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# Non-linear interaction of picosecond acoustic pulses with a paramagnetic crystal

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**Abstract.** Non-linear propagation of ultra-short lateral circularly polarized strain pulses in a paramagnetic crystal of cubic symmetry localized in an external magnetic field at super-low temperature has been studied. The direction of the magnetic field is parallel to the direction of pulse propagation and the fourth-order symmetry axis. Under certain physical conditions pulse propagation is described by the 'derivative non-linear Schrödinger equation', which belongs to the equations integrated by the inverted scattering transform method. The phenomenon of full reflection of acoustic pulses on the paramagnetic crystal surface is predicted. The possibility of continuous parametric increase of frequency of a high-power acoustic signal during its passage through the paramagnetic crystal has been shown. This frequency increase can be controlled through changes in the power of the incoming signal. In this direction in principle one can obtain 10-fold frequency increase of an initial pulse. As an example the crystal MgO with impurities of paramagnetic ions  $\text{Co}^{2+}$  is considered.

## 1. Introduction

Generation of picosecond acoustic pulses under laboratory conditions (Akhmanov and Gusev 1992) gave rise to a number of theoretical papers dedicated to the interaction of such pulses with paramagnetic crystals (Sazonov and Yakupova 1992, Sazonov 1992, 1993a). High-power picosecond lateral linearly polarized strain pulses have been investigated by Sazonov (1992, 1993a). However, spin–lattice interaction may lead to the rotation of strain-wave polarization plane (Tucker and Rampton 1972, Denisenko 1971).

Picosecond acoustic pulses are video pulses, i.e. contain nearly a single period of oscillations. So, herein the slowly varying envelope approximation cannot be applied for a wave equation. The purpose of the present paper is to investigate the propagation of lateral circularly polarized acoustic pulses of picosecond time duration during the interaction of the spin system  $S = \frac{1}{2}$  of paramagnetic atoms with crystal strains. As a physical realization of this model, the crystal MgO containing Kramers' doublet impurities of the paramagnetic ions  $\text{Co}^{2+}$  may be proposed (Tucker 1966).

Let a lateral strain pulse propagate parallel to the magnetic field  $H_0$ , directed along the fourth-order symmetry axis of a cubic symmetry crystal. The magnetic field causes Zeeman splitting of the Kramers doublet into two sublevels. This direction may be taken for the  $z$  axis. The general Hamiltonian of the system investigated can be written in the form:

$$H = \int (H_a + H_s + H_{\text{int}}) d^3r \quad (1)$$

where  $H_{a(s)}$  is the Hamiltonian density of an acoustic field (spin system) and  $H_{\text{int}}$  is the Hamiltonian density of the interaction of spins with an acoustic field. In our case

$$H_a = \frac{P^2}{2\rho} + \frac{1}{2}\rho a^2 \left| \frac{\partial \mathbf{u}}{\partial z} \right|^2 + \frac{1}{4}\rho a^2 \lambda \left| \frac{\partial \mathbf{u}}{\partial z} \right|^4 + \frac{1}{2}\rho a^2 \beta_a \left| \frac{\partial^2 \mathbf{u}}{\partial z^2} \right|^2 \quad (2)$$

$$H_s = \hbar \omega_0 S_z \quad (3)$$

$$H_{\text{int}} = \hbar \omega_0 F \left( \frac{\partial \mathbf{u}}{\partial z} + \beta_s \frac{\partial^3 \mathbf{u}}{\partial z^3} \right) S. \quad (4)$$

Here  $\mathbf{u} = (u_x, u_y)$ ,  $\mathbf{P} = (P_x, P_y)$  are displacement and strain field pulse vectors respectively,  $a$  is the velocity of lateral sound,  $\lambda$  is the fourth-order dimensionless anharmonicity parameter (cubic anharmonicity is absent in the case of lateral strain pulses (Kosevich and Kovalev 1989)),  $\beta_a$  is the parameter of spatial dispersion of acoustic field (for a one-dimensional lattice  $\beta_a = \hbar^2/12$ , where  $\hbar$  is the distance between the nearest atoms of a crystal (Sazonov 1992)),  $\rho$  is the average density of the medium,  $\omega_0$  is the Zeeman splitting frequency of the Kramers doublet,  $\hbar$  is the Planck constant,  $F$  is the lateral component of spin-acoustic interaction tensor, and  $S_x, S_y, S_z$  are the spin-density operators satisfying commutation relationships of the following type:

$$[S_k(\mathbf{r}), S_j(\mathbf{r}')] = i\varepsilon_{kjm} S_m(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}'). \quad (5)$$

Here  $k, j, m = x, y, z$ ,  $\varepsilon_{kjm}$  is the absolute antisymmetrical tensor,  $\beta_s$  is the parameter of spatial dispersion caused by non-local spin action on a strain field and back (for a one-dimensional lattice we have  $\beta_s = \hbar^2/6$  (Sazonov 1992)).

Expression (4) for  $H_{\text{int}}$  is a generalization of the expression considered in Denisenko (1971) on the case of short pulses when the effect of discrete crystal structure is essential ( $\beta_s \neq 0$ ).

Since the spatial length scale of a strain pulse  $l$  fulfils the inequality  $l \gg \hbar$ , then  $|\beta_s \partial^3 \mathbf{u} / \partial z^3| \ll |\partial \mathbf{u} / \partial z|$ .

To obtain equations describing the strain field we will use the Hamiltonian formalism:

$$\frac{\partial \mathbf{u}}{\partial t} = \frac{\delta H}{\delta \mathbf{P}} \quad \frac{\partial \mathbf{P}}{\partial t} = -\frac{\delta H}{\delta \mathbf{u}}. \quad (6)$$

After quantum averaging on spin states we find that

$$\frac{\partial^2 \varepsilon}{\partial t^2} - a^2 \frac{\partial^2 \varepsilon}{\partial z^2} - \beta_a a^2 \frac{\partial^4 \varepsilon}{\partial z^4} - 3\lambda a^2 \frac{\partial^2}{\partial z^2} (|\varepsilon|^2 \varepsilon) = F \frac{n}{\rho} \hbar \omega_0 \left( \frac{\partial^2 S}{\partial z^2} + \beta_s \frac{\partial^4 S}{\partial z^4} \right) \quad (7)$$

where  $\varepsilon = \varepsilon_{xz} + i\varepsilon_{yz}$ ,  $\varepsilon_{xz} = \partial u_x / \partial z$ ,  $\varepsilon_{yz} = \partial u_y / \partial z$  are the tensor components of crystal strain,  $nS = \langle S_x \rangle + i \langle S_y \rangle$ ,  $n$  is the concentration of paramagnetic atoms in a crystal, and  $\langle \dots \rangle$  is the quantum averaging operation.

Using the Heisenberg presentation for spin operators, we find that

$$\partial S / \partial t = i\omega_0 (S - F \bar{\varepsilon} W) \quad (8)$$

$$\partial W / \partial t = -\omega_0 F \text{Im}(\bar{\varepsilon} S^*) \quad (9)$$

where  $W = \langle \sigma_z \rangle / n$ ,  $\bar{\varepsilon} = \varepsilon + \beta_s \partial^2 \varepsilon / \partial z^2$ .

## 2. Full reflection of a strain pulse

Let us consider the case when

$$\omega_0 \tau_p \gg 1 \tag{10}$$

where  $\tau_p$  is characteristic temporal duration of a strain pulse.

Inequality (10) indicates the slow change of pulse parameters compared with spin precession in a magnetic field. It is clear that the pulse interacts weakly with the spin system and in this sense it propagates in the quasitransparency regime in non-linear medium. Then from (8) it is easy to understand that inequality (10) is equivalent to the condition

$$F^2 |\tilde{\varepsilon}|^2 \ll 1. \tag{11}$$

Having, for instance,  $F \sim 10$  (Tucker 1966) we find that  $|\tilde{\varepsilon}| \ll 0.1$ . Corresponding pressure inside the pulse is  $P \sim \rho a^2 |\tilde{\varepsilon}| \ll 5 \times 10^9$  Pa at  $\rho \simeq 5$  g cm<sup>-3</sup>,  $a \sim 3 \times 10^5$  cm s<sup>-1</sup>. We hope that strain pulses with pressures  $P \sim 10^9$ – $10^{10}$  Pa will be generalized under experimental conditions in the near future (Akhmanov and Gusev 1992).

By analogy with the papers of Crisp (1970) and Belenov *et al* (1991) the solution of equation (8), written in the form

$$S = -i\omega_0 F \int_0^\infty \tilde{\varepsilon}(t - t', z) W(t - t', z) \exp(i\omega_0 t') dt' \tag{12}$$

can be expanded in terms of small parameters  $\mu_1 \equiv (\omega_0 \tau_p)^{-1}$  and  $\mu_2 \equiv \hbar/l$ :

$$S = F \left( \tilde{\varepsilon} W - \frac{i}{\omega_0} \frac{\partial}{\partial t} (\varepsilon W) - \frac{1}{\omega_0^2} \frac{\partial^2}{\partial t^2} (\varepsilon W) + \dots \right). \tag{13}$$

Substitute (13) into (9), keeping the first two terms of the expansion. Moreover, subject to the condition (11), we will assume that  $W = W_\infty \equiv W(t = -\infty)$  in the right-hand part of equation (9). Then upon integration we obtain

$$W \sim W_\infty (1 - F^2 |\varepsilon|^2 / 2). \tag{14}$$

After substitution of (14) into (13), subject to (11), we find that

$$S \simeq W_\infty F \left( \varepsilon - \frac{1}{2} F^2 |\varepsilon|^2 \varepsilon - \frac{i}{\omega_0} \frac{\partial \varepsilon}{\partial t} - \frac{1}{\omega_0^2} \frac{\partial^2 \varepsilon}{\partial t^2} + \beta_s \frac{\partial^2 \varepsilon}{\partial z^2} \right). \tag{15}$$

Substitute (15) into (7). Then

$$\begin{aligned} \frac{\partial^2 \varepsilon}{\partial t^2} - \frac{a^2}{N^2} \frac{\partial^2 \varepsilon}{\partial z^2} &= \beta_a a^2 \frac{\partial^4 \varepsilon}{\partial z^4} + \left( 3\lambda a^2 - F^4 \frac{n}{2\rho} \hbar \omega_0 W_\infty \right) \frac{\partial^2}{\partial z^2} (|\varepsilon|^2 \varepsilon) \\ &\quad - F^2 \frac{n}{\rho} \hbar \omega_0 W_\infty \frac{\partial^2}{\partial z^2} \left( \frac{i}{\omega_0} \frac{\partial \varepsilon}{\partial t} + \frac{1}{\omega_0^2} \frac{\partial^2 \varepsilon}{\partial t^2} - 2\beta_s \frac{\partial^2 \varepsilon}{\partial z^2} \right). \end{aligned} \tag{16}$$

Here we introduce the refractive index

$$N = (1 + F^2 n \hbar \omega_0 W_\infty / \rho a^2)^{-1/2}. \tag{17}$$

An acoustic 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:  $\omega_j \gg \omega_0$  and  $F_j \ll F$ , where  $\omega_j$  is the quantum transition frequency from one of the Kramers' doublet sublevels to one of the remote quantum levels,  $F_j$  is the spin-elastic coupling constant for the corresponding quantum transition. The very large strains would be likely to mix states from higher levels into the Kramers doublet. But if inequalities (10) and (11) are valid, we may keep in each expression of (15) type, corresponding to the  $j$ th transition with account for replacements  $\omega_0 \rightarrow \omega_j$ ,  $F \rightarrow F_j$  and  $W_\infty \rightarrow -W_{1,2}$ , only the first term, neglecting other terms. Here  $W_{1,2}$  is the initial population probability of one of the Kramers' doublet sublevels. Then we can take into account the remote quantum level mixing by using the following replacement in (17):  $1 \rightarrow 1 - \delta$ . Here  $\delta$  is a positive constant. If  $H_0 = 0$ , then  $\omega_0 = 0$  and  $\omega_j \neq 0$ . Therefore, this constant is very slightly dependent on the external magnetic field value. In the case of thermodynamic equilibrium

$$W_\infty = -\tanh(\hbar\omega_0/2k_B T) \quad (18)$$

where  $k_B$  is the Boltzmann constant and  $T$  is the absolute medium temperature.

Then if

$$\tilde{q} \equiv (F^2 n \hbar \omega_0 / M n_0 a^2) \tanh(\hbar\omega_0/2k_B T) > 1 - \delta \quad (19)$$

where  $M$  and  $n_0$  are the mass of a crystal lattice loop and concentration of molecules forming a crystal lattice respectively (mass and concentration of MgO), the acoustic wave cannot propagate in a paramagnetic crystal. In this case the strain pulse must experience full reflection on the surface of the paramagnetic crystal.

The constant of spin-elastic coupling for  $\text{Co}^{2+}$  in MgO may attain values  $F \sim 10^2$  (Tucker 1966). Considering also that  $M \sim 10^{-23}$  g,  $n_0/n \sim 10$ ,  $a \sim 10^5$  cm s $^{-1}$ ,  $\hbar\omega_0/k_B T \gg 1$  and  $\delta \ll 1$ , we find that condition (19) is fulfilled when  $\omega_0 > 10^{11}$  s $^{-1}$ . Such Zeeman splitting frequencies are usually used in electron paramagnetic resonance (EPR) spectroscopy.

### 3. Acoustic soliton

In further discussion we will investigate the case when  $N$  takes only real values. The latter is true if  $q < 1$ . In the paper of Sazonov and Yakupova (1992) this condition may serve as the condition under which a bound state of two strain components of a steady-state video pulse can be formed.

The right-hand part of (16) is proportional to positive degrees of the small parameters  $\mu_1$  and  $\mu_2$ . So we can reduce this equation deflating the derivative order (Belenov *et al* 1991):

$$\begin{aligned} \frac{\partial \varepsilon}{\partial t} - i \frac{N^2 - 1}{2\omega_0 N^2} a^2 \frac{\partial^2 \varepsilon}{\partial \zeta^2} + \frac{a}{2} \left( 3\lambda N + F^2 \frac{N^2 - 1}{2N} \right) \frac{\partial}{\partial \zeta} (|\varepsilon|^2 \varepsilon) \\ + \frac{Na}{2} \left[ \beta_a + \frac{N^2 - 1}{N^2} \left( \frac{a^2}{N^2 \omega_0^2} - 2\beta_s \right) \right] \frac{\partial^3 \varepsilon}{\partial \zeta^3} = 0. \end{aligned} \quad (20)$$

Here  $\zeta = z - at/N$ .

The second and fourth terms in (20) describe different dispersion effects, whereas the third term deals with non-linearity. The relationship between the fourth and second terms can be estimated as:

$$\max\{\omega_0\tau_p, (\omega_0\tau_p)^{-1}\omega_0^2h^2/a^2\}.$$

Let  $\omega_0 \sim 10^{11} \text{ s}^{-1}$ ,  $h \sim 10^{-8} \text{ cm}$ ,  $a \sim 3 \times 10^5 \text{ cm s}^{-1}$ ,  $\tau_p \sim 100 \text{ ps}$ . Then  $\omega_0\tau_p \sim 10$ ,  $(\omega_0\tau_p)^{-1}(\omega_0h/a)^2 \sim 10^{-6}$ . Consequently, the fourth term on the left-hand side of equation (20) can be neglected. In this case we have the 'derivative non-linear Schrödinger equation' (DNLS):

$$i\frac{\partial \varepsilon}{\partial t} + \frac{N^2 - 1}{2\omega_0 N^2} a^2 \frac{\partial^2 \varepsilon}{\partial \zeta^2} + i\frac{a}{2} \left( 3\lambda N + F^2 \frac{N^2 - 1}{2N} \right) \frac{\partial}{\partial \zeta} (|\varepsilon|^2 \varepsilon) = 0 \quad (21)$$

which is integrated by the inverse scattering transform method (Kaup and Newell 1978).

Here the dispersion mechanism caused by non-local temporal relationship between the strain field and spins substantially affects the pulse dynamics. The spatial dispersion mechanisms induced by discrete crystal structure are not essential here.

To seek the one-soliton solution of equation (21) we will write  $\varepsilon$  in the form:

$$\varepsilon = \psi \exp(i\varphi) \quad (22)$$

where  $\psi$  and  $\varphi$  are real.

Substituting (22) into (21) and separating the real part from the imaginary one, we obtain

$$\frac{\partial \psi}{\partial t} + \frac{N^2 - 1}{2\omega_0 N^2} a^2 \left( 2 \frac{\partial \psi}{\partial \zeta} \frac{\partial \varphi}{\partial \zeta} + \psi \frac{\partial^2 \varphi}{\partial \zeta^2} \right) + \frac{3a}{2} \left( 3\lambda N + F^2 \frac{N^2 - 1}{2N} \right) \psi^2 \frac{\partial \psi}{\partial \zeta} = 0 \quad (23)$$

and

$$\psi \frac{\partial \varphi}{\partial t} - \frac{N^2 - 1}{2\omega_0 N^2} a^2 \frac{\partial^2 \psi}{\partial \zeta^2} + \frac{N^2 - 1}{2\omega_0 N^2} a^2 \psi \left( \frac{\partial \varphi}{\partial \zeta} \right)^2 + \frac{3\lambda N a}{2} \psi^3 \frac{\partial \varphi}{\partial \zeta} + F^2 \frac{N^2 - 1}{4N} a \psi^3 \frac{\partial \varphi}{\partial \zeta} = 0. \quad (24)$$

We will see the solution to equations (23) and (24) in the form

$$\varepsilon = \varepsilon(\zeta + ct) \quad \varphi = \omega t + \Phi(\zeta + ct) \quad c = \text{const.} \quad (25)$$

This solution corresponds to pulses propagating along the  $z$  axis at a speed of  $v = a/N - c$  and rotating in the transverse plane. In this case equation (23) possesses the motion integral:

$$\frac{c}{2} \psi^2 + \frac{N^2 - 1}{2\omega_0 N^2} a^2 \psi^2 \varphi' + \frac{3}{8} N a \left( 3\lambda + F^2 \frac{N^2 - 1}{2N^2} \right) \psi^4 = A \quad (26)$$

where the prime designates the derivative with respect to  $\zeta + ct \equiv z - vt$ . Since at  $t \rightarrow \pm\infty$ ,  $\varepsilon \rightarrow 0$ , then  $A = 0$ .

Expressing  $\varphi'$  in terms of  $\psi$  from (26) and substituting the corresponding expression into (24), subject to (25), we will find that

$$\begin{aligned} \psi'' = & \frac{2\omega_0 N^2}{(N^2 - 1)a^2} \left( \omega - \frac{c^2}{2a^2} \frac{\omega_0 N^2}{N^2 - 1} \right) \psi - \frac{2\omega_0^2 N c}{(N^2 - 1)^2 a^3} \left( 9\lambda + F^2 \frac{N^2 - 1}{4N} \right) \psi^3 \\ & - 3 \left( \frac{\omega_0 a N}{8(N^2 - 1)} [6\lambda N^2 + F^2(N^2 - 1)] \right)^2 \psi^5. \end{aligned} \quad (27)$$

The solution in terms of a solitary phase has the following form:

$$\psi = \psi_0 \left( \frac{1 + [1 + (kl)^{-2}]^{1/2}}{1 + [1 + (kl)^{-2}]^{1/2} \cosh[2(z - vt)/l]} \right)^{1/2} \quad (28)$$

$$\varphi = \omega t - k(z - vt) - 6 \tan^{-1} \{ q \tanh[(z - vt)/l] \} \quad (29)$$

where

$$\psi_0 = \frac{2}{l} \left( \frac{2(N^2 - 1)a}{\omega_0 k N [6\lambda N^2 + F^2(N^2 - 1)]} \frac{1}{1 + [1 + (kl)^{-2}]^{1/2}} \right)^{1/2} \quad (30)$$

$$v = \frac{a}{N} \left( 1 - \frac{N^2 - 1}{N} \frac{k}{\omega_0} a \right) \quad (31)$$

$$\omega = \frac{N^2 - 1}{2\omega_0 N^2} a^2 \left( k^2 + \frac{1}{l^2} \right) \quad q = \frac{[1 + (kl)^{-2}]^{1/2} - 1}{[1 + (kl)^{-2}]^{1/2} + 1}. \quad (32)$$

Solution (28) and (29) depends on two free parameters. For instance,  $l$  and  $k$  can be chosen as such. If  $kl \gg 1$ , we have the envelope soliton. In this limit solution (28)–(32) formally coincides with the one-soliton solution of the ‘non-linear Schrödinger equation’ (NLS). In the case when  $kl \sim 1$  we have from (28)–(32) a video soliton, i.e. a soliton that contains nearly a single period of acoustic oscillations.

The analysis of (28) and (29) shows that for inequality (10) and (11) to be valid the following conditions must be fulfilled:  $\omega/\omega_0 \ll 1$ .

The strain tensor components  $\varepsilon_{xz}$  and  $\varepsilon_{yz}$  corresponding to the pulse of the form of (22), (28)–(32) are shown in figure 1 for a fixed moment of time. At other moments of time the components  $\varepsilon_{xz}$  and  $\varepsilon_{yz}$  change their configurations in an accompanying framework due to the rotation of the polarization plane. However, the scale length of strain localization defined by the parameter  $l$  remains unchangeable.

As follows from physical considerations a pulse of the form of (28) and (29) can be stable only at  $v < a/N$ . However, formally solution (28) and (29) assumes the case  $v > a/n$  at  $6\lambda N^2 + F^2(N^2 - 1) < 0$  (see (30) and (31)). From (30) it follows that to overcome the velocity barrier  $v = a/N$ , an infinitely large density of strain energy is needed, which contradicts the initial assumption made in inequality (11). Thus, the pulse (28) and (29) can be formed at

$$6\lambda N^2 + F^2(N^2 - 1) > 0. \quad (33)$$

If  $W_\infty > 0$  (a non-equilibrium case), then  $N^2 < 1$ . In the absence of lattice anharmonicity the formation of pulses of the form of (28) and (29) turns out to be impossible.

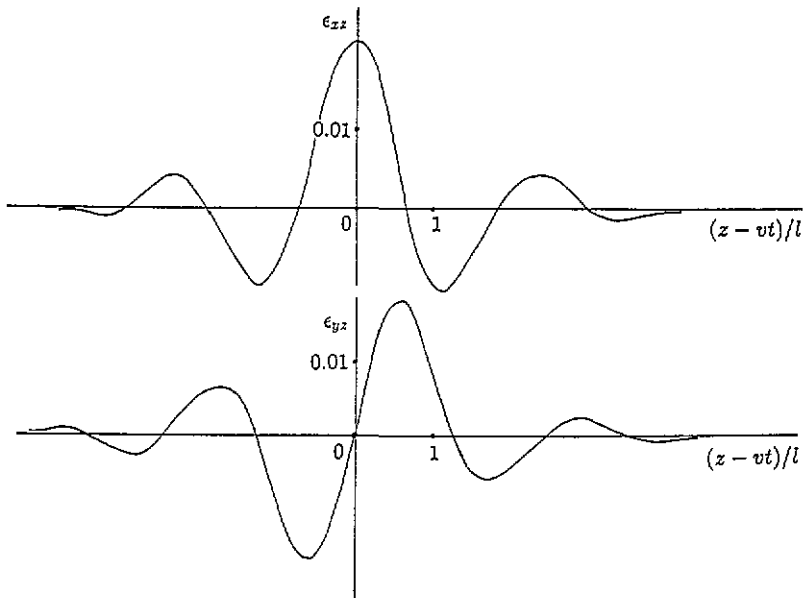


Figure 1. Instantaneous profile of the circularly polarized soliton components governed by equation (21);  $N^2 = 2$ ,  $\lambda = 10$ ,  $F = 10$ ,  $a = 3 \times 10^5$  cm s $^{-1}$ ,  $\omega_0 = 10^{11}$  s $^{-1}$ ,  $k = 10^5$  cm $^{-1}$ ,  $l = 3 \times 10^{-5}$  cm.

However, a 'rigid' anharmonicity ( $\lambda > 0$ ) makes this formation possible when fulfilling condition (33). At this we have that  $k < 0$  and  $\omega < 0$ . In the case of 'soft' anharmonicity ( $\lambda < 0$ ) and in the absence of paramagnetic centres the formation of a strain soliton is impossible as well. However, the presence of these centres favours the soliton formation in an equilibrium case ( $N^2 > 1$ ). Thus, the availability of two different non-linearity mechanisms can lead to the formation of solitary circularly polarized pulses, when in the absence of one of the mechanisms such formation proves to be impossible.

#### 4. Continuous parametric frequency increase

Consider the generation of high frequencies of a high-power acoustic pulse as a result of its interaction with a paramagnetic crystal. Let the pulse be so strong that the following condition is valid:

$$F^2 |\bar{\varepsilon}|^2 \gg 1, |\omega_0^{-1} \partial \varphi / \partial t|^2. \quad (34)$$

We consider also that the influence of the remote quantum levels on the quantum transition between Kramers' doublet sublevels is still slight.

At  $F \sim 10^2$  this condition is met for pulses with the relative strain  $|\varepsilon| > 0.03$ . Only very pure paramagnetic crystals without any impurities and dislocations are able to bear such relative strains. The inequality  $F^2 |\bar{\varepsilon}|^2 \gg |\omega_0^{-1} \partial \varphi / \partial t|^2$  indicates the slowness of the pulse polarization plane rotation compared with quantum spin transitions.

Introduce new variables  $f$  and  $L$ :

$$\bar{\varepsilon} = f \exp(i\varphi) \quad S = iL \exp(i\varphi). \quad (35)$$



Then the set of equations (8) and (9) takes the form

$$\partial L / \partial t = i(\omega_0 - \partial \varphi / \partial t)L - fW \quad (36)$$

$$\partial W / \partial t = \text{Re}(fL^*). \quad (37)$$

If (34) is fulfilled the first term of the right-hand part of (36) can be ignored. Then the functions  $f$  and  $L$  are real and we have solutions of system (36) and (37) in the form

$$W = W_\infty \cos \theta L = W_\infty \sin \theta \quad (38)$$

where

$$\theta = \omega_0 \int_{-\infty}^t f(t') dt.$$

Consequently,

$$s = iW_\infty \exp(i\varphi) \sin \theta. \quad (39)$$

Solutions (38) and (39) describe the nutation effect in a super-strong field: slow spin rotation in an equatorial plane at a frequency of  $\omega \equiv \partial \varphi / \partial t$  is imposed on fast spin rotation in a meridional plane of the Bloch sphere ( $|s|^2 + W^2 = W_\infty^2$ ) at a frequency of  $\Omega \equiv \omega_0 F f$ . In a specific case when  $\omega, f = \text{const}$  at the outlet of a paramagnetic crystal, we have two modes with frequencies:

$$\omega_{1,2} = \omega_0 F f \pm \omega \sim \Omega \sim I^{1/2} \quad (40)$$

where  $I$  is the pulse intensity. Since inequalities (34) are fulfilled then  $\omega_{1,2} \gg \omega$ .

Having  $\omega \simeq \omega_0$ ,  $F \sim 10^2$ ,  $f \sim |\varepsilon| \sim 0.1$ , we obtain a 10-fold increase in acoustic signal frequency. Further increase in relative strain with the aim of increasing the frequency transformation coefficient may lead to crystal destruction. Note that one can continuously control the frequency values  $\omega_1$  and  $\omega_2$  at the outlet of a circularly polarized signal by changing the inlet signal intensity. In the case of linearly polarized pulses at the outlet of a paramagnetic crystal, a discrete harmonic series of the initial frequency  $\omega$  is generated (Sazonov 1993a). Here at the growth of pulse intensity applied on a sample the energy is transferred to higher harmonics  $2\omega, 3\omega, \dots$ . Similar calculations for the case of linearly polarized and circularly polarized optical pulses interacting with a two-level non-resonant medium are given by Belenov *et al* (1991) and by Sazonov (1993b) respectively.

The effectiveness of the process of frequency parametric increase can be estimated upon calculating its cross section  $\sigma$ . According to the definition we have

$$\sigma = J^{-1} |\partial W / \partial t|$$

where  $J$  is the density of phonon flux of an initial signal. It is obvious that  $J \sim au / \hbar \omega$ , where  $u = 0.5 \rho a^2 |\varepsilon|^2$  is the energy density of a strain field. At  $\Omega = \text{const}$  after time averaging and using (18) we find that

$$\sigma \sim \frac{\hbar \omega_0^2 F^2}{\rho a^3} \frac{\omega}{\Omega} \tanh \left( \frac{\hbar \omega_0}{2k_B T} \right). \quad (41)$$

It is understandable that the process under consideration may be sufficiently effective at  $T < \hbar\omega_0/2k_B$ . Substituting here  $\omega_0 \sim 10^{10} \text{ s}^{-1}$ , we find that  $T < 0.1 \text{ K}$ . These temperatures are important moreover to avoid a strong attenuation of a coherent signal at the expense of its scattering on hot non-coherent phonons of a crystal. At  $F \sim 10^2$ ,  $\rho \simeq 5 \text{ g cm}^{-3}$ ,  $a \sim 3 \times 10^5 \text{ cm s}^{-1}$ ,  $\omega/\Omega \sim 0.1$  we have that  $\sigma \sim 10^{-21} \text{ cm}^2$ . If the concentration of paramagnetic atoms in a crystal is  $n \sim 10^{22} \text{ cm}^{-3}$ , then for an average distance of a free run of pulse phonon  $\bar{l}$  we obtain  $\bar{l} \sim (\sigma n)^{-1} \sim 10^{-1} \text{ cm}$ . Obviously if  $\Delta > \bar{l}$ , where  $\Delta$  is the thickness of a crystal in the direction of initial signal propagation, then practically all the initial signal is subject to the frequency transformation. For the numerical instance under consideration, a crystal with  $\Delta = 1 \text{ cm}$  will suffice. Note that the increase in ratio  $\Omega/\omega \sim \omega_{1,2}/\omega$  leads to cross section decrease. Consequently, a crystal of greater thickness should be used.

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