ASYMPTOTIC BI-SOLITON IN DIATOMIC CHAINS

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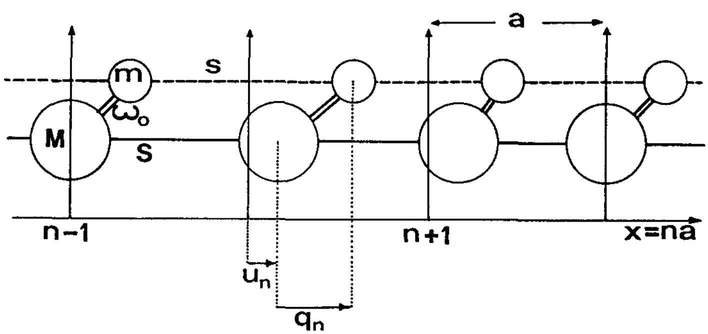
Abstract

The light scattering in a diatomic chain of nonlinearily coupled oscillators is studied on the basis of classical Hamiltonian equation of motion in the continuum limit. The basic process is a *localized Brillouin scattering* and we prove that the nonlinear interaction of the light-wave with the phonon-wave results in a strong localization and a mutual trapping of the acoustic wave and the reflected light wave. This is shown to corresponds to the exchange of a given *acoustic particle* whose energy and momentum depend only on the elastic parameters of the chain. We conclude that the nonlinear coupling induces the existence of a new energy level which value does not depend on the initial condition or any other external constraint or parameter. The asymptotic state consists in a sonic wave front followed by two localized structures which eventually coalease onto the wave front.

This report is a shortened version of [1] completed with a more detailed discussion of the asymptotic behavior of the generic solution and some figures.

We study the nonlinear effects in one-dimensional sytems of coupled oscillators, which is an essential problem in physics and biophysics (hydrogen-bond systems) [2]. Many phenomena (proton conductivity in ice [3], anomalous infrared absorption in crystalline acetanilide (ACN) [4], lossless energy transport along alpha-helical proteins [5]) are suspected to be related to the nonlinear nature of the interaction of two different types (HF vs BF) of vibrations along quasi one-dimensional chains of atoms or molecules.

We consider here the classical model of a diatomic chain of nonlinearily coupled oscillators represented as the following diatomic chain (the vertical axis represent a reference equilibrium frame), with the three elastic parameters $\omega_0, S/M$ and s/m:



The oscillators u_n (low frequency) represent the motion of the molecule in the crystal (acoustic phonons) and they are associated with the hamiltonian:

$$H_L = \frac{1}{2}\sum_n M\dot{u}_n^2 + S(u_{n+1} - u_n)^2 + S(u_n - u_{n-1})^2 \tag{1}$$

where M is the mass and S the spring constant accounting for the hydrogen bond. The index L stands for *Lattice.*

The oscillators q_n (high frequency) represent the motion of one atom (or group of atoms) of the molecule (optical mode) and they are associated with the hamiltonian:

$$H_V = \frac{1}{2}\sum_n m[\dot{q}_n^2 + \omega_0^2 q_n^2] + s(q_{n+1} - q_n)^2 + s(q_n - q_{n-1})^2 \tag{2}$$

where ω_0 is the frequency of the isolated oscillator and where the spring constant s allows for the propagation of the vibration q_n along the chain. Here the index V stands for *Vibration.*

Both oscillators are nonlinearily coupled through the following interaction hamiltonian [5, 6, 7]

$$H_I = \frac{1}{2}\sum_n \frac{C_1}{2}(u_{n+1} - u_{n-1})q_n^2 + C_2[(u_{n+1} - u_n)q_{n+1} + (u_n - u_{n-1})q_{n-1}]q_n \tag{3}$$

where C_1 and C_2 are the interaction constants.

The equations of motion result in the following coupled system

$$M\ddot{u}_n = -\frac{\partial H}{\partial u_n}, \quad m\ddot{q}_n = -\frac{\partial H}{\partial q_n}, \quad H = H_L + H_V + H_I. \tag{4}$$

The masses M and m, together with the spring constants S and s, are in quite different scales which allows us to define a scaling parameter ϵ as (see also (12) below):

$$\epsilon^2 = \frac{S}{s}\frac{m}{M}. \tag{5}$$

Now we can precisely state the problem we are concerned with: describe the nonlinear dynamics of the scattering of light (the incident radiation is represented by a forced oscillation of q_n at, say, $n = +\infty$) at a frequency close to ω_0, in the case when $\epsilon \ll 1$.

The process which we consider is the Brillouin back-scattering of an incident wave (ω, k) according to the following selection rules:

$$\omega = \Omega + \omega_r, \qquad k = K + k_r, \tag{6}$$

where (Ω, K) is the sound wave (with dispersion law $\Omega = vK$ for small wave numbers, consistently with the continuum limit later adopted) and (ω_r, k_r) is the back-scattered wave (therefore $k_r \simeq -k$, and hence $K \simeq 2k$).

We will prove that the reflected wave is localized, and hence the back-scattered wave is not directly observable, bounded to the acoustic wave, and that the three waves do obey (6) according to the following actual distribution:

$$\begin{array}{ccccc} \omega & = & \Omega^{(0)} - \Omega_B & + & \omega_r^{(0)} + \Omega_B \\ k & = & K^{(0)} - K_B & + & k_r^{(0)} + K_B \\ \textit{Incident} & & \textit{Acoustic} & & \textit{Reflected} \end{array} \tag{7}$$

where we have defined

$$\Omega^{(0)} = vK^{(0)}, \quad K^{(0)} = 2k, \quad \omega_r^{(0)} = \omega - \Omega^{(0)}, \quad k_r^{(0)} = -k, \tag{8}$$

$$\Omega_B = 2\epsilon^2\omega, \qquad K_B = \Omega_B/v. \tag{9}$$

The word *incident* indicates a wave propagating from right to left (the input zone is $n = +\infty$), and ω is the *free* parameter.

As a consequence, the nonlinear coupling of the vibration q_n with the acoustic phonon u_n results in the exchange of the energy Ω_B and momentum K_B (obeying the dispersion law of the acoustic wave), which then can be thought of as representing a *binding acoustic particle.* The shift in frequency ($\Omega_B \simeq 2\epsilon^2\omega_0$) depends *only* on the elastic constants of the chain and *not* on the coupling constants or any other adjustable parameter as the energy of the acoustic soliton.

Starting with the equation of motion (4), we first go to the continuum limit and obtain the following system of coupled wave equations:

$$q_{tt} - c^2 q_{xx} + \omega_0^2 q = -\alpha u_x q, \tag{10.a}$$

$$u_{tt} - v^2 u_{xx} = \beta q_x q, \tag{10.b}$$

where we have defined

$$c^2 = a^2\frac{s}{m}, \; v^2 = a^2\frac{S}{M}, \; \alpha = (C_1 + 2C_2)\frac{a}{m}, \; \beta = (C_1 + C_2)\frac{a}{M}, \tag{11}$$

and we have now

$$\epsilon \; = \; \frac{v}{c} \; \Rightarrow \; \Omega_B = 2(\frac{v}{c})^2\omega. \tag{12}$$

The above system is identical to the equation discussed in [7] where it is obtained without the approximation of a nearest-neighbour interaction.

The usual approach of a coupled system like (9-10) is to assume a quasi stationary acoustic wave $u(x,t)$ so that $u_x \propto q^2$. Then the slowly varying envelope of the wave $q(x,t)$ evolves according to the nonlinear Schrödinger equation (NLS). As a consequence, if a soliton solution to NLS is assumed (first problem: the mechanism of the *soliton creation* is not described) it has a non-zero phase and the resulting phase of the wave $q(x,t)$ is shifted from its value $\Omega^{(0)}$ of an amount depending on the soliton energy (second problem: the soliton energy is a *free* parameter).

Here we perform a multiscal analysis [8] on the system (9-10) by looking for small amplitude, slowly varying envelope solutions under the form:

$$q(x,t) = \epsilon a_1(\xi,\tau)\exp[i\phi_1] + \epsilon a_2(\xi,\tau)\exp[i\phi_2] + c.c. +$$

$$+ \sum_{n=2}^{\infty} \epsilon^n a_1^{(n)}(\xi,\tau)\exp[in\phi_1] + \epsilon^n a_2^{(n)}(\xi,\tau)\exp[in\phi_2] + c.c. \tag{13.a}$$

$$u(x,t) = \epsilon\Psi_1(\xi,\tau)\exp[i\phi] + \epsilon^2\Psi_0(\xi,\tau) +$$

$$+ \sum_{n=2}^{\infty} \epsilon^n \Psi_n(\xi,\tau)\exp[in\phi] + c.c. \tag{13.b}$$

$$\phi_1 = \omega t + kx, \; \phi_2 = \omega_r t + k_r x, \; \phi = \Omega t + Kx$$

$$\xi = \epsilon x, \ \tau = \epsilon^2 t. \tag{13.c}$$

Note that the dispertion relations for the wave $q(x,t)$ read

$$\omega^2 = \omega_0^2 + c^2 \ k^2, \tag{14}$$

$$\omega_r^2 = \omega_0^2 + c^2 \ k_r^2. \tag{15}$$

The above expressions (13) are inserted in (10) and the leading orders in ϵ give, after averaging over fast oscillations (which means practically: look at the coefficients of $\exp[i\phi]$):

$$\Psi_\tau - c\Psi_\xi = \gamma a_1 a_2^*,$$
$$a_{1,\xi} = \Psi a_2, \qquad a_{2,\xi} = \Psi^* a_1. \tag{16}$$

We have set hereabove

$$\Psi = \frac{\alpha}{c^2}\Psi_1, \quad \gamma = \frac{\alpha\beta'}{2c^3}, \quad \beta = \epsilon\beta', \tag{17}$$

(the last definition is consistent with (11) and (5)), and we have used the selection rules (6) together with (14), (15) and $\Omega = vK \equiv \epsilon cK$ from the very definition of ϵ.

The relation (14) determines k from the input ω. The relation (15) together with (6) (and $\Omega = vK$) can be solved and we obtain

$$K = [2k - 2\epsilon^2\frac{\omega}{v}][1 - 2k\epsilon^2]^{-1}, \tag{18}$$

which implies the relations (7),(8) and (9) for small k, at order $k\epsilon^2$.

Since we are dealing with the problem of the absorption of an incident (electromagnetic) wave (ω, k) at, say $x = +\infty$, with given amplitude A, we complete the system (14) with the following boundary values

$$a_1(\xi,\tau) \to A, \ (\xi \to +\infty), \quad a_2(\xi,\tau) \to 0, \ (\xi \to -\infty), \tag{19}$$

and an initial condition $\Psi(\xi, 0)$ in $L^2(\Re)$. It is worth remarking that, due to (19), the system (16) *implies* the property

$$\Psi(\xi,0) \to 0, (\xi \to \pm\infty) \Rightarrow a_2(\xi,0) \to 0, (\xi \to +\infty). \tag{20}$$

The essential point is now that the system (16) with boundary values (19) is *integrable* in the sense of the spectral transform theory [9] (it has actually a structure similar to that of the well known "self- induced- transparency" equation governing the light pulse propagation in a two level system [10]). In the spectral transform theory, the system (16) is said to have a *singular dispersion law* [11] and we have developed the general formalism for such classes of nonlinear coupled evolutions on the basis of the $\bar\partial$ problem formulation of the spectral transform [12] (the most general class of integrable evolutions of type (16) is displayed in [13]).

We have proved in [2] that, for any initially localized initial acoustic wave $\Psi(\xi, 0)$, the general solution of (16) has the following *universal* asymptotic form for $\tau \to +\infty$: it vanishes for $\xi + c\tau < 0$, and, for $\xi + c\tau > 0$ it obeys

$$a_1 \to \ A[1 + |\eta(\xi,\tau)|^2][1 - |\eta(\xi,\tau)|^2]^{-1}, \tag{21}$$

$$a_2 \to 2iA\eta(\xi,\tau)[1-|\eta(\xi,\tau)|^2]^{-1}, \tag{22}$$

$$\Psi \to -4i\sqrt{\frac{\Delta\tau}{\xi+c\tau}}\,\bar{\eta}(\xi,\tau)[1-|\eta(\xi,\tau)|^2]^{-1}, \tag{23}$$

where the function $\eta(\xi,\tau)$ is given by

$$\eta(\xi,\tau) = 2i\rho\frac{\sqrt{\pi y}}{\Delta\tau}[\sum_{n=1}^{N-1} A_n y^{-n} + \mathcal{O}(y^{-N})]\ \exp[y/2],$$

$$A_0 = 1,\quad A_1 = -27/16,\quad A_2 = 8385/2^9,\quad A_3 = -2589825/2^{13},$$

$$y(\xi,\tau) = 8\sqrt{\Delta\tau(\xi+c\tau)},\quad \Delta = |A|^2\gamma/4. \tag{24}$$

It is then an easy task to go back to the physical solution $u(x,t), q(x,t)$ of the system (10) by using successively (17), (13) and (12). So we have at first order in ϵ and for large t and for $x > -vt$:

$$q(x,t) = \epsilon A \exp[i(\omega t + kx)]\frac{1+|\eta|^2}{1-|\eta|^2}+$$

$$+2i\epsilon A \exp[i(\omega_r^{(0)} + \Omega_B)t + i(k_r^{(0)} + K_B)x]\frac{\eta}{1-|\eta|^2} + \text{c.c.} \tag{25}$$

$$u(x,t) = -4i\epsilon\frac{c^2}{\alpha}\sqrt{\frac{\Delta\tau}{\xi+c\tau}}\exp[i(\Omega^{(0)} - \Omega_B)t + i(K^{(0)} - K_B)x]\frac{\bar{\eta}}{1-|\eta|^2} + \text{c.c.} \tag{26}$$

while for $x < -vt$

$$q(x,t) = \epsilon A \exp[i(\omega t + kx)],\quad u(x,t) = 0. \tag{27}$$

The above behaviour of the solution allows us to deduce the following properties of the system of coupled wave equations (10):

1- The frequencies and wave numbers of the waves q and u do obey the relation (7): the scattered part of the electromagnetic wave q and the acoustic wave u have exchanged the *acoustic particle* (Ω_B, K_B).

2- Although these asymptotic behaviours present singularities (for $|\eta|^2 = 1$), the solution (q,u) itself is never singular (the saddle point expansion becomes exact only for t strictly infinite).

3- The localized wave $u(x,t)$ has been *created* out of the continuous spectrum and it has the structure of a sonic wave front followed by two supersonic localized coherent structures which eventually reach the wave front. The scattered light wave is bounded to these localized structures.

We have drawn in the figures 1 to 3 the acoustic wave $u(x,t)$ in the rest frame of sound wave (moving at velocity $-v$) for the following choices of the parameters:

$$v = 3800ms^{-1},\quad c = 56000ms^{-1},\quad K = 100cm^{-1}$$

$$\gamma|A|^2 = 0.1\ S.I.,\quad \alpha c^{-2} = 1\ S.I.\quad \rho = 0.01$$

at different times (from 10 to 10^4).

These parameters have been chosen for representing the crystal of acetanilide. These values imply that the nonlinear binding mode has a frequency of $15cm^{-1}$. Actually the value of c (velocity of the propagation of the Amide-I vibration) has been inferred from the value of Ω_B. The good point is that this value of c lies in the range of acceptable values.

For the fig. 1 we have taken the saddle point expansion at order 1 (that is $N = 1$ in (24)), for the fig. 2 at order 2 and 3 for the fig. 3. Then it is clear that indeed there is a bisoliton (or bicaviton) stucture, which appears better at order three.

Finally the proof that the expansion converges as $t \to \infty$ can be inferred from the figure 4 where we have drawn (solid lines) the curves

$$f_N(y) = [\sum_{n=1}^{N} A_n y^{-n}]^2\, y \tag{28}$$

(see (24)) for different N. The dashed line is the curve

$$(\frac{\gamma|A|^2}{8\rho\sqrt{\pi}})^2 \tau^2 e^{-y}. \tag{29}$$

The intersections of (28) with (29) give the positions of the singularities of $u(x, t)$.

It is clear that all curves (28) have for $N \geq 1$ two asymptotes, and therefore that at any order *except the order* 0, there exists a time t_m such that, for $t > t_m$ the solution is as close as we want to the solution at next order. This proves both the convergence of the series and the fact that the asymptotic solution consists *always* in **two** nonlinear coherent structures, see also [14].

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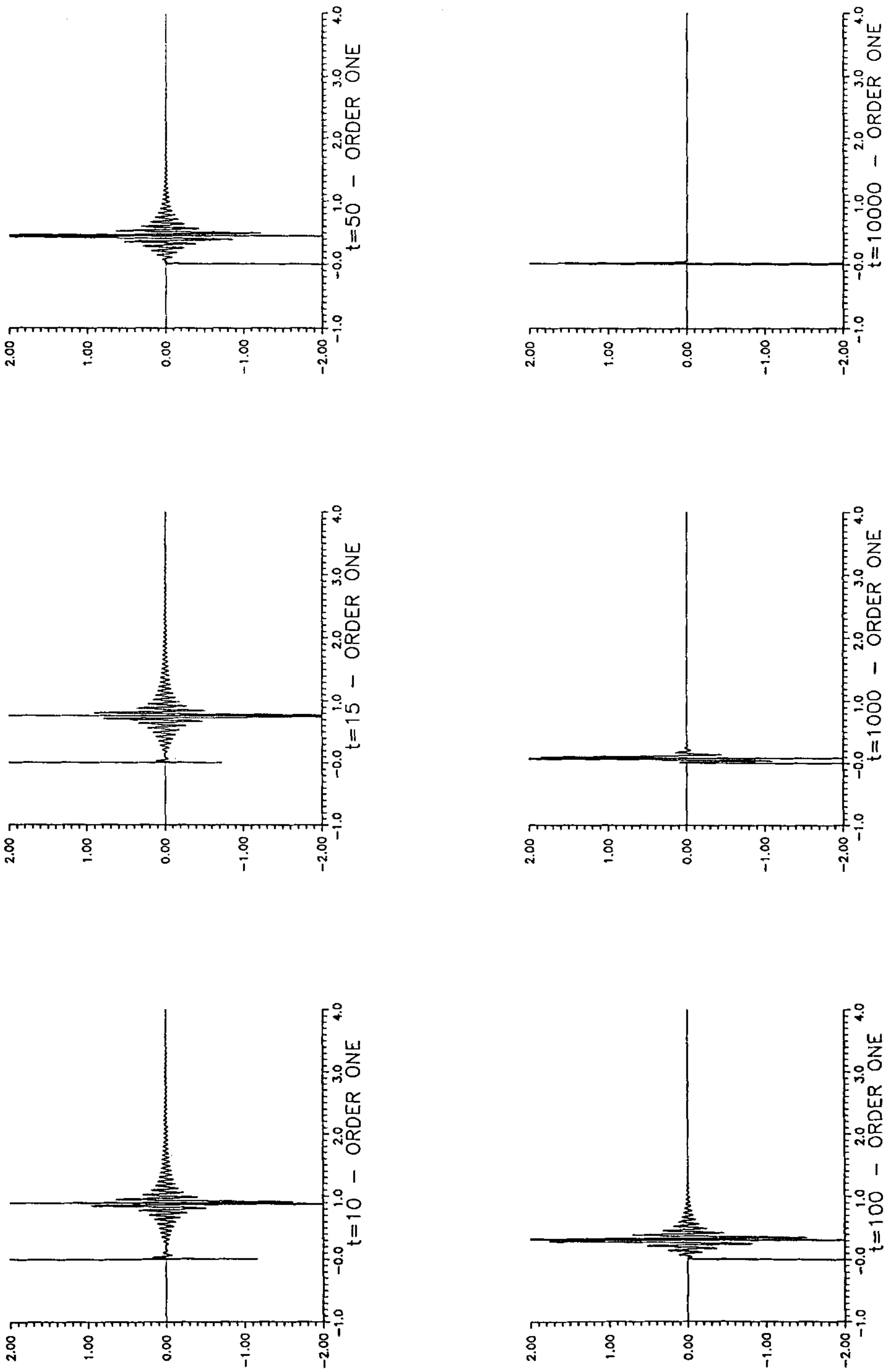
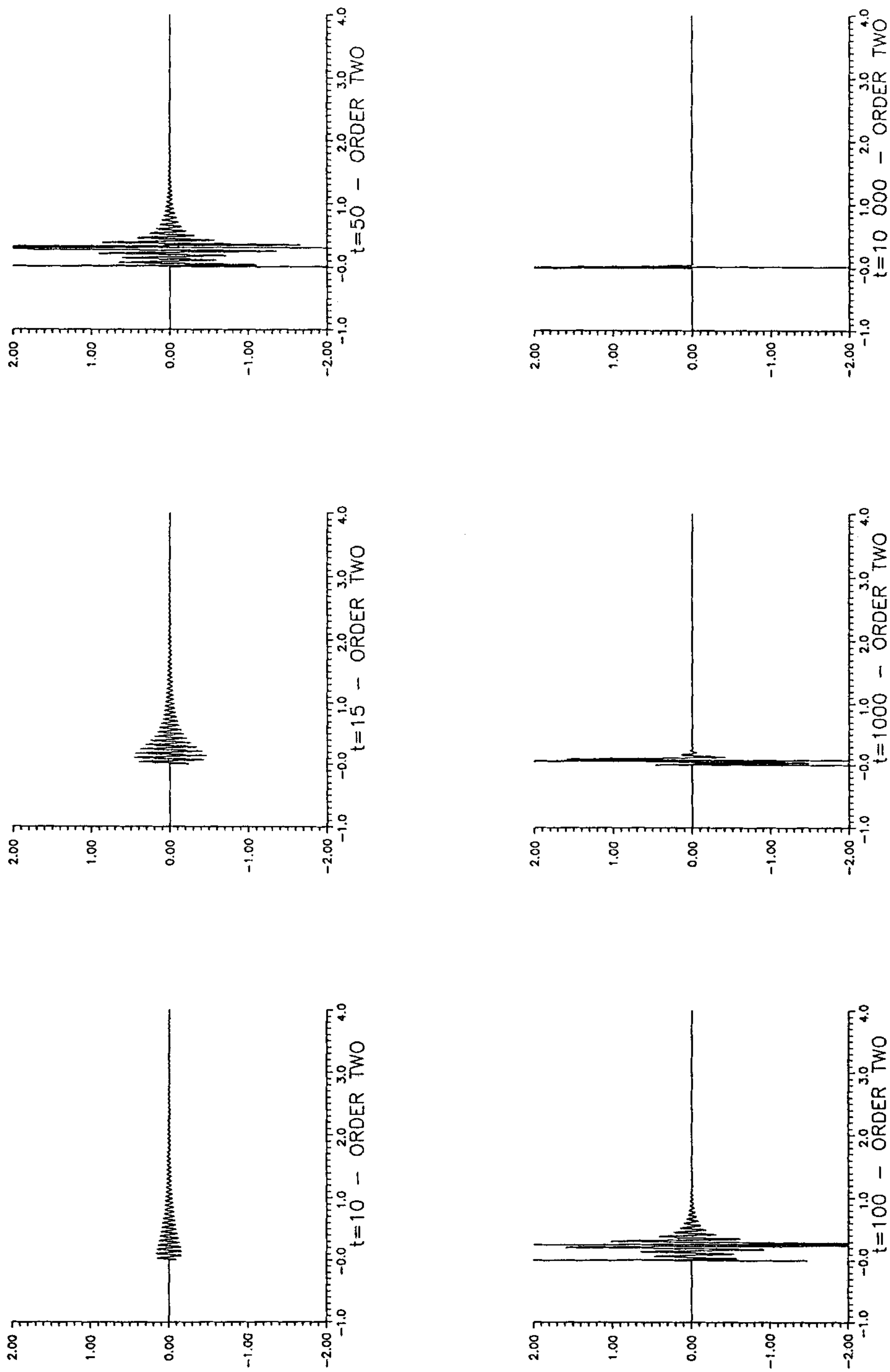


t=50 - ORDER ONE
t=15 - ORDER ONE
t=10 - ORDER ONE
t=10000 - ORDER ONE
t=1000 - ORDER ONE
t=100 - ORDER ONE
2.00
1.00
0.00
-1.00
-2.00
-1.0
-0.0
1.0
2.0
3.0
4.0

FIG. 1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t=15 - ORDER TWO
t=10 - ORDER TWO
t=10 000 - ORDER TWO
t=1000 - ORDER TWO
t=100 - ORDER TWO

FIG. 2

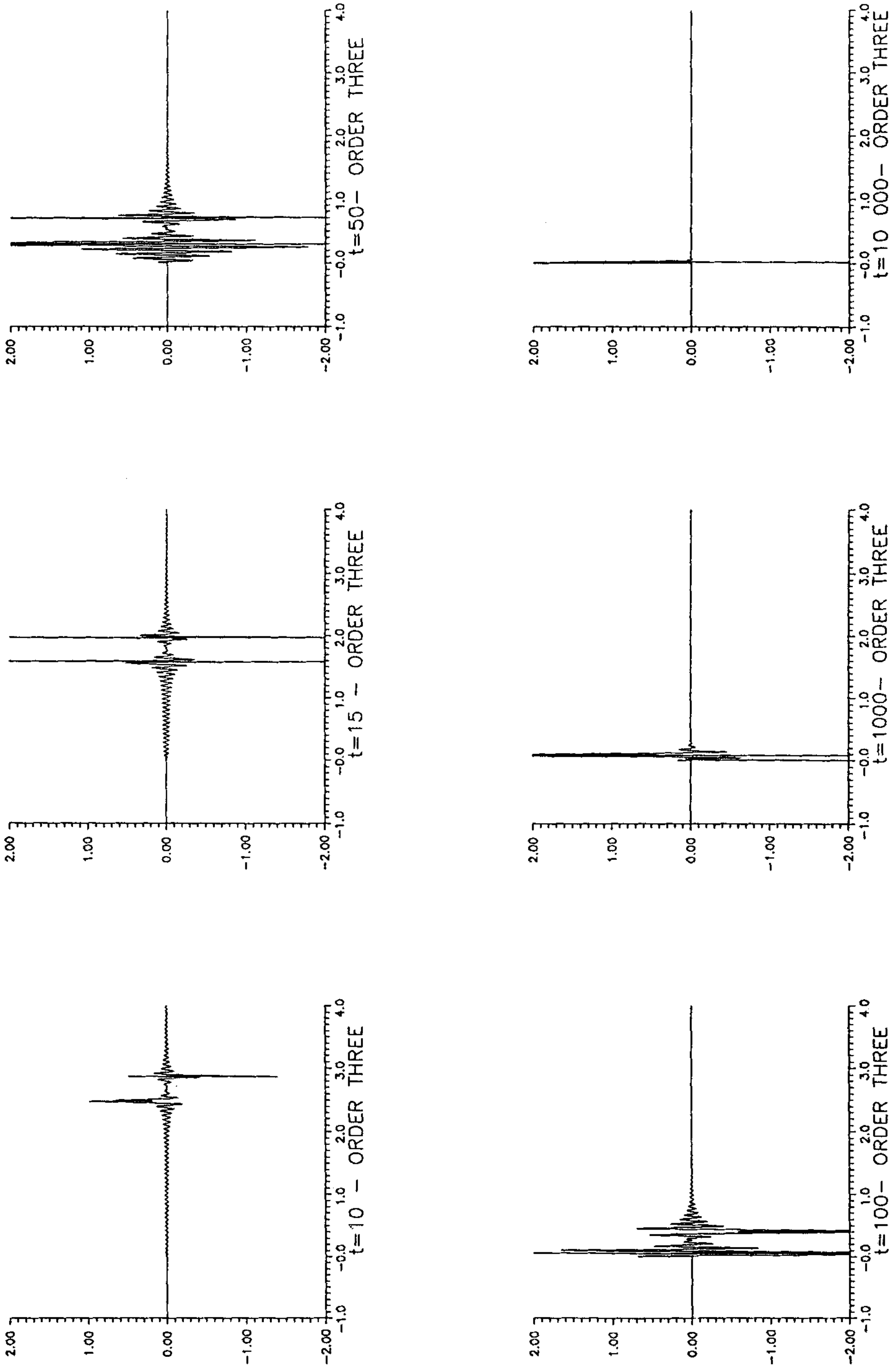
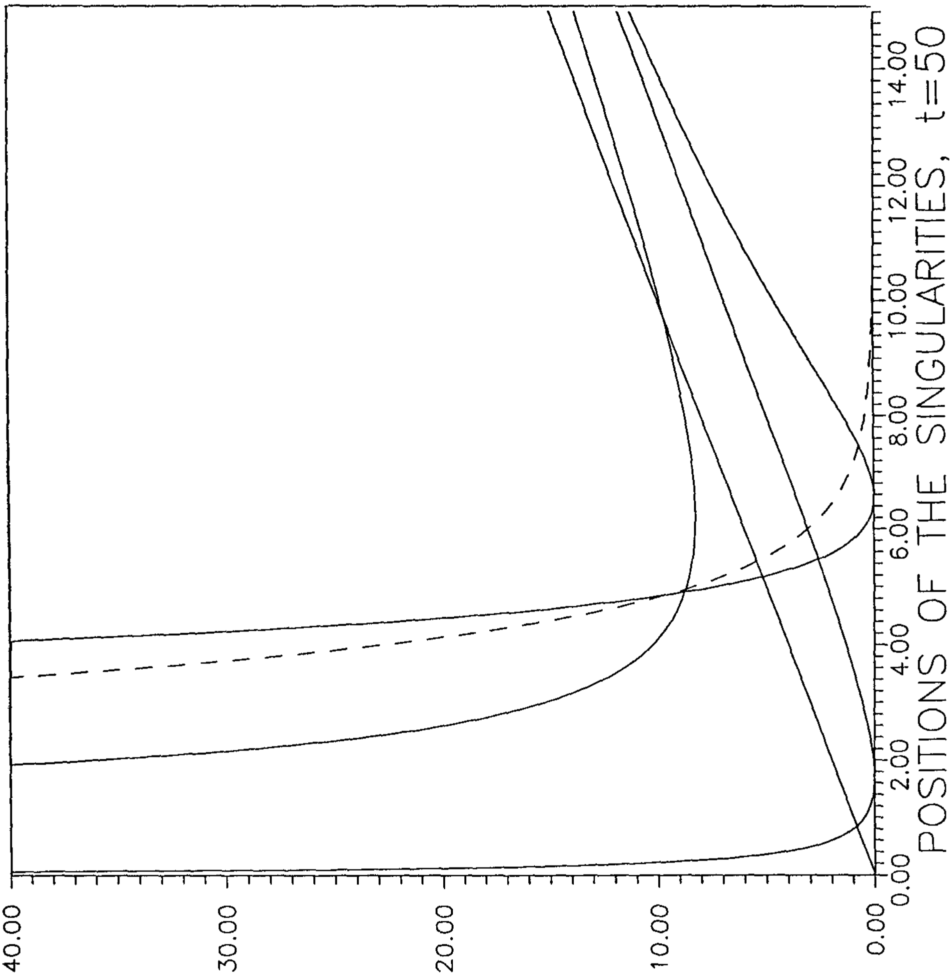

t=10 - ORDER THREE
t=15 - ORDER THREE
t=50- ORDER THREE
t=100- ORDER THREE
t=1000- ORDER THREE
t=10 000- ORDER THREE
2.00
1.00
0.00
-1.00
-2.00
-1.0
-0.0
1.0
2.0
3.0
4.0

FIG. 3


POSITIONS OF THE SINGULARITIES, t=50
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6.00
8.00
10.00
12.00
14.00
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10.00
20.00
30.00
40.00

FIG 4