Molecular Bose-Einstein condensation and bound states in lithium niobate type ferroelectrics

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The modal dynamics of both dark and bright solitons in lithium niobate is already established in Klein-Gordon lattice. For small oscillations, the modal dynamics is characterized as bound states, revealed via Associated Legendre polynomial. Bose Einstein condensation exclusively takes place around bosonic particles having different wave functions within the bound states. This paper tried to explore BEC in the realm of bound state for better understanding of switching phenomenon of devices. The pairing and interplay between the dark and bright solitons occur with their effect on the condensation. The bound state disappears after a critical frequency. The quasi-particles then become phonons in the unbound states that propagate through the domains.

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1. Introduction

The most outstanding experimental discovery in recent times is Bose-Einstein condensation (BEC) in 1995 [1]. This discovery has triggered both theoretical and experimental works on this fascinating topic of research. The reasons for these vigorous activities are: a) it gives an opportunity to open a new window for a macroscopic view of quantum mechanics, and b) it makes such studies most lucrative in the field of matter-wave relations. On the latter issue, experiments show unusual excitations within the wave that is known as solitons. The unusual properties of such quasi-particles, i.e. both bright and dark solitons (henceforth called bosonic particles or simply particles), in a condensate would allow us to manipulate them in periodic or other potential. There is some evidence for the formation of both dark and bright solitions in the condensate, but the concept of bound state still remains somewhat illusive, despite a lot of activities on nonlinear optical systems that are important for many devices. This gives us motivation to study bound state with a connection to BEC. Further, we find out both lower and upper bounds in relation to frequency and the extent of condensation in the bound state. Although we use data on lithium niobate ferroelectrics for general theoretical study, it can also be extended to other relevant systems, e.g. in magnon system with two-well Landau potential [2]. This effort is made through Klein-Gordon (K-G) equation which on perturbation gives rise to nonlinear Schrodinger equation (NLSE) that is a variant of Gross-Pitaevskii equation (GPE), which is popular and commonly used for studying BEC.

K-G equation was developed earlier [3] through vibrational principle taking various relevant energies in the

Landau-Ginzburg (L-G) potential in the continuum Hamiltonian. Recently, this was extended to ferroelectrics using one dimensional array of domains to probe localization for intrinsic localized modes [4]. An effective mechanism of 'energy localization' in K-G lattice, arising out of the discreteness and non-integrability of the system, was presented by Bang and Peyrard [5]. Very recently, a perturbation on the continuum K-G model through progressive wave NLSE [6] showed both bright and dark solitons with 'discrete energy levels', estimated via hypergeometric function. This dark soliton with low energy and lower velocity is not visible as it is part of the complex solution indicating the presence of an energy-gap. In this communication, for these solitons, our main focus lies on the bound state in the low frequency limit in the context of BEC. A critical frequency is also shown beyond which the phonon takes over in the unbound states in the high frequency regime.

There are excellent reviews on BEC in the vast ocean of literature, notably Ref [1]. An excellent work was done by Kivshar et al on solitons in nonlinear optics [7]; the dynamical generation and control of both bright and dark solitons in matter-wave BEC in optical lattices were extensively studied by several authors (see Ref. [7]). In this context, a review on dark solitons in atomic BEC by Frantzeskakis and Kevrekedis et al [8] also needs a mention. In atomic optics, the formation of coherent molecular BEC was explained in one-dimension by mean field theories of parametric nonlinearities that convert two solitons to one (and vice-versa) for non-integrable equations [9]; molecular BEC was invoked for a new type of reaction between molecules (multispecies) to engineer condensates of heavier molecules where macroscopic occupation of single molecular quantum state gives rise to

the coherent bosonic stimulation (see Ref. [9]). BEC was also reported on 10^5 Li₂ molecules in an optical trap with spin mixture of fermionic Li atoms by measuring a collective excitation mode [10]. This gives us further motivation to explore BEC in the realm of bound state that might help towards better understanding of switching phenomenon in a vast area of devices including many nano devices. Now, let us look at the potential and the bound states.

2. Theoretical development

The free energy density for the order parameter (P) can be written in L-G form: $G = -(\alpha_1/2)P^2 + (\alpha_2/4)P^4$. Where, α 's are Landau coefficients and here P is also a function of space. Now, $\frac{\partial G}{\partial P} = E = -\alpha_1 P + \alpha_2 P^3$. Here, E is the intrinsic field

and all the terms are non-dimensional; The relevant values in the context of a ferroelectric system is given in Ref. [4,6]. The 2^{nd} derivative of Landau energy is:

$$g''(P) = E/E_c = -\alpha_1 + 3\alpha_2 P^2$$
 (1)

Where, E_c is the switching field in kV/cm. Let us consider an idealized one-dimensional array of N identical rectangular domains along the x direction. Between the neighboring domains, there is domain wall and nearest neighbor coupling (K) is considered. For the mode dynamics of the extended modes and modes that are localized, nonlinear K-G equation relating P against space (x) and time (t) with a non-dimensional driving field (E_0) is [**3,6**]:

$$\frac{\partial^2 P}{\partial t^2} - \bar{K} \frac{\partial^2 P}{\partial x^2} - \bar{\alpha}_1 P + \bar{\alpha}_2 P^3 - E_0 = 0 \qquad (2)$$

K-G equation is a well-known equation of mathematical physics that exhibits a variety of interesting properties with applications in different physical systems [5,11]. K-G equation is useful for both dark and bright discrete breathers that throw light on quantum localization [11-13]. For highly localized modes in K-G lattice, bright soliton solutions have also been used for nonlinear dynamics of DNA [5]. Due to the localization, the length scale of excitation assumes more significance that obviously drives us to the nano domain, whose importance in the field of solid state physics cannot be denied. Next, let us go for the solutions: In the continuum limit, let P be the solution of Eq. (2) that is replaced by P = P(x) + f(x, t). Here, P(x) and f(x,t) are the functions of x and (x,t) respectively. From physics point of view this combination describes a periodic kink which, a priori, can experience the presence of phonons about its center of mass regardless of its dynamical property [14]. Thus, the resulting eigenvalue equation will be governed by a linearized problem. Let us write the space dependent equation:

$$-\bar{K}\frac{\partial^2 P(x)}{\partial x^2} - \bar{\alpha_1}P(x) + \bar{\alpha_2}P^3(x) = 0$$
(3)

Before describing our main focus area, let us briefly talk about a different approach of using Lame equation for the bound states [15]. Although the context is different, by deriving Jacobi elliptic function from Eq. (3) and using Eq. (1) as the index \rightarrow 1, we get a series of bound states, but their stability could not be assured with the Jacobian form of Lame equation (see *Appendix A*). Here, we use Associated Legendre Polynomial (ALP) to reveal a much richer physics by showing stable "upper and lower bounds". To note that for light-induced waveguide Segev et al. [16] used ALP function for modal composition of incoherent spatial solitons in nonlinear Kerr medium. Now, we go for the spatio-temporal equation:

$$\frac{\partial^2 f(x,t)}{\partial t^2} - \bar{K} \frac{\partial^2 f(x,t)}{\partial x^2} + g''(P)f(x,t) - E_0 = 0$$
(4)

Here, P(x) given in Eq. (3) is the static single 'kink' solution (not discussed here) with a form: $P(x) = \tanh qx$, where, $q = \sqrt{\bar{\alpha_1}/(2\bar{K})}$. Using this

form of P(x) and taking $\overline{\alpha}_1 \approx \overline{\alpha}_2$ in Eq. (1), we have

$$g''(P) = -\bar{\alpha_1} + 3\bar{\alpha_2} \tanh^2 qx = 2\bar{\alpha_1} \left(1 - (3/2) \sec h^2 qx \right)$$
(5)

g''(P) varies mainly in the region of the kink centre (assumed to be at x=0) and approaches a constant value (taken to be unity) far from the kink centre, and also g''(P) < 0 at x = 0. For small oscillations ($E_0 \approx 0$), f(x,t) is written as: $f(x,t) = \psi(x)e^{-i\omega t}$ (ω = angular frequency). Hence, the eigenvalue equation is:

$$\left(\bar{K}\frac{\partial^2}{\partial x^2} + 3\bar{\alpha_1}\sec h^2 qx\right)\psi = X\psi \qquad (6)$$

X is the eigenvalue of the system defined as: $X = (2\bar{\alpha_1} - \omega^2)$. Eq. (6) is identical with the Schrodinger equation for a particle moving in onedimensional potential well (g''(P)). This is considered as a variant of GPE. So, the bound and unbound states can be observed for this potential. For bosonic particles, let us introduce ALP to estimate the frequency of nondegenerate states with different wave functions. For soliton dynamics, in the low frequency range, the bound state will predominate, when condensation takes place and in the higher range, the phonon mode takes over in the unbound state, and here these are the main issues. Let us introduced a new variable: $z = \tanh qx$, then Eq. (6) becomes

$$(1-z^2)\frac{\partial^2 \psi}{\partial z^2} - 2z\frac{\partial \psi}{\partial z} + \left(n(n+1) - \frac{m^2}{1-z^2}\right)\psi = 0 \quad (7)$$

Where, $m^2 = 2X / \overline{\alpha_1} = 4 - (2\omega^2 / \overline{\alpha_1})$. The solution of Eq. (7) is: $\psi = p_2^m(z)$. In the bound state, ω is denoted as $\underline{\omega}_b$ and wave function ψ as ψ_b . If $0 \le \omega_b \le \sqrt{2 \, \alpha_1}$, then 'm' is real and ALP is only valid if n=2. The existence of different states is considered in the bound state in this limited range of frequency or energy. Here, all our solutions are 'real' and 'stable' in terms of interplay between the mode index and the frequency. When the bound state frequency, , 'm' will be imaginary. Under this condition, ALP is not valid, i.e. the bound state disappears. This is considered as a "critical" limit of frequency for the 'upper bound'. The impact of this critical limit on condensation is shown at the end of Section III.

3. Results and Discussion

The 'lower bound' of the non-degenerate state is at $\omega_b = 0$ for which the wave function (ψ_b) with translation symmetry gives rise to Goldstone mode (GM) for m = 2:

$$\psi_b = P_2^2(z) = 3\sec h^2 qx \tag{8}$$

To note that bright solitons predominate. This wave function for bosonic particles is not shown here, as it simply shows a typical Gaussian band. The wave functions for other symmetries of GM were not worked out to remain within our main focus on the bound state and BEC formation. As the frequency increases to: $\omega_b > \sqrt{(3\bar{\alpha}_1)/2}$, the system starts showing polarization within a band of $m = \pm 1$, whose wave functions are:

$$\psi_b = P_2^1(z) = 3\tanh qx \sec hqx \tag{9}$$

$$\psi_b = P_2^{-1}(z) = -(1/6) \tanh qx \sec hqx$$
 (10)

To note that both dark and bright solitons exist, and a pairing or coupling has started in the system. A small number of particles become polarized in opposite directions with the above value of eigenfrequency. Wave functions, as per Eq. (9) and (10), are shown in **Fig. 1a** and **Fig. 1b** respectively indicating that these behaviors manifest in both + and - directions starting at zero. Finally, the wave function for the 'upper bound' of the non-degenerate state with the limiting frequency

$$\omega_b = \sqrt{2 \, \alpha_1}$$
 is:
 $\psi_b = (1/2)(3 \tanh^2 q x - 1)$ (11)



Fig. 1: The wave function (ψ_b) for the quasi-particle, (a) As per Eqs. (9), when m = +1. The behavior is seen to move towards positive direction starting at zero. (b) As per Eqs. (10), when m = -1. The behavior is seen to move towards opposite direction starting at zero, compared to that in Fig. 1a.

To note that dark solitons have completely taken over, before going to the unbound state. After clearly observing both lower and upper bounds [Eq. (8) and (11)], we extend

1887

our results for both dark and bright solitons through a compact operator that is designed to evaluate the frequency for a chosen wave function. Following Eq. (6), the eigenvalue equation can be written as: $L\psi_b = X\psi_b$.

Here, $X = (2\bar{\alpha_1} - \omega^2)$ is the eigenvalue of the wave function ψ_b operated by the operator L as:

$$L = \frac{X}{2} \left(L_{+} L_{-} + L_{-} L_{+} \right)$$
(12)

Where,
$$L_{-} = \sqrt{\bar{K}/X} \left[\sqrt{(3\bar{\alpha_{1}}/\bar{K})} \sec hqx + i\frac{\partial}{\partial x} \right]$$

and $L_{+} = \sqrt{\bar{K}/X} \left[\sqrt{(3\bar{\alpha_{1}}/\bar{K})} \sec hqx - i\frac{\partial}{\partial x} \right].$

With this compact form, let us deal with the solitons as a preamble to our discussion on the BEC formation and number density.

For static dark soliton, the above eigenvalue equation contributes to the wave function with translation symmetry as: $\psi_b = \sqrt{N_0} \tanh qx$. This spatial relation is valid in the bound state within the corresponding frequency. Here, N_0 is the total number of bosonic particles. From the above operator (Eq. 12), this idea leads to the dispersion relation for small oscillations:

$$\omega_b = 2\alpha_1 \tanh^2 qx \tag{13}$$

The solution of Eq. (4) leads to the spatio-temporal wave function: $f_b(x,t) = \sqrt{N_0} \tanh qx.e^{-i\omega t}$. The dark soliton wave function and the frequency (Eq. 13) are plotted against spatial dimension in **Fig. 2a**. For BEC formation around bosonic particles at very low frequency, it is noted from **Fig. 2a** that the frequency curve touches the dark soliton wave function at zero frequency giving the signature of BEC, and also the dip of frequency curve does not cover up the dark soliton wave function (but merely touches it). Let us now describe the bright soliton.



Fig. 2: For small oscillations in the bound state for a coupling value= 4, (a) The frequency dispersion relation and the dark soliton wave function behavior. The frequency curve is seen to touch the wave function for the dark soliton at zero frequency giving the signature of BEC. Below and above this coupling value, the frequency curve does not coincide with the dark soliton behavior. (b)The frequency curve is seen to fully cover up the wave function peak for the bright soliton indicating full condensation at Goldstone Mode frequency.

The bright soliton wave function is: $\Psi(x) = \sqrt{N_0}$ sechqx using the above operator (Eq. 12) with the spatial dispersion relation:

$$\omega_b^2 = 2\bar{\alpha_1} - 2\bar{\alpha_1} \sec h^2 q x - 1/2 = 2\bar{\alpha_1} \tanh^2 q x - 1/2$$
(14)

It is seen that the energy is lower than that of dark soliton. Now, the solution of equation (4) is: $f_b(x,t) = \sqrt{N_0} \sec hqx.e^{-i\omega t}$. Similar to **Fig. 2a**, here we also plot bright soliton wave function and the frequency (Eq. 14) in **Fig. 2b**. This shows that the dip of frequency is able to fully cover up the peak of the bright soliton wave function. As shown below, bright soliton is totally condensed, since its energy is lower than that of dark soliton by -(1/2), comparing Eq. (14) and (13). These figures seem to indicate a 'pairing' of two solitons. Various possibilities of conversion from dark to bright solitons (and vice-versa) and pairing within them occur in the bound state. Having discussed both dark and bright solitons, and their 'interplay', let us show the evidence of BEC.

It is considered that within the limited frequency range, the condensation takes place, as per the dispersion relations. Due to bosonic particles within the bound state, their amplitude can be expressed by number density. In the context of matter-wave relation, as the frequency starts increasing, the dark solitons seem to ride on the back of bright solitons, which are totally condensed at zero frequency. Hence, some particles are considered as remaining outside the 'condensate'. The probability of finding these "free particles" is: $f_b^* f_b = N_0 - N$, and by taking the dark soliton wave functions we derive $N_0 \tanh^2 qx = f_b^* f_b = N_0 - N$. Where, N is the condensed particle density. This is re-written as: $[1 - (N/N_0)] = \tanh^2 qx$. Below BEC transition (for a finite temperature) using the dispersion relation (Eq. 13), the form of BEC relating number density and the frequency is:

$$(N/N_0) = 1 - (\omega_b^2/2\alpha_1)$$
 (15)

Although dimers of bosons decay with time, the fermionic ⁶Li dimers show remarkable stability with lifetimes far longer than those for elastic collisions and thermalization [10,17]. In important nonlinear optical systems, such as lithium niobate or tantalate, as per Ref. [11], the pentavalent niobium or tantalum atoms are sitting in their positions (in terms of the double-well Landau potential) due to cooperative effect, whereas Li atom tends to play a tricky (?) role in that it goes up and down from its equilibrium position and thereby, intuitively speaking, it may drive the bosonic particles for some kind of conversion to take place. This might inspire the 'experimentalists' to work with larger molecules (multispecies) containing lithium [9,10]. As shown below, when the frequency increases the particles in the condensate decreases (Eq. (15)). Here, the Landau coefficient (read, nonlinearity) assumes more significance. The number density of BEC is shown against frequency in **Fig. 3.** It is seen that at $\omega_b = \sqrt{2\overline{\alpha_1}}$, at a value of Landau coefficient of 353.42 (equivalently at 0.133 mole% niobium antisite defect at a switching field of 40kV/cm) [6], N/N_0 goes to zero at upper bound. For 'quantum breathers' by quantizing discrete K-G Hamiltonian with Bosonic operators, a quantum pinning transition was observed at 40 kV/cm under periodic boundary condition in the 'phonon hopping coefficient' vs. impurity plot in lithium niobate [12].

At Goldstone mode $(\omega_b = 0)$, all the bright solitons get condensed (**Fig. 2b**) in the bound state as per Eq. (8). For $\omega_b = \sqrt{\alpha_1}$, $N = N_0/2$, which means that half of the number density of particles is condensed, i.e. the number of uncondensed particles (dark solitons) starts increasing. As the frequency in the system further increases to: $\omega_b = \sqrt{(3/2)\alpha_1}$, $1/4^{\text{th}}$ of particles is condensed and $3/4^{\text{th}}$ of the number density are still in the bound state, also revealing polarized particles in the opposite directions at $m=\pm 1$, as per ALP formalism. This could be construed as the origin of polarization vis-à-vis bound state just below

the upper bound.



Fig. 3: The number density of BEC is shown against frequency. It is seen that at $\omega_b = \sqrt{2\overline{\alpha_1}} = 26.59$ (nondimensional) that has to be divided by 10^9 second. This frequency is for Landau coefficient of 353.42, when the fraction of BEC condensation (N/N₀) goes to zero, and this frequency is considered as the 'critical limit' beyond which there is no more condensation of bosonic particles in the system, i.e. at or beyond the upper bound of the bound states, when phonon propagation starts in the unbound state.

Finally, at criticality, $\omega_{b} = \sqrt{2 \, \dot{\alpha_{1}}}$, the bound state totally disappears, and no particle can be condensed anymore in the system. Thus, in the context of BEC, the ALP formalism (Eq. 8 to 11) is quite noteworthy. At this criticality, the spatial extent (i.e. equivalent to dimer-dimer distance=0.6a as per Petrov et al [10], where *a*=scattering length) is estimated to be 104 nm, and 15 nm for 75% condensation; for Li₂ molecular BEC, a varied from 116 to 185 nm, as per Jochim et al [10]. Note that with decreasing condensation, a increases thereby indicating the dark solitons are going apart. With the compact operator, the number density of bosons is clearly related to the frequency, which has not been attempted before, and BEC exclusively forms within this bound state. Above critical limit, the phonon is dominant in the system in the unbound state. In the wave function of phonon, if we put the propagation constant k=0, we get back the bound state solution at the "critical limit" (Eq. 11), which implies "localization" at lower energy that again indicates a richer physics (see Appendix B).

4. Conclusions

Bose-Einstein condensation is shown in the realm of bound state that disappears after a critical frequency and thereafter no more condensation in the system. Within the bound state, the 'interplay' between the dark and bright solitons increases the former reducing the condensate density. Also, the phonons start propagating within the system in the unbound state at higher energy, when scattering becomes important.

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Appendix-A: Here, we could also use Lame equation by taking the solution of Eq. (3) as:

$$P(x) = l \sqrt{\frac{2}{(1+l^2)}} Sn\left(\sqrt{\frac{2\bar{\alpha_1}}{\bar{K}(1+l^2)}}x, l\right)$$
(3a)

Here, *Sn* is the 'Jacobi elliptic function' of modulus *l* with $0 \le l \le 1$. The above equation is a periodic kink soliton that is static. In the limit $l \rightarrow 1$, for the infinite length kink solutions, the elliptic function becomes hyperbolic function as: P(x) = tanh qx (*q* defined in the text). Such solutions can be obtained by using the 2nd derivative of the Landau energy (Eq. (1)), and we could get a series of bound states by deriving Jacobian form of 'Lame equation, while the question of their stability could not be tackled.

Appendix-B: Having shown BEC in the lower regime of frequency in the bound state, it is natural to look for what happens in the unbound state. After the above critical limit of frequency, the switchover of bound \rightarrow unbound state takes place and all the quasi-particles as 'phonons' will propagate in a given domain region in the system and through the domain wall. The phonons will propagate through the system with strong dispersion relation as:

$$\omega_P^2 = 2\bar{\alpha_1} + k^2 \bar{K} q^2 \tag{16}$$

Where *k* is the propagation constant and now ω_b has to be replaced by phonon frequency (ω_P). It is convenient to use Eq. (16) in Eq. (6) that produces phonon solutions for $0 \le k \le \infty$ without using the normalized constant of $1/\sqrt{(3\pi)}$ as:

$$\psi_{P} = (1/\sqrt{1+k^{2}}) (3 \tan h^{2}qx - 3ik \tanh qx - (1+k^{2})) e^{ikqx}$$
(17)

This solution indicates propagation through onedimensional array of domains in x direction. The complex conjugate of the above function for waves that propagate through the system along the x direction is expressed as:

$$\psi_{P}^{*} = (1/\sqrt{1+k^{2}}) (3 \tan h^{2}qx' + 3ik \tanh qx' - (1+k^{2})) e^{-ikqx'}$$
(18)

In the wave function of phonons in the unbound state, if we put k= 0, we get the bound state solution at the "critical limit" (Eq. 11), which implies "localization" at lower energy that again indicates a richer physics. When phonons pass through the nonlinear optical medium, they encounter scattering in the vicinity of domain wall. Here, the Green function has a role to play in explaining this scattering in which phonons get shifted from one point to another point that should be the subject matter of future work in revealing the switching behavior in such systems.

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