Introduction.— In biological systems, a driven nonequilibrium condensation phenomenon which was often compared with the Bose–Einstein condensation (BEC) was hypothesized by Fröhlich in the late 1960s [1–4]. While BEC is a low-temperature equilibrium phenomenon whose discovery was only recently realized as the cooling technology advances [5, 6], the Fröhlich condensate is intrinsically an out-of-equilibrium condensation of collective vibrations at the lowest mode driven by an external energy supply and is expected to occur at much higher temperatures [7]. The proposal has stimulated a wide range of studies on the collective modes in nonequilibrium systems such as quasi-equilibrium magnon condensation at room temperature [8, 9], the polariton condensation in photonic systems [10–14] as well as nonequilibrium phase transitions [15, 16]. Recently, due to the rapid progress of terahertz technology, many experimental and analytical works have been carried out recently in this direction and signals of the long-anticipated nonequilibrium condensation were finally observed [17, 18]. More experimental investigations in both biological and non-biological systems have been proposed such as using lysozymes and the bovine serum albumin (BSA) proteins with spectroscopy and an array of membranes coupled with an optomechanical cavity [19, 20].

Though Fröhlich’s theory of nonequilibrium condensation dates back more than half a century ago, quantum and statistical properties of the system near the transition remain largely unknown. The rate equations in Fröhlich’s reasoning were constructed heuristically by requiring a Bose–Einstein distribution of the phonons in the absence of energy sources [1, 2]. It does not provide information on how systems behave during transitions to condensates. The microscopic theory was formulated by Wu and Austin through the quantum Hamiltonian and finite-temperature Green’s function techniques [21–24]. They concluded the same rate equations and confirmed the possibility of detecting the phonon condensation in biological systems even at room temperature. The rate equations suggest the emergence of the cooperative self-organization when the input energy is high and the chemical potential approaches the lowest phonon energy in the branch of vibrational modes [23, 24], while leaving certain essential questions pertaining to the properties of the transition unanswered. In this Letter, we study the statistics of the phonons with an explicit aim to reveal the transition behavior of the Fröhlich condensate and the statistics near the transition. We show for the first time the analytical proof of the transition behavior of the condensate. The common lore for predicting the feature of Fröhlich condensate is to make an analogy of the BEC. Nonetheless, a priori assumptions of the condensate according to the BEC should be carefully scrutinized. As pointed out in Ref. [19], Fröhlich condensation is a different nonequilibrium phenomenon whose mechanism is more suitable compared to that of a single mode maser rather than the atomic BEC. Besides, the calculation in [19] using a classical pumping field shows a continuous crossover between the two phases. Tough some critical pump strength can be defined artificially as a reference scale at which the external input is roughly strong enough to generate the condensate, there is no qualitative distinction across the two phases. On the other hand, recent experiments using BSA proteins have indicated not only the existence of the collective oscillations of the biomolecules but also a sign of nonequilibrium phase transition when a control parameter exceeds a critical threshold value [18]. Like many researches on Fröhlich condensate, the analysis in Ref. [18] also adopts a semi-classical method approximated by dequantizing the Wu-Austin Hamiltonian to obtain the classical equations of motion. However, such treatment renders the analysis nearly intractable and difficult to retrieve information.

In this Letter, we adopt a full quantum treatment of the nonequilibrium system with the surrounding solvents and external fields. We show analytically that the transition from the nonequilibrium steady state with no condensate to the condensate state is a second-order phase transition in the large-N limit regardless of certain approximations used in precedent researches. Unlike BEC, the condensation process does not require extremely low temperature but a high-temperature energy input. Besides the main goal, we also demonstrate the statistics and the fluctuations profiles of the condensate phonons near the phase transition point.

Driven Dissipative System.—The appearance of Fröhlich condensate is believed to be a general consequence of the following three conditions: dissipation to the thermal bath, external energy source, and nonlinearity. For the terahertz vibrations inside protein molecules such as phonon modes in DNA
or BSA proteins, the surrounding solvents function approximately as a thermal bath. The nonlinear coupling between the system vibration modes and the surrounding medium in the second quantization form gives rise to the Hamiltonian formulated by Wu and Austin [21–24]. The system we consider is modeled by a collection of \( N \) oscillators in a narrow bandwidth \( \omega_j \in \hat{I}_{sys} \) which corresponds to the normal modes of the electromagnetic vibrations. The vibrational modes are annihilated by the operator \( a_i \). The normal modes interact with the surrounding biological system with the excitation energies \( \Omega_k \) and the associated annihilation operator \( b_k \), as well as the external source with frequency \( \Omega \).

\[ V_{int} = \hbar \sum_i \sum_k f_{ik} a_i^\dagger b_k e^{i(\omega_k - \Omega) t} + \hbar \sum_{i,j,k} g_{ijk} a_i^\dagger a_j b_k e^{i(\omega_k - \omega_j - \Omega) t} + \hbar \sum_i \omega_i a_i^\dagger a_i + \text{h.c.} \]  

where \( f_{ik} \) and \( g_{ijk} \) are the coupling constants between the molecular vibrations and the solvent, and \( \omega_i, \Omega_k \) and \( \Omega_k \) denote the frequencies associated with the \( i \)-th vibrational mode of molecules, the solvent modes and the energy source. The Hamiltonian has been criticized for the unbounded potential energy from below [26]. This does not concern us here as the Hamiltonian is only used as an effective approximation in the interested domain of energy instead of a fundamental description.

The rate equation of the condensate can be derived directly from the above interaction Hamiltonian. The details of the derivation can be found in the Supplementary Material. We use the standard techniques of the full quantum master equation with Born and Morkov approximations which returns formally the equations of motion for the whole system. We then reduce the equations of motion of the density operator to only the \( l \)-th state. After rearranging and contracting the repeated indices, we obtain the rate equation of phonons at mode \( \omega_l \) in reminiscence of the heuristic equation given by Fröhlich:

\[ \langle n_l \rangle = \langle \bar{n}_{\omega_l} - \langle n_l \rangle \rangle + \Lambda \langle n_{\omega_l} - \langle n_l \rangle \rangle + \chi \left\{ \sum_{j,l,j} \langle \bar{n}_{\omega_j}\langle n_j - n_l \rangle \rangle + \langle n_j n_l + n_l \rangle \right\} \]

where \( \phi = 2\pi \int_0^{\infty} D(\omega) \), \( \Lambda = 2\pi \int_0^{\infty} D(\omega) \), \( \chi = 2\pi \int_0^{\infty} D(\omega) \) are the rates of dissipation, energy input and energy redistribution, respectively. \( D(\omega) \) is the density of states of the surrounding environment. The rate equation of the total number of phonons \( \bar{N} = \sum_{\omega = 0}^D a_i^\dagger a_i \) can be further calculated to be the following: \( \langle \bar{N} \rangle \approx \frac{(D + 1)(\phi \bar{n} + \Lambda n_{ext})}{\phi + \Lambda} \).

where \( \bar{n} \) is the occupation at frequency \( \omega_0 \) of the solvent and \( n_{ext} \) is the occupation of the external source at \( \omega_0 \). Notice that the nonlinear term does not contribute to the total phonon number and only contributes to the energy redistribution. At the steady state, the phonon number at mode \( \omega_0 \) can be formally expressed as

\[ \langle n_0 \rangle = \frac{\phi \bar{n}_0 + \chi(\bar{n}_0 + 1)((\langle N \rangle - \langle n_0 \rangle) + \Lambda n_{ext})}{\phi + \Lambda - \chi((\langle N \rangle - \langle n_0 \rangle - D\bar{n}_0))} \]

It is easy to check that by setting the nonlinear coupling \( \chi = 0 \), the mean value of phonon number at mode \( \omega_0 \) becomes the weighted average of thermal distributions due to the solvent and the external source: \( \langle n_0 \rangle = \frac{\phi \bar{n}_0 + \Lambda n_{ext}}{\phi + \Lambda} \). This is expected from the secular approximation used in the equations of motion which ignores the off-diagonal subdominant contribution [27–29]. To obtain Eq. (4), we have assumed the decorrelation approximation between the \( n_0 \) and \( n_i \) namely \( \langle n_0 n_i \rangle \approx \langle n_0 \rangle \langle n_i \rangle \) and \( \langle n_0^2 \rangle = \langle n_0 \rangle \), in the limit of large molecule numbers, the solution of the mean condensate number is:

\[ \langle n_0 \rangle = \frac{N - D\bar{n}}{2} + \frac{\sqrt{(N - D\bar{n})^2 + 4(\bar{n} + 1)N}}{2} \]

It is easy to see that the fraction of the condensate becomes \( \langle n_0 \rangle/N \rightarrow 0 \) for the case \( n_{ext} \leq \bar{n} \). When \( n_{ext} > \bar{n} \), we have:

\[ \langle n_0 \rangle/N \approx \frac{\Lambda}{\phi \bar{n} + \Lambda n_{ext}}(n_{ext} - \bar{n}) \]

which suggests a continuous phase transition at \( n_{ext} = \bar{n} \) and gives the critical exponent \( \beta = 1 \).

Condensate statistics and fluctuations without decorrelation approximation. The above approach to solve for the approximate mean value of condensate phonons is a reminiscence of Fröhlich’s equation. However, one can argue that the decorrelation approximation used in obtaining the formula is heavily invalidated by the coherence in the vibrational modes as well as fluctuations in each mode. Therefore, in the case of situations with strong coherence or near the transition point, such approximations are not applicable and a new approach needs to apply. To gain insight into such systems, we study the quantum statistics of phonons in the condensate state assuming only the decorrelation between the total number of phonons \( N \) and the condensate phonons \( \langle n_0 \rangle \), namely, \( \langle n_0 N \rangle \approx \langle n_0 \rangle \langle N \rangle \). This approximation is reasonable since the total phonon number is a conserved quantity under the interaction Hamiltonian. The statistics of the condensate contains not only the information about the mean value of the phonon numbers in the condensate but also its distribution. One of the important features of the distribution is captured by its the
Mandel-Q parameter defined as $Q = \rho - 1$, where $\rho$ is the Mandel parameter. For any classical probability distribution it's distance from the classical Poisson distribution is characterized by $Q$. For simplicity, we denote the probability of finding $n$ phonons in the condensate. For one specific $n_0$, the diagonal element has the physical interpretation of the probability of finding $n_0$ phonons on the lowest mode. The steady state solution for the distribution of the condensate phonons can be solved exactly. For details, please refer to Supplementary Material. For simplicity, we denote the probability as $P(m)$ where $m \in \{0, 1, 2, 3, \ldots \}$, then the reduced equations of motion of the system have simple solutions as follows:

$$P(m) = P(0) \left(1 + \frac{1}{\bar{n}_s}\right)^m \frac{(N - m)_{(m)}}{(N + D - m)_{(m)}},$$

where $(q)_{(m)}$ is the rising Pochhammer symbol, $N$ is defined as

$$N = N + 1 + \frac{\phi \bar{n}_s + \Lambda n_c}{\chi (\bar{n}_s + 1)} \quad \text{and} \quad D = D + \frac{\phi (\bar{n}_s + 1) + \Lambda (n_c + 1)}{\chi \bar{n}_s} - 1.$$  

The probability of zero condensate phonon $P(0)$ can be determined by the normalization condition $\sum_m P(m) = 1$, which gives:

$$P(0)^{-1} = \begin{aligned} 
{\begin{array}{c}
2F_1 \left(1, -N; -D - N + 1; 1 + \frac{1}{\bar{n}_s}\right), \\
\end{array}}
\end{aligned}$$

where $2F_1 \left(1, -N; -D - N + 1; 1 + \frac{1}{\bar{n}_s}\right)$ is the Gauss hypergeometric function of type $(2, 1)$. As a reminder, $D$ is the total number of oscillators in the system and $N$ is the total number of phonons in all modes given in Eq. [3]. The mean phonon numbers in the condensate can be calculated by the summation $\langle n_0 \rangle = \sum_m mP(m)$. The fraction of the phonon condensate $\langle n_0 \rangle/N$ is given as follows:

$$\frac{\langle n_0 \rangle}{N} = \frac{N(1 + \frac{1}{\bar{n}_s})}{(D + N - 1)_{(D + N - 1)} 2F_1 \left(1, -N; -D - N + 1; 1 + \frac{1}{\bar{n}_s}\right)/N}.$$

In deriving Eq. [7] for $\delta n < 0$, we have treated the total phonon number $N$ as its mean value given in Eq. [5]. The second moment of condensate phonon number takes the analytic form:

$$\langle n_0 \rangle^2 = \frac{N(1 + \frac{1}{\bar{n}_s})^2 2F_2 \left(2, 1 - N; -D - N + 2; 1 + \frac{1}{\bar{n}_s}\right)}{(D + N - 1)_{(D + N - 1)} 2F_1 \left(1, -N; -D - N + 1; 1 + \frac{1}{\bar{n}_s}\right)}.$$

The Mandel Q parameter defined by

$$Q = \frac{\text{Var}[n_0]}{\text{E}[n_0]} - 1 = \langle (n_0^2) \rangle - \langle n_0 \rangle^2 \langle n_0 \rangle - 1$$

can be directly computed from the above expressions [see Fig. 1].

Phase transition. For the Gauss hypergeometric function given in Eq. [2], $2F_1(a, b; c; d)$ is bounded when the ratio $\frac{c}{b} < d$ and it converges to a finite positive value as $b \to \infty$. Notice that in the large-$N$ limit, the total phonon number and the total vibrational modes are related by $D \approx \frac{1}{(\bar{n}_s + \delta n)}N$ where $\delta n = \frac{\Lambda}{\phi \bar{n}_s} (n_c - \bar{n}_s)$. Therefore, in this case the fraction of condensate phonons is reduced to:

$$\frac{\langle n_0 \rangle}{N} = \frac{(1 + \frac{1}{\bar{n}_s})_{(D + N - 1)} 2F_1 \left(1, -N; -\left(1 + \frac{1}{\bar{n}_s + \delta n}\right)N; 1 + \frac{1}{\bar{n}_s}\right)}{(1 + \frac{1}{\bar{n}_s + \delta n})_{(D + N - 1)} 2F_1 \left(1, -N; -\left(1 + \frac{1}{\bar{n}_s + \delta n}\right)N; 1 + \frac{1}{\bar{n}_s}\right)}.$$

It can be shown that for large integer values of $N$ and $\delta n < 0$, the ratio of the hypergeometric functions is finite and the fraction vanishes:

$$\frac{\langle n_0 \rangle}{N} \to 0 \quad \text{for} \quad \delta n < 0 \text{ and } N \in \mathbb{Z}^+.$$
Notice that in large-$N$ reduces to the same as the surrounding solvent: For $\delta = 0$, $T_{cr} = T_{solvent}$ in the case of thermal distribution of the external source. In other words, when the external source has an effective temperature lower than that of the surrounding medium no condensate will form. The formation starts when the external source has a higher effective temperature. Therefore, the Fröhlich condensate is a result of the high-power nonequilibrium driving and is drastically different from the BEC where in most situations condensation appears only in the low temperature limit with low degrees of dissipation. On the other hand, the emergence of the condensation also infers the off-diagonal long-range order in the system indicated by the Penrose-Onsager criterion \cite{31, 32}. In the long-distance limit, the single-particle density matrix becomes $\rho(x, x') = \langle \psi(x) \psi(x') \rangle \to n_s$, where $n_s$ stands for the density of the condensate. The emergence of the condensate, though obtainable from the dequantized or semi-classical equations, suggests a strong enhancement of non-classicality of the system.

The phase behavior was studied in the case of a classical pumping field where the pump coupling $V_p = \sum_x F_x(t) a_x e^{-i\omega x t}$ is featured by a broad spectrum $(F_x(t))^2 = \frac{1}{2} \delta(t-t')$ with the single control parameter $r$ denoting the pumping rate \cite{19}. However, the issue with the incoherent classical pumping is that it is qualitatively similar to an external source at an infinitely high temperature. The only free parameter is the coupling. As we have discussed, the coupling influences only quantitatively the fraction of the condensate rather than the condition of the initiation of the phase transition in the large-$N$ (large molecule number) limit. Therein many of the critical properties at low temperatures and near the transition are erased. For instance, the critical energy supply at which the condensation emerges, in this situation, approaches zero in the large-$N$ limit: $r = \frac{\phi}{D^2T_1} (1 + \frac{\phi}{\chi}) D^\to 0$. The meaning of it is that the condensate phonons at the lowest mode start to accumulate from the zero pumping and increase with the pumping field until saturation. As shown in Fig. 2 and Fig. 3, the condensation will emerge regardless the types of irradiating energy but the transition to the condensation phase becomes a continuous crossover with no real distinction between the phases in this case.

In Fig. 1 we show that the Mandel $Q$ parameter reaches the maximal value in the transition point $n_{ex} = \bar{n}_s = 16$ and

\[ n_{ex} \sim \frac{(D - 1) \Lambda - 2\phi}{(D + 1)\Lambda} \bar{n}_s + \left( \frac{\phi + \Lambda}{\chi} + n_{cr} - 1 \right) \frac{\phi + \Lambda}{(D + 1)\Lambda}. \]

\[ n_{ex} = \bar{n}_s, \]
decays rapidly away from the phase transition point. For extremely large energy input, \( Q \) becomes negative and it suggests a sub-Poissonian statistics similar to that in the resonance fluorescence of a single atom. In Fig. 2 the fraction of condensate phonons as the occupation of the external source is simulated. For relatively small total phonon numbers (e.g. \( N = 200 \)), the transition from the ordinary nonequilibrium phase to the condensation phase is similar to a continuous crossover due to the finiteness of the system. In particular, in this case the condensation fraction is nonzero even for zero external energy input due to the finiteness of the system and the thermal equilibrium with the surrounding solvent. However, in the large-\( N \) case, the transition becomes sharp and the fraction is negligible at zero input, which is expected at the thermodynamic limit. This agrees with the findings reported in a recent experiment [18].

We compare the solution with the decorrelation approximation Eq. (5) and the solution reported in a recent experiment [18]. We compare the solution with the decorrelation approximation Eq. (5) and the solution without the assumption Eq. (9). The difference maximizes near the transition point (for \( D = 200 \), the critical point is at \( n_c \approx 17.4 \)). The numerical simulations show the transition agrees with our analysis according to Eq. (15). In Fig. 3 we show that in the case of a classical incoherent broad-spectrum pumping, such transition will not appear.

Conclusions.—It has been long hinted that molecules go through a phase transition to the condensate states during the Fröhlich process. In this Letter, we developed a full quantum treatment of the Fröhlich condensation and the critical phenomenon. In particular, we presented an analytical proof of the phase transition based on Wu-Austin Hamiltonian and outlined the conditions for witnessing such transitions. We analyzed the phonon distribution and fluctuations near the transition point and showed that the phase transition is only realizable in the large-\( N \) limit. In biological systems, this condition can be naturally realized and this analysis demonstrates the viability of broad experimental prospect to observe the phase transition of the molecular vibrations into a coherent quantum state in biological systems. Recent experiments have made tremendous progress in the nonequilibrium condensation in biological systems, and signs of such phase transition as shown in Fig. 2 were already witnessed for the first time. Further experimental investigations and smoking-gun evidences are in need to exclude all potential artifacts and to confirm the finding. It is hoped that this will open a new door for further explorations of out-of-equilibrium collective oscillations in biomolecules.

---

wangxh@ucas.ac.cn
jin.wang.1@stonybrook.edu

1] H. Fröhlich, Long-range coherence and energy storage in biological systems, International Journal of Quantum Chemistry 2, 641 (1968).

2] H. Fröhlich, Bose condensation of strongly excited longitudinal electric modes, Physics Letters A 26, 402 (1968).

3] H. Fröhlich, Long range coherence and the action of enzymes, Nature 228, 1093 (1970).

4] H. Fröhlich, Theoretical physics and biology, in Biophysical Coherence and Response to External Stimuli edited by H. Fröhlich (Springer Berlin Heidelberg, Berlin, Heidelberg, 1988) pp. 1–24.

5] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell, Observation of bose-einstein condensation in a dilute atomic vapor, science 269, 198 (1995).

6] K. B. Davis, M.-O. Mewes, M. R. Andrews, N. J. van Druten, D. S. Durfee, D. Kurn, and W. Ketterle, Bose-einstein condensation in a gas of sodium atoms, Physical review letters 75, 3969 (1995).

7] Y. M. Bunkov and G. Volovik, Spin superfluidity and magnon bose-einstein condensation, KH Bennemann and JB Ketterson, International Series of Monographs on Physics 156, 253 (2013).

8] S. O. Demokritov, V. E. Demidov, O. Dzyapko, G. A. Melkov, A. A. Serga, B. Hillerbrands, and A. N. Slavin, Bose–einstein condensation of quasi-equilibrium magnons at room temperature under pumping, Nature 443, 430 (2006).

9] A. Chunak, G. Melkov, V. Demidov, O. Dzyapko, V. Safonov, and S. Demokritov, Bose-einstein condensation of magnons under incoherent pumping, Physical review letters 102, 187205 (2009).

10] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. J. Keeling, F. Marchetti, M. Szymańska, R. André, J. Staehli, et al., Bose–einstein condensation of exciton polaritons, Nature 443, 409 (2006).

11] F. Manni, K. G. Lagoudakis, B. Pietka, L. Fontanesi, M. Wouters, V. Savona, R. André, and B. Deveaud-Plédran, Polariton condensation in a one-dimensional disordered potential, Physical Review Letters 106, 176401 (2011).

12] H. Deng, H. Haug, and Y. Yamamoto, Exciton-polariton bose-einstein condensation, Reviews of modern physics 82, 1489 (2010).

13] Z. Zhang, S. Zhao, and D. Lei, Quantum fluctuations and coherence of a molecular polariton condensate, arXiv preprint arXiv:2204.13528 (2022).

14] J. Bloch, I. Carusotto, and M. Wouters, Non-equilibrium bose–einstein condensation in photonic systems, Nature Reviews Physics , 1 (2022).

15] H. Haken, Cooperative phenomena in systems far from thermal equilibrium and in nonphysical systems, Reviews of modern physics 47, 67 (1975).

16] H. Haken, Synergetics. An Introduction. Nonequilibrium Phase Transitions and Self-organization in Physics, Chemistry, and Biology (Springer, 1977).

17] J. R. Reimers, L. K. McKemmish, R. H. McKenzie, A. E. Mark, and N. S. Hush, Weak, strong, and coherent regimes of fröhlich condensation and their applications to terahertz medicine and quantum consciousness, Proceedings of the National Academy of Sciences 106, 4219 (2009).

18] I. Nardecchia, J. Torres, M. Lechelon, V. Giliberti, M. Ortolani, P. Nouvel, M. Gori, Y. Meriguet, I. Donato, J. Preto, et al., Out-of-equilibrium collective oscillation as phonon condensation in a model protein, Physical Review X 8, 031061 (2018).

19] Z. Zhang, G. S. Agarwal, and M. O. Scully, Quantum fluctuations in the fröhlich condensate of molecular vibrations driven far from equilibrium, Physical Review Letters 122, 158101 (2019).

20] X. Zheng and B. Li, Fröhlich condensate of phonons in optomechanical systems, Physical Review A 104, 043512 (2021).

21] T. Wu and S. Austin, Bose condensation in biosystems, Physics Letters A 64, 151 (1977).

22] T. Wu and S. Austin, Bose-einstein condensation in biological
systems, Journal of theoretical biology 71, 209 (1978).
[23] T. Wu and S. Austin, Cooperative behavior in biological sys-
tems, Physics Letters A 65, 74 (1978).
[24] T. Wu and S. J. Austin, Fröhlich’s model of bose condensa-
tion in biological systems, Journal of Biological Physics 9, 97 (1981).
[25] J. Preto, Semi-classical statistical description of fröhlich con-
densation, Journal of biological physics 43, 167 (2017).
[26] H. Bolterauer, Elementary arguments that the wu-austin hamil-
tonian has no finite ground state (the search for a microscopic foun-
dation of fröhlichs theory), Bioelectrochemistry and bioen-
tergetics 48, 301 (1999).
[27] H.-P. Breuer, F. Petruccione, et al., The theory of open quantum systems (Oxford University Press on Demand, 2002).
[28] X. Wang and J. Wang, Nonequilibrium effects on quantum cor-
relations: Discord, mutual information, and entanglement of a two-fermionic system in bosonic and fermionic environments, Physical Review A 100, 052331 (2019).
[29] X. Wang and J. Wang, The effect of nonequilibrium entropy production on the quantum fisher information and correlations, Quantum Information Processing 21, 1 (2022).
[30] L. Mandel, Squeezed states and sub-poissonian photon statistics, Physical Review Letters 49, 136 (1982).
[31] O. Penrose and L. Onsager, Bose-einstein condensation and liq-
uid helium, Physical Review 104, 576 (1956).
[32] C. N. Yang, Concept of off-diagonal long-range order and the quantum phases of liquid he and of superconductors, Reviews of Modern Physics 34, 694 (1962).

SUPPLEMENTARY MATERIAL

MODELS AND RATE EQUATIONS

We start from the Wu-Austin Hamiltonian to model the collective oscillations of biomacromolecules in the solvent:
\[ H = \sum_i \hbar \omega_i a_i^\dagger a_i + \sum_i \hbar \Omega_i b_i^\dagger b_i + \sum_i \hbar \Omega_i p_i^\dagger p_i + \hbar \sum_i \sum_k f_{ik} a_i^\dagger b_k e^{i(\omega_i - \Omega_k)} + \hbar \sum_i \sum_j g_{ij} a_i^\dagger a_j b_i e^{i(\omega_i - \omega_j)} + \hbar \sum_i \sum_k \lambda_{ik} p_i^\dagger p_k e^{i(\omega_i - \Omega_k)} + \text{h.c.}, \]
(17)
where \( a_i \) is the annihilation operator for the vibrational modes, \( b_i \) is the annihilation operator for the surrounding biological system (solvent), and \( p_i \) is for the external source. In most biological settings, the solvent contains a large degrees of freedom with a short decoherence time and can be modeled as the Markovian bath. With this Hamiltonian, we can write the equation of motion for the density matrix of the system using the standard techniques:
\[ \dot{\rho} = -\frac{1}{\hbar^2} \int_0^\infty dt' \text{Tr}_b \{ [V_{\text{int}}(t), [V_{\text{int}}(t-t'), \rho_b \otimes \rho_s(t)]] \} \]
(18)
where \( \rho_s \) is the reduced density matrix of the system, \( \rho_b \) is the density matrix of the bath, and \( V_{\text{int}} \) is the interaction Hamiltonian in the second line of Eq. (17). Since we will be dealing with only the system density matrix from now on and we omit the subscript “s” from the reduced density matrix \( \rho_s \). After some algebra, we obtain the equations of motion for the system density operator:
\[ \dot{\rho} = \frac{\phi}{2} \sum_{j=0}^D \{ (\bar{n}_{\omega_j} + 1) (a_{s j}^\dagger a_{s j}^\dagger a_{s j} a_{s j}) + \bar{n}_{\omega_j} (a_{s j}^\dagger a_{s j}^\dagger a_{s j}^\dagger a_{s j}) \} \\
+ \frac{\Lambda}{2} \sum_{j=0}^D \{ (\bar{n}_{\omega_j} + 1) (p_{s j}^\dagger p_{s j}^\dagger p_{s j} - p_{s j}^\dagger p_{s j}) + \bar{n}_{\omega_j} (p_{s j}^\dagger p_{s j} - p_{s j}^\dagger p_{s j}) \} \\
+ \frac{\chi}{2} \sum_{j=1}^D \sum_{j'=0}^{j-1} \{ (\bar{n}_{\omega_{j'}} + 1) (a_{s j'}^\dagger a_{s j'}^\dagger a_{s j} - a_{s j'}^\dagger a_{s j} a_{s j'} a_{s j}) + \bar{n}_{\omega_{j'}} (a_{s j'}^\dagger a_{s j'} a_{s j}^\dagger a_{s j} - a_{s j'}^\dagger a_{s j} a_{s j'} a_{s j}) \} + \text{h.c.}, \]
(19)
where \( \bar{n}_{\omega} = [\exp(\hbar \omega/k_B T) - 1]^{-1} \) is the occupation number of the bath at frequency \( \omega \), and \( \phi = 2\pi f_{\omega}^e \mathcal{D}(\omega), \Lambda = 2\pi f_{\omega}^d \mathcal{D}(\omega), \chi = 2\pi f_{\omega}^\omega \mathcal{D}(\omega) \) are the rates of pumping, dissipation (one-phonon) and energy redistribution (two-phonon), respectively. \( \mathcal{D}(\omega) \) is the density of states of the bath. To concentrate on the collective behavior of condensation, we further reduce the system density matrix to the condensate state:
\[ \rho_{\text{con}} := \sum_{[n_k]} \langle n_k | \rho | n_k \rangle, \quad [n_k] = \{ n_1, n_2, \ldots, n_D \}, \]
(20)
where \( \{ n_k \} \) is the configuration of the excited states phonons. Hereafter, we further omit the subscripts for convenience. The elements of the condensate density matrix are \( \rho_{\phi_{\omega},n} = \sum_{\{n_k\}} \langle n_I | n_k \rangle | \rho | n_I \rangle \langle n_k | \). Then the mean phonon number at mode \( \omega_j \) is \( \langle n_I \rangle = \sum_{n_I=0}^{\infty} n_I \rho_{\phi_{\omega},n_I} \). The rate equations of phonon numbers reads

\[
\langle \hat{n}_I \rangle = \sum_{n_I=0}^{\infty} n_I \rho_{\phi_{\omega},n_I} ,
\]

where

\[
\dot{\rho}_{\phi_{\omega},n} = \sum_{\{ n_k \}} \langle n_I, \{ n_k \} | \rho | n_I, \{ n_k \} \rangle ,
\]

\[
= \phi \sum_{x=0}^{D} \sum_{\{ n_k \}} \sum_{\{ n_k \}} \left[ \left( \bar{n}_{\omega_x} + 1 \right) \left( a_x^\dagger a_x^\dagger - a_x^\dagger a_x \right) + \bar{n}_{\omega_x} \left( a_x^\dagger a_x^\dagger - a_x a_x^\dagger \right) \right] | \varphi \rangle | n_I, \{ n_k \} \rangle
\]

\[
+ \Lambda \sum_{x=0}^{D} \sum_{\{ n_k \}} \sum_{\{ n_k \}} \left[ \left( \bar{n}_{\omega_x} + 1 \right) \left( p_x^\dagger p_x^\dagger - p_x p_x \right) + \bar{n}_{\omega_x} \left( p_x^\dagger p_x - p_x p_x^\dagger \right) \right] | \varphi \rangle | n_I, \{ n_k \} \rangle
\]

\[
+ \chi \sum_{x=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} \sum_{\{ n_k \}} \left[ \left( \bar{n}_{\omega_x} + 1 \right) \left( a_x^\dagger a_x a_j^\dagger a_j + a_x^\dagger a_x^\dagger a_j^\dagger a_j \right) + \bar{n}_{\omega_x} \left( a_x^\dagger a_x a_j^\dagger a_j^\dagger - a_j^\dagger a_x a_j^\dagger a_x \right) \right] | \varphi \rangle | n_I, \{ n_k \} \rangle .
\]

The calculation of the above rate equation can be greatly simplified if we decompose the triple summations proportional to \( \chi \) by the following rule:

\[
\sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} = \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}}
\]

\[
= \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}}
\]

\[
= \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}}
\]

\[
= \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}} + \sum_{s=1}^{D} \sum_{j=0}^{D} \sum_{\{ n_k \}}
\]

It is easy to check that only the last two summations in the last line of Eq. (22) contribute to the \( \dot{\rho}_{\phi_{\omega},n_I} \) and the first two summations are identically zero. After some manipulation of algebra, we can find the expression of Eq. (22) to be the following,

\[
\dot{\rho}_{\phi_{\omega},n} = (\phi \langle \hat{n} \rangle + 1 + \Lambda (n_{\text{ex}} + 1)) \left( n_I + 1 \right) \rho_{\phi_{\omega}+1,n_I+1} - n_I \rho_{\phi_{\omega},n} \right) + (\phi \langle \hat{n} \rangle + \Lambda n_{\text{ex}} + 1) \left( n_I + 1 \right) \rho_{\phi_{\omega}+1,n_I+1} - n_I \rho_{\phi_{\omega},n} \right) + (\phi \langle \hat{n} \rangle + \Lambda n_{\text{ex}} + 1) \left( n_I + 1 \right) \rho_{\phi_{\omega}+1,n_I+1} - n_I \rho_{\phi_{\omega},n} \right)
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]

\[
= \sum_{j=0}^{D} \sum_{\{ n_k \}} \left[ \langle n_I + 1 | a_j^\dagger a_j | n_I + 1; n_j \rangle | \varphi \rangle | n_I + 1; n_j \rangle \right]
\]
Using the dynamics of the state Eq. (24), we obtain the rate equation as the following:

\[
\langle \dot{n}_t \rangle = (\phi(n_t + 1) + \Lambda(n_{ex} + 1)) \sum_{n_t=0}^{\infty} n_t (n_t + 1) \rho_{n_{ex}+1,n_t+1} - n_t \rho_{n_t,n_t} + (\phi n_t + \Lambda n_{ex}) \sum_{n_t=0}^{\infty} n_t (n_t \rho_{n_{ex}+1,n_t} - (n_t + 1) \rho_{n_{ex},n_t}) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} n_t (n_t \nu_{\omega,j} \langle n_t - 1; \{n_{j}\} | \rho | n_{j} - 1; \{n_{j}\} \rangle - (n_t + 1) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
+ \bar{n}_{\omega,j} \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} n_t (n_t + 1) \langle n_{j} | \rho | n_{j} \rangle - n_t (n_{j} | \rho | n_{j} \rangle) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \sum_{\{n_{j}\}} n_t (n_t + 1) \langle n_{j} | \rho | n_{j} \rangle - n_t (n_{j} | \rho | n_{j} \rangle) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \sum_{\{n_{j}\}} n_t (n_t + 1) \langle n_{j} | \rho | n_{j} \rangle - n_t (n_{j} | \rho | n_{j} \rangle) \\
= (\phi \bar{n}_{\omega} + \Lambda n_{ex}) (n_t + 1) - \langle \phi \bar{n}_{\omega} + 1 + \Lambda(n_{ex} + 1) \rangle \langle n_t + 1 \rangle + \chi \sum_{j=1}^{D} [\bar{n}_{\omega,j} + 1] \langle n_{j} | \rho | n_{j} \rangle + \bar{n}_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle + \bar{n}_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle)
\]

To simplify the rate equation \langle \dot{n}_t \rangle, we apply the following identity:

\[
\sum_{n_t=0}^{\infty} n_t \dot{\rho}_{n_t,n_t} = \sum_{n_t=0}^{\infty} (n_t - 1) \dot{\rho}_{n_{t-1},n_t} = \sum_{n_t=0}^{\infty} (n_t + 1) \dot{\rho}_{n_t+1,n_t+1},
\]

and rewrite the rate equation as follows:

\[
\langle \dot{n}_t \rangle = (\phi(n_t + 1) + \Lambda(n_{ex} + 1)) \sum_{n_t=0}^{\infty} ((n_t + 1)^2 - (n_t + 1)) \rho_{n_{ex}+1,n_t+1} - n_t^2 \rho_{n_t,n_t} \\
+ (\phi n_t + \Lambda n_{ex}) \sum_{n_t=0}^{\infty} ((n_t + 1)^2 \rho_{n_{ex}+1,n_t} - (n_t^2 + n_t) \rho_{n_t,n_t}) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} ((n_t - 1)^2 n_t \nu_{\omega,j} \langle n_t - 1; \{n_{j}\} | \rho | n_{j} - 1; \{n_{j}\} \rangle - (n_t^2 + n_t) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
+ \bar{n}_{\omega,j} \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} ((n_t + 1) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle - (n_t + 1) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
+ \bar{n}_{\omega,j} \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} ((n_t + 1)^2 \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle - (n_t^2 + n_t) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} ((n_t + 1) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle - (n_t + 1) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
+ \bar{n}_{\omega,j} \sum_{\{n_{j}\}} \sum_{n_{j}=0}^{\infty} ((n_t + 1)^2 \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle - (n_t^2 + n_t) \nu_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle) \\
\]

After some manipulation, we have:

\[
\langle \dot{n}_t \rangle = (\phi(n_t + 1) + \Lambda(n_{ex} + 1)) \left(\langle n_t^2 - n_t \rangle - \langle n_t \rangle \right) + (\phi n_t + \Lambda n_{ex}) \left(\langle n_t^2 + 2n_t + 1 \rangle - \langle n_t^2 + n_t \rangle \right) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \left(\langle n_t (n_t^2 + 2n_t + 1) \rangle - \langle n_t (n_t^2 + n_t) \rangle \right) + \bar{n}_{\omega,j} \left(\langle (n_t + 1)n_t^2 - n_t(n_t + 1) \rangle - \langle n_t^2(n_t + 1) \rangle \right) \\
+ \chi \sum_{j=1}^{D} (\bar{n}_{\omega,j} + 1) \left(\langle (n_t + 1)n_t^2 \rangle - \langle n_t(n_t + 1) \rangle \right) + \bar{n}_{\omega,j} \left(\langle n_t(n_t + 1)^2 \rangle - \langle (n_t^2 + n_t)n_t \rangle \right) \\
\]

\[
= (\phi \bar{n}_{\omega} + \Lambda n_{ex}) (n_t + 1) - (\phi (\bar{n}_{\omega} + 1) + +\Lambda(n_{ex} + 1)) \langle n_t + 1 \rangle + \chi \sum_{j=1}^{D} [\bar{n}_{\omega,j} + 1] \langle n_{j} | \rho | n_{j} \rangle + \bar{n}_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle + \bar{n}_{\omega,j} \langle n_{j} | \rho | n_{j} \rangle)
\]
To recapitulate, the rate equation of phonon numbers at mode $\omega_j$ is shown to be:

$$
\langle n_j \rangle = \phi [\bar{n}_{\omega_j} - \langle n_j \rangle] + \Lambda [n_{ex} - \langle n_j \rangle] + \chi \left( \sum_{j \neq i} [\bar{n}_{\omega_j} \langle n_j - n_i \rangle + \langle n_j n_i \rangle] + \sum_{j < l} [\bar{n}_{\omega_j} \langle n_j - n_l \rangle - \langle n_j n_l \rangle] \right),
$$

(29)

then the rate equation of the total number of phonons $N = \sum_{\omega \in \Omega} n_{\omega}$ can be calculated under the approximations $\bar{n}_{\omega_l} \approx \bar{n}_{\omega_0} \equiv \bar{n}$, $\bar{n}_{\omega_j} \approx \bar{n}_{\omega_0} \equiv \bar{n}$:

$$
\langle N \rangle = (D + 1)(\phi \bar{n} + \Lambda n_{ex}) - (\phi + \Lambda)\langle N \rangle + \chi \left( \sum_{j=1}^{D} \sum_{l=1}^{D} \langle n_j (n_l + 1) \rangle - \sum_{l=1}^{D} \sum_{j=0}^{D-1} \langle (n_j + 1) n_l \rangle \right).
$$

(30)

Here, the nonlinear term vanishes identically. Therefore, in the NESS the total number of phonons is:

$$
\langle N \rangle = \frac{(D + 1)(\phi \bar{n} + \Lambda n_{ex})}{\phi + \Lambda}.
$$

(31)

In particular, the rate equation for the condensate phonon number $n_0$ according to Eq. (28) is:

$$
\langle n_0 \rangle = \phi [\bar{n} (\langle n_0 \rangle + 1) - \langle n + 1 \rangle (\langle n_0 \rangle)] + \Lambda [n_{ex} (\langle n_0 \rangle + 1) - \langle n_{ex} + 1 \rangle (\langle n_0 \rangle)] 
+ \chi \left( \sum_{j=1}^{D} \sum_{l=1}^{D} \langle n_j (n_l + 1) (\langle n_0 \rangle + 1) - \bar{n}_{\omega_j} (\langle n_0 \rangle + 1) \rangle \right)
$$

(32)

where in the $\chi$-part in Eq. (32), the first term describes the process that the mode $\omega_0$ is pumped by the mode $\omega_s$, together with the dissipation by the bath mode $\omega_s - \omega_0$. The reverse process is described by the second term. At the steady state, the phonon number at mode $\omega_0$ can be formally expressed as:

$$
\langle n_0 \rangle = \frac{\phi \bar{n} + \chi (\bar{n} + 1) (\langle N_s \rangle + \Lambda n_{ex})}{\phi + \Lambda - \chi (\langle N_s \rangle - \Lambda n_{ex})} 
= \frac{\phi \bar{n} + \chi (\bar{n} + 1) (\langle N \rangle - \langle n_0 \rangle)}{\phi + \Lambda - \chi (\langle N \rangle - \langle n_0 \rangle)}.
$$

(33)

The nonlinear terms proportional to $\chi$ is essential in the condensation process. It is easy to check that by setting $\chi = 0$, the mean value of phonon number at mode $\omega_0$ is given by the thermal average of the environment

$$
n_0 = \frac{\phi \bar{n} + \Lambda n_{ex}}{\phi + \Lambda},
$$

(34)

which reduces to the thermal distribution in the case of single bath.

**STATISTICS AND FLUCTUATIONS**

To study the statistics and the fluctuations of the phonon condensate, we will only assume the decorrelation between the condensate state $n_0$ and total phonon number $N$, $\langle n_0 N_T \rangle \approx N_T \langle n_0 \rangle$, rather than the decorrelation approximation $\langle n_j n_l \rangle \approx \langle n_j \rangle \langle n_l \rangle$ as used in Fröhlich’s original work. This is due to the fact that the total phonon number $N$ is solely dictated by pumping and dissipation (one phonon processes), instead of the nonlinear two-phonon process. We proceed by defining the density matrix for the mode $\omega_0$

$$
\rho_{n_0 n_0} = \sum_{|n_0|} \langle n_0, |n_0| |n_0, |n_0| \rangle, \quad |n_j| = \{n_1, n_2, \cdots, n_D\}.
$$

(35)

Restrained only to the lowest vibrational state $|\omega_0\rangle$, Eq. (24) gives

$$
\dot{\rho}_{n_0 n_0} = (\phi (\bar{n} + 1)) \left( \langle n_0 + 1 \rangle \rho_{n_0 + 1, n_0 + 1} - n_0 \rho_{n_0, n_0} \right) + (\phi \bar{n}) \left( \langle n_0 \rangle \rho_{n_0 + 1, n_0 + 1} - (n_0 + 1) \rho_{n_0, n_0} \right) 
+ (\Lambda (n_{ex} + 1)) \left( \langle n_0 + 1 \rangle \rho_{n_0 + 1, n_0 + 1} - n_0 \rho_{n_0, n_0} \right) + (\Lambda n_{ex}) \left( \langle n_0 \rangle \rho_{n_0 + 1, n_0 + 1} - (n_0 + 1) \rho_{n_0, n_0} \right) 
+ \chi \sum_{j=1}^{D} \sum_{|n_j|} \langle n_0 \rangle \left( n_0 \langle n_0 \rangle - 1 \right) \left( n_j \langle n_0 \rangle - 1 \right) - (n_0 + 1) n_j \langle n_0 \rangle \langle n_j \rangle \right),
$$

(37)
Using the smooth spectrum approximation,
\[
\sum_{s=1}^{D} (\bar{n}_{w_s} + 1)(n_s)_{n_0} = (\bar{n} + 1)((N) - n_0), \quad \text{and} \quad \sum_{s=1}^{D} \bar{n}_{w_s}(n_s + 1)_{n_0} = \bar{n}((N) - n_0 + D),
\] (38)

one has the following equations
\[
\dot{\rho}_{n_0, n_0} = - (\phi \bar{n} + \Lambda n_{e x} + \chi(\bar{n} + 1)((N) - n_0))(n_0 + 1)\rho_{n_0, n_0}
+ (\phi \bar{n} + \Lambda n_{e x} + \chi(\bar{n} + 1)((N) - n_0 + 1))n_0\rho_{n_0 - 1, n_0 - 1}
- (\phi(\bar{n} + 1) + \Lambda(n_{e x} + 1) + \chi \bar{n}((N) - n_0 - 1 + D))(n_0 + 1)\rho_{n_0 + 1, n_0 + 1}.
\] (39)

Now that we have reduced the density matrix into the lowest diagonal mode \(\rho_{n_0, n_0} = \sum_{n_0} \langle n_0 | \rho \rangle \langle n_0 | \phi \rangle \langle n_0 | n_0 \rangle\), where \(\{n_1, n_2, \ldots, n_0\}\) is the configuration of the excited states and \(n_0\) is the variable denoting the phonon number in the condensate. For simplicity, we denote the probability as \(P(m)\) where \(m \in \{0, 1, 2, 3, \ldots\}\). To solve the phonon distribution function \(P(n_0)\) at the nonequilibrium steady state, notice that we can regroup Eq. (39) into two equivalent equations:
\[
(\phi \bar{n} + \Lambda n_{e x} + \chi(\bar{n} + 1)((N) - n_0))(n_0 + 1)\rho_{n_0, n_0} = (\phi \bar{n} + \Lambda(n_{e x} + 1) + \chi \bar{n}((N) - n_0 - 1 + D))(n_0 + 1)\rho_{n_0 + 1, n_0 + 1} = 0,
\]
\[
(\phi \bar{n} + \Lambda n_{e x} + \chi(\bar{n} + 1)((N) - n_0 + 1))n_0\rho_{n_0 - 1, n_0 - 1} = (\phi(\bar{n} + 1) + \Lambda(n_{e x} + 1) + \chi \bar{n}((N) - n_0 + D))(n_0 + 1)\rho_{n_0 + 1, n_0 + 1} = 0.
\] (40)

Notice that the series of equations of different \(n_0\)’s are now related by geometric progression whose solution for \(m\) condensate phonons is:
\[
P(m) = P(0) \left(1 + \frac{1}{\bar{n}}\right)^m \frac{(N + 1 + \phi \bar{n} + \Lambda n_{e x})_{(m)}}{(N + D + \phi(\bar{n} + 1) + \Lambda(n_{e x} + 1))_{(m)}} = P(0) \left(1 + \frac{1}{\bar{n}}\right)^m \frac{\Gamma(N + 1 + \phi \bar{n} + \Lambda n_{e x})}{\Gamma(N + D + \phi(\bar{n} + 1) + \Lambda(n_{e x} + 1))}.
\] (41)

Then, we redefine variables as the following:
\[
N = N + 1 + \frac{\phi \bar{n} + \Lambda n_{e x}}{\chi(\bar{n} + 1)} \equiv N + \alpha = \frac{(D + 1)(\phi \bar{n} + \Lambda n_{e x})}{\phi + \Lambda} + \alpha
\]
\[
D = D + \frac{\phi(\bar{n} + 1) + \Lambda(n_{e x} + 1)}{\chi \bar{n}} - \frac{\phi \bar{n} + \Lambda n_{e x}}{\chi(\bar{n} + 1)} - 1 \equiv D + \beta.
\] (43)

After substitution of variables, the probability distribution can be rewritten simply as:
\[
P(m) = P(0) \left(1 + \frac{1}{\bar{n}}\right)^m \frac{(N - m)_{(m)}}{(N + D - m)_{(m)}} = P(0) \left(1 + \frac{1}{\bar{n}}\right)^m \frac{\Gamma(N) \Gamma(N + D - m)}{\Gamma(N - m) \Gamma(N + D)}.
\] (45)

The probability of zero condensate \(P(0)\) can be determined by the normalization condition \(\sum_m P(m) = 1\). This summation is the power series expansion of the hypergeometric function given as follows:
\[
P(0)^{-1} = _2F_1 \left(1, -N; -D - N + 1; 1 + \frac{1}{\bar{n}}\right),
\] (46)
where \( _2F_1 \left( 1, -N; -D - N + 1; 1 + \frac{1}{N} \right) \) is the Gauss hypergeometric function of type \((2, 1)\). As a reminder, \( D \) is the total number of oscillators in the system and \( N \) is the total number of phonons in all modes in the NESS which is a function of \( D \).

The mean phonon numbers in the condensate can be calculated by the summation \( \langle n_0 \rangle = \sum_m mP(m) \), and we obtain:

\[
\langle n_0 \rangle = \frac{N(1 + \frac{1}{D})}{(D + N - 1)} _2F_1 \left( 2, 1 - N; -D - N + 2; 1 + \frac{1}{N} \right).
\]

Similarly, we can calculate the mean of the second moment of the condensate phonon numbers. The summation returns:

\[
\langle n_0^2 \rangle = \frac{N(1 + \frac{1}{D})}{(D + N - 1)} _2F_2 \left( 2, 2, 1 - N; 1, -D - N + 2; 1 + \frac{1}{N} \right).
\]

The Mandel Q parameter \( Q \) is the case of large \( \bar{n} \). For example, we can look at the mean phonon number in the condensate

\[
\langle \bar{n} \rangle = \frac{1}{N} \sum_{n=0}^{N} nP(n),
\]

This is further equivalent to the statement that there exists a positive integer \( M \in N \) such that

\[
\lim_{{N \to \infty}} P(m)/P(m - 1) < 1 \quad \text{for all} \quad m > M.
\]

Using Eq. (45), we have:

\[
\frac{P(m)}{P(m - 1)} = \frac{\bar{n} + 1}{\bar{n}} \cdot \frac{\bar{n}}{N - m} \cdot \frac{(N - m - 1)_{m-1}}{(N - (m - 1))_{m-1}}.
\]

where definitions of \( N \) and \( D \) are given in Eq. (44). Using the definitions of \( N \) and \( D \) and after some manipulation, we have the inequality from \( P(m)/P(m - 1) < 1 \) as:

\[
\frac{(D + 1)\Lambda}{\phi + \Lambda} (n_{ex} - \bar{n}) < \frac{\phi + \Lambda}{\chi} - 2\bar{n} + m - 1.
\]

It is obvious that in the large-\( D \) limit the condition of no condensate is that the external source occupation is below the occupation of the bath, i.e., \( n_{ex} < \bar{n} \). Therefore, in the large-\( D \) limit,

\[
\frac{\langle n_0 \rangle}{N} = 0, \quad \text{when} \quad n_{ex} < \bar{n}.
\]

Furthermore, we simultaneously obtain the condition for condensation to emerge, which is \( P(n_{cr} + 1) > P(n_{cr}) \) for some \( n_{cr} \geq 1 \). The inequality gives the condensation condition

\[
n_{ex} > \frac{(D - 1)\Lambda - 2\phi}{(D + 1)\Lambda} \bar{n} + \left( \frac{\phi + \Lambda}{\chi} + m - 1 \right) \frac{\phi + \Lambda}{(D + 1)\Lambda}.
\]

Similarly, in the large \( D \) limit, the critical number density reduces to:

\[
n_{ex,cr} = \bar{n}.
\]
This proves the finiteness of the hypergeometric functions in the case 
where 

hence

notice that

where the subscript \((b)_{(a-1)}\) is the rising Pochhammer symbol defined by \((b)_{(a-1)} = b(b+1)...(b+m-1)\). Notice that \((m+1)_{(a-1)}\) is the polynomial of \(m\) of order \(a - 1\). For \(\xi > d\), we can assume without loss of generality that \(\frac{\xi}{\xi} \leq k < 1\). In this case, one can notice that

\[
\frac{(b-m+1)_{(a-1)}}{(c)_{(a-1)}} d^m < \frac{(b)^{m}}{(c)^{m}} d^m \leq k^m.
\]

Therefore, the summation

\[
_2F_1(a, -b; -c; d) = \sum_m (m+1)_{(a-1)} \frac{(b-m+1)_{(a-1)}}{(c)_{(a-1)}} d^m < \sum_m (m+1)_{(a-1)} k^m
\]

is convergent. The boundedness of such summation can be easily proved since there exists an \(M \in \mathbb{N}\) such that for \(m > M\),

\[
(m+1)_{(a-1)} < (1/k)^{m/2},
\]

hence

\[
\sum_m (m+1)_{(a-1)} k^m < \sum_m k^{m/2}.
\]

This proves the finiteness of the hypergeometric functions in the case \(\xi > d\) and \(a, b, c, d \in \mathbb{N}\). One may notice that in the limit \(N \to \infty\),

\[
\frac{n_0}{N} = \frac{N(1 + \frac{1}{n})_2F_1(2, 1 - N; -D - N + 2; 1 + \frac{1}{n})}{(D + N - 1)_2F_1(1, -N; -D - N + 1; 1 + \frac{1}{n})}/N
\]

\[
\to \frac{(1 + \frac{1}{n})_2F_1(2, 1 - N; -D - N + 2; 1 + \frac{1}{n})}{(D + N)_{_2F_1}(1, -N; -D - N + 1; 1 + \frac{1}{n})}. \tag{62}
\]

For the case of \(\delta n = \frac{N}{\delta N} \langle n_{ex} - \bar{n} \rangle < 0\), the ratio of the second and the third entries becomes \(\frac{N}{\delta N} > 1 + \frac{1}{n}\). Therefore, the ratio between the above two hypergeometric functions is bounded, and the fraction of the condensate becomes \(\frac{n_0}{N} \to 0\) in the large-\(N\) limit. One can similarly show that the limit \(\delta n \to 0\) exists and \(\lim_{\delta n \to 0} \frac{n_0}{N} \to 0\). On the other hand, notice that

\[
_2F_1(a+1, -b+1; -c+1; d) = \frac{\sum_m (m+1)_{(a)} (b-m+1)_{(a-1)} d^m}{\sum_m (m+1)_{(a-1)} (c-1)_{(a-1)} d^m} = \frac{\sum_m (m+a)(1-m)_{(a-1)} f(m)}{\sum_m f(m)}, \tag{63}
\]

where \(f(m)\) is the distribution function of \(m\) defined by

\[
f(m) = \frac{(m+1)_{(a-1)} (b-m+1)_{(a-1)}}{m!} (c)_{(a-1)} d^m.
\]

For the case we consider here, from Eq. [54] we know that the peak of the distribution is roughly at

\[
m_p = n_{ex} - \left(\frac{(D+1)\Lambda - 2(\phi + \Lambda)}{(D+1)\Lambda + \phi + \Lambda}\right) \frac{(D+1)\Lambda}{\phi + \Lambda} - \frac{\phi + \Lambda}{\chi} + 1. \tag{65}
\]
In the large-$D$ limit,

$$m_p \sim (n_{ex} - \bar{n}) \frac{\Lambda}{\phi + \Lambda} D,$$

(66)

which is approximately proportional to $D$. Therefore, in the zeroth order approximation,

$$\frac{\,_{2}F_{1}(2, 1 - N; -D + N - 2; 1 + \frac{1}{n})}{\,_{2}F_{1}(1, -N; -D - N + 1; 1 + \frac{1}{n})} = m_p (1 - \frac{m_p}{N})(1 + \frac{m_p}{N + 2}) = \delta n (1 - \frac{\delta n}{\bar{n} + \delta n})(1 + \frac{\delta n}{\bar{n} + \delta n + 1}) D.$$

(67)

The ratio of the two hypergeometric functions in Eq. (47) scales linearly with $D$. The fraction of the condensate given by Eq. (47) is then approximated as:

$$\langle n_0 \rangle \approx \frac{(1 + \frac{1}{n})}{(\bar{n} + \delta n + 1)} (1 - \frac{\delta n}{\bar{n} + \delta n})(1 + \frac{\delta n}{\bar{n} + \delta n + 1}) \delta n.$$

(68)

Two important messages can be derived from the above calculation: the mean fraction of the condensation is nonzero in the limit $D \to \infty$ for the case $(n_{ex} - \bar{n}) > 0$ indicating the phase transition in the large-$D$ limit, and the critical exponent of the phase transition is $\beta = 1$. 

---