Exciton-Phonon Dynamics with Long-Range Interaction

Nick Laskin*

**TopQuark Inc., Toronto, ON, M6P 2P2, Canada*
(e-mail: nlaskin@rocketmail.com)

Abstract: Exciton-phonon dynamics on a 1D lattice with long-range exciton-exciton interaction has been introduced and elaborated. The long-range interaction leads to a nonlocal integral term in the motion equation if we go from discrete to continuous space. It has been found that in some particular cases for power-law interaction the integral term can be expressed through a fractional order derivative. In other words, the non-locality originating from the long-range exciton interaction results in dynamic equations with space derivatives of fractional order. New theoretical frameworks to study non-linear quantum dynamics with long-range interaction have been established. Those frameworks are: fractional generalization of Zakharov system, Hilbert-Zakharov system, Hilbert-Ginzburg-Landau equation and nonlinear Hilbert-Schrödinger equation. We also discuss non-linear fractional Schrödinger equation and fractional Ginzburg-Landau equation.

Keywords**:** Davydov's Hamiltonian, Exciton-phonon dynamics, Long-range interaction, Non-linear fractional differential equations.

1. INTRODUCTION

Dynamic lattice models are widely used to study a broad set of physical phenomena and systems. In the early 1970's a novel mechanism for the localization and transport of vibrational energy in certain types of molecular chains was proposed by A.S. Davydov (Davydov, 1973). He pioneered the concept of the *solitary excitons* or the *Davydov soliton* (Davydov and Kislukha, 1973). Our focus is analytical developments on quantum 1D exciton-phonon dynamics with power-law long-range exciton-exciton interaction $J_{n,m} = J/|n-m|^{-s}$, $(s > 0)$ for excitons located at the lattice sites n and m. In addition to the well-known interactions with integer values of s, some complex media can be described by fractional values of s (see, for example, references in (Zaslavsky *et al*., 2006). Using the ideas developed at first in (Laskin and Zaslavsky, 2006), we elaborate the Davydov model for the exciton-phonon system with a fractional power-law exciton-exciton interaction. It has been shown that 1D lattice exciton-phonon dynamics in the long-wave limit can be effectively presented by the general system of two coupled equations for exciton and phonon dynamic variables. The dynamic equation describing the exciton subsystem is the fractional differential equation, which is manifestation of non-locality of interaction, originating from the long-range interaction term. From this general system of two coupled equations we obtain the following theoretical frameworks to study non-linear quantum dynamics with long-range interaction: non-linear fractional Schrödinger equation, fractional Ginzburg-Landau equation, fractional generalization of Zakharov system, Hilbert-Zakharov system, Hilbert-Ginzburg-Landau equation and nonlinear Hilbert-Schrödinger equation.

The paper is organized as follows. In Sec.2 we generalize Davydov's Hamiltonian for the case of long-range power-law exciton-exciton interaction. The system of two coupled discrete equations of motion for exciton and phonon subsystems have been found using the Davydov anzatz. Transformation to the system of two continuous equations of motion has been performed in the long wave limit. Sec.3 focuses on new nonlinear fractional differential equations resulting from our general approach to study the 1D exciton-phonon system with long-range interaction. In conclusion, we outline our new developments.

2. LATTICE EXCITON-PHONON HAMILTONIAN WITH LONG-RANGE INTERACTION

2.1 Davydov's Hamiltonian

To model 1D quantum lattice dynamics with long-range exciton-exciton interaction we follow (Davydov, 1991) and consider a linear, rigid arrangement of sites with one molecule at each lattice site. The Davydov's Hamiltonian reads

$$H = H_{ex} + H_{ph} + H_{int}, \tag{1}$$

Here H_{ex} is the Hamiltonian operator of the exciton system, which describes dynamics of intra-molecular excitations or simply excitons, H_{ph} is phonon Hamiltonian operator, which describes molecular displacements or, in other words, the lattice vibrations, and H_{int} is the exciton-phonon operator, which describes interaction of an exciton with the lattice vibrations. The exciton Hamiltonian is

$$H_{ex} = \varepsilon \sum_{n=-\infty}^{\infty} b_n^+ b_n - \sum_{n,m=-\infty}^{\infty} J_{n.m} b_n^+ b_m, \qquad (2)$$

where b_n^+ is creation and b_n is annihilation operators of an exciton on the *n* site. Operators b_n^+ and b_n satisfy the relations $[b_n, b_m^+]=\delta_{n,m}$, $[b_n, b_m]=0$, $[b_n, b_m^+]=0$. Parameter ε is exciton energy on the site, $J_{n,m}$ is the exciton transfer matrix, which describes exciton-exciton interaction between sites *n* and *m*. To extend Davydov's model and go beyond the nearest-neighbour interaction we introduce the power-law interaction between excitons on sites *n* and *m*

$$J_{n,m} = J_{n-m} = \frac{J}{|n-m|^s}, \quad n \neq m, \qquad (3)$$

where J is the interaction constant, parameter s covers different physical models; the nearest-neighbor approximation (s=∞), the dipole-dipole interaction (s=3), the Coulomb potential (s=1). Our main interest will be in fractional values of *s* that can appear for more sophisticated interaction potentials attributed to complex media.

The phonon Hamiltonian H_{ph} is

$$H_{ph} = \frac{1}{2} \sum_{n=-\infty}^{\infty} \left(\frac{\hat{p}_n^2}{m} + w(\hat{u}_{n+1} - \hat{u}_n)^2 \right), \qquad (4)$$

where w is the elasticity constant of the 1D lattice and $\hat{u}_n$ is the displacement operator from the equilibrium position of the site *n*, $\hat{p}_n$ is the momentum operator of the site *n* and *m* is molecular mass.

Finally, the exciton-phonon Hamiltonian H_{int} is

$$H_{int} = \chi \sum_{n=-\infty}^{\infty} (\hat{u}_{n+1} - \hat{u}_n) b_n^+ b_n, \qquad (5)$$

with coupling constant χ. Further, aiming to obtain a system of classical dynamic equations for the exciton-photon system under consideration, we introduce Davydov's ansatz.

2.2 Davydov's anzatz and motion equations

To study system (1) we introduce quantum state vector $|\phi(t)>$ following (Davydov, 1973, 1991), (Scott, 1992)

$$|\phi(t)> = |\Psi(t)>|\Phi(t)>, \qquad (6)$$

where quantum vectors $|\Psi(t)>$ and $|\Phi(t)>$ are defined by

$$|\Psi(t)> = \sum_n \psi_n(t) b_n^+ |0>_{ex}, \qquad (7)$$

and

$$|\Phi(t)> = \exp\left\{ -\frac{i}{\hbar} \sum (\xi_n(t)\hat{p}_n - \eta_n(t)\hat{u}_n \right\} |0>_{ph}, \qquad (8)$$

here $\hbar$ is the Planck's constant, $|0>_{ex}$ and $|0>_{ph}$ are vacuum states of the exciton and phonon subsystems and $\xi_n(t)$ is the diagonal matrix element of the displacement operator $\hat{u}_n$ in the basis defined by (6), while $\eta_n(t)$ is diagonal matrix element of the momentum operator $\hat{p}_n$ in the same basis,

$$\xi_n(t) = <\phi(t)|\hat{u}_n|\phi(t)>, \qquad \eta_n(t) = <\phi(t)|\hat{p}_n|\phi(t)>,$$

The state vector $|\phi(t)>$ satisfies the normalization condition

$$<\phi(t)|\phi(t)> = \sum_n |\psi_n(t)|^2 = N,$$

with $|\psi_n(t)|^2$ being the probability to find exciton on the n^{th} site and *N* is the total number of excitons.

Therefore, the study of dynamics of the exciton-photon system (1) can be performed in terms of the functions $\psi_n(t)$, $\xi_n(t)$ and $\eta_n(t)$. In other words, Davydov's ansatz defined by (6)-(8) allows us to go from the quantum Hamiltonian operator introduced by (1) to the Hamiltonian function developed below. In the basis of the vectors $|\phi(t)>$, the Hamiltonians H_{ex}, H_{ph}, and H_{int} become the functions of classical dynamic variables $\psi_n(t)$, $\psi_n^*(t)$, $\xi_n(t)$ and $\eta_n(t)$

$$<\phi(t)|H_{ex}|\phi(t)> = H_{ex}(\psi_n, \psi_n^*) =$$

$$\varepsilon \sum_{n=-\infty}^{\infty} \psi_n^*(t)\psi_n(t) - \sum_{n,m=-\infty}^{\infty} J_{n-m} \psi_n^*(t)\psi_m(t), \qquad (9)$$

and

$$<\phi(t)|H_{ph}|\phi(t)> = H_{ph}(\xi_n, \eta_n) =$$

$$\frac{1}{2} \sum_{n=-\infty}^{\infty} \left(\frac{\eta_n^2}{m} + w(\xi_{n+1} - \xi_n)^2 \right), \qquad (10)$$

and

$$<\phi(t)|H_{int}|\phi(t)> = H_{int}(\psi_n, \psi_n^*; \xi_n, \eta_n) =$$

$$\chi \sum_{n=-\infty}^{\infty} (\xi_{n+1} - \xi_n)\psi_n^*(t)\psi_n(t), \qquad (11)$$

From (9)-(11) we obtain the system of dynamic equations in discrete space for $\psi_n(t)$, $\xi_n(t)$ and $\eta_n(t)$,

$$i\hbar \frac{\partial \psi_n(t)}{\partial t} = \Lambda \psi_n(t) -$$

$$- \sum_{\substack{m \\ (n \neq m)}} J_{n-m} \psi_m(t) + \chi(\xi_{n+1}(t) - \xi_n(t))\psi_n(t), \qquad (12)$$

$$\frac{\partial \xi_n(t)}{\partial t} = \frac{\eta_n(t)}{m}, \qquad (13)$$

and

$$\frac{\partial \eta_n(t)}{\partial t} = w(\xi_{n+1}(t) - 2\xi_n(t) + \xi_{n-1}(t)) +$$

$$+\chi(|\psi_{n+1}(t)|^2 - |\psi_n(t)|^2), \tag{14}$$

where the constant Λ is

$$\Lambda = \varepsilon + \frac{1}{2}\sum_{n=-\infty}^{\infty}\left(m\left(\frac{\partial \xi_n(t)}{\partial t}\right)^2 + w(\xi_{n+1}(t) - \xi_n(t))^2\right).$$

Substituting $\eta_n(t)$ from (13) into (14) yields

$$m\frac{\partial^2 \xi_n(t)}{\partial t^2} = w(\xi_{n+1}(t) - 2\xi_n(t) + \xi_{n-1}(t)) +$$

$$+\chi(|\psi_{n+1}(t)|^2 - |\psi_n(t)|^2). \tag{15}$$

Our focus now is the system of two coupled discrete dynamic equations (12) and (15).

2.3 From lattice to continuum

To go from the discrete to continuum version of (12) and (15) let us introduce

$$\varphi(k,t) = \sum_{n=-\infty}^{\infty} e^{-ikn}\psi_n(t), \qquad v(k,t) = \sum_{n=-\infty}^{\infty} e^{-ikn}\xi_n(t),$$

where $\psi_n(t)$ is related to $\varphi(k,t)$ as

$$\psi_n(t) = \frac{1}{2\pi}\int_{-\pi}^{\pi} dk e^{ikn}\varphi(k,t),$$

and $\xi_n(t)$ is related to $v(k,t)$ as

$$\xi_n(t) = \frac{1}{2\pi}\int_{-\pi}^{\pi} dk e^{ikn} v(k,t),$$

and k can be considered as a wave number. In the long wave limit when the wavelength exceeds the intersite scale a (let's put for simplicity a=1) we may consider $\varphi(k,t)$ as a $k^{\underline{\text{th}}}$ Fourier component of continuous function $\psi(x,t)$, $\psi_n(t) \xrightarrow[k\to 0]{} \psi(x,t)$ and $v(k,t)$ as a $k^{\underline{\text{th}}}$ Fourier component of function $\xi(x,t)$, $\xi_n(t) \xrightarrow[k\to 0]{} \xi(x,t)$. That is the functions $\psi(x,t)$ and $\varphi(k,t)$ are related each other by the Fourier transform

$$\psi(x,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk e^{ikx}\varphi(k,t), \quad \varphi(k,t) = \int_{-\infty}^{\infty} dx e^{ikx}\psi(x,t),$$

and similarly for $\xi(x,t)$ and $v(k,t)$,

$$\xi(x,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk e^{ikx} v(k,t), \quad v(k,t) = \int_{-\infty}^{\infty} dx e^{ikx}\xi(x,t).$$

Therefore, we conclude that in the long wave limit (12) and (15) become continuous equations of motion

$$i\hbar\frac{\partial \psi(x,t)}{\partial t} = \lambda\psi(x,t) -$$

$$-\int_{-\infty}^{\infty} dy \partial_x K(x-y)\partial_x \psi(x,t) + \chi\frac{\partial \xi(x,t)}{\partial x}\psi(x,t), \tag{16}$$

and

$$m\frac{\partial^2 \xi(x,t)}{\partial t^2} = w\frac{\partial^2 \xi(x,t)}{\partial x^2} + 2\chi\frac{\partial |\psi(x,t)|^2}{\partial x}, \tag{17}$$

where the kernel $K(x)$ in (16) has been introduced as

$$K(x) = \frac{1}{\pi}\int_{-\infty}^{\infty} dk e^{ikx}\frac{G(k)}{k^2},$$

with the function $G(k)$ defined by

$$G(k) = J(0) - J(k), \qquad J(k) = \sum_{\substack{n=-\infty \\ n\neq 0}}^{\infty} e^{-ikn} J_n,$$

here J_n is given by (3) and finally, $\lambda = \Lambda - J(0)$.

Thus, we obtained a new system of coupled dynamic equations (16) and (17) which model 1D exciton phonon dynamics with long-range exciton interaction (3). The field $\psi(x,t)$ describes the exciton subsystem and the field $\xi(x,t)$ describes the phonon subsystem. Equation (16) is the integro-differential equation while equation (17) is the differential one. The integral term in (16), which is a manifestation of non-locality of interaction, comes from the long-range interaction term in the Hamiltonian (2).

3. FRACTIONAL DIFFERENTIAL EQUATIONS TO STUDY EXCITON-PHONON DYNAMICS

To transform the system (16), (17) into the system of coupled differential equations of motion we use the properties of function $G(k)$ at the limit $k\to 0$, which can be obtained from the asymptotics of the polylogarithm (Laskin and Zaslavsky, 2006)

$$G(k) \sim \frac{\pi J}{\Gamma(s)\sin(\pi(s-1)/2)}|k|^{s-1}, \quad 2 \le s < 3, \tag{18}$$

$$G(k) \sim -Jk^2 \ln k, \quad s = 3, \tag{19}$$

$$G(k) \sim \frac{J\zeta(s-2)}{2}k^2, \quad s > 3, \tag{20}$$

where $\Gamma(s)$ is Γ-function, $\zeta(s)$ is the Riemann zeta function and coefficient D_s is defined by

$$D_s = \frac{\pi J}{\Gamma(s)\sin(\pi(s-1)/2)}. \tag{21}$$

It is seen from (18) that the fractional power of k occurs for interactions with $2 \le s < 3$ only. In the coordinate space fractional power of $|k|$ gives us the fractional Riesz derivative of order s-1 (Samko *et al*., 1993), (Saichev and Zaslavsky, 1997), and we come to a fractional differential equation

$$i\hbar\frac{\partial\psi(x,t)}{\partial t}=\lambda\psi(x,t)-$$

$$-D_s\partial_x^{s-1}\psi(x,t)+\chi\frac{\partial\xi(x,t)}{\partial x}\psi(x,t),\quad 2\le s<3,\qquad(22)$$

here ∂_x^{s-1} is the Riesz fractional derivative

$$\partial_x^{s-1}\psi(x,t)=-\frac{1}{2\pi}\int_{-\infty}^{\infty}dke^{ikx}\,|k|^{s-1}\,\varphi(k,t).$$

Thus, our main result is the new system of coupled equations (17) and (22) to study one-dimensional exciton-phonon dynamics with long-range interaction on 1D lattice.

Now let us introduce and briefly discuss a few new general theoretical frameworks which come out from our approach to study one dimensional lattice quantum dynamics with long-range intersite interaction. They are: fractional generalization of Zakharov system, non-linear fractional Schrödinger equation, fractional Ginzburg-Landau equation, Hilbert-Zakharov system, nonlinear Hilbert-Schrödinger equation, and fractional Hilbert-Ginzburg-Landau equation.

3.1 Fractional generalization of Zakharov system

Introducing new variable $\sigma(x,t)=\frac{\partial\xi(x,t)}{\partial x}$ turns (17) and (22) into the following new system of equations for the fields $\psi(x,t)$ and $\sigma(x,t)$,

$$i\hbar\frac{\partial\psi(x,t)}{\partial t}=\lambda\psi(x,t)-$$

$$-D_s\partial_x^{s-1}\psi(x,t)+\chi\sigma(x,t)\psi(x,t),\quad 2\le s<3,\qquad(23)$$

and

$$\left(\frac{\partial^2}{\partial t^2}-v^2\frac{\partial^2}{\partial x^2}\right)\sigma(x,t)=\frac{2\chi}{m}\frac{\partial^2}{\partial x^2}|\psi(x,t)|^2,\qquad(24)$$

where $v=\sqrt{\omega/m}$ is the velocity of sound. This system of two equations can be considered as a fractional generalization of the Zakharov system introduced in 1972 to study the Langmuir waves propagation in an ionized plasma (Zakharov, 1972).

3.2 Non-linear fractional Schrödinger equation

Assuming the existence of a stationary solution $\partial\xi(x,t)/\partial t=0$ in the system of (17) and (22) results in the following fractional differential equation for wave function $\psi(x,t)$,

$$i\hbar\frac{\partial\psi(x,t)}{\partial t}=\lambda\psi(x,t)-$$

$$-D_s\partial_x^{s-1}\psi(x,t)-\frac{2\chi}{\omega}|\psi(x,t)|^2\,\psi(x,t),\quad 2\le s<3,\qquad(25)$$

which can be rewritten in the form of nonlinear fractional Schrödinger equation,

$$i\hbar\frac{\partial\phi(x,t)}{\partial t}=-D_s\partial_x^{s-1}\phi(x,t)-\frac{2\chi}{\omega}|\phi(x,t)|^2\,\phi(x,t),\qquad(26)$$

where $2\le s<3$ and the wave function $\phi(x,t)$ is related to the wave function $\psi(x,t)$ by

$$\phi(x,t)=\exp\{i\lambda t/\hbar\}\psi(x,t).\qquad(27)$$

It follows from (20) that for $s>3$, (26) turns into the non-linear Schrödinger equation

$$i\hbar\frac{\partial\phi(x,t)}{\partial t}=-\frac{J\zeta(s-2)}{2}\partial_x^2\phi(x,t)-\frac{2\chi}{\omega}|\phi(x,t)|^2\,\phi(x,t).$$

where $\partial_x^2=\partial^2/\partial x^2$.

Finally, let's note that the linear fractional Schrödinger equation in one and three dimensions have been developed at first in (Laskin, 2000a,b,c, 2002). Three quantum mechanical problems were studied in these papers for physical applications of developed fractional quantum mechanics; a quantum particle in an infinite potential well, fractional quantum oscillator, and fractional Bohr atom. The energy spectra for these three fractional quantum mechanical problems were found using the linear fractional Schrödinger equation.

3.3 Fractional Ginzburg-Landau equation

In the case of propagating waves we can search for the solution of system (17) and (22) in the form of travelling waves; $\psi(x,t)=\psi(x\text{-v}t)$, and $\xi(x,t)=\xi(x\text{-v}t)$, where v is velocity of the wave. From (17) and (22) let's go to (23) and (24) and substitute $\psi(x,t)=\psi(\zeta)$, and $\sigma(x,t)=\sigma(\zeta)$, where $\zeta=(x\text{-v}t)$. It is easy to see that a solution of (24) is

$$\sigma(x,t)=\frac{2\chi}{m(\mathrm{v}^2-v^2)}|\psi(\zeta)|^2\,.\qquad(28)$$

Then (23) results in nonlinear equation

$$i\hbar\mathrm{v}\frac{\partial\psi(\zeta)}{\partial\zeta}=\lambda\psi(\zeta)-$$

$$-D_s\partial_\zeta^{s-1}\psi(\zeta)+\gamma|\psi(\zeta)|^2\,\psi(\zeta),\quad 2\le s<3,\qquad(29)$$

where γ is the nonlinearity parameter

$$\gamma=\frac{2\chi}{m(\mathrm{v}^2-v^2)}.\qquad(30)$$

Equation (29) can be considered as a fractional generalization of the well known Ginzburg-Landau equation. Fractional Ginzburg-Landau equation has been initially proposed in (Weitzner and Zaslavsky, 2003).

3.4 Hilbert-Zakharov system

It follows from (18) that in the case s=2 the function $G(k)$ at the limit $k \rightarrow 0$ takes the form

$$G(k) \sim \pi J \, | \, k \, |, \quad s = 2.$$

Hence, (23) becomes

$$i\hbar \frac{\partial \psi(x,t)}{\partial t} = \lambda \psi(x,t) -$$

$$- \pi J \mathscr{H} \{\partial_x \psi(x,t)\} + \chi \sigma(x,t) \psi(x,t), \quad s = 2, \qquad (31)$$

here $\mathscr{H}$ is the Hilbert integral transform defined by

$$\mathscr{H} \{\varphi(x,t)\} = P \int_{-\infty}^{\infty} dy \, \frac{\varphi(y,t)}{y - x},$$

where P stands for the Cauchy principal value of the integral.

We will call the system of equations (31) and (24) as the Hilbert-Zakharov system

3.5 Nonlinear Hilbert-Schrödinger equation

In the case when s=2 and $\partial \xi(x,t)/\partial t$=0, the system of (16), (17) results in the following non-linear quantum mechanical equation for wave function $\psi(x,t)$,

$$i\hbar \frac{\partial \psi(x,t)}{\partial t} = \lambda \psi(x,t) -$$

$$- \pi J \mathscr{H} \{\partial_x \psi(x,t)\} + \frac{2\chi}{m} | \, \psi(x,t) \, |^2 \, \psi(x,t). \qquad (32)$$

Introducing wave function $\phi(x,t)$ related to the wave function $\psi(x,t)$ by means (27), brings the Hilbert-Schrödinger equation

$$i\hbar \frac{\partial \phi(x,t)}{\partial t} = -\pi J \mathscr{H} \{\partial_x \phi(x,t)\} - \frac{2\chi}{\omega} | \, \phi(x,t) \, |^2 \, \phi(x,t). \quad (33)$$

3.6 Hilbert-Ginzburg-Landau equation

In the case when s=2, let's search for the solution of the system (31), (24) in the form of travelling waves, $\psi(x,t)$= $\psi(x$-v$t)$, and $\xi(x,t)$= $\xi(x$-v$t)$, where v is velocity of the wave. The solution of (24) has the form of (27). Thus, (31) results in

$$i\hbar \mathrm{v} \frac{\partial \psi(\zeta)}{\partial \zeta} = \lambda \psi(\zeta) - \pi J \mathscr{H} \{\partial_x \psi(\zeta)\} + \gamma \, | \, \psi(\zeta) \, |^2 \, \psi(\zeta), \quad (34)$$

where γ is the nonlinearity parameter introduced by (03). We will call (34) as the Hilbert-Ginzburg-Landau equation.

4. CONCLUSIONS

It has been shown that the long-range power-law interaction leads, in general, to a nonlocal integral term in the equation of motion if we go from discrete to continuous space. In some particular cases for power-law interaction with non-integer power s the integral term can be expressed through the fractional order derivative. That is, non-locality originating from the long-range interaction reveals the dynamics in the form of space derivatives of fractional order.

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