INTERPLAY OF QUANTUM PHASES AND NON-LINEARITY
IN THE NON-ADIABATIC DIMER

V.M. KENKRE and H.-L. WU
Department of Physics and Astronomy, University of New Mexico, Albuquerque, NM 87131, USA

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We study the interplay of initial phase relations between the amplitudes in a dimer with the non-linearity in its evolution. The system investigated represents a moving quasiparticle interacting strongly with lattice vibrations. The study focuses on relaxing the traditional adiabaticity approximation, recovers earlier (adiabatic) results in appropriate limits and results in new findings.

Strong interactions with lattice vibrations can lead to profound modifications in the transport of quasiparticles such as electrons or excitons moving in solids. Recent approaches to this problem have been based on discrete non-linear quantum evolution equations [1–9] resulting, in most cases, from an “adiabatic” approximation in which it is assumed that the time scales of the two coupled processes, viz. vibrational relaxation and quasiparticle motion, are disparate. In a recent study [10], we have reported some novel consequences of dropping this adiabatic approximation. That study focused on initial conditions in which the quasiparticle is localized at a single site. In this Letter we remove the restriction of initial localization and examine the interplay of arbitrary initial quantum phases with the non-linearity in the evolution.

We begin with the coupled evolution equations for the moving quasiparticle and the vibrating oscillators used by Scott and his collaborators [1–3], modify the equation obeyed by the oscillators by introducing damping and removing dispersion to get

\[ i \frac{dc_n}{dt} = V(c_{n+1} + c_{n-1}) + E x_n c_n, \]  \[(1)\]

\[ d^2 x_n / dt^2 + \alpha (dx_n / dt) + \omega^2 x_n = - \text{const} \times |c_n|^2, \]

\[(2)\]

\[ \text{assume that the damping coefficient } \alpha \text{ in the vibrational equation (2) is large enough to justify the neglect of the second derivative of the } x \text{'s and consequently to characterize the evolution of the oscillators.} \]

\[ x \text{ towards their equilibrium positions through a single characteristic "vibrational relaxation" time, viz. } \alpha / \omega^2 = 1 / T, \text{ and restrict our study to the dimer in which } n \text{ takes on the values 1, 2 only. Our point of departure is thus} \]

\[ i \frac{dc_1}{dt} = V c_2 + E x_1 c_1, \]

\[ i \frac{dc_2}{dt} = V c_1 + E x_2 c_2, \]

\[ dx_1 / dt + \Gamma x_1 = - (\chi / E) |c_1|^2, \]

\[ dx_2 / dt + \Gamma x_2 = - (\chi / E) |c_2|^2, \]

\[(3)–(6)\]

where \( c_1 \) and \( c_2 \) are the respective amplitudes for the quasiparticle to be on the two sites 1 and 2. \( V \) describes the intersite transfer interaction for the quasiparticle, \( x_1 \) and \( x_2 \) are the two oscillator displacements, \( \Gamma \) is the (single) vibrational relaxation rate mentioned above, and \( E \) and \( \chi \) are constants.

The first step in our analysis is the conversion of (3)–(6) into evolution equations for the density matrix elements \( \rho_{11}, \rho_{22}, \rho_{12}, \rho_{21} \) which describe the quasiparticle, and the suitably normalized difference of the displacements \( y(t) = -(E / \chi) (x_1(t) - x_2(t)) \) which describes the oscillators. The quasiparticle equations are

\[ d\rho_{12} / dt = i V \rho_{12} + i x \rho_{21}, \]

\[ d\rho_{21} / dt = -i V \rho_{21} - i x \rho_{12}, \]

\[ dp / dt = 2i V \rho_{12} - \rho_{21}, \]

\[ \text{where } \rho_{12} \text{ and } \rho_{21} \text{ are the density matrix elements representing the probability of finding the quasiparticle on sites 1 and 2, respectively.}\]
where $p$ is the probability difference $p_{11}-p_{22}$. The next step is to obtain from them an equation for $y$, which is coupled to the evolution of $y$.

$$ \frac{dp(t)}{dt} + 4V^2 p = 2V \chi p(t) \left( \rho_{12} + \rho_{21} \right) \chi_0 $$

$$ -\chi y(t) \int_0^t ds \, y(s) \left[ \frac{dp(s)}{ds} \right] $$

$$ \chi \frac{dp(t)}{dt} + \Gamma y(t) = \Gamma p(t) $$

where the subscript 0 denotes the initial value.

If the vibrational relaxation rate $\Gamma$ in (11) is infinitely large, one may substitute $y = p$ in (11), notice that the integral can be evaluated to yield the characteristic cubic term in $p$ (see e.g. eq. (2.1) in ref. [8]), and obtain the results of the usual adiabatic analysis. We will present the (new) consequences of non-infinite $\Gamma$ in two stages. First we shall assume that, although non-vanishing, $\Gamma$ is smaller than all characteristic times in the system and present, on the basis of this assumption, an analytical investigation of the dynamics of the non-adiabatic dimer. We will then return to the case of arbitrary $\Gamma$ and present numerical solutions of the evolution.

The Laplace transform of (11) is

$$ \hat{y}(\epsilon) = \frac{y(0)}{\epsilon + \Gamma} + \frac{p(\epsilon)}{1 + \epsilon/\Gamma} \exp(-\epsilon/\Gamma). $$

If $\Gamma$ is smaller than all characteristic times of the system, and if one is interested in the evolution described by (12) only at long times ($t \gg 1/\Gamma$), one may expand $(1 + \epsilon/\Gamma)^{-1}$ in powers of $\epsilon/\Gamma$ and retain the lowest term. At long times, the initial value term in (12) may be neglected, to yield

$$ \hat{y}(\epsilon) \approx p(\epsilon) \exp(-\epsilon/\Gamma). $$

The oscillator displacement difference $y(t)$ thus follows the quasi-particle probability difference $p(t)$ with a phase lag.

$$ y(t) \approx p(t-1/\Gamma) \approx p(t) - \left(1/\Gamma \right)dp(t)/dt. $$

We then obtain a closed damped non-linear equation for the probability difference valid for arbitrary initial conditions but for long times:

$$ \frac{dp(t)}{dt} + D(dp(t)/dt) = A' p - Bp^3. $$

Eq. (15) differs from the adiabatic counterpart to be found in refs. [5,7,9] through: (i) the existence of the non-linear damping coefficient $D$, which, in the present case, ensures the evolution of the non-adiabatic dimer to stationary states, and (ii) the difference between $A'$ and its adiabatic counterpart $A$:

$$ D = \chi^2/2\Gamma \left[ (\rho_{12}^2 - \rho_{12}^0)^2 - (4V \chi)(\rho_{12} + \rho_{21}) \right]. $$

$$ A' = A + \frac{\chi^2}{\Gamma} \int_0^t ds \left[ \frac{dp(s)}{ds} \right]^2. $$

The particular case of initial localization on one of the two sites is described by $p_x = 1$ and $(\rho_{12} + \rho_{21})_0 = 0$, and has been analyzed in great detail in ref. [10], where it has also been shown why an equation such as (15) will drive the quasiparticle to its stationary state. Here, we shall find the stationary states explicitly from (15). For this purpose we take the system to be placed in one of those states initially so that $(\rho_{12} + \rho_{21})_0$ is the stationary value $\rho_{12} + \rho_{21}$ and put the $p$ derivatives to zero for all times. The result is that $A' = A$ and the stationary states are exactly those of the adiabatic dimer first found by Eilbeck et al. [3]. They satisfy $p^2 = A/B$ and their amplitudes $c_i$ and $c_2$ are given by [3,8]

$$ (1/\sqrt{2})(1 \pm [(1 - (2V \chi)^2)^{1/2}]/2). $$

We thus see that the non-adiabatic dimer is driven to the stationary states of the adiabatic dimer, no matter what the initial conditions are.

In order to view explicitly the approach of the non-adiabatic dimer to the stationary states of the adiabatic dimer, as well as other aspects of the evolution, we return to the exact equations of motion (3)–(6) and present in figs. 1–3 their numerically obtained solutions for arbitrary $\Gamma$ and for short as well as long times. Striking effects of the interplay of initial phases and non-linearity are observed.

In all the figures shown, the initial oscillator displacements are zero. In fig. 1, the non-linearity $\chi$ and the vibrational relaxation rate $\Gamma$ are 5 and 1 respectively (in units of $\chi$), and the initial value of the probability on one of the sites, $P_x(0)$, is 0.51. In fig. 1a, the two site amplitudes are initially in phase and in fig. 1b, they differ by $\pi$. In other words, $c_1(0) = \sqrt{0.51}$ in both cases but $c_2(0) = -\sqrt{0.49}$ in fig. 1a and $-\sqrt{0.49}$ in fig. 1b. Two features should be
noticed. The probability of the initially occupied site tends to its stationary state value 0.958 for long times in both cases. And the influence of the initial phase is pronounced: when the amplitudes are initially in phase, wild oscillations are seen in the probability. Fig. 1 also shows the oscillator displacement evolution (dotted lines) along with the probabilities (solid lines). The adiabaticity approximation would take the oscillators as following the probabilities instantaneously.

Whereas the extreme limits (0 and π) of the initial phases are considered in Fig. 1, the full intermediate variation is depicted in Fig. 2. The parameters are $V=2.5$, $\Gamma/V=10$, $P_1(0)=0.8$. The 3-d plot in Fig. 2 shows the considerable influence that initial phases have on the evolution. Among the features to be noticed are the approach to the stationary states for long times, the so-called “repulsion effect” derived in Refs. 9.

approach to the stationary state with $P_1(\infty) = 0.8$ after an initial increase in the oscillation amplitude. The dashed line (initial phase difference $\frac{1}{2}\pi$) shows the "repulsion effect" in that the initial tendency of the probability is to increase rather than to decrease in value [9]. It exhibits the distorted evolution characteristic of Jacobian elliptic functions, and approaches the stationary state value 0.2. The switching effect as one goes from 0 to $\frac{1}{2}\pi$ is evident. The dotted line (initial phase difference $\pi$) shows the switching effect again, the stationary state approached being the one with probability 0.8. The oscillations are much more subdued in this case. The details of the repulsion effect mentioned above may be found in ref. [9] where it has been analyzed for the adiabatic dimer. The additional observation from fig. 2b is the initial increase in the oscillations particularly noticeable in the solid line (0 initial phase). The source of this antidamping phenomenon, which is present in the 0 phase difference case but absent in the $\pi$ phase difference case, may be found in (15).

The initial phase difference determines the sign of the damping coefficient $D$, the factor $(\rho_{12} + \rho_{21})_0$ being positive and negative in the two respective cases.

In fig. 3 ($\chi/V = 2.5$, $T/V = 30$, $P_1(0) = 0.7$) we study the relation between the initial condition analysis of the non-adiabatic dimer carried out here (solid lines) and its adiabatic counterpart in ref. [9] (dotted lines). The initial phase difference between the site amplitudes is 0 in fig. 3a and $\pi$ in fig. 3b. In the out-of-phase case of fig. 3b we see the repulsion effect in both the adiabatic and non-adiabatic dimers: the perpetual oscillation described by the Jacobian function in the former case and the approach to the stationary state value 0.8 in the latter case. On the other hand, in the in-phase case, we see considerable difference in the evolution: the non-adiabatic dimer shows an initial increase in the probability oscillation amplitude followed by an approach to the stationary state.

The new results presented in this investigation are eqs. (10), (15)–(18) and their consequences exhibited in figs. 1–3. They extend our analysis of the non-adiabatic dimer to initially non-localized states. Applications to several experimentally observable situations will be reported elsewhere.
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References