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Fröhlich and Davydov regimes in the dynamics of dipolar oscillations of biological membranes

H. Bolterauer
Institut für Theoretische Physik, Justus-Liebig-Universität Giessen, 6300 Giessen, Germany

J. A. Tuszyński
Department of Physics, University of Alberta, Edmonton, Alberta, Canada T6G 2J1

M. V. Satarić
Faculty of Technical Sciences, 21000 Novi Sad, Serbia, Yugoslavia

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In this paper we present a model describing the dynamics of biological membranes. The description presented here is based both on Frölich's earlier conjectures and on the actual biophysical data concerning the structure, composition, and functions of biological membranes. A model Hamiltonian is proposed that involves the oscillations of both the lipid head groups and hydrocarbon chains of the membrane. The presence of dielectrically active material, mainly water, is also accounted for. It is subsequently demonstrated that, depending on the form of the coupling constant between head- and tail-group oscillations, two limiting regimes may occur. The Fröhlich regime is manifested by Bose condensation in the space of dipole oscillation frequencies. This results in self-focusing in the frequency domain. On the other hand, the Davydov regime is associated with spatial localization of the polarized state and leads to soliton formation. Thermal dissipation of the membrane's energy and damping effects are also examined.

I. INTRODUCTION

The molecular membrane that bounds both the cytoplasm of the cell as a whole and the individual organelles contained within it is a structure common to all living systems in nature. All communication of a cell or an organelle with its environment is carried out across the membrane. That includes ion transport processes, diffusive transport of small molecules such as H_2O and CO_2, as well as active transport of large molecules such as lipoproteins [1].

The membrane is essentially a lipid bilayer with a variety of other molecules (e.g., globular proteins, cholesterol) embedded therein and forming a complex interacting system. Such membrane macromolecules as α helices and β sheets, for example, are instrumental in transmembrane transport processes. The generally accepted fluid mosaic model [2] envisages the membrance as comprised mainly of lipid molecules of several types. The most numerous membrane constituents, phospholipids, consist of a polar head group based on a phosphate and attached through a glycerol moiety to two hydrocarbon chains containing 14–20 carbon atoms. The electric charge in the head group makes it hydrophylic, while the oily hydrocarbon chains are hydrophobic. Thus, in water, phospholipids form bilayered lamina 40–50 Å thick, with the polar head groups facing outwards and the tails inwards, away from the water (Fig. 1). The lamina close into stable, spherical vesicles to completely isolate the chains from the membrane's hydrated environment (see Fig. 2). Each of the two lipid layers of the membrane may have an independent phospholipid composition.

The membrane's rigidity and configuration change with temperature (see Fig. 3). In particular, at a narrow temperature range, the membrane undergoes a sudden pronounced loss in packing density becoming more fluidlike above this characteristic temperature T_c, with individual lipids undergoing Brownian motion and exchanging with each other on the order of a microsecond. The fluidlike quality of the membrane is enhanced by the presence of cholesterol molecules amidst hydrocarbon chains, since they reduce chain-chain attraction.

This paper is organized as follows. In Sec. II we outline the basic features of the Fröhlich theory. In Sec. III a model Hamiltonian is introduced to describe the dynamics of head-group dipoles. This is based on earlier proposals of Fröhlich. Section IV deals with the dynamics of membrane tail groups, while Sec. V addresses the question of the interactions between head and tail groups.

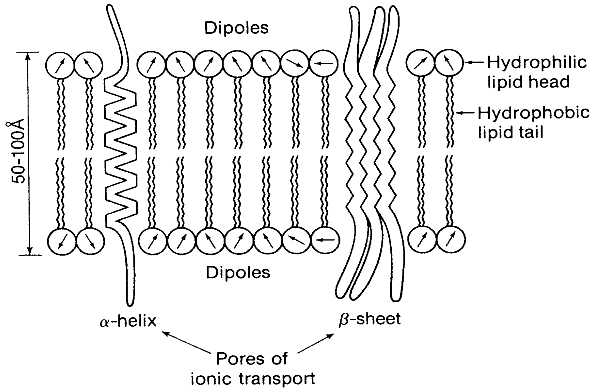

FIG. 1. Schematic illustration of the membrane as a lipid bilayer.

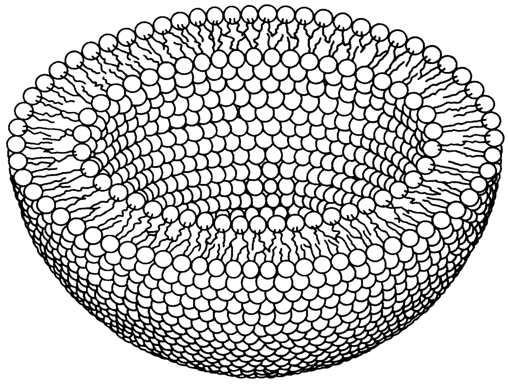

FIG. 2. Graphical representation of the spherical membrane.

Having set up the effective Hamiltonian of the membrane we then derive an equation of motion for the dipolar oscillations of head groups. Two limiting regimes are then discussed, both of which yield exactly solvable equations. These two limiting cases: the Fröhlich regime and the Davydov regime are then extensively analyzed in separate Sections. Discussion and conclusions then follow.

II. FRÖLICH THEORY

Fröhlich [3–5] argued that metabolically active biological cells should exhibit long-range coherence manifested by Bose condensation of longitudinal elastic vibrations of membrane dipoles into a narrow (microwave) frequency band. The crucial physical assumptions made by Fröhlich about the living membrane can be summarized as follows.

(i) A high degree of order (mainly in terms of functional organization but also spatial arrangement) exists. The membrane is viewed as a system of interacting oscillating dipoles placed in a polarizable medium. Both the geometry of the dipole network and the dynamical properties of the medium are expected to play an essential role in establishing long-range dynamical order in the system.

(ii) A sufficient supply of metabolic energy is required to render the transformation to a metabolically active state a nonequilibrium symmetry-breaking transition. Energy pumping is known to occur incoherently at specific membrane locations (ion channels) and is realized through classical electromagnetic forces. The energy may then be redistributed throughout the membrane via mode-mode interactions.

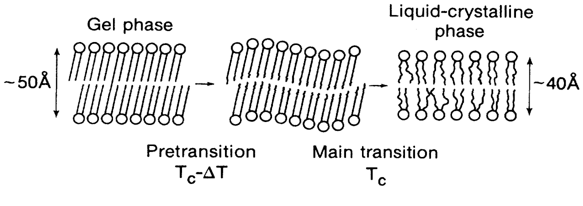

FIG. 3. A typical sequence of transitions experienced by the hydrocarbon chains of the membrane.

(iii) A heat bath, which includes the numerous elastic and electric membrane modes that do not condense and the degrees of freedom of the environment, provides a reservoir of thermal energy. If the energy pumps were to be shut off, or if nonlinearity caused by dipole-dipole interactions were not strong enough, most of the cell's energy would be thermalized.

(iv) The existence of a sufficiently strong transmembrane potential difference is required to electrically polarize membrane molecules. A potential difference of about 10–100 mV is known to exist across biological membranes resulting in an electric field of $(1-20)\times 10^6$ V/m, which may possibly induce an electret state in parts of the membrane.

The ensuing dynamical picture involves high-frequency longitudinal dipolar oscillations of membrane segments with displacements perpendicular to the surface [3]. Their frequency can be estimated as $\omega \equiv 2\pi\nu = (3-8)\times 10^{11}$ Hz. Several specific mechanisms are capable of producing oscillations in this range [4]: (a) dipole oscillations within the lipid head groups; (b) vibrations of ionic double layers; (c) dipole oscillations within large molecules such as DNA, RNA, proteins, and hydrogen-bonded amides; and (d) plasmon-type excitations of ions freed in chemical reactions. These various modes of oscillation are expected to interact among themselves, giving rise to narrow bands of coherent oscillations.

Various experiments were earlier described [3] that appear to demonstrate the sensitivity of metabolic processes to certain frequencies of electromagnetic radiation. All these experiments reveal biological effects that cannot be explained as simply due to heating or the direct action of an applied field but require a nonlinear and nonequilibrium approach [3]. However, at the present time it cannot be stated that the Fröhlich theory has been conclusively confirmed by experiment. Moreover, to the best of our knowledge, no satisfactory microscopic formulation of the Fröhlich theory has been presented to date either. This is in spite of many efforts [6–12] directed towards putting the rather phenomenological framework used by Fröhlich on a more rigorous microscopic basis. There is a particular need for a realistic physical model that would be applicable to a specific biological system or phenomenon. This would undoubtedly help in parallel experimental attempts to confirm Fröhlich's predictions.

This paper's objective is to present a realistic microscopic model of a typical biological membrane envisaged in light of Fröhlich's ideas. We wish to arrive at a set of criteria for the membrane dipoles to form a nonequilibrium metastable polarized state. It is expected that the system comprised of the membrane itself, the energy pumps, and the environment can be adequately described by an appropriate Hamiltonian. The membrane will be modeled as a collection of phospholipids. The hydrocarbon tails will be considered separately from the lipid heads, which is based on the hierarchy of relaxation times. Interactions between these two parts will be added at the end so that the tails predominantly determine the equilibrium phase of the membrane while the heads are crucial to the dynamics of the system.

III. DYNAMICS OF LIPID HEAD GROUPS

The lipid head groups of the cell membrane are less massive and appear to have fewer degrees of freedom than the tail groups. They are also more regular in their structure. It is believed [4] that their dipole moment oscillations play a dominant role in the membrane dynamics. The polar character of the head groups is due to the presence of a positive phosphorus ion at one end and a negative oxygen ion at the other end in the phosphate group. Interactions of these dipoles among themselves and between them and polar water molecules hold the membrane together establishing a temperature-dependent lateral pressure in each of the monolayers.

However, according to Fröhlich [3–5], dynamic interactions between these dipoles are more important for metabolic processes than the static van der Waals forces required for membrane formation. Following Fröhlich [3-5], then, we assume that within the cell membrane the head-group dipoles oscillate at high frequencies and interact among themselves via a combination of elastic and electric forces, as will be elaborated on below. In Fig. 4(a) we have shown the two main head-group vibrational modes: the C=N stretch and the P—O stretch. We believe that the latter one is more important in the process of dipolar mode condensation that will be discussed below. However, these and many other modes (e.g., the C=O bond oscillation) may play important roles in membrane dynamics, and their identification should be verified experimentally.

Following earlier calculations performed by Fröhlich [3] for the interactions between oscillating dipoles, the model Hamiltonian for the head-group dipoles is postulated as

$$H_1=\sum_l \left[\frac{p_l^2}{2m_l}+\tfrac{1}{2}m_l\Omega_l^2u_l^2\right]-\tfrac{1}{2}\sum_{l,m\neq l} u_lT_lu_m \ , \quad (3.1)$$

where l enumerates the head groups within a monolayer, Ω_l are the individual frequencies of dipolar oscillations, u_l are dipole displacements from their equilibrium positions (r_l),p_l are the associated momenta, and T_{lm} is the interaction energy. Fröhlich [3] derived an expression for T_{lm} as

$$T_{lm}(\omega)=\gamma^2e^2(Z_lZ_m)^{1/2}/[mR_{lm}^3\epsilon'(\omega)] \ , \quad (3.2)$$

where γ is a constant on the order of unity which depends on the geometry of the system, e is the elementary charge, Z_l is the number of elementary charges in the lth group, and

$$R_{lm}=|R_l-R_m| \quad (3.2a)$$

is the instantaneous separation between group l and group m, so that

$$R_l=r_l+u_l \ . \quad (3.2b)$$

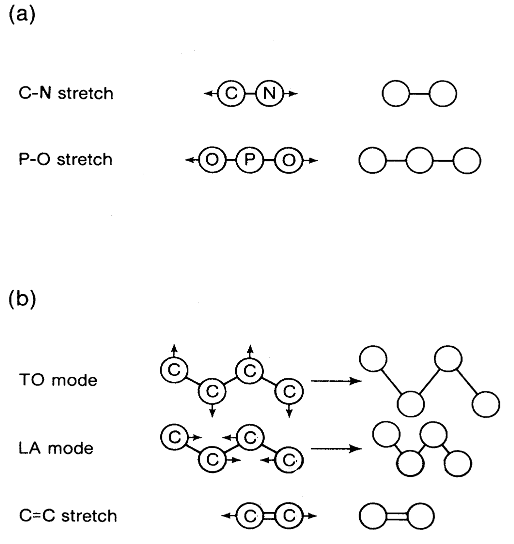

FIG. 4. The predominant vibrational modes of (a) the head groups and (b) the chains of the biological membrane.

Henceforth, it will be assumed that both the numbers of elementary charges and the masses are identical for all groups, i.e.,

$$Z_l=Z_m=Z, \quad m_l=m_m=m \ . \quad (3.2c)$$

The real part of the frequency-dependent dielectric constant of the cell has been denoted by $\epsilon'(\omega)$, with ω being the frequency of electromagnetic radiation in the medium (predominantly water). It is believed that the dynamic dielectric constant may play an extremely important role in frequency-specific mechanisms occurring at a cell level. These mechanisms could provide clues to a number of unexplained trigger effects such as those present in growth, division, and recognition. However, at this early state of theoretical development it is very difficult to provide reliable microscopic modeling of such processes. A calculation of $\epsilon'(\omega)$ for biological systems has been undertaken before [13] but the level of complexity is too great to make use of these results in the present model. Instead, we have graphically illustrated the expected type for several substances (see Fig. 5). In Fig. 5(a) we have shown the plot of $\epsilon'(\omega)$ for myoglobin [14]. Figure 5(b) illustrates it for muscle tissue, and, finally, Fig. 5(c) for water [15]. The main difference between water and biological material is in the number of plateaus of $\epsilon'(\omega)$, which are labeled as α, β, γ, δ, etc., in biological substances. There exist rather narrow crossover regions between these plateaus, and it is conceivable that they may be related to sharp absorption maxima, as will be discussed later in this paper.

Returning to our physical model, the dielectric constant of a biological cell may be in the first instance approximated as a linear combination of contributions due to the membrane, bound water, and free water [13]. Free water is by far the most important component constituting the bulk of the cell's mass and having the relaxation time in the range of interest. Thus, approximating $\epsilon'(\omega)$ by its free-water estimate and using the Debye equation,

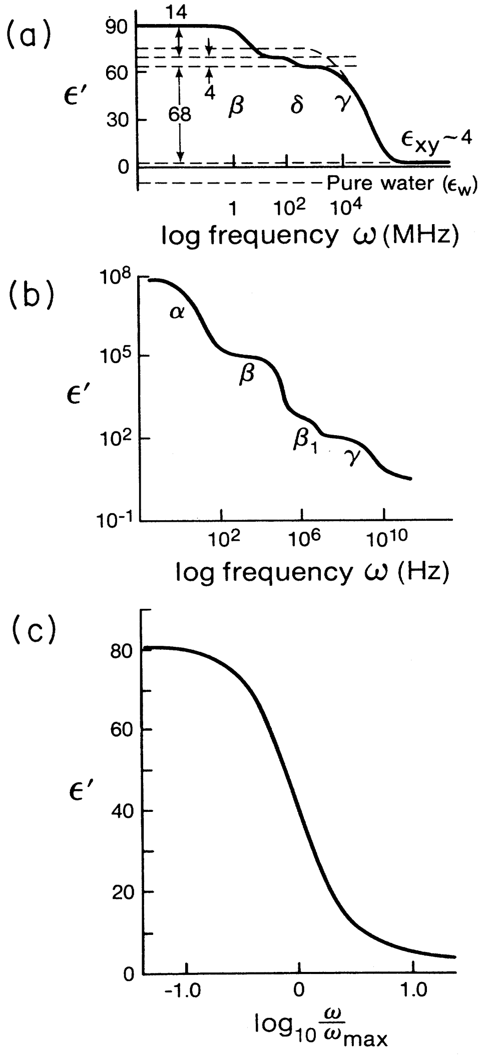

FIG. 5. (a) Dispersion curve for a 10% solution of myoglobin at 20 °C, following Ref. [14]; (b) relative permittivity of muscle tissue, following Ref. [14]; and (c) the permittivity of water at 20 °C where $\omega_{\max}=17.2$ GHz, following Ref. [15].

gives [16]

$$\epsilon'(\omega)=\epsilon_\infty+(e_s-e_\infty)/(1+\omega^2\tau^2)\ , \tag{3.3}$$

where ϵ_s is the static dielectric constant $\epsilon_s=\epsilon'(0)$, ϵ_∞ is the optical dielectric constant $\epsilon_\infty=\epsilon'(\infty)$, and τ is the relaxation time in seconds. At room temperature [16]

$$\epsilon_\infty=4.5,\quad \epsilon_s=78.5,\quad \tau=0.9\times10^{-11}\text{s}\ . \tag{3.4}$$

It should be noted that the electric fields of the order of magnitude present in the membrane do not drastically affect the dielectric properties of the constituent water [14]. Furthermore, raising the temperature close to the upper physiological limit of 40 °C results in ϵ_∞ dropping by about 10% and τ decreasing by almost 50% [14]. Using the head-group Hamiltonian H_1 of Eq. (3.1), the Euler-Lagrange equations are

$$m\left[\frac{d^2}{dt^2}+\Omega_l^2\right]u_l(t)=-\sum_{m(\neq l)}\int_{-\infty}^{t}dt'T_{lm}(t-t')u_m(t')\ , \tag{3.5}$$

which is an integro-differential equation of motion for $u_l(t)$ with a time-dependent interaction term arising from the frequency dependence of T_{lm} in Eq. (3.2). Fourier transforming the latter expression yields an explicit form of $T_{lm}(t)$ as

$$T_{lm}(t)=\frac{\gamma^2e^2Z}{mR_{lm}^3}\left[\frac{\sqrt{2\pi}}{\epsilon_\infty}\delta(t)+\left[\frac{\pi}{2\epsilon_s\epsilon_\infty^3\tau^2}\right]^{1/2}(\epsilon_\infty-\epsilon_s)\times\exp\left[-\frac{\sqrt{\epsilon_s/\epsilon_\infty}}{\tau}t\right]\right]. \tag{3.6}$$

The first term in the square brackets above is a contact term and the second is an exponential decay term that describes time-delay effects resulting from the frequency dependence of dipole-dipole interactions.

Equation (3.5) can be readily Fourier transformed using the convolution theorem and the causality condition of $T_{lm}(t-t')$ to give

$$m(\Omega_l^2-\Omega^2)u_l(\Omega)=\sum_{m,l(\neq m)}T_{lm}(\Omega)u_m(\Omega)\ , \tag{3.7}$$

where $u_m(\Omega)$ is the Fourier transform of $u_m(t)$. Note that Ω_l denotes the oscillation frequency of an *individual* head group located at site l, while we have reserved the symbol Ω for the resultant *collective* oscillation frequency of the entire system of head-group dipoles. The value of Ω is affected both by the dipole-dipole interactions within the membrane and by the state of the polarizable medium, especially water.

Assuming, for simplicity, that the positions of head groups in equilibrium are ordered regularly on a one-dimensional lattice with position vectors $\mathbf{r}_l$ we determine Ω as an eigenfrequency of the system of coupled oscillating dipoles. The corresponding matrix is

$$H_{lm}\equiv\Omega_l^2\delta_{lm}-(1-\delta_{lm})T_{lm}(\Omega)\ , \tag{3.8}$$

whose eigenfrequency problem can, in principle, always be solved yielding the squares of eigenfrequencies Ω^2. First, we define the Fourier transforms of instantaneous lattice displacement as

$$\tilde{u}_k\equiv\sum_l u_l\exp(ikr_l)\ , \tag{3.9}$$

where k is a reciprocal-lattice vector. Then, the dispersion relation is found for collective-mode eigenfrequencies Ω that form bands labeled by k, so that

$$\begin{aligned}\Omega_k^2&=\frac{1}{L}\sum_{l,n(\neq l)}\exp[ik(r_l-r_n)]\left[\Omega_0^2\delta_{l,n}-(1-\delta_{l,n})\frac{T_{ln}}{m}\right]\\&=\Omega_0^2-\frac{1}{m}\sum_{l\neq0}T_{l0}\exp(ikr_l^0)\ .\end{aligned} \tag{3.10}$$

Here, L is the length of the lattice, $r_l^0 \equiv r_l - r_0$, and the lattice site with $m=0$ has been chosen as a reference point. Also, Ω_0 denotes the value of the frequency of dipole oscillations corresponding to $k=0$.

In order to proceed further with the model we now make an assumption that the predominant effect on the frequency dependence of $T_{lm}(\Omega)$ is due to the presence of constituent water molecules vibrating with a characteristic frequency ω. This seems justified in view of the fact that, depending on the type of biological material, water makes up between 75% and 90% of its mass. Furthermore, since for $\omega \leq 10^{11}$ Hz in water $0 < \omega^2\tau^2 < 1$, the real part of the dielectric constant, can be approximated by

$$\epsilon'(\omega) \cong \frac{\epsilon_s - \epsilon_\infty}{1+\omega^2\tau^2} , \tag{3.11}$$

which is accurate to within 10%. Therefore, the interaction term T_{lm} can be estimated as

$$T_{lm}(\omega) \cong A(1+\omega^2\tau^2)(r_l - r_m)^{-3} , \tag{3.12}$$

where

$$A = \gamma^2 e^2 Z / m(e_s - e_\infty) . \tag{3.13}$$

Consequently, the dispersion relation for the band of collective dipole oscillations Ω_k is now

$$\Omega_k^2 = \Omega_0^2 - [A(1+\omega^2\tau^2)/m] \sum_{l=1}^{\infty} (r_l^0)^{-3} \exp(ikr_l^0) . \tag{3.14}$$

The summation in Eq. (3.14) can be evaluated using a continuum approximation such that

$$I_0 \equiv \int_{\epsilon = r_0 k}^{\infty} \frac{\cos x}{x^3} dx , \tag{3.15}$$

where $r_0 = |r_l - r_{l\pm 1}|$ denotes the equilibrium lattice spacing. This yields

$$\Omega_k^2 \cong \Omega_0^2 + \alpha(1+\omega^2\tau^2)k^2 , \tag{3.16}$$

where

$$\alpha = -2AI_0/mr_0 . \tag{3.17}$$

Moreover, under realistic physical conditions we may expect that $\alpha k^2 \ll \Omega_0^2$ so that the final formula for Ω_k is found as

$$\Omega_k \cong \Omega_0 + \frac{\alpha}{2\Omega_0}(1+\omega^2\tau^2)k^2 . \tag{3.18}$$

Two comments are now in order. First, generalizing this approach to higher dimensional lattices would basically affect the value of I_0 through the presence of a Jacobian in the integrand of Eq. (3.15). This would tend to diminish the value of I_0 and thus α, as the dimensionality of the lattice would increase to $D=2$ and then to $D=3$. Second, using typical orders of magnitude one finds $\alpha \sim 10^{20}$, and expecting Ω_0 to be on the order of 10^{11} Hz translates into the condition that the corresponding wavelengths should exceed 10^9 sites (very long) for $\alpha k^2 \ll \Omega_0^2$ to be satisfied.

In the final part of this section we intend to recast the head-group Hamiltonian H_1 into a second quantized form, which would take full account of the properties found so far. The procedure we use is a standard one and it first involves Fourier transforming the site displacements and conjugate momenta u_l and p_l into $\tilde{u}_q$ and $\tilde{p}_q$, respectively. Then, a second quantized annihilation operator a_k is defined through

$$a_k \equiv (2m\Omega_k)^{-1/2}(m\Omega_k \tilde{u}_k + i\tilde{p}_k^\dagger) , \tag{3.19}$$

with the creation operator $a_k^\dagger$ being the hermitian conjugate of a_k. Thus, the head group Hamiltonian H_1 can now be recast into the diagonalized form

$$\begin{aligned} H_1 &= \sum_k \hbar\Omega_k a_k^\dagger a_k \\ &= \sum_k \hbar\Omega_0 a_k^\dagger a_k + J(\omega) \sum_k k^2 a_k^\dagger a_k , \end{aligned} \tag{3.20}$$

where

$$J(\omega) = \frac{\hbar\alpha}{2\Omega_0}(1+\omega^2\tau^2) . \tag{3.21}$$

It should be emphasized again that Ω_k represents the band of characteristic dipolar oscillation frequencies of the cell's head-group subsystem as a whole, while ω is the oscillation frequency of the medium's polar molecules, mainly H_2O. Thus the Hamiltonian of Eq. (3.20) can be thought of as an effective Hamiltonian.

Finally, it is worth noting that a virtually equivalent approach (at least in the continuum limit) can be adopted by assuming the presence of hopping (dispersive) terms in the head-group Hamiltonian, so that it is simply postulated that

$$H_1 = \sum_k [\hbar\Omega_0 a_k^\dagger a_k - \tilde{J} a_k^\dagger (a_{k-1} + a_{k+1})] . \tag{3.22}$$

Here, $\tilde{J}$ represents the hopping parameter, which is related to J but not identical to it. This form of H_1 shows that the energy that may be stored in a given k mode is allowed to spread in momentum space through hopping. This approach was first used in an earlier publication [17], without an elaborate analysis, that preceded its appearance here. In the present paper we shall adopt either Eq. (3.20) or (3.22) for H_1, depending on the type of coupling limit used between the head groups and the hydrocarbon chains of the membrane. We feel that both these forms are *a priori* equally feasible and so we shall not discriminate against either of them. Rather, we shall separately examine the possible outcomes based on these two formulations.

IV. DYNAMICS OF HYDROCARBON CHAINS

The biophysics of hydrocarbon chains of the membrane has been extensively investigated in the past. Most of the statistical properties of the membrane can be calculated from a postulated Ising-type Hamiltonian [18]. Consequently, a phase transition takes place at T_c from a stiffer gel phase ($T < T_c$) to a liquid crystal phase ($T > T_c$), as shown in Fig. 3. A pretransition usually occurs at $T_c - \Delta T$, within several degrees of T_c, where chains develop some "gauche" rotations near their free ends and the lattice structure becomes monoclinic. In addition to these static features, a large number of vibrational modes exist and several of them have been shown in Fig. 4(b). As with any structural change, the dynamics of instantaneous positions of lattice sites is coupled to the critical behavior of the mean displacement. A large body of literature exists [19] in which Landau-Ginzburg models are employed for the description of structural transitions that may occur in a biological membrane. We shall adopt some aspects of this approach.

Below T_c, the most relevant features of the tail dynamics can be reflected by a quartic Landau-Ginzburg type of Hamiltonian

$$H_2=\tfrac{1}{2}\sum_{n=1}^{N}\left[(M\dot{x}_n^2+K\rho_n^2)+\frac{2E_0}{a^4}(x_n^2-a^2)^2\right], \quad (4.1)$$

where x_n is the instantaneous position of the nth chain group, M is its mass, K is the effective stiffness constant, ρ_n represents the relative displacement of two neighboring sites, i.e.,

$$\rho\equiv 2a-|x_n-x_{n-1}|, \quad (4.2)$$

and $2a$ is the equilibrium separation between them. The bistable quartic "on-site" potential proportional to the potential barrier E_0 becomes unistable *above* T_c. Thus, in the high-temperature range, the Hamiltonian can be approximated harmonically as

$$H_2\cong\tfrac{1}{2}\sum_{n=1}^{N}(M\dot{x}_n^2+K\rho_n^2). \quad (4.3)$$

It is easy to show that the thus approximated Hamiltonian can be recast as

$$H_2\cong 2NKa^2-2Ka\sum_{n=1}^{N}|x_n-x_{n=1}|+K\sum_{n=1}^{N}x_n^2 -K\sum_{n=1}^{N}x_nx_{n-1}. \quad (4.4)$$

Using periodic boundary conditions a simplified form is found for H_2 as

$$H_2=H_0+\sum_{n=1}^{N}\left[\frac{\pi_n^2}{2M}+\sum_{n'=1}^{N}V_{nn'}x_nx_{n'}\right], \quad (4.5)$$

where $\pi_n=M\dot{x}_n$ denotes the associated momentum at the nth site, and the coupling constant is found as

$$V_{nn'}=\begin{cases}2K & \text{if } n'=n\\ -K & \text{if } n'=n\pm 1\\ 0 & \text{otherwise}.\end{cases} \quad (4.6)$$

The constant-energy shift is $H_0=2NaK|a-L/N|$. Fourier transforming x_n and π_n into $\tilde{x}_q$ and $\tilde{\pi}_q$, respectively, yields

$$H_2=H_0+\sum_q\left[\frac{1}{2M}\tilde{\pi}_q\tilde{\pi}_q^\dagger+\tfrac{1}{2}M\nu_q^2\tilde{x}_q\tilde{x}_q^\dagger\right], \quad (4.7)$$

where ν_q is the eigenfrequency of the phonon mode with a wave number q. It is well known [20] that in the case of acoustic phonons the dispersion relation is

$$\nu_q=2\sqrt{K/M}\,\sin(q/2), \quad (4.8)$$

and thus the asymptotic behavior close to $q=0$ is proportional to q: $\nu_q\,\alpha\,q$. On the other hand, optical phonon modes [20] obey a different dispersion law, namely,

$$\nu_q\cong\nu_0+\nu_2q^2+\cdots. \quad (4.9)$$

In the sections that follow we shall investigate both of these possibilities and their consequences on the head-group dipole dynamics.

Finally, defining the second quantized annihilation and creation operators for the chain phonons through

$$A_q\equiv(2M\nu_q)^{-1/2}(M\nu_q\tilde{x}_q+i\tilde{\pi}_q^\dagger) \quad (4.10a)$$

and

$$A_q^\dagger\equiv(2M\nu_q)^{-1/2}(M\nu_q\tilde{x}_q^\dagger-i\tilde{\pi}_q), \quad (4.10b)$$

respectively, we can express H_2 in second-quantized form as follows:

$$H_2=\sum_q\hbar\nu_qA_q^\dagger A_q+V_0, \quad (4.11)$$

where V_0 is a constant-energy shift.

A similar procedure could be readily applied to the original chain-group Hamiltonian of Eq. (4.1) that is relevant below T_c. Harmonic approximation applied to the neighborhood of the double minima would lead to a form analogous to Eq. (4.11), which is diagonal. The obvious difference would be that oscillations would be with respect to different equilibrium positions ($x_n=\pm a$ not $x_n=0$). Moreover, the various constants would have different values. The relevant coupling constant $V_{nn'}$ would now be

$$V_{nn'}=\begin{cases}2(K-E_0/a^2) & \text{if } n'=n\\ -K & \text{if } n'=n\pm 1\\ 0 & \text{otherwise}.\end{cases} \quad (4.12)$$

Consequently, the eigenfrequencies ν_q would become

$$\nu_q=2\left[\frac{K}{M}\right]^{1/2}\left[\left[1-\frac{E_0}{Ka^2}\right]-\cos q\right]^{1/2}, \quad (4.13)$$

so that the wave numbers exceeding $\arccos(1-E_0/Ka^2)$ would be damped. The onset of criticality would be brought about by the vanishing of $(K-E_0/a^2)$.

As shown in Fig. 4(b) and discussed earlier, a number of variational modes in the hydrocarbon chains may be of importance in this dynamical model. Hence, strictly speaking, another label should be introduced, in addition to the wave number k, which would designate the type of a vibrational mode of the chain under consideration. In the following section the discussion will concentrate on the interactions between head and chain groups of the membrane.

V. INTERACTIONS BETWEEN TAILS AND HEADS

Lipid head groups exert lateral pressure on the tails, thereby affecting the equilibrium properties of the membrane as a whole. However, in the dynamical picture the tails appear as nearly "frozen" and they determine the equilibrium spacings between the head groups as a function of temperature.

Expanding the interaction energy between two head groups T_{lm} linearly with respect to the deviations from the equilibrium positions of the tails x_l leads to the following contribution to the Hamiltonian:

$$H_{12}=-\tfrac{1}{2}\sum_l u_l T_{l,l+1} u_{l+1} , \tag{5.1}$$

with

$$T_{l,l+1}=\frac{\gamma^2 e^2 Z}{m\epsilon'(\omega)R^3_{l,l+1}} , \tag{5.2}$$

where

$$R_{l,l+1}=R_0+u_l-u_{l+1}+x_l , \tag{5.3}$$

and x_l denotes the displacement of the lth tail group. To the lowest order of approximation in perturbation calculations the effect of this term on the energy of the system is

$$\Delta H=\frac{\partial H_{12}}{\partial R_{l,l+1}}x_l$$
$$=-\tfrac{1}{2}\sum_l u_l u_{l+1}\left[-3\frac{\gamma^2 e^2 Z}{m\epsilon'(\omega)R^4_{l,l+1}}\right]x_l . \tag{5.4}$$

Then, putting $R_{l,l+1}\cong R_0$ (i.e., the equilibrium spacing) to evaluate the terms in large parentheses above gives

$$\Delta H=\frac{3}{2}\frac{\gamma^2 e^2 Z}{m\epsilon'(\omega)R_0^4}\sum_l u_l u_{l+1} x_l . \tag{5.5}$$

Following the results of Secs. III and IV we now introduce the second-quantized operators, in momentum space, for the head- and tail-group displacements:

$$u_l=\sum_k\left[\frac{\hbar}{2mN\Omega_k}\right]^{1/2}(a_k e^{ikR_0 l}+a_k^\dagger e^{-ikR_0 l}) , \tag{5.6a}$$

$$u_{l+1}=\sum_q\left[\frac{\hbar}{2mN\Omega_q}\right]^{1/2}(a_q e^{iqR_0(l+1)}+a_q^\dagger e^{-iqR_0(l+1)}) , \tag{5.6b}$$

$$x_l=\sum_s\left[\frac{\hbar}{2mN\nu_s}\right]^{1/2}(A_s e^{isR_0 l}+A_s^\dagger e^{-isR_0 l}) . \tag{5.6c}$$

Then the product term becomes

$$u_l u_{l+1} x_l=\sum_l\sum_{k,q,s}\left[\frac{\hbar}{2N}\right]^{3/2}(\Omega_k\Omega_q\nu_s)^{-1/2}\{a_k a_q^\dagger A_s \exp[iklR_0-i(l+1)R_0 q+iR_0 ls]$$
$$+a_k^\dagger a_q A_s^\dagger \exp[iR_0 lk-i(l+1)R_0 q-iR_0 ls]\}/m\sqrt{M} , \tag{5.7}$$

where we have only included the terms that conserve the number of head-group excitations, $\mathcal{N}=\sum_k a_k^\dagger a_k$. However, it has been shown [21] that for Hamiltonians exemplified by Eq. (5.1) the relative deviation in the number of excited quasiparticles compared with their average value $\langle\mathcal{N}\rangle$ satisfies the inequality

$$\frac{\mathcal{N}-\langle\mathcal{N}\rangle}{\langle\mathcal{N}\rangle}\ll 1 .$$

This is the extent to which we did not take into account the contribution due to the nonconserving parts of the Hamiltonian. Finally, then, the interaction part of the membrane Hamiltonian can be written as

$$\Delta H=\sum_{k,q}(\chi_{k,q}A_{k-q}^\dagger a_k^\dagger a_q+\text{H.c.}) , \tag{5.8}$$

where the coupling constant has been calculated as

$$\chi_{k,q}=\left[\frac{3}{2}\right]\left[\frac{\hbar}{2N}\right]^{3/2}\frac{\gamma^2 e^2 Z}{m\sqrt{M}\,\epsilon'(\omega)R_0^4}(\Omega_k\Omega_q\nu_{k-q})^{1/2} , \tag{5.9}$$

since momentum conservation (i.e., normal processes only) has been assumed for the processes of head-group phonon scattering off the tail-group phonons.

It is worth commenting now on the k dependence of χ, which enters through (a) dipolar oscillations of the head groups Ω_k and Ω_q, (b) phonon modes of the tails ν_{k-q}, and (c) the dielectric constant of the medium $\epsilon'(\omega)$ whose frequency is expected to be close to both Ω_k and Ω_q. Furthermore, the latter two frequencies are expected to be very close to each other, i.e., $\Omega_k\cong\Omega_q$. Consequently, $\nu_{k-q}\cong\nu_0$. This has an important bearing on $\chi_{k,q}$. If the coupling is predominantly due to acoustic phonons in the tail group, then $\chi_{k,q}$ develops a singularity at $k=q$, so we can approximately write

$$\chi_{k,q}^{\text{acoustic}}\cong\chi_k\delta(k-q) . \tag{5.10}$$

On the other hand, for optical phonons in the chain-group subsystem, $\chi_{k,q}$ is only weakly dependent on k, and in the first order of approximation it can be assumed that

$$\chi_{k,q}^{\text{optical}}\cong\chi_0=\text{const} , \tag{5.11}$$

whose value is estimated at $k=q=0$.

Finally, it should also be pointed out that a more complicated (and realistic) form of the dielectric constant

$\epsilon'(\omega)$, such as the ones shown in Figs. 5(a) and 5(b) for biological materials, could selectively reinforce the coupling constant $\chi_{k,q}$ at particular frequency levels. These special frequencies would correspond to the crossover regions between the neighboring plateaus. Thus one could envisage an approximately deltoidal regime for $\chi_{k,q}$ at sharp frequency values corresponding to crossovers, while a predominantly constant regime would dominate vast ranges of frequencies in the plateau regions. We are therefore strongly motivated to study both these limiting cases separately in the two sections that follow.

VI. THE FRÖLICH REGIME

Many papers have been devoted to a critical examination of Fröhlich's idea of Bose condensation of elementary excitations in biological systems [22–24]. From Refs. [22] and [23] it follows that the creation of order inside living matter may be described in terms of Fröhlich's electric dipole waves. In the dipole set considered by Fröhlich, an externally supplied flow of energy exceeding a necessary threshold populates vibrational modes. A giant electric polarization wave sweeps through the living matter, producing long-range correlations and coherence thereby creating functional order inside the biological system.

In this section we present a model of self-focusing in the frequency domain for a biological membrane. The basis for the model Hamiltonian has been provided in previous sections. Our starting point is to consider $H=H_1+H_2+\Delta H$ using Eqs. (3.22), (4.11), and (5.8), and to use a slightly more general notion, so that [17]

$$H=\sum_k \hbar\Omega_0 a_k^\dagger a_k+\sum_q \hbar\nu_q A_q^\dagger A_q-\sum_k \tilde{J} a_k^\dagger(a_{k-1}+a_{k+1}) + \sum_{k',k'',k'''} \chi_{k',k'',k'''}(A_{k'}a_{k''}a_{k'''}^\dagger+A_{k'}^\dagger a_{k'''}^\dagger a_{k''}) , \tag{6.1}$$

where the form of χ will be specified later in accordance with Eq. (5.10). The first step is to use the Heisenberg equations of motion for the two sets of operators, which result in

$$i\hbar\dot{A}_k=\hbar\nu_k A_k+\sum_{k'',k'''}\chi_{kk'',k'''}a_{k'''}^\dagger a_{k''} \tag{6.2}$$

and

$$i\hbar\dot{a}_k=\hbar\Omega_0 a_k-\tilde{J}(a_{k-1}+a_{k+1}) + \sum_{k',k''} a_{k''}(\chi_{k'kk''}A_{k'}+\chi_{k'k''k}A_{k'}^\dagger) . \tag{6.3}$$

The time scales for microscopic transitions between the numerous excited states of the lipid tails vary from $(10^{-10}-10^{-11})$ s for the lowest-energy states to $(10^{-6}-10^{-7})$ s for the highest-energy states. Since the living cells predominantly reside in the fluidlike phase ($T>T_c$), the longest time scale prevails. Thus the time-dependent behavior of the tails appears to be rather insignificant in the microwave frequency range upon which Fröhlich focused, while the dynamics of polar head groups will be all important there. On this basis we adopt a type of Born-Oppenheimer approximation and set

$$\frac{dA_k}{dt}\cong 0 , \tag{6.4}$$

relative to the time scale of head-group oscillations. This results in

$$A_k=-\frac{1}{\hbar\nu_k}\sum_{k'',k'''}\chi_{kk''k'''}a_{k'''}^\dagger a_{k''} , \tag{6.5}$$

and, consequently, Eq. (6.3) becomes

$$i\hbar\dot{a}_k=\hbar\Omega_0 a_k-\sum_{p,q,l,m}[(\hbar\nu_p)^{-1}(\chi_{pkq}\chi_{pml} + \chi_{pqk}\chi_{plm})a_l^\dagger a_m a_q] - \tilde{J}(a_{k-1}+a_{k+1}) . \tag{6.6}$$

We now invoke the deltoidal regime of Eq. (5.10), which greatly simplifies the effective equation of motion for the operator a_k,

$$i\hbar\dot{a}_k=\hbar\Omega_0 a_k-\frac{2\chi_k^2}{\hbar\nu_k}a_k^\dagger a_k a_k-\tilde{J}(a_{k-1}+a_{k+1}) . \tag{6.7}$$

We now assume a product ansatz for the wave function $|\psi\rangle$ of the entire membrane

$$|\psi\rangle\cong|\psi_{\text{heads}}\rangle|\psi_{\text{tails}}\rangle , \tag{6.8}$$

with a coherent Glauber-state ansatz for both $|\psi_{\text{heads}}\rangle$ and $|\psi_{\text{tails}}\rangle$. Defining the corresponding field translations as

$$\langle\psi|A_k|\psi\rangle\equiv\Lambda_k \tag{6.9}$$

and

$$\langle\psi|a_k|\psi\rangle\equiv\alpha_k , \tag{6.10}$$

we can now deal with these classical c-number quantities rather than the underlying quantum operators. The resultant equation of motion for α_k is virtually identical to (6.7)

$$i\hbar\dot{\alpha}_k=\hbar\Omega_0\alpha_k-J(\alpha_{k-1}+\alpha_{k+1})-\frac{2\chi_k^2}{\hbar\nu_k}|\alpha_k|^2\alpha_k , \tag{6.11}$$

but we do not have to deal with commutation relations anymore. Furthermore, we may seek a stationary solution of this discrete version of the nonlinear Schrödinger equation in k space by assuming its form as

$$\alpha_k(t)=\exp(-iE_s t/\hbar)\alpha_k(0) . \tag{6.12}$$

Expanding $\alpha_{k\pm1}$ in a Taylor series around α_k yields the continuum limit

$$-\tilde{J}\frac{\partial^2\alpha_k(0)}{\partial k^2}=(E_s-\hbar\Omega_0)\alpha_k(0)+\left[\frac{2\chi_0^2}{\hbar\nu_0}\right][\alpha_k(0)]^3 , \tag{6.13}$$

where the values of χ_0 and ν_0 are the estimates of χ_k and ν_k at $k=0$, respectively. The soliton solution of Eq. (6.13) is then found as

$$\alpha_k(0)=\alpha_0\operatorname{sech}(\lambda k) , \tag{6.14}$$

where

$$\alpha_0=\pm[-(E_s-\hbar\Omega_0)\hbar\nu_0/\chi_0^2]^{1/2} \tag{6.15a}$$

and

$$\lambda=[-(E_s-\hbar\Omega_0)/\tilde{J}\,]^{1/2} \; . \tag{6.15b}$$

Thus with $\tilde{J}>0$ it is required that $E_s<\hbar\Omega_0$ and $\nu_0>0$ for this k-space soliton to exist. Furthermore, the width of this localized pulse is proportional to λ^{-1}, i.e., increases with $\sqrt{\tilde{J}}$. It should be kept in mind that imposing the normalization condition on the number of quasiparticles contained in the soliton, i.e.,

$$N=\sum_k N_k=\sum_k |\alpha_k|^2 \; , \tag{6.16}$$

also defines the soliton energy E_s in an implicit way. The solution itself can be interpreted as a form of self-focusing in the reciprocal space so that the probability distribution of dipole oscillations peaks at $k=0$ and its width is proportional to $\sqrt{\tilde{J}}$. This may be seen as a manifestation of Fröhlich's Bose condensation effect, as it describes a coherent vibration of a macroscopic number of the membrane dipole modes within a narrow frequency range [see Fig. 6(a)].

VII. DAVYDOV REGIME

Here, we consider the opposite limiting case. We assume the long-wavelength approximation and neglect the dispersion in the nonlinear part of the Hamiltonian, i.e., we take the Hamiltonian as

$$H=\sum_k [\hbar\Omega_0+J(\omega)k^2]a_k^\dagger a_k+\sum_q \hbar\nu_q A_q^\dagger A_q +\rho(\omega)\sum_{k,q}(A_{k-q}^\dagger a_k^\dagger a_q+\text{H.c.}) \; , \tag{7.1}$$

which is based on Eqs. (3.20), (4.11), and (5.8). Here, the coupling constant $\rho(\omega)$ is approximated by

$$\rho(\omega)\cong\frac{3}{2}\frac{\gamma^2e^2Z\hbar^{3/2}}{m\sqrt{MN}R_0^4\Omega_0\nu_0^{1/2}\epsilon'(\omega)} \; , \tag{7.2}$$

where Ω_0 and ν_0 are the ground-state frequencies for the head and tail groups, respectively.

First, the Heisenberg equation of motion for the tail-group operators yields

$$i\hbar\frac{\partial A_s}{\partial t}=\hbar\nu_s A_s+\rho(\omega)\sum_k a_k^\dagger a_{k-s} \; . \tag{7.3}$$

Again, on the basis of the previously discussed hierarchy of relaxation times we assume that the tail-group dynamics is practically negligible, so that $\partial A_s/\partial t=0$ and consequently

$$A_{k-q}^\dagger\cong-\frac{\rho(\omega)}{\hbar\nu_0}\sum_s a_{s-k+q}^\dagger a_s \; . \tag{7.4}$$

Substituting this result into the model Hamiltonian H of Eq. (7.1) yields the effective head-group Hamiltonian

$$H_1^{\text{eff}}=\sum_k \epsilon_k a_k^\dagger a_k-\frac{\kappa}{N}\sum_{k,k',q} a_k^\dagger a_{k'-q}^\dagger a_{k-q}a_{k'} \; , \tag{7.5}$$

where

$$\epsilon_k=\hbar\Omega_0-\frac{N\rho^2(\omega)}{\hbar\nu_0}+Jk^2 \tag{7.6a}$$

and

$$\kappa=\frac{9\gamma^4e^4Z^2\hbar^2}{4m^2MR_0^8[\epsilon'(\omega)]^2\Omega_0^2\nu_0^2} \; . \tag{7.6b}$$

This type of Hamiltonian is fairly well known in field theory and it describes Bose particles interacting through a δ potential in real space. Since $\kappa>0$ and the sign premultiplying the interaction terms in Eq. (7.5) is negative, we may expect condensation to occur somewhat analogously to Cooper-pair formation in superconductivity. Furthermore, assuming that κ depends only on the frequency of the medium but is otherwise constant we have an opposite situation to that discussed in Sec. VI. Namely, the interactions here are constant in k space and deltoidal in real space. The Fröhlich regime involved constant interactions in real space and deltoidal interactions in k space.

In order to obtain a clustered bound state [25] we perform the following semiclassical procedure. Let the state of the system described by Eq. (7.5) be represented by the wave function

$$|\varphi(t)\rangle=\prod_q|\alpha_q(t)\rangle \; , \tag{7.7}$$

where the coherent-state vectors $|\alpha_q(t)\rangle$ are defined by the property that $a_q|\alpha_q(t)\rangle=\alpha_q(t)|\alpha_q(t)\rangle$ for all of the

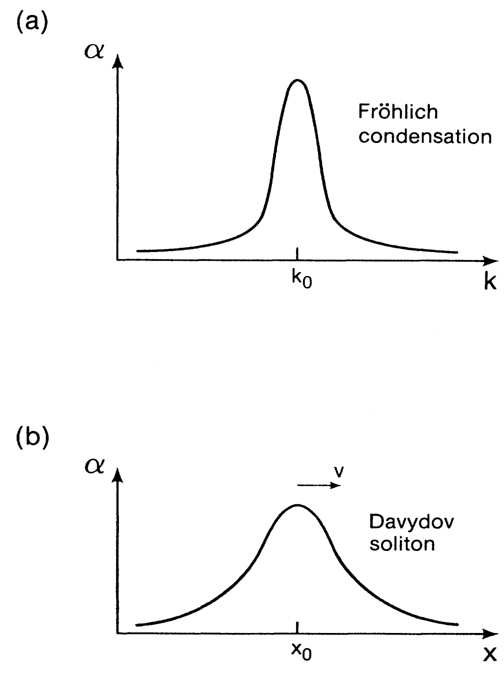

FIG. 6. An illustration of (a) frequency-domain self-focusing as exemplified by Fröhlich condensation and (b) the spatial localization phenomenon as exemplified by the Davydov-like soliton.

annihilators a_q. Then we construct the classical Hamilton function [26]

$$\mathcal{H}=\langle \psi(t)|H_H|\psi(t)\rangle \ ,$$

or explicitly

$$\mathcal{H}=\sum_k \mathcal{E}_k|\alpha_k|^2-\frac{\kappa}{N}\sum_{k,k'q}\alpha_k^*\alpha_{k'-q}^*\alpha_{k-q}\alpha_{k'} \ . \qquad (7.8)$$

The amplitude $\alpha_k(t)$ can be treated as a generalized coordinate with the corresponding generalized momentum $i\hbar\alpha_k^*(t)$. The equation of motion for this generalized dynamical variable is simply

$$i\hbar\frac{\partial\alpha_q(t)}{\partial t}=\frac{\partial\mathcal{H}}{\partial\alpha_q^*(t)} \ , \qquad (7.9a)$$

or

$$i\hbar\frac{\partial\alpha_q(t)}{\partial t}=\mathcal{E}_q\alpha_q(t)-\frac{\kappa}{N}\sum_{k,k'}\alpha_{k-k'}^*(t)\alpha_{q-k'}(t)\alpha_k(t) \ , \qquad (7.9b)$$

which is an expression of the variational principle.

If we multiply Eq. (7.9b) by $N^{-1/2}\exp(iqx)$ and sum over q we obtain

$$i\hbar\sum_q\frac{\partial\alpha_q(t)}{\partial t}N^{-1/2}\exp(iqx)=E_0\sum_q\alpha_qN^{-1/2}\exp(iqx)+J(\omega)\sum_qN^{-1/2}q^2\alpha_q\exp(iqx)$$
$$+\kappa N^{-3/2}\sum_{k,k'}\alpha_{k-k'}^*\alpha_{q-k'}\alpha_k\exp(iqx) \ , \qquad (7.10)$$

where

$$E_0=\hbar\Omega_0-\frac{N\rho^2(\omega)}{\hbar v_0} \ .$$

We can now Fourier transform [26,27] the various quantities back into direct space according to

$$\alpha(x,t)=N^{-1/2}\sum_q\alpha_q(t)\exp(iqx) \ , \qquad (7.11a)$$

$$-\frac{\partial^2\alpha(x,t)}{\partial x^2}=N^{-1/2}\sum_q q^2\alpha_q(t)\exp(iqx) \ , \qquad (7.11b)$$

$$|\alpha(x,t)|^2\alpha(x,t)$$
$$=N^{-3/2}\sum_{k,k'}\alpha_{k-k'}^*(t)\alpha_{q-k'}(t)\alpha_k(t)\exp(iqx) \ , \qquad (7.11c)$$

so that Eq. (7.10) becomes the well-known cubic nonlinear Schrödinger equation (CNSE)

$$i\hbar\frac{\partial\alpha(x,t)}{\partial t}=E_0\alpha(x,t)+J(\omega)\frac{\partial^2\alpha(x,t)}{\partial x^2}$$
$$+\kappa|\alpha(x,t)|^2\alpha(x,t) \ . \qquad (7.12)$$

We assume here that due to a constant and large enough external energy supply a sufficiently high population of coherent head modes is achieved, so that we can use the normalization condition

$$\int_{-\infty}^{+\infty}|\alpha(x,t)|^2dx=\mathcal{N} \ , \qquad (7.13)$$

where $\mathcal{N}$ represents the occupation number of interacting quantum modes.

The envelope of a clustered bound state of $\mathcal{N}$ bosons has the bell-shaped form often referred to as the clustered "boson droplet" and is illustrated in Fig. 6(b).

$$\alpha(x,t)=\left[\mathcal{N}\frac{\mu}{2}\right]^{1/2}\frac{\exp\{i[k_s(x-x_0)-\omega_st]\}}{\cosh[(\mu/R_0)(x-x_0-vt)]} \ . \qquad (7.14)$$

The corresponding parameters are defined as follows:

(i) x_0 is the coordinate of the cluster's center.

(ii) k_s is the cluster's quasi wave number given by

$$k_s=\frac{R_0m^2(\epsilon_s-\epsilon_\infty)}{2I_0Z\gamma^2e^2(1+\omega^2\tau^2)}Nv \ . \qquad (7.15)$$

(iii) v is the group velocity of the cluster.

(iv) The cluster's amplitude parameter μ and its inverse width l are, respectively,

$$\mu=\frac{R_0\kappa}{2J(\omega)}\mathcal{N}, \quad l=\frac{\pi R_0}{\mu} \ . \qquad (7.16)$$

Bearing in mind that $\mathcal{N}$ interacting excitations must be spread along several lattice sites, it is easy to see that the cluster's width satisfies the inequality $l/R_0 \gg 1$, which justifies the use of a continuum approximation imposing the following constraint:

$$1 \ll \mathcal{N} \ll \frac{2J(\omega)}{R_0\kappa} \ . \qquad (7.17)$$

This threshold condition predominantly depends on the dielectric characteristics of the membrane and linearly increases with the frequency of head-group oscillations.

The energy of the cluster is

$$\hbar\omega_s=E^*+\tfrac{1}{2}m^*v^2 \ , \qquad (7.18)$$

where

$$E^*=E_0\mathcal{N}\left[1-\frac{\kappa^2}{24E_0J(\omega)}\mathcal{N}^2\right] \ , \qquad (7.19)$$

and the effective mass m^* is inversely proportional to the transfer matrix element $J(\omega)$ and directly proportional to the number of bound quasiparticles

$$m^*=\frac{\hbar^2}{2J(\omega)}\mathcal{N} \ . \qquad (7.20)$$

It follows from Eq. (7.19) that upon increasing $\mathcal{N}$ the binding energy per particle decreases in proportion to $\mathcal{N}^2$. The particle density $\rho(x,t)=|\alpha(x,t)|^2$ from Eq. (7.14) increases in proportion to $\mathcal{N}^2$, but a collapse of the cluster is prevented by the condition (7.17). One additional reason preventing the occurrence of a collapse is the fact that increasing the density of quasiparticles enhances three-body repulsive forces, which begin to act over small distances.

It is very useful to transform the envelope (7.14) into the reciprocal space by Fourier transforming it through

$$\alpha_k(t)=R_0^{-1}\int_{-\infty}^{+\infty}\alpha(x,t)\exp(-ikx)dx \tag{7.21}$$

to get

$$\alpha_k(t)=\frac{\pi}{\sqrt{2\mu}}\,\frac{\exp[i(k_s-k)x_0+i\omega_k t]}{\cosh[(\pi R_0/2\mu)(k_s-k)]}\ , \tag{7.22}$$

where

$$\omega_k=kv-\omega_s\ . \tag{7.23}$$

This form is very convenient because the time dependence is involved only through the oscillatory factor $\exp(i\omega_k t)$ and will be used in Sec. VIII in a procedure to calculate the cluster's damping factor. It should also be mentioned that the above results are in accordance with many papers devoted to the Bose condensation in other areas of condensed-matter physics [27–29].

To summarize, in this section we have demonstrated that, in the regime of the coupling constant being independent of the wave number, a Davydov-type soliton formation occurs. This means that a clustered bound state is formed that is localized in space and has remarkable stability properties. This may lead to the emergence of particular regions in the membrane that are polarized and are coupled to an elastic deformation.

VIII. DYNAMIC DAMPING AND THE EFFECTS OF THERMALIZATION

A nonzero temperature behavior of the system may be found only by introducing thermal fluctuations. It should be noted that a temperature decrease does not improve the ordering of the biological system, as would be expected from the fact that fluctuations also decrease. On the contrary, in living systems the dynamical order disappears when temperatures decrease well below the room temperature.

The purpose of this section is to examine the influence of thermal fluctuations on the dynamical behavior of the clustered Boson "droplet." We consider a total system composed of the oscillations of head groups coupled with the tails, as well as a thermal reservoir, including their mutual interactions. Therefore, the corresponding Hamiltonian consists of three parts,

$$H=H_H+H_B+H_{HB}\ , \tag{8.1}$$

where H_H is the head-group Hamiltonian (3.20). The reservoir is seen as a collection of harmonic oscillators,

$$H_B=\sum_p \hbar\omega_p C_p^\dagger C_p\ , \tag{8.2}$$

and H_{HB} stands for an interaction between the two subsystems,

$$H_{HB}=\frac{1}{\sqrt{N}}\sum_{p,k}G_p a_{k+p}^\dagger a_k(C_p+C_{-p}^\dagger)\ , \tag{8.3}$$

where $C_p^\dagger, C_p$ are the creation and annihilation operators of the reservoir's degrees of freedom, respectively. The interaction energy parameter is given by

$$G_p=\sigma\sqrt{\hbar/2M_0\omega_p}\ , \tag{8.4}$$

where σ is a coupling constant and the linear dispersion relation is

$$\omega_p=c_0 p \tag{8.5}$$

where c_0 is the characteristic sound velocity of the reservoir while M_0 is the mass of one molecule of the reservoir (for example, an H_2O molecule). All these operators are assumed to be time independent. It is shown in the Appendix that, under the influence of the heat bath, the resultant equation of motion for the amplitude $\alpha(x,t)$ is now

$$\begin{aligned} i\hbar\frac{\partial\alpha}{\partial t}=&(E_0+\Delta E)\alpha-R_0^2J(\omega)\frac{\partial^2\alpha(x,t)}{\partial x^2}\\ &-G|\alpha(x,t)|^2\alpha(x,t)-\frac{i\hbar}{2}\Gamma\alpha(x,t)+F_f(x,t)\ , \end{aligned} \tag{8.6}$$

representing a cubic nonlinear Schrödinger equation with damping and friction terms. The last term on the right-hand side of the above equation represents the transformed energy of the fluctuation forces

$$F_f=N^{-1/2}\sum_p G_p(C_p+C_{-p}^\dagger)\exp(ipx)\alpha^{(0)}(x,t)\ , \tag{8.7}$$

which depends on the unperturbed solution $\alpha^{(0)}(x,t)$ of the CNSE (7.14) and on the thermal reservoir's operators.

If we use the simple phase-shift transformation

$$\alpha(x,t)=\exp\left[-\frac{\Gamma}{2}t\right]\Phi(x,t)\ , \tag{8.8}$$

we can easily obtain a new and simpler form of the perturbed CNSE,

$$\begin{aligned} i\hbar\frac{\partial\Phi(x,t)}{\partial t}=&(\mathcal{E}_0+\Delta E)\Phi(x,t)-R_0^2J(\omega)\frac{\partial^2\Phi(x,t)}{\partial x^2}\\ &-G|\Phi(x,t)|^2\Phi(x,t)+F_f\ . \end{aligned} \tag{8.9}$$

In the first stage, for the sake of simplicity and due to its smallness, we can neglect the fluctuation force energy, considering semiquantitative features of the problem only. Keeping in mind that the envelope of the clustered bound state $\alpha(x,t)$ must obey the normalization condition (7.13) it is easy to see that the function $\Phi(x,t)$ can have the form

$$\Phi(x,t)=\left[\mathcal{N}\frac{\mu}{2}\right]^{1/2}\frac{\exp[i(kx-\omega_s t)]}{\cosh[(\mu/R_0)e^{-\Gamma t}(x-x_0-vt)]}\ , \tag{8.10}$$

so that the envelope function becomes

$$\alpha(x,t)=[\mathcal{N}\mu\exp(-\Gamma t)] \times\frac{\exp[i(kx-\omega_s t)]}{\cosh[(\mu/R_0)\exp(-\Gamma t)(x-x_0-vt)]} . \tag{8.11}$$

Therefore, our result is that damping causes a decrease of the envelope's amplitude as a function of time through

$$\mu(t)=\mu_0\exp(-\Gamma t), \quad \mu_0=\frac{\kappa R_0}{2J(\omega)}\mathcal{N} . \tag{8.12}$$

On the other hand, the region of the cluster's localization is given by

$$l(t)=\frac{\pi R_0}{\mu_0}\exp(+\Gamma t) , \tag{8.13}$$

so that it increases exponentially with time. Bearing in mind that the damping factor Γ lost its k dependence we can explicitly calculate its value as

$$\Gamma=\frac{2\pi}{\hbar^2}\frac{R_0}{\mu}\int_{-\pi/R_0}^{+\pi/R_0}dp\,|G(p)|^2[(n_p+1)\delta(\omega_s-vp) +n_p\delta(\omega_s+vp)] \tag{8.14}$$

or

$$\Gamma=\frac{\pi}{\hbar^2}R_0\left|G\left[\frac{\omega_s}{v}\right]\right|^2\left[n\left[\frac{\omega_s}{v}\right]+\frac{1}{2}\right] . \tag{8.15}$$

From the definitions we have

$$\left|G\left[\frac{\omega_s}{v}\right]\right|^2=\sigma^2\frac{\hbar}{2M_0c_0\omega_s}v \tag{8.16}$$

and

$$n\left[\frac{\omega_s}{v}\right]=\left[\exp\left[\frac{\hbar\omega_s\frac{c}{v}}{k_BT}\right]-1\right]^{-1} . \tag{8.17}$$

The general solution thus becomes

$$\Gamma(v,t)=2\gamma v\left\{\left[\exp\left[\frac{\hbar\omega_s c_0}{k_BTv}\right]-1\right]^{-1}+\frac{1}{2}\right\} , \quad \gamma=\frac{R_0\sigma^2}{\hbar M_0\omega_s} . \tag{8.18}$$

In Figs. 7(a) and 7(b) we have shown the dependence of Γ/γ on velocity v and temperature T, respectively. It is clear that for a stationary ($v=0$) cluster that the damping factor disappears. For greater velocities of the cluster the condition

$$\hbar\omega_s c_0 \gg k_BTv \quad \text{or} \quad (E_0+\tfrac{1}{2}m^*v^2)c_0 \gg k_BTv , \tag{8.19}$$

is valid, and it follows from Eq. (8.18) that

$$\Gamma(v,t)=2\tilde{\gamma}v\left[\exp\left[-\frac{\hbar\omega_s c_0}{k_BTv}\right]+\frac{1}{2}\right]\approx\tilde{\gamma}v . \tag{8.20}$$

In this case the damping increases linearly with the cluster's velocity and it is almost independent of temperature. The equation of motion for a cluster of effective mass m^* is then

$$m^*\frac{dv}{dt}=-\Gamma v \quad \text{or} \quad m^*\frac{dv}{dt}=-\tilde{\gamma}v^2 . \tag{8.21}$$

This kind of velocity dependence is quite analogous to the equation of motion of a classical particle in a viscous medium when the velocity is greater than some critical

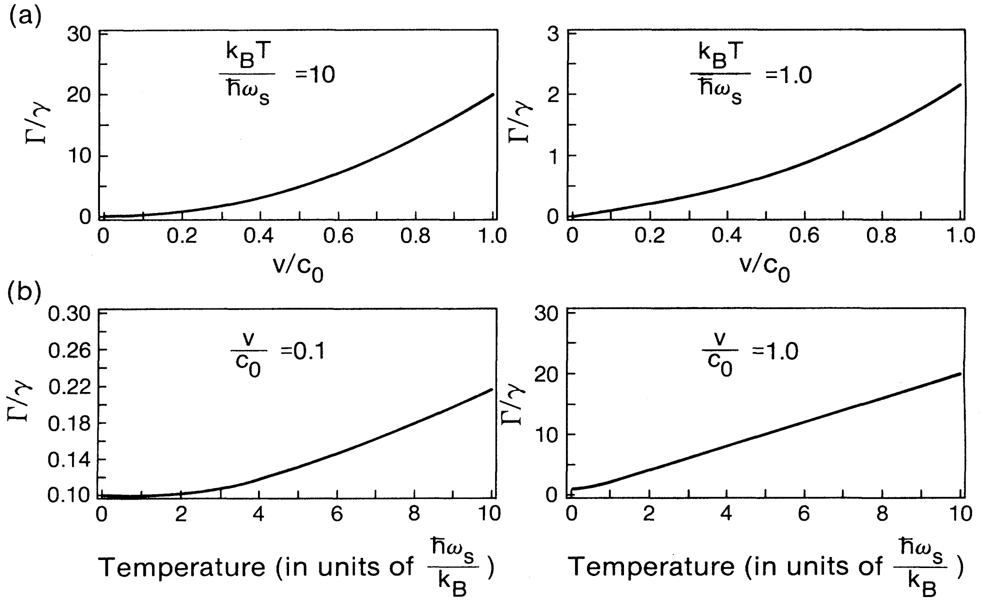

FIG. 7. A plot of the damping constant Γ as a function of (a) velocity and (b) temperature.

value. In this case a similar condition imposes the inequality

$$v > 4\frac{k_B T}{m^* c_0} \ . \tag{8.22}$$

The amplitude of the cluster is now given as

$$\mu(t) = \mu_0 \exp\left[-\frac{\tilde{\gamma}}{2} v t\right] , \tag{8.23}$$

so that fast clusters extend very rapidly while slow ones better preserve their form in the process of propagation.

IX. DISCUSSION AND CONCLUSIONS

In this paper we have developed a microscopic model of the metabolically active biological membrane. We have based our presentation on Fröhlich's ideas but maintained close contact with the actual data concerning the function and structure of biological membranes. Interactions within the hydrocarbon tails of the membrane are known to lead to a number of thermodynamic phases and determine the equilibrium spacings between the various groups. Head groups have been modeled using a dynamical picture of high-frequency dipolar vibrations, which, under special conditions, may lead to extended modes. This is facilitated by the presence of water as a dielectrically sensitive medium.

Interactions between the head groups and lipid tails have been shown to lead to strongly nonlinear equations of motion for the operators describing creation and annihilation of dipole oscillation modes. In fact, under realistic conditions, which include incoherent pumping and weak dispersion, a k-space analog of the nonlinear Schrödinger equation has been derived, provided the head-tail interactions are deltoidal in reciprocal space. This is manifested by the self-focusing of head group oscillations, resembling very much the phenomenon of Fröhlich's coherence. In the opposite regime, namely, when the interactions are highly localized in real space, we have shown that the result is soliton formation in the actual space. The equation describing it is the nonlinear Schrödinger equation. This equation has been previously applied by Davydov [30] to model the energy transfer in quasilinear molecular chains. Most notably, a stable localized solution is known to exist, called the Davydov soliton, in applications to molecular chains [30], which has a macroscopic mass in our case.

We have subsequently developed a picture of a solitonlike clustered bound state representing Bose condensation in the membrane. In Sec. VIII the effects of finite temperature have been shown to lead to damping and dissipation. The damping coefficient strongly depends on the velocity of propagation.

In conclusion, we have demonstrated the feasibility of highly nonlinear phenomena in biological membranes, including the presence of long-range coherence and analogs of Davydov's solitons in the form of clustered bound states. Moreover, such important factors as temperature and dissipative forces have been explicitly included in our formulation of the problem. However, much still remains to be done to determine specific structural requirements for such effects.

It is very important to stress the probability of both regimes, depending on the prevailing conditions. That might include a particular frequency range, the type of phonon mode being excited, and the state of the chain system. Also, depending on a particular application, the membrane or its subsystem may give preference to one regime or the other. For example, some modes may be involved in arranging coherence at a given frequency, say, in the microwave range, as a means of providing an "internal clocking" mechanism for the cell. On the other hand, segments of the membrane, such as α channels and β sheets may have a tendency to prefer the Davydov regime in order to facilitate transmembrane transport of ions and energy. Also, potential gradients at specific membrane locations could be very useful in creating local attractive or repulsive forces which might be frequency specific.

We feel that the present model contains a number of important features, which qualify it as a good starting point towards a more general theory of the membrane's metabolic state. It includes both parts of the membrane in a single Hamiltonian. Finally, the result obtained here and indicating the possible existence of a standing soliton in momentum space has, to the best of our knowledge, not been previously presented.

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APPENDIX

The Heisenberg equation of motion for an arbitrary dynamical variable $A(t)$ is given by

$$\frac{dA(t)}{dt} = \frac{i}{\hbar}[H, A(t)] = iLA(t) , \tag{A1}$$

where L denotes the Liouville operator corresponding to H. The solution of Eq. (A1) has the usual form

$$A(t) = e^{iLt} A(0) , \tag{A2}$$

so that

$$\frac{dA(t)}{dt} = e^{iLt} iLA(0) \ . \tag{A3}$$

In order to transform Eq. (A3) into the form of the Heisenberg-Langevin equation, we need to recast the operator e^{iLt} into a form including relaxation terms. To eliminate irrelevant variables associated with the reservoir we introduce a time independent projection operator $\mathcal{P}$ by the relation

$$\mathcal{P}A(t) = \langle A(t) \rangle_B = \mathrm{Tr}[\rho_B A(t)] , \tag{A4}$$

where ρ_B is a suitably chosen equilibrium density operator of the reservoir

$$\mathcal{P}A(t) = \mathrm{Tr}\left[\frac{1}{Q_B} \exp\left[-\beta \sum_p \hbar\omega_p C_p^\dagger C_p\right] A(t)\right] , \tag{A5}$$

with $\beta=1/k_BT$ and Q_B denoting the statistical sum. Then, the equation of motion (A3) can be written as

$$\frac{dA(t)}{dt}=e^{iLt}\mathcal{P}iLA(0)+e^{iLt}(1-\mathcal{P})iLA(0)\ . \qquad \text{(A6)}$$

We wish to eliminate the operator $e^{iLt}(1-\mathcal{P})$ as far as possible and to extract the factor $e^{iLt}\mathcal{P}$ on the left end of each main term. In accordance with the procedure developed by Hashitsume and co-workers [31–33] we transformed the formula (A6) as follows:

$$\frac{dA(t)}{dt}=e^{iLt}iLA(0)+e^{iLt}\int_0^\infty d\tau\, e^{-iL_s\tau}\langle iL_{HB}e^{i(L_H+L_B)\tau}iL_{HB}\rangle_B A(0)+R(t)\ . \qquad \text{(A7)}$$

The indices S and SB correspond to the relevant parts of the energy operator of Eq. (8.1). The symbol $\langle\ \rangle_B$ denotes the average with respect to the degrees of freedom of the thermal reservoir. The term $R(t)$ represents the quantum-fluctuation force, which has the general form

$$R(t)=(1-\mathcal{P})\exp[i(L_H+L_B)t]iL_{HB}A(0)\ . \qquad \text{(A8)}$$

Equation (A7) has a pure quantum-mechanical character. This fact presents an obstacle in our efforts to involve solitonic excitations because the latter solutions follow from a semiclassical description of the subsystem of head modes. For this reason it seems worthwhile to try and adopt the above equation for use within a solitonic framework. In order to apply a suitable procedure we construct the Hamilton function

$$\mathcal{H}=\langle\varphi(t)|H_H+H_B+H_{HB}|\varphi(t)\rangle\ , \qquad \text{(A9)}$$

where the eigenfunction can be chosen in the usual form

$$|\varphi(t)\rangle=\prod_k \alpha_k(t)a_k^\dagger|0\rangle\ , \qquad \text{(A10)}$$

so that we have

$$\begin{aligned}\mathcal{H}=&\sum_k \mathcal{E}_k|\alpha_k(t)|^2-\frac{\kappa}{2N}\sum_{k,k'q}\alpha_q^*(t)\alpha_{k-q}(t)\alpha_{k'-q}^*\alpha_k(t)\\&+\frac{1}{\sqrt{N}}\sum_{k,p}G_p\alpha_{k+p}^*(t)\alpha_q(t)(C_p+C_{-p}^\dagger)\\&+\sum_p \hbar\omega_p C_p^\dagger C_p\ . \qquad \text{(A11)}\end{aligned}$$

The above functional is classical with respect to the head mode's amplitude, while being quantual with respect to reservoir excitations. The equation of motion for the variable $A(\alpha_k,\alpha_k^*)$ is then the following classical equation of motion:

$$\frac{\partial A}{\partial t}=-\frac{i}{\hbar}\sum_k\left[\frac{\partial\mathcal{H}}{\partial\alpha_k^*}\frac{\partial A}{\partial\alpha_k}-\frac{\partial\mathcal{H}}{\partial\alpha_k}\frac{\partial A}{\partial\alpha_k^*}\right]=iLA\ . \qquad \text{(A12)}$$

If we simply put $A(\alpha_k,\alpha_k^*)=\alpha_k$, we get

$$\frac{\partial\alpha_k(t)}{\partial t}=-\frac{i}{\hbar}\frac{\partial H}{\partial\alpha_k}=iL\alpha_k(t)\ . \qquad \text{(A13)}$$

For quantum operators C_p we have the quantum equations of motion

$$\frac{dC_p}{dt}=\frac{i}{\hbar}[H,C_p]=iLC_p(t)\ . \qquad \text{(A14)}$$

In order to obtain the time evolution of the solitonic amplitude $\alpha_k(t)$ we calculate the terms of Eq. (A7) step by step

$$iL_{HB}\alpha_k(t)=-\frac{i}{\hbar}\frac{\partial\mathcal{H}_{HB}}{\partial\alpha_k^*}=-\frac{i}{\hbar}\sum_k G_p\alpha_{k-p}(0)[C_p(0)+C_{-p}^\dagger(0)]\ , \qquad \text{(A15)}$$

$$e^{i(L_H+L_B)\tau}iL_{HB}\alpha_k(0)=-\frac{i}{\hbar}\sum_k G_p\alpha_{k-p}(0)e^{i\omega_{k-p}\tau}(C_pe^{-i\omega_p\tau}+C_{-p}^\dagger e^{i\omega_p\tau})\ , \qquad \text{(A16)}$$

$$\begin{aligned}iL_{HB}e^{i(L_H+L_B)\tau}iL_{HB}\alpha_k(0)=&-\frac{i}{\hbar^2}\frac{1}{N}\sum_{p,p'}G_pG_{p'}\alpha_{k-p-p'}(0)e^{i\omega_{k-p}\tau}(C_pe^{-i\omega_p\tau}+C_{-p}^\dagger e^{i\omega_p\tau})[C_p(0)+C_{-p}^\dagger(0)]\\&+\frac{i}{\hbar^2}\frac{1}{N}\sum_{k'p}|G_p|^2e^{i\omega_{k-p}\tau}(e^{i\omega_p\tau}-e^{-i\omega_p\tau})\alpha_{k'-p}^*(0)\alpha_{k'}(0)\alpha_{k-p}(0)\ , \qquad \text{(A17)}\end{aligned}$$

where

$$\omega_p=pv-\omega_s\ , \qquad \text{(A18)}$$

and we recall that v represents the velocity of the clustered bound state, while $\hbar\omega_s$ is its energy. After averaging with respect to the reservoir's density matrix, the expression under the integration sign in Eq. (A7) becomes

$$e^{-iL_H\tau}\langle iL_{HB}e^{i(L_H+L_B)\tau}L_{HB}\rangle_B\alpha_k(0)=\langle\Omega(t)\rangle\ , \tag{A19}$$

where

$$\begin{aligned}\langle\Omega(\tau)\rangle=&-\frac{i}{\hbar^2}\frac{1}{N}\sum_p|G_p|^2[(n_p+1)e^{i(\omega_{k-p}-\omega_k-\omega_p)\tau}+n_pe^{i(\omega_{k-p}-\omega_k+\omega_p)\tau}]\alpha_k(0)\\&+\frac{i}{\hbar^2}\frac{1}{N}\sum_{p,k'}|G_p|^2e^{i(\omega_{k'-p}-\omega_{k'})}(e^{i\omega_p\tau}-e^{-i\omega_p\tau})\alpha^*_{k'-p}(0)\alpha_{k'}(0)\alpha_{k-p'}(0)\ .\end{aligned} \tag{A20}$$

The next step is the integration with respect to τ, which gives

$$\begin{aligned}\int_0^\infty e^{-iL_s\tau}\langle\Omega(\tau)\rangle\alpha_k(0)d\tau=&-\frac{\pi}{\hbar^2}\frac{\alpha_k(0)}{N}\sum_p|G_p|^2[\delta(\omega_k-\omega_{k-p}+\omega_p)(n_p+1)+\delta(\omega_k-\omega_{k-p}-\omega_p)n_p]\\&-\frac{i}{\hbar^2}\frac{\alpha_k(0)}{N}\sum_p|G_p|^2\left[(n_p+1)\widetilde{\mathcal{P}}\left[\frac{1}{\omega_k-\omega_{k-p}+\omega_p}\right]+n_p\widetilde{\mathcal{P}}\left[\frac{1}{\omega_k-\omega_{k-p}-\omega_p}\right]\right]\ ,\end{aligned} \tag{A21}$$

where

$$n_p=\left[\exp\left[-\frac{\hbar\omega_k}{k_BT}\right]-1\right]^{-1}\ . \tag{A22}$$

The first term on the right-hand side of Eq. (A21) represents the damping factor caused by the emission and absorption of a quantum of energy from the heat reservoir. The second term on the right-hand side, including the principal values of the corresponding integrals denoted by $\widetilde{\mathcal{P}}$ involves an energy shift of the clustered bound state. The nonlinear dynamical equation arising from Eq. (A7) now has the form

$$\begin{aligned}i\hbar\frac{\partial\alpha_k}{\partial t}=&(\mathcal{E}_k+\Delta\mathcal{E}_k)\alpha-\frac{\kappa}{N}\sum_{p,k'}\alpha^*_{k'-q}\alpha_{k'}\alpha_{k-q}\\&-\frac{i\hbar}{2}\Gamma_k\alpha_k+R_k\ ,\end{aligned} \tag{A23}$$

where the damping Γ_k is expressed as follows:

$$\begin{aligned}\Gamma_k\frac{2\pi}{\hbar^2N}\sum_p|G|^2[&(n_p+1)\delta(\omega_k-\omega_{k-p}-\omega_p)\\&+n_p\delta(\omega_k-\omega_{k-p}+\omega_p)]\ ,\end{aligned} \tag{A24}$$

and the energy shift ΔE_k is

$$\begin{aligned}\Delta E_k=\frac{-i}{\hbar N}\sum_p|G_p|^2\Bigg[&(n_p+1)\widetilde{\mathcal{P}}\left[\frac{1}{\omega_k-\omega_{k-p}+\omega_p}\right]\\&+n\widetilde{\mathcal{P}}\left[\frac{1}{\omega_k-\omega_{k-p}-\omega_p}\right]\Bigg]\ ,\end{aligned} \tag{A25}$$

while the energy of fluctuation forces is

$$R_k(t)=(1-\mathcal{P})\exp[i(L_H+L_B)t]iL_{HB}\alpha_k(0)\ . \tag{A26}$$

It is easy to see that due to relation (A20) we have that the arguments $\omega_k-\omega_{k-p}\pm\omega_p=2pv\pm\omega_s$ are really independent of k. This fact facilitates the transition to the continuum limit so that after performing the Fourier transform, Eq. (A23) becomes

$$\begin{aligned}i\hbar\frac{\partial\alpha(x,t)}{\partial t}=&(\mathcal{E}_0+\Delta E)\alpha(x,t)-R_0^2J(\omega)\frac{\partial^2\alpha(x,t)}{\partial x^2}\\&-G|\alpha(x,t)|^2\alpha(x,t)-\frac{i\hbar}{2}\Gamma\alpha(x,t)+F_f(x,t)\ ,\end{aligned} \tag{A27}$$

representing a damped CNSE.

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