1)(u+u_2)]^{1/2} \tag{18}$$

where $u = R - R_\infty$, $u_1 = 2R_\infty + 2\omega_0/\beta_x$, $u_2 = 2R_\infty + 2\omega_0/\beta_y$.

Soliton-like behaviour in (18) is attained under the condition $u_1u_2 < 0$. We therefore let $u_1 < 0$, $u_2 > 0$, i.e.

$$\omega_0/\beta_x < |R_\infty| < \omega_0/\beta_y. \tag{19}$$

Integrating (18) with regard to (19) we obtain

$$R = |R_\infty| - 2\omega_0/\beta_x - 2\omega_0(1/\beta_y - 1/\beta_x)[\tanh^2\theta/(q^2 - \operatorname{sech}^2\theta)]. \tag{20}$$

Here $\theta = \xi/\tau$,

$$q^2 = \omega_0(\beta_x - \beta_y)/\beta_y(\beta_x|R_\infty| - \omega_0) \qquad \tau^{-1} = \sqrt{\omega_0(1 - \beta_y/\beta_x)(\beta_x|R_\infty| - \omega_0)}. \tag{21}$$

From (20) it is seen that $R \to -|R_\infty|$ at $t \to \pm\infty$. Consequently, the effective spin of a paramagnetic centre after the passage of a strain video pulse, as in the case of SIT (Denisenko 1971), returns to its initial state. However, in contrast with SIT, when the maximum spin inversion is $R_{\max} = |R_\infty|$, in a particular case, the spin excitation is partial since $R_{\max} = |R_\infty| - 2\omega_0/\beta_x$. Using (16), (17), (20) and (12) we find that

$$U = 2\omega_0\sqrt{(1/\beta_y)(1/\beta_y - 1/\beta_x)[(\omega_0 - \beta_y|R_\infty|)/(\beta_x|R_\infty| - \omega_0)]}$$
$$\times [\operatorname{sech}\theta/(q^2 - \operatorname{sech}^2\theta)] \tag{22}$$

$$V = -2\sqrt{(\omega_0/\beta_y)(\omega_0/\beta_y - |R_\infty|)}[(\tanh\theta\operatorname{sech}\theta)/(q^2 - \operatorname{sech}^2\theta) \tag{23}$$

$$\epsilon_{xz} = \epsilon_{xz}^0[\operatorname{sech}\theta/(q^2 - \operatorname{sech}^2\theta)] \tag{24}$$

$$\epsilon_{yz} = \epsilon_{yz}^0[(\tanh\theta\operatorname{sech}\theta)/(q^2 - \operatorname{sech}^2\theta) \tag{25}$$

where

$$\epsilon_{xz}^0 = -(2/F_x)\sqrt{(\beta_x/\beta_y)(\beta_x/\beta_y - 1)(\omega_0 - \beta_y|R_\infty|)/(\beta_x|R_\infty| - \omega_0)}$$

$$\epsilon_{yz}^0 = (2/\omega_0F_y)\sqrt{\omega_0\beta_y(\omega_0/\beta_y - |R_\infty|)}.$$

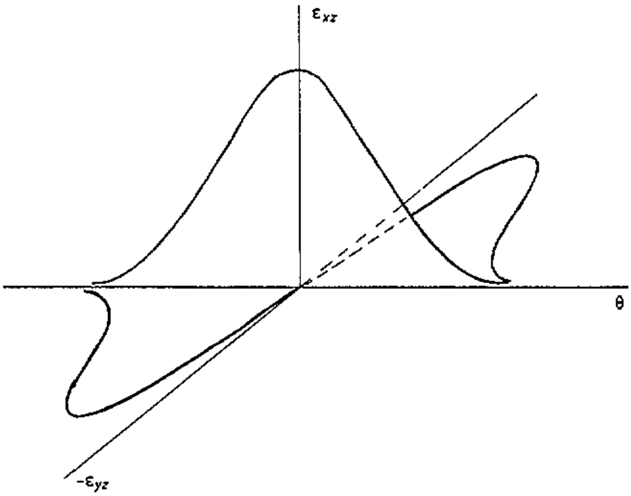

Figure 1. Bound state of the lateral components ϵ_{xz} and ϵ_{yz} of the strain tensor in the light of (25) and (26).

The solutions (24) and (25) are presented in figure 1.

The double inequality (19), which is necessary for the existence of solitary video pulses, imposes the following restrictions on the velocity of propagation:

$$a_x^2 - (n\hbar\omega_0/\rho)F_x^2|R_\infty| < v^2 < a_y^2 - (n\hbar\omega_0/\rho)F_y^2|R_\infty|. \qquad (26)$$

It is straightforward to understand that if

$$\omega_0/\beta_x > |R_\infty| > \omega_0/\beta_y$$

then the corresponding solution is obtained from (20)–(25) by virtue of the substitutions

$$\beta_x \leftrightarrows \beta_y \qquad U \leftrightarrows -V \qquad \epsilon_{xz} \leftrightarrows \epsilon_{yz}.$$

Instead of the double inequality (26) we have

$$a_x^2 - (n\hbar\omega_0/\rho)F_x^2|R_\infty| > v^2 > a_y^2 - (n\hbar\omega_0/\rho)F_y^2|R_\infty|.$$

4. Analysis of solutions and conclusion

A strain video pulse during its propagation interacts with a spin system. Hence $v < \max\{a_x, a_y\}$. If $\alpha a_j^2 - F_j^2 > a_k^2$, where $\alpha = \rho/\hbar\omega_0 n|R_\infty|$, $a_j = \max\{a_x, a_y\}$, $a_k = \min\{a_x, a_y\}$, then $v > \min\{a_x, a_y\}$. In the opposite case $v < \min\{a_x, a_y\}$.

If $\alpha a_j^2 < F_j^2$, then a bound state of two lateral polarizations of strain video pulse cannot be formed. This imposes a condition on the value of external magnetic field, since $\hbar\omega_0 = g_{zz}\beta_0 B_0$.

Let N be the concentration of atoms which form a crystal matrix. Then the necessary condition for forming a solitary strain video pulse is

$$\hbar\omega_0|R_\infty| < (N/nF_j^2)Ma_j^2 \tag{27}$$

where M is the atomic mass of a crystal lattice. The constants of spin–elastic coupling may attain values $F_j \simeq 10$–10^3 (Tucker 1966). Let $M \simeq 10^{-23}$ g, $N/n \simeq 10$, $a_j \simeq 3\times10^5$ cm s^{-1} and $|R_\infty| \simeq 1$ so that $\omega_0 < 10^{11}$–10^{15} s^{-1}. The Zeeman splitting frequency $\omega_0 \simeq 10^{11}$ s^{-1} corresponds to a magnetic field $B_0 \simeq 1$ T. Such magnetic fields are usually used in EPR spectroscopy; so there exists an opportunity for breaking the inequality (27). Thus, by varying the value of the magnetic field B_0, one may make qualitative changes in the character of the propagation of a strain video pulse in the system of paramagnetic centres.

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