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## Soliton interactions in gyrotropic crystals

K T Stoychev, M T Primatarowa and R S Kamburova

Institute of Solid State Physics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria

E-mail: stoychev@issp.bas.bg (K T Stoychev), prima@issp.bas.bg (M T Primatarowa) and krad@issp.bas.bg (R S Kamburova)

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**Abstract.** The interactions of conjugate circularly polarized vibrational solitons in gyrotropic molecular crystals are investigated. Their dynamics is governed by a system of coupled non-linear Schrödinger equations which are solved analytically and numerically. Depending on the initial conditions and the material parameters, different evolutionary patterns are obtained corresponding to unbound or bound one- and two-soliton solutions. For structures with  $C_4$  and  $D_4$  symmetries and weak gyrotropic properties, an intensive energy exchange between the modes takes place and under certain conditions the fast mode becomes unstable.

### 1. Introduction

Solitary waves (solitons) formed of interacting quasiparticles in condensed media have been widely investigated. They have been predicted and studied for vibron [1–5], exciton [6–8], polariton [9–12] and magnon [13–15] excitations. Almost all investigations to date refer to solitons associated with non-degenerate excitations such as those with polarization normal to the optical axis in uniaxial gyrotropic crystals. A challenging task is the study of solitons formed of degenerate excitations in gyrotropic crystals. The latter exhibit natural optical activity expressed in general by a different response to right and left circularly polarized light. It is manifested in optical rotation and circular dichroism, light scattering, luminescence and fluorescence (for comprehensive reviews of molecular optical activity, see e.g. [16, 17]).

Traditionally, optical activity has been studied in the visible and the near-ultraviolet regions where it is associated with electronic transitions. Vibrational optical activity, studied more recently, is related to transitions between the vibrational levels of the molecules and provides valuable information for biochemistry and biophysics. In [18] a theory of the non-linear dynamics of intramolecular vibrations in uniaxial gyrotropic crystals was developed. The transverse normal modes propagating along the optical axis are circularly polarized. The chirality of the structure yields different dispersion laws and hence different phase and group velocities for the left- and right-handed modes. The anharmonicity of the vibrations leads in general to three types of non-linear interaction between the conjugate circular modes. The first one is between excitations with the same handedness and governs the formation of isolated partial solitons. The other two describe non-linear cross-interactions between the modes. One is elastic and conserves the energy of each mode, while the other is inelastic and allows energy exchange between the modes. Reference [18] focuses on the effects associated with the elastic cross-interaction which is present in chiral structures with arbitrary axial symmetry. It is shown that under certain conditions it can lead to coupling of the conjugate partial pulses

into one- or two-soliton bound states. The present paper extends the investigations of [18] by taking into account the inelastic cross-interaction between the partial solitons, which is present in gyrotropic crystals belonging to point group  $C_4$  or  $D_4$ . This interaction can influence considerably the bound soliton states as well as the processes of scattering of partial solitons.

The organization of the paper is as follows. In section 2 the non-linear equations of motion which govern the dynamics of conjugate circular solitons are derived. In the semi-continuum limit they transform into a system of coupled non-linear Schrödinger equations. The role of the elastic non-linear cross-interaction for the partial soliton dynamics is reviewed in section 3. Analytical solutions for the unbound and bound one-soliton states are presented, as well as numerical solutions corresponding to bound two-soliton states. A qualitative explanation of the observed effects is given. The role of the inelastic (exchange) cross-interaction for the evolution of conjugate partial pulses is studied in detail in section 4. An approximate analytical bound soliton solution is obtained in the case of weak exchange interaction. The evolution and instabilities of the partial pulses for strong inelastic interaction are studied numerically. The main results of the paper are summarized in section 5.

## 2. General consideration

In the present paper we shall investigate the dynamics and interactions of solitons associated with circular vibrations propagating along the optical axis in uniaxial gyrotropic crystals. Biaxial crystals and uniaxial crystals in directions different from the optical axis exhibit linear birefringence and linear dichroism which are 2–3 orders of magnitude stronger than the circular dichroism and can obscure the effects of the latter. Thus the crystal classes which favour the observation of circular dichroism are the trigonal classes 32 and 3, the tetragonal classes 4,  $\bar{4}$  and 422, the hexagonal classes 6 and 622, and the cubic classes 23 and 432. For wave vectors  $k = 0$  the normal modes transform within the irreducible representations of the point group of the crystal. The optical phonons are then either non-degenerate (A mode) with polarization along the optical axis, or doubly degenerate (E mode), polarized perpendicularly to the optical axis. For  $k \neq 0$  the degeneracy is split linearly in  $k_z$  where  $z$  is directed along the optical axis [19]. The dipole and multipole moments associated with the two modes are also different.

From a microscopic point of view the optical activity is related to the chirality (dissymmetry) of the structure which excludes improper rotation axes, i.e. centres of inversion, reflection planes and rotation–reflection axes. Thus optical activity in crystals may derive from inherent molecular chirality and/or from chirality due to the arrangements of the molecules along a helix within the unit cell. In the first case the molecules are essentially three-dimensional and dissymmetric (with an internal chiral structure), and their dimensions cannot be neglected compared to the wavelength of light. In the second case the molecules can be achiral and the gyrotropy arises from the crystalline structure. The microscopic theory of both types of optical activity in molecular crystals in the excitonic part of the spectrum has been developed in [20]. Vibrational circular dichroism of the second type has been observed in [21].

In the present paper we shall consider for simplicity gyrotropic crystals with one molecule per unit cell where the gyrotropy originates from the chiral structure of the molecules. With some effort the present investigation can be extended to crystals with more than one molecule per unit cell and to chiral polymers. We shall also consider the case where the symmetry point group of the molecules coincides with that of the crystal.

Let  $q_r$  and  $q_l$  be two normal modes of E-mode optical phonons with  $k = 0$  which transform as  $(x + iy)$  and  $(x - iy)$ . Following [22] we can introduce creation and annihilation operators for right ( $A_n^\dagger, A_n$ ) and left ( $B_n^\dagger, B_n$ ) circular vibrations in molecule  $n$ , which are related to the

corresponding normal coordinates and conjugate momenta through

$$\begin{aligned} q_{rn} &= (A_n^\dagger + B_n)/\sqrt{2} & q_{ln} &= (B_n^\dagger + A_n)/\sqrt{2} \\ p_{rn} &= (B_n^\dagger - A_n)/\sqrt{2} & p_{ln} &= (A_n^\dagger - B_n)/\sqrt{2}. \end{aligned} \quad (1)$$

The operators  $A_n$  and  $B_n$  obey the usual boson commutation rules:

$$\begin{aligned} [A_n, A_m^\dagger] &= [B_n, B_m^\dagger] = \delta_{n,m} \\ [A_n, B_m^\dagger] &= [A_n, B_m] = [A_n, A_m] = [B_n, B_m] = 0. \end{aligned} \quad (2)$$

The energy operator of molecule  $n$  in the harmonic approximation is

$$H_n = \hbar\omega_0(A_n^\dagger A_n + B_n^\dagger B_n) \quad (3)$$

where  $\omega_0$  is the frequency of the degenerate modes. The intramolecular vibrations in crystals are usually strongly localized and possess narrow energy bands. They can be described well within the Heitler–London approximation [20]. Thus the resonant interaction between molecules  $n$  and  $m$  associated with exchange of left and right circular vibrations can be written as

$$H_{nm} = M_{nm}^r (A_n^\dagger A_m) + M_{nm}^l (B_n^\dagger B_m). \quad (4)$$

The matrix elements  $M_{nm}^r$  and  $M_{nm}^l$  in general do not coincide. They include parts corresponding to the electric dipole–electric dipole intermolecular interaction, which are real, symmetric and equal for right and left circular excitations ( $\text{Re } M_{nm}^r = \text{Re } M_{mn}^r = \text{Re } M_{nm}^l = \text{Re } M_{mn}^l = M_{nm}^s$ ). In chiral systems, however, there are also non-vanishing parts of the matrix elements corresponding to the electric dipole–magnetic dipole interaction and the electric dipole–electric quadrupole interaction. These parts are imaginary, antisymmetric and change sign for excitations with different handedness ( $\text{Im } M_{nm}^r = -\text{Im } M_{mn}^r = \text{Im } M_{nm}^l = -\text{Im } M_{mn}^l = M_{nm}^{as}$ ) [20, 16]. Thus the intermolecular interaction operator can be rearranged as

$$H_{nm} = M_{nm}^s (A_n^\dagger A_m + B_n^\dagger B_m) + iM_{nm}^{as} (A_n^\dagger A_m - B_n^\dagger B_m). \quad (5)$$

The difference between the intermolecular interactions for left and right circular excitations is the cause of the optical activity within our model. The magnetic dipole and the electric quadrupole moments are much smaller than the electric dipole moment, and hence  $|M_{nm}^{as}/M_{nm}^s| \ll 1$ . Thus the gyrotropic effects are in general weaker than the dispersion effects associated with  $M_{nm}^s$ ; however, for short pulses and large distances they can cause considerable changes in the evolutionary patterns.

The anharmonicity of the intramolecular vibrations is usually weak and in the non-linear part of the Hamiltonian we can keep only the phonon-conserving terms [23]. The lowest-order terms which obey this condition are quartic. In order to write them down explicitly, we have to take into account the axial symmetry of the system. The normal coordinates  $q_{rn}$  and  $q_{ln}$  transform as the basis functions of a pair of complex-conjugate representations of the symmetry point group of the crystal. Thus the product  $q_{rn}q_{ln}$  and all of its powers are invariant in all point groups with axial symmetry. For this reason, the anharmonic part of the Hamiltonian for uniaxial gyrotropic crystals will always contain quartic terms of the type  $q_{rn}^2 q_{ln}^2$ . With the help of (1) this gives

$$H_{anh} = (g_1/2) \sum_n (A_n^\dagger A_n^\dagger A_n A_n + B_n^\dagger B_n^\dagger B_n B_n) + g_2 \sum_n A_n^\dagger B_n^\dagger A_n B_n. \quad (6)$$

It is important to note that this anharmonic part of the Hamiltonian commutes with the number-of-particles operators of each mode  $\sum_n A_n^\dagger A_n$  and  $\sum_n B_n^\dagger B_n$ . The first term in (6) describes non-linear interaction between quasiparticles of the same type. The second term in (6) describes an elastic interaction (scattering) of conjugate quasiparticles in which their individual numbers are conserved.

However, it is not difficult to see that in crystals belonging to the point groups  $C_4$  or  $D_4$ , anharmonic terms of the type  $q_{rn}^4$  and  $q_{ln}^4$  are also invariant. With the help of (1) they yield the following anharmonic Hamiltonian:

$$H'_{anh} = (g_3/2) \sum_n (A_n^\dagger A_n^\dagger B_n B_n + B_n^\dagger B_n^\dagger A_n A_n). \quad (7)$$

This Hamiltonian commutes with the operator of the total number of particles  $\sum_n (A_n^\dagger A_n + B_n^\dagger B_n)$  but not with the individual number-of-particles operators. Thus (7) describes non-linear cross-interactions in which quasiparticles of one type are transformed into quasiparticles of the other type and vice versa. These interactions allow energy exchange between the two conjugate modes and will be called inelastic.

The total Hamiltonian for circular vibrations in gyrotropic crystals belonging to point groups  $C_4$  or  $D_4$  thus becomes

$$\begin{aligned} H = \hbar\omega_0 \sum_n (A_n^\dagger A_n + B_n^\dagger B_n) + \sum_{n,m} [(M_{nm}^s + iM_{nm}^{as}) A_n^\dagger A_m + (M_{nm}^s - iM_{nm}^{as}) B_n^\dagger B_m] \\ + (g_1/2) \sum_n (A_n^\dagger A_n^\dagger A_n A_n + B_n^\dagger B_n^\dagger B_n B_n) + g_2 \sum_n A_n^\dagger B_n^\dagger A_n B_n \\ + (g_3/2) \sum_n (A_n^\dagger A_n^\dagger B_n B_n + B_n^\dagger B_n^\dagger A_n A_n). \end{aligned} \quad (8)$$

Further on we shall consider a one-dimensional case with nearest-neighbour interaction. The non-vanishing matrix elements of the intermolecular interaction operator can be represented as

$$\begin{aligned} M_{nn+1}^s = M_{n+1n}^s = M \\ M_{nn+1}^{as} = -M_{n+1n}^{as} = -\gamma \quad M, \gamma \text{ real.} \end{aligned} \quad (9)$$

The equations of motion for the operators  $A_n$  and  $B_n$  are

$$\begin{aligned} i\hbar \frac{\partial A_n}{\partial t} = \hbar\omega_0 A_n + M(A_{n+1} + A_{n-1}) - i\gamma(A_{n+1} - A_{n-1}) \\ + g_1 A_n^\dagger A_n A_n + g_2 B_n^\dagger B_n A_n + g_3 A_n^\dagger B_n B_n \\ i\hbar \frac{\partial B_n}{\partial t} = \hbar\omega_0 B_n + M(B_{n+1} + B_{n-1}) + i\gamma(B_{n+1} - B_{n-1}) \\ + g_1 B_n^\dagger B_n B_n + g_2 A_n^\dagger A_n B_n + g_3 B_n^\dagger A_n A_n. \end{aligned} \quad (10)$$

Averaging (10) with a wave function involving on-site coherent states:

$$\begin{aligned} |\Psi(t)\rangle = \prod_n |\alpha_n(t)\rangle \otimes |\beta_n(t)\rangle \\ A_n |\alpha_n(t)\rangle = \alpha_n(t) |\alpha_n(t)\rangle \quad B_n |\beta_n(t)\rangle = \beta_n(t) |\beta_n(t)\rangle \end{aligned} \quad (11)$$

we obtain the following system of coupled non-linear equations for the complex amplitudes  $\alpha_n(t)$  and  $\beta_n(t)$ :

$$\begin{aligned} i\hbar \frac{\partial \alpha_n}{\partial t} = \hbar\omega_0 \alpha_n + M(\alpha_{n+1} + \alpha_{n-1}) - i\gamma(\alpha_{n+1} - \alpha_{n-1}) + (g_1 |\alpha_n|^2 + g_2 |\beta_n|^2) \alpha_n + g_3 \alpha_n^* \beta_n^2 \\ i\hbar \frac{\partial \beta_n}{\partial t} = \hbar\omega_0 \beta_n + M(\beta_{n+1} + \beta_{n-1}) + i\gamma(\beta_{n+1} - \beta_{n-1}) + (g_1 |\beta_n|^2 + g_2 |\alpha_n|^2) \beta_n + g_3 \beta_n^* \alpha_n^2. \end{aligned} \quad (12)$$

The soliton dynamics is determined completely by equations (12) which have been solved numerically. We have considered wide solitons ( $L \gg 1$ ) for which discreteness effects are negligible and we have used periodic boundary conditions. Discreteness effects for narrow solitons have been analysed explicitly in [7].

An analytical solution of (12) in the case of weak anharmonicity and long pulses can be sought in the form of amplitude-modulated waves with slowly varying envelopes:

$$\begin{aligned} \alpha_n(t) &= e^{i(k_1 n - \omega_1 t)} \varphi_n(t) \\ \beta_n(t) &= e^{i(k_2 n - \omega_2 t)} \psi_n(t) \end{aligned} \tag{13}$$

where  $k_{1,2}$  and  $\omega_{1,2}$  are the wavenumbers and the frequencies of the carrier waves (the lattice constant equals unity). Within the semi-discrete approach [24] the system (12) reduces to

$$\begin{aligned} i\hbar \frac{\partial \varphi}{\partial t} &= (\epsilon_1 - \hbar\omega_1)\varphi + b_1 \frac{\partial^2 \varphi}{\partial x^2} - 2i \frac{\partial b_1}{\partial k_1} \frac{\partial \varphi}{\partial x} + (g_1 \varphi^2 + g_2 \psi^2)\varphi + g_3 e^{2i(\Delta k x - \Delta\omega t)} \psi^2 \varphi \\ i\hbar \frac{\partial \psi}{\partial t} &= (\epsilon_2 - \hbar\omega_2)\psi + b_2 \frac{\partial^2 \psi}{\partial x^2} - 2i \frac{\partial b_2}{\partial k_2} \frac{\partial \psi}{\partial x} + (g_1 \psi^2 + g_2 \varphi^2)\psi + g_3 e^{-2i(\Delta k x - \Delta\omega t)} \varphi^2 \psi \end{aligned} \tag{14}$$

where

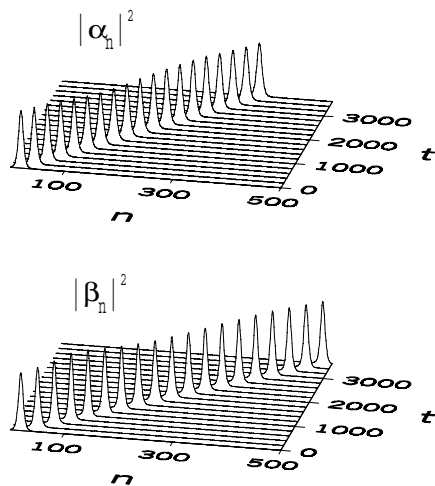
$$\begin{aligned} \epsilon_{1,2} &= \hbar\omega_0 + 2b_{1,2} & b_{1,2} &= M \cos k_{1,2} \pm \gamma \sin k_{1,2} \\ \Delta k &= k_2 - k_1 & \Delta\omega &= \omega_2 - \omega_1. \end{aligned} \tag{15}$$

### 3. Effects of the elastic cross-interaction

Without the cross-interaction terms ( $g_2 = g_3 = 0$ ), the system (14) decomposes into two uncoupled non-linear Schrödinger equations for the conjugate circular amplitudes. They possess the following bright-soliton solutions:

$$\begin{aligned} \varphi(x, t) &= \varphi_0 \operatorname{sech} \frac{x - v_1 t}{L_1} & \psi(x, t) &= \psi_0 \operatorname{sech} \frac{x - v_2 t}{L_2} \\ \varphi_0^2 &= \frac{2b_1}{gL_1^2} & \psi_0^2 &= \frac{2b_2}{gL_2^2} & \hbar\omega_1 &= \epsilon_1 + \frac{b_1}{L_1^2} & \hbar\omega_2 &= \epsilon_2 + \frac{b_2}{L_2^2} \\ b_1 &= M \cos k + \gamma \sin k & b_2 &= M \cos k - \gamma \sin k \\ v_1 &= -2\hbar^{-1}(M \sin k - \gamma \cos k) & v_2 &= -2\hbar^{-1}(M \sin k + \gamma \cos k) \end{aligned} \tag{16}$$

where  $\varphi_0, \psi_0; L_1, L_2;$  and  $v_1, v_2$  are the amplitudes, widths and velocities of the two solitons. This solution describes non-interacting conjugate pulses, propagating with equal carrier wavenumbers and different velocities and shapes (figure 1).



**Figure 1.** Evolution of non-interacting partial pulses with  $k_1 = k_2 = 0.6$ ,  $M = -0.1$  and  $\gamma = 0.01$ . The length is measured in lattice constants and the time in  $\omega_0^{-1}$ .

When an attractive elastic non-linear cross-interaction between the pulses exists ( $g_2 < 0$ ,  $g_3 = 0$ ), the system (14) possesses an exact analytical solution of the form:

$$\begin{aligned}\varphi(x, t) &= \varphi_0 \operatorname{sech} \frac{x - vt}{L} \\ \psi(x, t) &= \psi_0 \operatorname{sech} \frac{x - vt}{L}\end{aligned}\quad (17)$$

with the following relations between the soliton parameters:

$$\begin{aligned}\Delta k &= 2 \arctan(-\gamma/M) & b_1 = b_2 = b & \epsilon_1 = \epsilon_2 = \epsilon \\ \omega_1 = \omega_2 &= \hbar^{-1} \left( \epsilon + \frac{b}{L^2} \right) & \varphi_0^2 = \psi_0^2 &= \frac{2b}{L^2(g_1 + g_2)} \\ v &= -2\hbar^{-1}(M \sin k_1 - \gamma \cos k_1) = -2\hbar^{-1}(M \sin k_2 + \gamma \cos k_2).\end{aligned}\quad (18)$$

The solution (17), (18) describes a soliton bound state which consists of partial pulses with identical shapes and velocities and different carrier wavenumbers. Depending on the values of the parameters and the initial amplitudes, a linearly polarized pulse can evolve to either an unbound or a bound one- or two-soliton state for the partial pulses.

The evolutionary pattern is controlled by the balance between the kinetic energy of the relative motion of the pulses

$$U_{kin} = -\frac{2\gamma^2 \cos k}{M} N_e \quad (19)$$

and the potential energy of their elastic interaction

$$U_{pot} = \frac{2}{3} g_2 \varphi_0^2 N_e \quad (20)$$

where

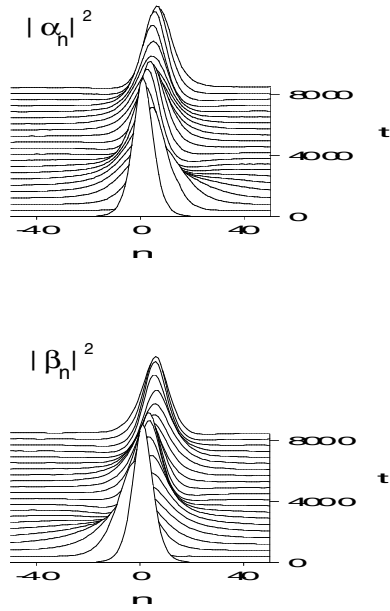
$$N_e = \int_{-\infty}^{\infty} \varphi^2(x, t) dx = \int_{-\infty}^{\infty} \psi^2(x, t) dx \quad (21)$$

gives the numbers of excited vibrational quanta of each circular mode which we have considered equal. This corresponds to the physically important case of a linearly polarized initial pulse whose evolution we have studied numerically. The condition for bound soliton states is

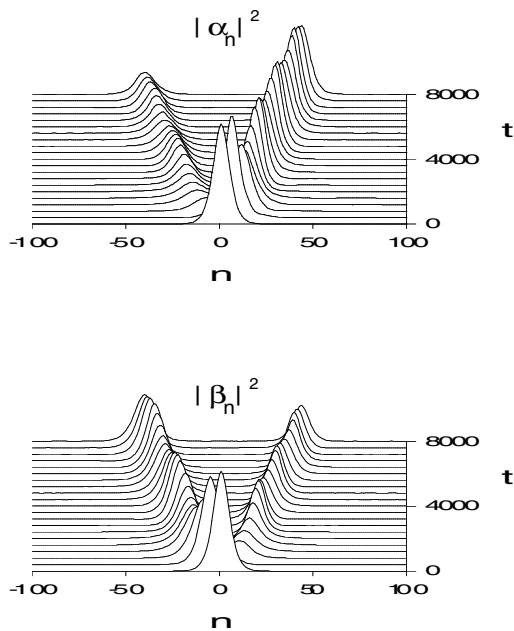
$$R = \left| \frac{U_{kin}}{U_{pot}} \right| = \left| \frac{3\gamma^2 \cos k}{g_2 M \varphi_0^2} \right| < 1. \quad (22)$$

In the case of strong gyrotropy and weak elastic cross-interaction, when the kinetic energy dominates over the potential one, an initial linearly polarized pulse decomposes into two circularly polarized pulses propagating with different velocities (16). In the opposite case, when the potential energy dominates ( $R < 1$ ), the partial pulses are locked together to form a soliton bound state (figure 2). The wavenumbers of the pulses are modified according to the analytical solution (18) and the energy in each mode is conserved. The coupling process is accompanied by amplitude and position oscillations of the partial pulses, and while the amplitude oscillations seem to decay over time, the position ones which are of the order of a few lattice constants are long-lasting. Similar breather-like behaviour of the coupled solution has been obtained in [25] for solitons in birefringent optical fibres.

In the intermediate region ( $R \sim 1$ ), where the kinetic energy nearly equals the potential one, the initial pulse decomposes into two pairs of coupled partial pulses (figure 3), which can be classified as a two-soliton bound state. This process can be explained qualitatively in the following way: the potential energy in this case is not sufficient for the coupling of the pulses into one-soliton bound states and a walk-off effect takes place. However, due to



**Figure 2.** Evolution of interacting partial pulses with initial  $k_1 = k_2 = 0.6$  into a bound one-soliton solution for  $M = -0.1$ ,  $\gamma = 0.01$  and  $g_1 = g_2 = -0.005$ . The frame of reference is moving with velocity  $v$ .



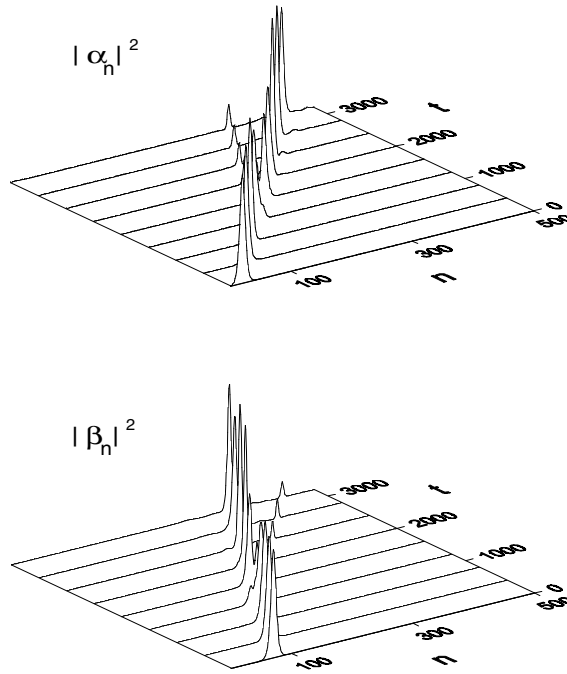
**Figure 3.** Evolution of the initial partial pulses into a two-soliton bound state for  $\gamma = 0.015$ . All other parameters are the same as in figure 2.

the non-linear cross-interaction, a portion of the energy splits off each partial pulse in the form of small-amplitude soliton which is locked to the conjugate large-amplitude soliton. The relative velocity of the two pairs is smaller than that of the uncoupled solution. Weaker initial amplitudes lead to weaker coupling and a strongly asymmetric two-soliton bound state and for  $R \gg 1$  the evolution corresponds to the uncoupled one-soliton solution.

We have investigated the stability of the partial solitons against collision in the presence of an attractive elastic cross-interaction. For  $g_2 = g_1$  and  $g_3 = 0$  the system (14) is completely



integrable [26, 27] and the solitons emerge from the collision process unchanged except for a phase shift. For  $|g_2| > |g_1|$  the integrability of (14) is destroyed and a two-soliton bound state is formed after the collision as shown in figure 4 ( $g_2 = -0.02$ ,  $g_1 = -0.005$ ). The evolution is similar to the one in figure 3; however, a larger potential energy is necessary in this case to overcome the relative stability of the uncoupled partial pulses.



**Figure 4.** Formation of a two-soliton bound state in a collision process with  $\gamma = 0.01$ ,  $g_1 = -0.005$  and  $g_2 = -0.02$ .

#### 4. Effects of the exchange cross-interaction

As mentioned above, in crystals belonging to the point groups  $C_4$  or  $D_4$  an additional non-linear interaction between the conjugate normal modes exists, which allows energy exchange between the modes. Vibrational circular dichroism in  $\alpha$ -NiSO<sub>4</sub>·6H<sub>2</sub>O and  $\alpha$ -ZnSeO<sub>4</sub>·6H<sub>2</sub>O crystals, belonging to the point group  $D_4$ , has been observed in [21], so in principle the soliton interactions studied in the present section can also be observed in such crystals. As the non-linear interactions between the pulses act only while substantial parts of them overlap, it can be expected that the exchange cross-interaction will considerably affect the bound soliton states but be less important for the unbound ones. This interaction yields a spatially periodic non-linear potential with a period  $\pi/\Delta k$ , which leads to amplitude modulation and energy exchange between the partial solitons. In the case of a weak-exchange cross-interaction ( $|g_3| \ll |g_1|, |g_2|$ ), we can seek an approximate solution of equations (14) in the form

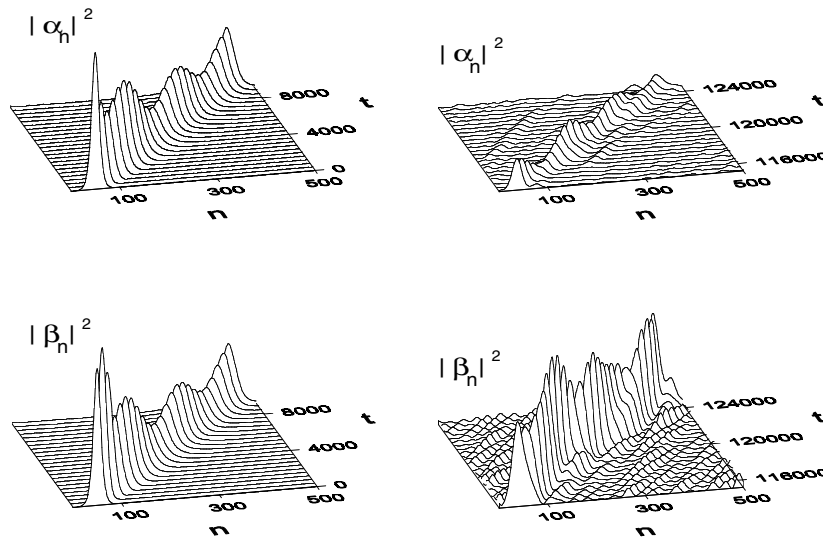
$$\begin{aligned}\varphi(x, t) &= \varphi_0(1 + \varphi_1(x, t)) \operatorname{sech} \frac{x - vt}{L} \\ \psi(x, t) &= \psi_0(1 + \psi_1(x, t)) \operatorname{sech} \frac{x - vt}{L}\end{aligned}\quad (23)$$

where  $\varphi_0$  and  $\psi_0$  are the bound soliton solutions (18) and  $\varphi_1$  and  $\psi_1$  are small compared to unity. Substituting (23) into (14) and keeping only terms linear in  $\varphi_1$  and  $\psi_1$ , the following solution can be obtained:

$$\begin{aligned}\varphi_1(x, t) &= \frac{g_3}{b_1} L^2 \psi_0^2 \cos(2 \Delta k x) \operatorname{sech}^2 \frac{x - vt}{L} \\ \psi_1(x, t) &= \frac{g_3}{b_2} L^2 \varphi_0^2 \cos(2 \Delta k x) \operatorname{sech}^2 \frac{x - vt}{L}.\end{aligned}\quad (24)$$

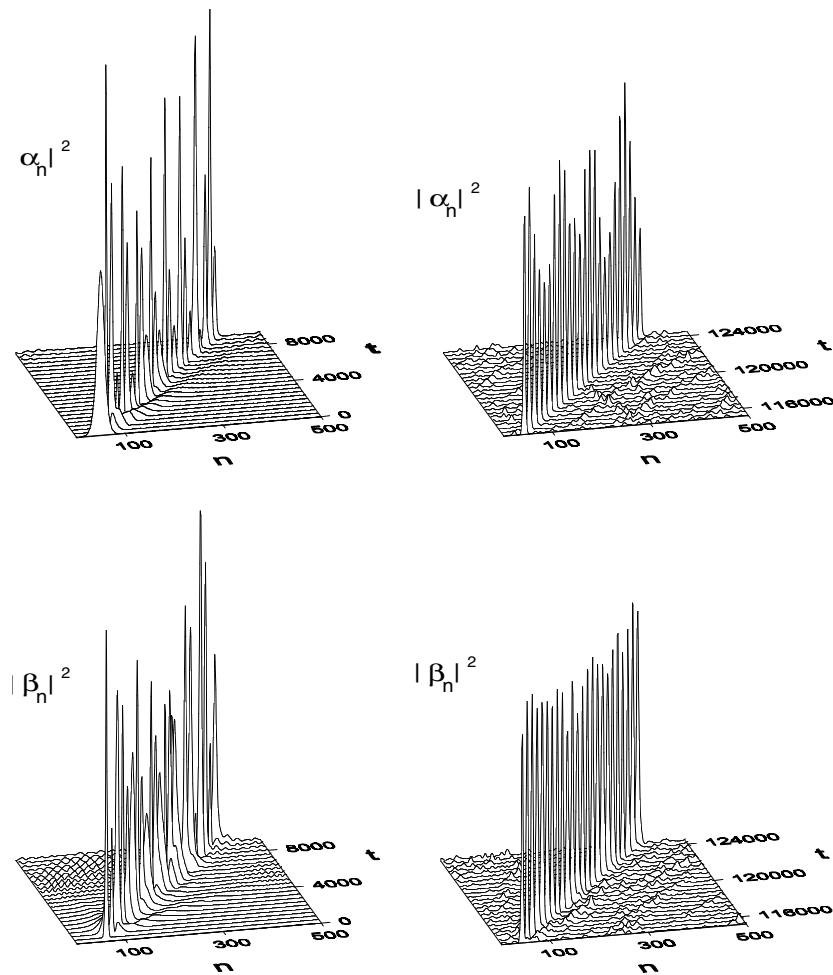
In the case of strong gyrotropy, the modulation length is short compared with the soliton width and the spatially oscillating cross-interaction terms are averaged over the soliton period. The coupled soliton solution is not affected by the exchange interaction and remains stable.

Much more interesting and of physical importance is the case of weak gyrotropy. The modulation length in this case is longer than the soliton width and this leads to periodic oscillations of the solitons' amplitudes and intensive energy exchange between the pulses. Figure 5 shows the evolution of a pair of conjugate partial pulses with  $\gamma = 0.001$ ,  $\Delta k = 0.02$  and equal amplitudes. Strong oscillations of the amplitudes of the partial pulses are observed in the initial moments. After some time, the fast mode (the one with larger phase velocity) decays, transferring most of its energy to the slower one. When only a fast-mode partial pulse is launched, it transfers most of its energy into a slow-mode soliton and decays. When a slow-mode pulse is launched it remains practically stable. Thus in all cases of  $N = 1$  initial amplitudes, the fast partial pulse becomes unstable over time and decays, while the slow one remains stable.



**Figure 5.** Instability of the fast mode due to the exchange interaction in the case of weak gyrotropy ( $\gamma = 0.001$ ) when two conjugate pulses are launched with  $g_1 = g_2 = g_3 = -0.005$ ,  $k_1 = 0.20$  and  $k_2 = 0.22$ .

A different picture is observed in the case of pulses with larger initial amplitudes corresponding to  $N = 2$  solutions. When a two-soliton fast pulse is launched (figure 6), it quickly excites the conjugate partial soliton and both modes remain stable and with equal amplitudes over large distances. Similar evolutionary patterns are observed when an  $N = 2$  slow partial pulse is launched, or when both modes are excited with two-soliton amplitudes. In all cases of  $N = 2$  amplitudes, strong oscillations are observed in the initial moments and a considerable



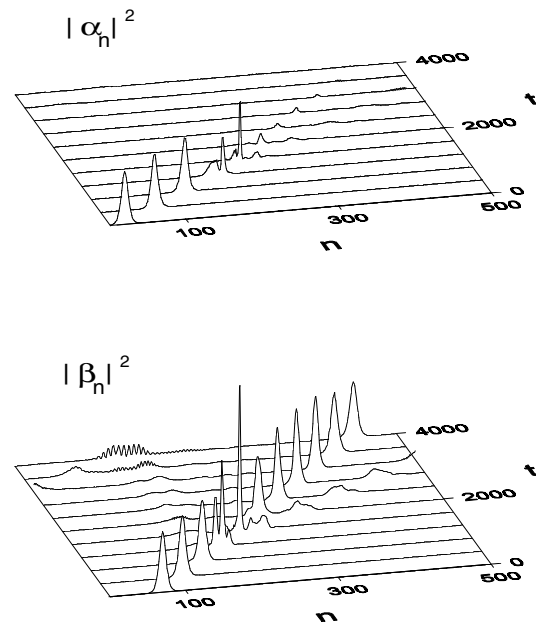
**Figure 6.** Redistribution of the energy and stability of the soliton bound state when an  $N = 2$  fast-mode pulse is launched. All other parameters are the same as in figure 5.

compression of the pulses takes place. Similar effects with interchanged space and time domains have been obtained in [25, 28–31] for the interaction of linearly polarized solitons in birefringent optical fibres and for elliptically polarized solitons in gyrotropic materials [32].

We investigated also the stability of the partial solitons against collisions in the presence of an exchange cross-interaction. For weaker interaction ( $|g_3| \sim |g_1|, |g_2|$ ), both solitons emerge from the collision process with only minor distortions. For stronger interaction however, the faster soliton transfers most of its energy to the slower one during the collision and decays (figure 7). To our knowledge this is the first demonstration of such an instability.

## 5. Conclusions

The dynamics and the stability of conjugate circularly polarized envelope solitons in gyrotropic molecular crystals are governed by the parameters of the initial pulses (amplitudes, widths, carrier wavenumbers) and the material parameters which determine the dispersion and



**Figure 7.** Instability of the faster partial soliton in a collision with a slower one in the case of strong exchange interaction.  $\gamma = 0.01$ ,  $g_1 = g_2 = -0.005$ ,  $g_3 = -0.02$  and  $k_1 = k_2 = 0.6$ .

the interactions of the partial waves. A group-theoretical analysis yields the allowed non-linear terms in the Hamiltonian for the different point groups of symmetry. The presence of gyrotropy tends to separate the pulses, while an attractive elastic interaction tends to keep them together. The balance between the kinetic energy of the relative motion of the solitons and the potential energy of their elastic interaction yields the possible evolutionary patterns—coupled or uncoupled one-soliton solutions when stronger inequalities hold, and coupled two-soliton solutions in the intermediate region where the kinetic energy nearly equals the potential one. Our results show that for weak gyrotropy and strong attraction, an initial linearly polarized pulse will propagate as a single pulse with a rotating plane of polarization, while in the opposite case it will decompose into a pair of conjugate circularly polarized pulses propagating with different velocities. In the intermediate case the initial pulse will decompose into a pair of elliptically polarized pulses with opposite handedness, walking off. Bound soliton states can also be formed in collision processes in the case of strong attraction.

The inelastic (exchange) cross-interaction, present in systems with  $C_4$  and  $D_4$  symmetries, is crucial for the coupled soliton solutions in the physically important case of weak gyrotropy. It leads to an instability of the fast mode for  $N = 1$  initial conditions. For  $N = 2$  initial conditions, both modes remain stable and the energy is redistributed equally between them. A strong exchange cross-interaction can also lead to an instability and decay of the faster soliton in a collision with a slower soliton with opposite handedness.

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