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Multiple stationary states and saturation effects for rotational polarons

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Abstract

The phenomenon of saturation of nonlinearity is studied via an investigation of the nature and stability of the stationary states of rotational polarons, which consist of quasiparticles interacting strongly with rotations such as those of rod-like molecules in liquid crystals. The basis is a generalized discrete nonlinear Schrödinger equation which has been introduced recently.

A polaron consists of an electron, or electronic or vibrational excitation, interacting strongly with an oscillatory coordinate such as the vibrational displacement of a molecule. If the oscillation is a rotation, as would be present in rod-like molecules in liquid crystals [1], the resulting rotational polaron can exhibit counterintuitive phenomena such as saturation of nonlinearity and destruction of selftrapping on *increasing* nonlinearity beyond a characteristic value [2–4]. The dynamics, i.e., the time evolution, of such entities has been studied earlier [2]. Our interest in this Letter is in investigating the stationary states of the rotational polaron on the basis of the general formalism given in Ref. [2]. The physical interest of our study is not only in the area of liquid crystals [1] but in any context in which there are partially ordered aggregates of molecules possessing directed shapes and performing angular oscillations around equilibrium directions.

Many features of ordinary polarons can be studied via the discrete nonlinear Schrödinger equation (DNLSE) [5–8]. The rotational polarons, on the other hand, evolve according to a generalized form of that equation introduced by Kenkre et al. [2,3]. The physics behind the generalized equation is based on the fact that no molecular oscillator obeys Hooke's law exactly and no interaction energy is truly linear in the oscillator displacement. We refer the reader to Refs. [2–4] for all details and begin here with the following coupled equations for the angular displacement θ_m of the rotating molecule at site m, and for the amplitude c_m that site m is occupied by the quasiparticle (electron/exciton)

$$i\hbar \frac{dc_m}{dt} = \sum_n V_{mn} c_n + E(\theta_m) c_m , \tag{1}$$

$$\frac{d^2\theta_m}{dt^2} + \omega^2 f(\theta_m) + RE'(\theta_m) |c_m|^2 = 0 . \tag{2}$$

Here V_{mn} are intersite transfer matrix elements describing the linear evolution of the quasiparticle among the states $|m\rangle$, R is a proportionality constant related to the moment of inertia of the rotating molecule, $E(\theta_m)$ is a generally nonlinear function of θ_m describing the quasiparticle–rotation interaction, and $\omega^2 f(\theta_m)$ is a generally nonlinear function describing the restoring force on the rotating molecule. The rotational polaron which we will study here is characterized by a sinusoidal

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restoring force and a sinusoidal interaction described through

$$f(\theta)=\frac{\sin(\Lambda\theta)}{\Lambda},\quad E(\theta)=\frac{E_0}{\Lambda}\sin(\Lambda\theta)\,, \tag{3}$$

and results [2–4], through the application of the standard argument of time scale disparity, to the GDNLSE

$$i\hbar\frac{dc_m}{dt}=\sum_n V_{mn}c_n-\frac{\chi|c_m|^2}{\sqrt{1+(\chi/\Delta)^2|c_m|^4}}c_m\,. \tag{4}$$

This GDNLSE reduces to the standard discrete nonlinear Schrödinger equation (DNLSE), for small χ/Δ but predicts substantially different transport features for nonnegligible values of χ/Δ. Here the nonlinearity χ and the saturation energy Δ are given respectively by E_0^2R/ω^2 and E_0/Λ. In the case of a dimer, i.e. a two-site system in which m, n can take only the values 1 and 2, it has been shown [2,3] that the probability difference p, defined through $p=\rho_{11}-\rho_{22}$, and the associated density matrix element combinations $q=i(\rho_{12}-\rho_{21})$, $r=(\rho_{12}+\rho_{21})$, obey

$$\frac{dp}{dt}=2Vq\,, \tag{5}$$

$$\frac{dq}{dt}=-2Vp+\frac{dg(p)}{dp}r\,, \tag{6}$$

$$\frac{dr}{dt}=-\frac{dg(p)}{dp}q\,, \tag{7}$$

where the function $g(p)$ is given by

$$g(p)=\Delta[\sqrt{(p+1)^2+(2\Delta/\chi)^2}+\sqrt{(p-1)^2+(2\Delta/\chi)^2}-2\sqrt{1+(2\Delta/\chi)^2}]\,. \tag{8}$$

Our interest here is in investigating the *stationary* states of the rotational polaron. The simplest manner of examining them is to put the right hand sides of (5)–(7) equal to zero and to use the fact that conservation of probability and the fact that the states we are studying are stationary yield $r=\pm\sqrt{1-p^2}$. The stationary state probability differences are thus obtained as the solutions of

$$2Vp\pm\sqrt{1-p^2}\,\frac{d}{dp}g(p)=0\,. \tag{9}$$

Nonzero solutions signify self-trapping. Of greater interest is the branch in (9) with a minus sign as it leads to self-trapping stationary states. Considering only that branch, we see that those stationary state probability differences p are given explicitly by

$$\frac{4V}{\chi}p-\sqrt{1-p^2}\left(\frac{1+p}{\sqrt{1+(\chi/2\Delta)^2(1+p)^2}}-\frac{1-p}{\sqrt{1+(\chi/2\Delta)^2(1-p)^2}}\right)=0\,. \tag{10}$$

Solutions of (10) show [2] that it is possible for an increase of nonlinearity to *destroy* self-trapping. Ordinary self-trapping [7,8] as would arise from the large saturation limit of (10), i.e. from the DNLSE, is represented by the onset of a bifurcation in the values of p as χ exceeds a characteristic value. The surprising new phenomenon of destruction of nonlinearity is manifested in this bifurcation disappearing as χ exceeds another characteristic value. This is seen in Fig. 1. The comparison of such a plot with that of the DNLSE has been given in Ref. [2]. It is possible to construct a potential $W(p)$ whose derivative is given by the left hand side of (10). The minima in such a plot obviously signify the stationary states. We have shown such a potential in Fig. 2. Whereas the ordinary DNLSE would result in a $W(p)$ that develops a two-well nature which persists for all larger values of nonlinearity once its onset has occurred, our new results, depicted graphically in Fig. 1a, are that increase of nonlinearity can force an additional minimum at $p=0$ giving $W(p)$ a

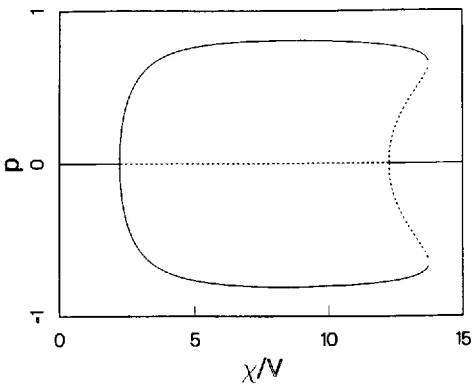

Fig. 1. Stationary state values of the probability difference p plotted as a function of the ratio χ/V. The onset of self-trapping around the value 2.5 of χ/V, and its disappearance for values beyond approximately 14, are seen clearly. Dashed lines represent metastable states and solid lines represent stable states. Multiple stationary states occur for $12.5<\chi/V<14$.

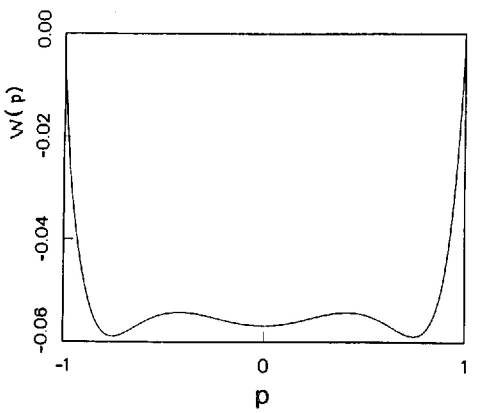

Fig. 2. Stationary state potential $W(p)$ as a function of the probability difference p for $\chi/2V=13$ and $\Delta/2V=4$ showing the existence of multiple stationary states each corresponding to a minimum of the potential. The central minimum is higher than the side minima. This signifies that the central stationary state ($p=0$) is metastable. Self-trapping states are present. Further increase of nonlinearity makes the central minimum dip below the side minima and destroys self-trapping.

three-well nature. The central minimum, it it is higher than the side minima as in Fig. 2, represents only a metastable stationary state. Increase of nonlinearity results in the minimum dipping below the side minima. This converts the *free* central state into a stable state, and self-trapping is destroyed.

We note in passing that an alternative method of obtaining the stationary states directly form the amplitude equation (4) has been given in Ref. [4]. We put

$$c_{1,2}(t)=C_{1,2}\,e^{i\omega t},\quad p=|C_1|^2-|C_2|^2, \tag{11}$$

where ω is the energy of the system, and obtain (10) as well as the energies ω explicitly.

We will now apply standard stability analysis to our system. At the stationary states, the left hand sides of (5)–(7) are zero and it is clear that $p=0$ is always a solution. The linear stationary state is then given by $p=q=0$, $r=\pm 1$. In order to study stability around these values, we let $p=\epsilon$, $q=\delta$, $r=\pm 1+\eta$, where ϵ, δ and η are small quantities. Retaining terms to first order in these small quantities, we have

$$\frac{d\epsilon}{dt}=2V\delta, \tag{12}$$

$$\frac{d\delta}{dt}=-2V\epsilon\{1+r_s(\chi/2V)[1+(\chi/2\Delta)^2]^{-3/2}\}, \tag{13}$$

$$\frac{d\eta}{dt}=0. \tag{14}$$

The eigenvalues of (12)–(14) can be calculated from the secular equation. In order to have the stationary state stable, the real parts of all eigenvalues have to be less than zero. This stability condition is thus

$$1+r_s(\chi/2V)[1+(\chi/2\Delta)^2]^{-3/2}>0. \tag{15}$$

The stationary state with $r_s=1$ always satisfies this condition. For $r_s=-1$, we have the transition condition

$$(\chi/2V)^2=[1+(\chi/2\Delta)^2]^3. \tag{16}$$

In the limit that the saturation parameter Δ is infinitely large, (16) yields the known DNLSE result [7,8] for the transition, viz., $\chi=2V$. Generally, the nonlinearity value at the transition is given by the solution of the cubic equation (16) which we rewrite in the form

$$z^3+3z^2+[3-(\Delta/V)^2]z+1=0,\quad z=(\chi/2\Delta)^2. \tag{17}$$

The equation has two real roots, one corresponding to the appearance and the other to the disappearance of the self-trapping stationary states. As Δ becomes smaller (starting from the infinitely large value corresponding to the DNLSE), these two roots merge into one and become imaginary. Once that happens, self-trapping stationary states cannot occur no matter what the value of the nonlinearity. From the standard analysis of cubic equations (see, e.g., Ref. [9]) it follows immediately that the condition for self-trapping stationary states to disappear no matter what the value of the nonlinearity χ, is

$$(\Delta/V)^2<\tfrac{27}{4}. \tag{18}$$

The study of stationary states that we have given in this note is important particularly in the investigation of optical absorption since the energy differences are directly related to the spectrum. While the effects we have predicted are difficult to observe for the case of a normal polaron since a huge distortion of the lattice is required to reach the characteristic level of saturation, they may indeed be observable in the context of the rotational polaron.

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