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Propagation of picosecond strain video pulses in a one-dimensional paramagnetic lattice

S V Sazonov

Pacific Oceanological Institute, Far Eastern Branch of Russian Academy of Sciences, 43 Baltiiskaya Street, Vladivostok 690 032, Russia

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Abstract. On the basis of a one-dimensional crystal lattice model the interaction of a lateral picosecond acoustic video pulse with a system of spins $S = \frac{1}{2}$ has been studied. The mechanisms of dispersion and non-linearity due to lattice structure, the oscillatory anharmonicity of the lattice and the spin–acoustic interaction are taken into account. Conditions on the lattice parameters which make a steady-state pulse of strain propagation possible are given. Amplification of a strain video pulse by a system of inverted spins is predicted.

1. Introduction

In recent years, developing laser equipment has made the generation of femtosecond light pulses possible (Auston *et al* 1984). Several theoretical studies dedicated to the interaction between ultrashort electromagnetic pulses and matter have been published (Belenov *et al* 1988, Belenov and Nazarkin 1990, Maimistov and Elyutin 1991, Nakata 1991, Sazonov 1991, Azarenkov *et al* 1991). The characteristic spatial length scale of such pulses is $l_e \simeq c\tau_p \simeq 10^{-4}$ cm (where c is the speed of light and τ_p is the temporal pulse duration). At the same time, because of the different mechanisms of opto-acoustic interaction, ultrashort light signals are able to generate picosecond acoustic pulses in solids (Akhmanov *et al* 1988, Gusev and Karabutov 1991). These acoustic signals are video pulses, i.e. contain strain waves of nearly one period of oscillation (Akhmanov *et al* 1988). Herein, as in the case of femtosecond light pulses, the slowly varying envelope approximation (Allen and Eberly 1975) is not applicable. The spatial length scale of such strain pulses is $l_s \simeq a\tau_p \simeq 10^{-6}$–$10^{-7}$ cm (where a is the velocity of sound in solids), that is two to three orders of magnitude less than the corresponding length scale of an optical femtosecond pulse. Since the value l_s is comparable with the interatomic distance in a crystal lattice, then the effects of spatial dispersion due to the lattice structure are important. The latter statement can be referred to femtosecond light pulses, since $l_e \gg h$, where h is the constant of a crystal lattice. Therefore, in further discussion we shall call this mechanism acoustic dispersion.

Picosecond acoustic pulses are very strong, their inside pressure attaining 1–100 kbars (Akhmanov *et al* 1988, Gusev and Karabutov 1991). Therefore, the anharmonicity effects of lattice vibrations may have great significance for the propagation of picosecond strain pulses in solids. We shall call the anharmonicity of lattice vibrations acoustic non-linearity.

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As is known, magnetic, optic and acoustic coherent spectroscopies developed simultaneously. Such phenomena as a photon echo (Kopvillem and Nagibarov 1963, Kurnit *et al* 1964), paramagnetic and nuclear magnetic resonances (Pake 1962) and optical self-induced transparency (McCall and Hahn 1969) have acoustic analogues (Al'tshuler 1952, Kopvillem 1963, Baranskii 1957, Menes and Bolef 1958, Tucker and Rampton 1972, Golenishchev-Kutuzov *et al* 1977, Shiren 1970, Denisenko 1971). So it is obviously relevant to enquire into the propagation of picosecond strain pulses in a paramagnetic lattice. The interaction between lattice oscillations and spins causes an interaction between temporal dispersion and non-linearity which also takes place on interaction between spins and an electromagnetic field. We shall call these mechanisms spin–acoustic dispersion and spin–acoustic non-linearity, respectively. Because of these mechanisms, the spectrum of physics phenomena in picosecond acoustics must be much richer than in the optics of these systems. This stimulates to a considerable extent the development of acoustic picosecond spectroscopy of paramagnetic and nuclear systems.

2. Basic model

Let us consider the propagation of lateral strain waves along the z axis in a one-dimensional lattice in the approximation of the interaction of nearest neighbours as the model. Let every atom of this lattice have a spin $S = \frac{1}{2}$. Following Jacobsen and Stevens (1963) the Hamiltonian of this system can be written in the following form:

$$H = \sum_j \left(\frac{P_j^2}{2M} + \frac{\kappa}{2}(u_{j+1} - u_j)^2 + \frac{\alpha}{4}(u_{j+1} - u_j)^4 + \hbar\frac{q}{h}(u_{j+1} - u_{j-1})S_x^j + \mu_0 B g_{\parallel} S_z^j \right). \tag{1}$$

Here M is the atomic mass, u_j and P_j are the displacement and the impulse of the jth atom along the x axis, respectively (the strain pulse is linearly polarized), κ is the coefficient of elasticity of a restoring force on the interaction of nearest neighbours, α is the quartic anharmonicity (in the case of lateral vibrations of a one-dimensional lattice the cubic anharmonicity is absent (Kosevich and Kovalev 1989)), $\hbar$ is the Planck constant and q is the constant of spin–lattice interaction. In the simplest case this interaction may be due to sound modulation of the zx component of the **g** Landé tensor (Jacobsen and Stevens 1963). B is the value of the external magnetic intensity directed along the z axis, μ_0 is the Bohr magneton, and S_x^j and S_z^j are the components of the spin operator belonging to the jth atom.

A video pulse is not resonant for any pair of quantum levels. Therefore, a two-level approximation can be applied here provided that these levels are at a sufficient distance from any other quantum levels. To illustrate this, two sublevels resulting from the S-state splitting of a paramagnetic atom in an external magnetic field can be considered.

From (1) after quantum averaging and employing the Hamiltonian and the Heisenberg formalisms, the following set of equations can be obtained:

$$M\ddot{u}_j = \kappa(u_{j+1} - 2u_j + u_{j-1}) + \alpha[(u_j - u_{j-1})^2 + (u_j - u_{j-1})(u_{j+1} - u_j)$$

$$+(u_{j+1}-u_j)^2](u_{j+1}-2u_j+u_{j-1})+\hbar(q/h)(R_{j+1}-R_{j-1}) \quad (2)$$

$$\dot{R}_j=-\omega_0 V_j \qquad \dot{V}_j=\omega_0 R_j-(q/h)(u_{j+1}-u_{j-1})W_j$$
$$\dot{W}_j=(q/h)(u_{j+1}-u_{j-1})V_j \quad (3)$$

where $R_j\equiv\langle S_x^j\rangle$, $V_j\equiv\langle S_y^j\rangle$, $W_j\equiv\langle S_z^j\rangle$, $\langle\ldots\rangle$ is the quantum average and $\omega_0=g_{\|}\mu_0 B/\hbar$.

Let us now employ a 'quasi-continuous approximation':

$$u_{j\pm1}=u\pm h\partial u/\partial z+(1/2!)h^2\partial^2 u/\partial z^2\pm(1/3!)h^3\partial^3 u/\partial z^3$$
$$+(1/4!)h^4\partial^4 u/\partial z^4\pm\ldots \quad (4)$$
$$R_{j\pm1}=R\pm h\partial R/\partial z+(1/2!)h^2\partial^2 R/\partial z^2\pm(1/3!)h^3\partial^3 R/\partial z^3+\ldots.$$

Here we shall preserve the derivatives over the first order only in the terms which occur in (2) and (3) linearly. Then we obtain

$$\partial^2 u/\partial t^2=a^2\partial^2 u/\partial z^2+(a^2h^2/12)\partial^4 u/\partial z^4+\delta h^2(\partial u/\partial z)^2\partial^2 u/\partial z^2$$
$$+\sigma\partial R/\partial z+\tfrac{1}{3}\sigma h^2\partial^3 R/\partial z^3 \quad (5)$$

$$\partial R/\partial t=-\omega_0 V \qquad \partial V/\partial t=\omega_0 R-\Omega W$$
$$\partial W/\partial t=\Omega V. \quad (6)$$

Here $\Omega=2q\epsilon$, $\epsilon\equiv\partial u/\partial z$ is the relative strain and $a=h(\kappa/M)^{1/2}$ is the velocity of sound of a linear wave in a long-wave approximation, $\delta=3\alpha h^2/M$ and $\sigma=\hbar q/M$. From (6) we find that

$$\partial^2 R/\partial t^2=-\omega_0^2 R+\omega_0\Omega W \quad (7)$$

$$\partial W/\partial t=-(\Omega/\omega_0)\partial R/\partial t. \quad (8)$$

Then, following Belenov *et al* (1988), assume that a pulse is so short that its duration is

$$\tau_p\ll\omega_0^{-1}. \quad (9)$$

Then one may write (7) approximately in the form

$$\partial^2 R/\partial t^2=\omega_0\Omega W. \quad (10)$$

Equations (8) and (9) are integrated using the arbitrary function $\theta(z,t)$:

$$W=W_\infty\cos\theta \qquad \partial R/\partial t=\omega_0 W_\infty\sin\theta \quad (11)$$

where $\theta=\int_{-\infty}^{t}\Omega(z,t')\,dt'$ is the spin inversion before the action of the strain pulse (in the case of thermodynamic equilibrium, $W_\infty<0$). It is evident that the function $\theta(z,t)$ is related to the displacement in the following manner:

$$\partial\theta/\partial t=2q\epsilon=2q\partial u/\partial z. \quad (12)$$

Using (11) and (12) we can rewrite (5) in the form

$$\partial^4\theta/\partial t^4-a^2\partial^4\theta/(\partial z^2\partial t^2)-(a^2h^2/12)\partial^6\theta/(\partial z^4\partial t^2)$$
$$-(\delta h^2/4q^2)[\partial^2/(\partial z\partial t)][(\partial\theta/\partial t)^2\partial^2\theta/(\partial z\partial t)]$$
$$=2q\sigma\omega_0 W_\infty(\partial^2/\partial z^2)(\sin\theta)+\tfrac{2}{3}q\sigma\omega_0 W_\infty h^2(\partial^4/\partial z^4)(\sin\theta). \quad (13)$$

3. Solutions and analysis

For $q = 0$, equation (13), subject to (12), is transformed into the modified Boussinesq equation

$$\partial^2 u/\partial t^2 - a^2\partial^2 u/\partial z^2 - (a^2h^2/12)\partial^4 u/\partial z^4 - \delta h^2(\partial u/\partial z)^2\partial^2 u/\partial z^2 = 0. \quad (14)$$

The strain solution described by this equation has a speed of $v > a$ (Bataille and Lund 1982). The corresponding solution is

$$\begin{aligned} \epsilon &= \partial u/\partial z = (1/h)\sqrt{2(v^2-a^2)/\delta}\,\mathrm{sech}[(t-z/v)/\tau_s] \\ \tau_s^{-1} &= (2v/h)\sqrt{3(v^2/a^2-1)}. \end{aligned} \quad (15)$$

If $l_s \gg h$, then we may ignore acoustic dispersion and acoustic non-linearity, i.e. $h = 0$ in equation (13). Also, ignoring the anharmonicity of lattice oscillations, we obtain

$$\partial^4\theta/\partial t^4 - a^2\partial^4\theta/(\partial z^2\partial t^2) = 2\hbar(q^2\omega_0 W_\infty/M)(\partial^2/\partial z^2)(\sin\theta). \quad (16)$$

Now it is easy to obtain the solution to a relative strain in the form of a steady-state pulse:

$$\begin{aligned} \epsilon &= \sqrt{2\hbar\omega_0|W_\infty|/M(a^2-v^2)}\,\mathrm{sech}[(t-z/v)/\tau_p] \\ \tau_p^{-1} &= |q|\sqrt{2\hbar\omega_0|W_\infty|/M(a^2-v^2)} \end{aligned} \quad (17)$$

where $W_\infty < 0$. During the propagation of an acoustic pulse, one may observe its absorption and reradiation by the spin system; consequently $v < a$. Let $\mu \equiv 2\hbar^3 q^2\omega_0/(Ma^2)^3 \ll 1$; then it is possible to introduce the local time $\tau = t - z/a$ and the slow coordinate $\xi = \mu z$ and expand equation (16) in terms of μ, neglecting the terms $\mu^2, \mu^3, \ldots$. Equation (16) is reduced to the sine–Gordon equation:

$$\partial^2\theta/(\partial\xi\partial\tau) = (M^2a^3/2\hbar^2)W_\infty \sin\theta. \quad (18)$$

For $W_\infty > 0$, equation (18) has a similar solution depending on $\xi\tau$ in the form of an oscillatory π-pulse (Lamb 1971, 1980, Belenov *et al* 1988). While propagating along the lattice the strain amplitude increases in proportion to z: $\epsilon \propto z$. This amplification occurs together with pulse compression, which is expressed as an increase in oscillation frequency in proportion to z. The effects caused by acoustic dispersion and acoustic non-linearity become considerable as amplification and compression increase. At present such an analysis has not been provided. For $W_\infty < 0$, equation (18) has solutions in the form of soliton pulses. The velocities of these pulses satisfy the condition $v \lesssim a$. By introducing a slow coordinate $\zeta = \lambda z$ where $\lambda = \delta h^2/a^2 \ll 1$ and a local time $\tau = t - z/a$, equation (14) can be reduced to the modified Korteweg–de Vries equation for ϵ which also possesses soliton solutions. The velocity of solitons here is $v \gtrsim a$.

In the presence of both types of dispersion and non-linearity we may have both $v < a$ and $v > a$. In the general case, it is difficult to study equation (13). A solution to (13) is sought in the form of a steady-state pulse depending on z and t as $t - z/v$. Then after twofold integration we find that

$$(1 - a^2/v^2)\theta'' - (a^2h^2/12v^4)\theta'''' - (\delta h^2/4q^2v^2)\theta'^2\theta''$$
$$= 2q\sigma\omega_0(W_\infty/v^2)\sin\theta + (2/3v^4)q\sigma\omega_0 W_\infty h^2(\sin\theta)''. \quad (19)$$

It is evident that a localized steady-state pulse must correspond to a full spin inversion ($W = +\frac{1}{2}$) and its subsequent restoration to the initial state $W = -\frac{1}{2}$. Otherwise, if a spin after the passage of a signal remains in a superpositional state, this will lead to its rotation and, as a consequence, to reradiation at a frequency ω_0. This statement corresponds to the *anzatz*

$$\theta' = \omega\sin(\theta/2). \quad (20)$$

Here both ω and v are the unknown constants subject to determination. Employing (20), (12) and (11), we find that

$$\epsilon = (\omega/2q)\,\mathrm{sech}[(\omega/2)(t - z/v)] \quad (21)$$

$$W = |W_\infty|\{1 - 2\tanh^2[(\omega/2)(t - z/v)]\} \quad (22)$$

$$R = 4(\omega_0/\omega)|W_\infty|\,\mathrm{sech}[(\omega/2)(t - z/v)]. \quad (23)$$

In addition, from (20) it follows that

$$\theta'' = (\omega^2/4)\sin\theta \qquad \theta'''' = (3\omega^4/32)\sin(2\theta) - (\omega^4/8)\sin\theta. \quad (24)$$

Substituting (20) and (24) into (19), equating the coefficients of $\sin\theta$ and $\sin(2\theta)$ to zero and employing simple algebraic transformations, we find that

$$\omega_\pm = (4q/a)\sqrt{2\hbar\omega_0|W_\infty|/M(1 - 4kv_\pm^2/a^2)} \quad (25)$$

$$v_\pm^2 = a^2[1 + 2k \pm \sqrt{(1 + 2k)^2 - 4(3 + \beta)k(1 - k)}]/8k(1 - k). \quad (26)$$

Here $k = \delta/q^2$ and $\beta = 16\alpha h^4\hbar\omega_0|W_\infty|/(Ma^2)^2$. From (25) it follows that for $\alpha > 0$ we have the condition on the velocity of pulse propagation:

$$v < a/2\sqrt{k}. \quad (27)$$

Analysis (24) shows that the condition (27) may be satisfied if

$$k < \tfrac{1}{4}. \quad (28)$$

Here $v_- < a$, $v_+ < a/2\sqrt{k}$. Thus for $k < \frac{1}{4}$ it is possible that the two strain pulses may exist in the form of (21). In addition, there exists another condition on the parameter β expressed by the positive discriminant in equation (26):

$$\beta \leqslant (1 - 4k)^2/4k(1 - k). \quad (29)$$

An analogous study shows that for $\alpha < 0$ the following statements are possible.

(1) $|k|$ is an arbitrary value, $|\beta| < 3$, and a single pulse exists with ω_+ and v_+.
(2) $|k| > 0.5$, $|\beta| > 3$, and the two pulses exist with both $\omega_\pm$ and $v_\pm$.

In both cases there is no limitation on the velocity of pulse propagation. Here the condition analogous to (29) must be satisfied:

$$|\beta| \leqslant (1 + 4|k|)^2/4|k|(1 + |k|). \quad (30)$$

In the absence of spin–lattice interaction when $k < 0$, steady-state solitary strain pulses cannot exist (see (13)). However, the spin–lattice interaction may advance the formation of such pulses. Note that in this case the video pulse duration $\tau_p \simeq \omega^{-1}$ increases as the velocity of its propagation increases.

4. Concluding remarks

The excitation of coherent strain pulses of duration $\tau_p \simeq 1$–100 ps and internal pressure $p \simeq 10^5$–10^3 bar is possible in a physical sense in solids. Let the density of a solid be $\rho \simeq 5$ g cm^{-3} and the velocity of sound in the solid be $a \simeq 3 \times 10^5$ cm s^{-1}. Then the pressure $p \simeq 10^3$ bar and $\tau_p \simeq 100$ ps correspond to $\epsilon = p/\rho a^2 \simeq 2\times10^{-3}$. Consequently, for $\tau_p \simeq 100$ ps and $p \simeq 10^3$ bar, both acoustic non-linearity and acoustic dispersion may be neglected. In this case, solutions (15) and (16) are valid. The pressure $p \simeq 10^5$ bar and the duration $\tau_p \simeq 1$ ps correspond to $\epsilon \simeq 0.1$ and $l_s \simeq 10^{-7}$ cm. Here acoustic non-linearity, acoustic dispersion with spin–acoustic non-linearity and spin–acoustic dispersion may be of considerable significance.

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