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INITIAL CONDITION EFFECTS IN THE EVOLUTION OF A NONLINEAR DIMER ☆

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The initial state analysis of the evolution of a nonlinear degenerate dimer shows that, in addition to the self-trapping transition, a new transition occurs while the particle is in the trapped region. This transition can be understood in part in terms of the behavior of a linear nondegenerate dimer, and is intimately related to the stationary states of the nonlinear dimer.

The discrete nonlinear Schrödinger equation [1–6], which describes the dynamics of quasiparticles in several nonlinear media, is

$$\mathrm{i}\,\mathrm{d}c_m/\mathrm{d}t=\sum_n V_{mn}c_n-\chi|c_m|^2c_m\,, \tag{1}$$

where c_m is the probability amplitude to find the particle at site m, V_{mn} is the intersite matrix element and χ is the nonlinearity parameter that describes an energy lowering of the particle when at site m due to polaronic or other nonlinear effects. The equation was recently solved [2] for a system involving two identical molecules, i.e. a dimer, and the consequences of the nonlinear evolution in experimentally observable quantities such as the neutron scattering function were studied [3]. The inital condition explored in these studies was that of extreme initial localization, i.e. one wherein the particle is initially entirely on one of the two sites of the dimer. In the present paper we discuss the motion of the particle in the more general case of arbitrary initial placement. We show that, as the nonlinearity and the initial conditions are varied, the particle undergoes two transitions. One of these is the "frequency transition", studied in ref. [2], that occurs when the frequency of the intersite oscillations becomes zero and the particle gets self-trapped. The other is a new "amplitude transition" that happens while the particle is in the trapped region. It is characterized by a decrease in the amplitude of the (trapped) motion until it becomes zero and a subsequent increase but with opposite sign that corresponds to a reversal of the phase of the evolution. This new transition can be understood in terms of the behavior of a *linear nondegenerate* dimer, is intimately related to the analysis of the stationary states of ref. [1], and is the primary content of this note.

The general solution for the probability difference $P(t)\equiv\rho_{11}(t)-\rho_{22}(t)$ arising from (1) for a dimer of sites 1 and 2 can be expressed [2,3] in terms of the jacobian elliptic functions cn and dn as

$$\begin{aligned}p(t)&=C\,\mathrm{cn}[(C\chi/2k)(t-t_0)\,|\,k]\\&=C\,\mathrm{dn}[(C\chi/2)(t-t_0)\,|\,1/k]\,.\end{aligned} \tag{2}$$

General expressions for C, t_0 and k may be found in ref. [2]. Here we will assume $(\mathrm{d}p/\mathrm{d}t)_0=0$. This is equivalent to placing the dimer initially in a pure state characterized by *real* off-diagonal matrix elements of the density matrix ρ. The solution (2) then becomes

$$p(t)=p_0\,\mathrm{cn}(p_0\chi t/2k\,|\,k)=p_0\,\mathrm{dn}(p_0\chi t/2\,|\,1/k)\,, \tag{3}$$

$$k^2=\frac{k_0^2p_0^2}{1+2k_0r_0}\,, \tag{4}$$

p_0 and r_0 being the initial values of p and $\rho_{12}+\rho_{12}$, respectively, and k_0 being the nonlinearity ratio $\chi/4V$.

The initial (real) amplitudes $c_1(0)$ and $c_2(0)$ can

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either have the same sign or opposite signs. In the former case, $r_0=+(1-p_0^2)^{1/2}$ whereas, in the latter, $r_0=-(1-p_0^2)^{1/2}$. We consider these cases separately.

Case (I): $r_0=+(1-p_0^2)^{1/2}$. Eq. (4) shows that, upon varying k_0 (i.e. $\chi/4V$) from 0 (linear dimer) to the value $k_0^c=[1+(1-p_0^2)^{1/2}]/p_0^2$, the *elliptic modulus k* varies from 0 to 1. The system executes complete oscillations between the two dimer sites. The motion is appropriately described by the cn function with decreasing frequency as k_0 increases. When k attains the critical value k_0^c, $k=1$, the frequency of oscillation becomes zero and the time evolution is given by $p(t)=p_0\,\mathrm{sech}(p_0\chi t/2)$. As k_0 increases beyond k_0^c, the value of k becomes larger than 1, and the particle finds itself trapped on one of the two sites of the dimer. The evolution is then described by the dn function.

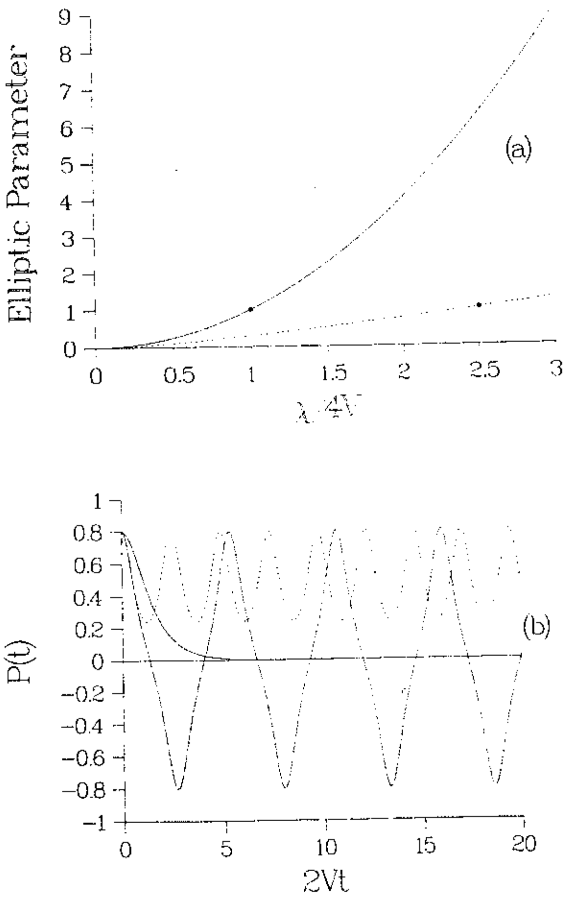

Fig. 1. $r_0>0$: In (a) the elliptic parameter k^2 is plotted as a function of $k_0=\chi/4V$ for $p_0=1$ (full line) and 0.8 (dotted line). The bullet (●) shows the position of the self-trapping transition. In (b) the time evolution of the probability difference $p(t)$ is plotted as a function of the time for $p_0=0.8$ and for different values of k_0: 2.3 (dashed line), 2.5 (full line) and 2.7 (dotted line).

While this behavior shows the "frequency transition" as in the analysis of Kenkre and Campbell [2], we see here how the amount of nonlinearity necessary for the self-trapping transition to occur varies with the initial condition. If initially the particle is completely localized ($p_0=1$), then $k_0^c=1$, if it is completely delocalized ($p_0=0$), $k_0^c=\infty$. In the latter case, self-trapping cannot occur. Furthermore, the more localized the particle is initially, the smaller is the degree of nonlinearity required to self-trap the particle. The explicit relation of χ and the initial conditions at the transition is given by (4) on putting $k=1$.

In fig. 1a, we plot the elliptic parameter k^2 as a function of $\chi/4V$. In fig. 1b, we plot the actual time evolution for several values of $\chi/4V$. The transition from free to self-trapped motion (cn to dn) is clearly visible.

Case (II): $r_0=-(1-p_0^2)^{1/2}$. This case is physically more interesting since it describes an entirely new behavior for the dimer while in the trapped region. The critical value k_0^c of k_0, for which $k=1$ is now $[1-(1-p_0^2)^{1/2}]/p_0^2$. As with the previous case, while k_0 remains smaller than k_0^c, the elliptic modulus is less than 1 and the particle executes periodic motion: $p(t)$ oscillates between p_0 and $-p_0$ following the cn function. When $\chi/4V$ attains the critical value k_0^c the self-trapping transition occurs, and for further increase of k_0 the particle is trapped (dn evolution). Unlike case (I), we see here, however, that a further increase in the nonlinearity χ can make $k_0=\frac{1}{2}(1-p_0^2)^{-1/2}$ which results in k becoming infinite. For this choice of the degree of nonlinearity and initial conditions the system finds itself initially in one of its stationary states [1,3,7]. The probability of occupation of the sites is then clearly time-independent.

A further increase of k_0 makes k^2 negative; upon applying the imaginary argument transformation [8] to the cn function in (3) we get

$$p(t)=p_0\,\mathrm{nc}[p_0\chi t/2|k|\,|(1+|k^2|)^{1/2}]\,. \tag{5}$$

Since the modulus of the nc function is greater than

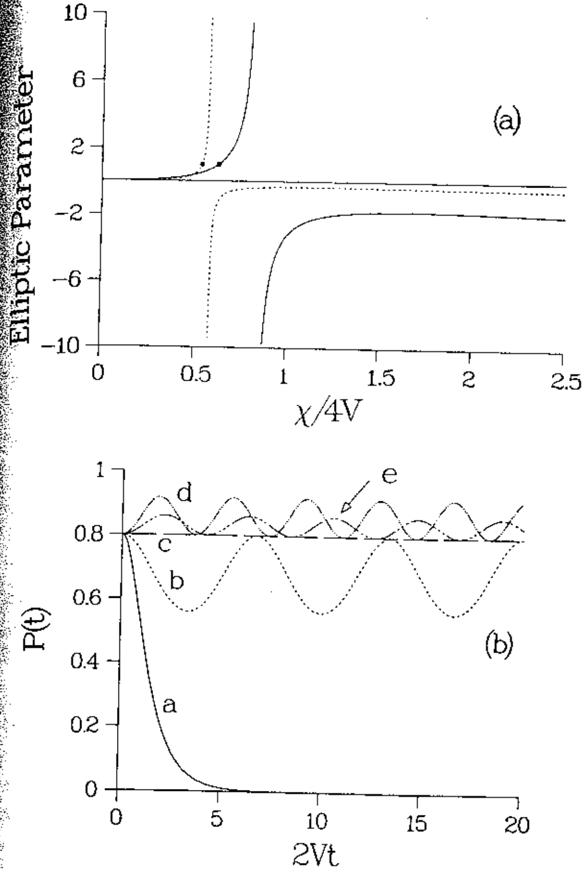

Fig. 2. $r_0<0$: In (a) the elliptic parameter k^2 is plotted as a function of $k_0=\chi/4V$ for $p_0=0.8$ (full line) and 0.5 (dashed line). The bullet (●) shows when the self-trapping transition occurs. In (b) $p_0=0.8$ and the values of k_0 are: (a) 0.625, self-trapping transition, (b) 0.7, the particle is trapped (dn-evolution), (c) 0.833, amplitude transition; the system occupies the stable stationary state, (d) 0.9 and (e) 1.0 (trapped-nd).

l for all these possible values of $|k|$, we now apply the reciprocal modulus transformation [8] to the nc function and finally arrive at the following expression for the time evolution:

$$p(t)=p_0 \times \mathrm{nd}[p_0\chi(1+|k^2|)^{1/2}t/2|k|\,|(1+|k^2|)^{-1/2}] \,. \quad (6)$$

While the particle continues to be trapped, the increase of k_0 beyond $\frac{1}{2}(1-p_0^2)^{-1/2}$ causes the elliptic parameter k^2 to increase to maximum whose value is $-p_0^2(1-p_0^2)^{-1}$ and then to decrease slowly to $-\infty$ as shown in fig. 2a. The time evolution as given by (6) is plotted in fig. 2b. Then cn→dn→nd transitions are displayed clearly.

We thus see that in case (II) an "amplitude" transition occurs on varying the degree of nonlinearity which is in addition to the "frequency" transition discussed in ref. [2]. The transition occurs in the self-trapped region of the nonlinear dimer and we have used the term "amplitude" transition to emphasize the fact that it entails the reduction of the amplitude of motion at the transition and its change in sign across it. The source of this additional transition can be understood through a comparison of the nonlinear dimer, for which the nonlinearity χ introduces an effective energy mismatch while in the trapped region [2], with the linear but nondegenerate dimer. For the latter, the difference in the probability of occupation of the two sites is given by:

$$p(t)=(1/\delta^2)[c_1\cos(2V\delta t)+c_2] \,, \quad (7)$$

$$c_1=p_0+(\Delta/2V)r_0 \,, \quad (8)$$

the quantities δ, c_2 and Δ being $c_2=(\Delta/2V)\times[p_0(\Delta/2V)-r_0]$, $\delta=[1+(\Delta/2V)^2]^{1/2}$ and the energy difference between the two sites respectively. Clearly for the value $\Delta/2V=-p_0/r_0=p_0/(1-p_0^2)^{1/2}$, $c_1=0$, $p(t)$ becomes equal to p_0 and the linear nondegenerate dimer is placed in one of its eigenstates. Further increase of $\Delta/2V$ changes the sign of c_1 and the motion reverses its phase. We note that a similar situation occurs and in the case of the nonlinear dimer where one of the stationary states involves a probability difference between the two sites, in sharp contrast to the linear (degenerate) case. The complete set of stationary states of the nonlinear dimer can be easily seen: one for $p_0=0$, and $r_0=+1$ and the other for $p_0=0$, $r_0=-1$ while $\chi<2V$. When $\chi>2V$ the stable stationary state can be reached for $r_0=-1/2k_0=2V/\chi$ and consequently for $p_0=[1-(2V/\chi)^2]^{1/2}$ [1,7].

The analysis that we presented for the initial condition dependence of the evolution of the nonlinear degenerate dimer proves itself extremely relevant in the case of a class of experiments that involve illumination of chemical solutions with linearly polarized light and the subsequent study of the fluorescence emission from the molecular aggregates that constitute the solution. In fact, as we will show in a forthcoming paper [9], the fluorescence depolarization experiments constitute a direct probe in the space defined through the different initial density matrix elements of the system and consequently identify an experimental domain where all the above described behavior can be directly tested.

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