

Quantum thermalization via multiwave mixing

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We discuss thermalization in a multimode quantum cavity under unitary evolution. According to general principles, an isolated system with quadratic couplings does not exhibit thermalization. However, we find that nonlinearities, here three-wave perturbation, typical for instance in superconducting Josephson systems, may lead to thermalization into a Bose-Einstein distribution of occupations of the modes. The temperature of this state is dictated by energy conservation in this closed system, and the thermal distribution is robust against weak disturbances. We discuss how our findings open up new avenues to experimentally probe fundamental assumptions of statistical physics in solid-state systems.

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The nature and origin of thermalization has been heavily researched since the birth of statistical physics, and more recently for isolated quantum systems [1–4]. Despite intensive theoretical efforts, experiments have until now been largely limited to those on cold atoms [5–7]. The very rapid development of quantum information processing devices based on superconducting resonators and qubits [8] and circuit QED systems opens a new, exciting prospect for experimental studies of quantum thermalization [9–11]. The high-quality factors of resonators and long coherence times of qubits now allow us to consider these systems also as quasi-isolated ones. Most importantly, these systems can be accurately designed, measured, and controlled by electric and magnetic fields, and their dynamics follow a Hamiltonian derivable precisely from the circuit layout. A key objective for the present study is to demonstrate that nonlinearities lead to thermalization, a known but hard to prove belief [12,13], and that this can be realized in circuits including Josephson junctions that are in essence nonlinear inductors. We present how a realistic nearly integrable system reaches a thermal state due to such nonlinearity. Although we consider here three-wave mixing exclusively, we believe that other types of multiphoton down- and upconversion can lead to thermalization as well, since they yield the possibility of energy redistribution via nonconserved photon number. As a particular example, we analyze a multimode cavity (see Fig. 1) that can be made of a superconducting coplanar wave resonator, or alternatively of an array of Josephson junctions. Naturally, a cavity coupled to a thermal bath as in Fig. 1(a) gets thermalized with populations of states dictated by the temperature of that bath, but here we study explicitly an isolated cavity, Fig. 1(b). Ideally its Hamiltonian is quadratic, which makes it integrable with the number of photons being a conserved quantity, thus not exhibiting

thermalization; see, e.g., [14]. Yet recent developments in the circuit QED area have introduced elements that lead to three-wave mixing type nonlinearities in the circuit [15–22]. This can be achieved by adding shifted cosine potentials due to asymmetries in superconducting Josephson junction loops in magnetic field, like in the SNAIL (Superconducting Nonlinear Asymmetric Inductive eLement) configuration [15]. Another recent strategy for three-wave mixing has been achieved by coupling a fluxonium qubit to a Josephson junction array [16].

We focus on a generic multimode cavity whose Hamiltonian $\hat{\mathcal{H}}$ can be written as

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{V}, \quad (1)$$

where $\hat{\mathcal{H}}_0 = \sum_{k=1}^N \hbar \Omega_k a_k^\dagger a_k$ is the noninteracting Hamiltonian. Here N is the number of available modes, and $a_k^\dagger, a_k$ are the (bosonic) creation and annihilation operators of the k th mode, respectively. The coupling between the modes, or for an open system that between a mode and environment, is described by $\hat{V}$. We take the eigenenergies of $\hat{\mathcal{H}}_0$ to be equidistant so that $\Omega_k = k\Delta\Omega$, where $\Delta\Omega$ is the free spectral range.

We are going to look at the evolution of the population operator for the j th level, $\hat{N}_j = a_j^\dagger a_j$, and especially its expectation value $n(\Omega_j) \equiv \langle \hat{N}_j \rangle$. To find the kinetic equation for $n(\Omega_j)$, we write for the time rate of change of the operator $\hat{N}_j$, $\dot{\hat{N}}_j = \frac{i}{\hbar} [\hat{V}, \hat{N}_j]$, and use the Kubo formula in the interaction picture to obtain

$$\dot{n}(\Omega_j) = -\frac{i}{\hbar} \int_{-\infty}^0 dt' \langle [\dot{\hat{N}}_j, \hat{V}_I(t')] \rangle_0, \quad (2)$$

where $\hat{V}_I(t) = e^{i\hat{\mathcal{H}}_0 t/\hbar} \hat{V} e^{-i\hat{\mathcal{H}}_0 t/\hbar}$, and $\langle \cdot \rangle_0$ denotes averaging in the noninteracting system.

Now the evolution of $n(\Omega_j)$ depends critically on the type of coupling $\hat{V}$ (besides the initial state of the cavity). If the modes are not coupled ($\hat{V} = 0$), the populations $n(\Omega_j)$ are trivially stationary, and determined only by the initialization.

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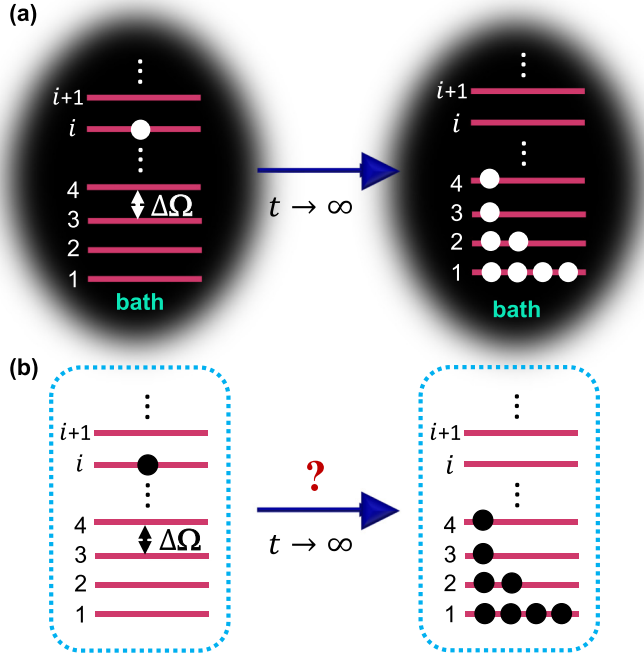


FIG. 1. The dynamics of a multimode quantum resonator. (a) Resonator coupled to a heat bath reaches a thermal state in the long-time limit. (b) Here we study the population dynamics of an isolated resonator.

Also, unsurprisingly, if the coupling is quadratic of the form

$$\hat{V} = \frac{1}{2} \sum_{p \neq q} g_{pq} (a_p + a_p^\dagger)(a_q + a_q^\dagger), \quad (3)$$

where g_{pq} are arbitrary coupling constants between modes p and q , there is no thermalization. This is expected from general arguments. We find this explicitly for our perturbative model as well, since the populations do not vary in time, i.e., $\dot{n}(\Omega_j) \equiv 0$, presented in the Supplemental Material [23].

On the contrary, ubiquitous nonlinearities do play a role in thermalization. In multiwave mixing, energy conservation can be secured in transitions unlike in coupling two modes only [Eq. (3)]. Experimentally common examples are three- and four-wave mixing. In basic three-wave mixing, a photon is down-converted into two, or vice versa. Formally the coupling then reads

$$\hat{V} = \sum_{p,q,r} (M_{pqr} a_p^\dagger a_q^\dagger a_r + M_{pqr}^* a_r^\dagger a_q a_p). \quad (4)$$

In what follows, we write the couplings in the form $M_{pqr} \equiv A_{pq} B_r$ for convenience and for physical reasons. We then find the kinetic equation using Eq. (2) as

$$\begin{aligned} \dot{n}(\Omega_j) = & \frac{4\pi}{\hbar^2} v_0 \left\{ \sum_{k=1}^N 2|A_{jk}|^2 |B_{j+k}|^2 [n(\Omega_j + \Omega_k) + n(\Omega_j)n(\Omega_j + \Omega_k) + n(\Omega_k)n(\Omega_j + \Omega_k) - n(\Omega_j)n(\Omega_k)] \right. \\ & \left. - \sum_{k=1}^{j-1} |A_{k,j-k}|^2 |B_j|^2 [n(\Omega_j) + n(\Omega_j)n(\Omega_k) + n(\Omega_j)n(\Omega_j - \Omega_k) - n(\Omega_k)n(\Omega_j - \Omega_k)] \right\}. \end{aligned} \quad (5)$$

For derivation of Eq. (5), see the Supplemental Material [23]. Here $v_0 = 1/\Delta\Omega$ is the density of equidistant modes valid for free photonic modes in the cavity. For the most general form of three-wave coupling, one may omit the factorized $|A_{pq}|^2 |B_r|^2$ and replace it by $|M_{pqr}|^2$ with proper indices in Eq. (5). We find easily that a Bose-Einstein distribution $n_B(\Omega)$ is a stationary solution of Eq. (5), i.e.,

$$\dot{n}(\Omega_j) \equiv 0 \text{ if } n(\Omega) = n_B(\Omega) \equiv \frac{1}{e^{\beta\hbar\Omega} - 1}. \quad (6)$$

This can be seen, since the following identities hold for $n_B(\Omega)$: $n_B(\Omega_j + \Omega_k) = n_B(\Omega_j)n_B(\Omega_k)/[1 + n_B(\Omega_j) + n_B(\Omega_k)]$, and $n_B(\Omega_j - \Omega_k) = n_B(\Omega_j)[1 + n_B(\Omega_k)]/[n_B(\Omega_k) - n_B(\Omega_j)]$. Based on these relations, all terms in the sums on the right-hand side of Eq. (5) vanish identically for any temperature $T = (k_B\beta)^{-1}$. Below we will find the actual value of T based on energy conservation. In general, a system without additional constraints beyond energy conservation relaxes to thermal equilibrium.

We can test whether the solution $n_B(\Omega)$ is stable by introducing small deviations $\delta n(\Omega_j)$ around this stationary solution. By a simplifying argument that there is deviation in one population only, we then find that

$$\frac{d\delta n(\Omega_j)}{dt} = -\frac{4\pi}{\hbar^2} v_0 \left\{ \sum_{k=1}^N 2|A_{jk}|^2 |B_{j+k}|^2 \frac{n_B(\Omega_k)[1 + n_B(\Omega_k)]}{1 + n_B(\Omega_j) + n_B(\Omega_k)} + \sum_{k=1}^{j-1} |A_{k,j-k}|^2 |B_j|^2 \frac{n_B(\Omega_k)[1 + n_B(\Omega_k)]}{n_B(\Omega_k) - n_B(\Omega_j)} \right\} \delta n(\Omega_j). \quad (7)$$

This equation can then be written as $\frac{d\delta n(\Omega_j)}{dt} = -\kappa_j \delta n(\Omega_j)$, where κ_j is positive for any nonvanishing coupling of form (4), securing stability of the stationary distribution. Numerical examples of the dynamics will be given below. They show the stability of the $n(\Omega_j)$ distribution, as all these populations approach exponentially toward the equilibrium one.

The remaining question is how the temperature of the system is determined. This in fact can be answered based on an elementary energy conservation argument for an isolated system such as ours. The internal energy of the initial state is

$$U_{\text{ini}} = \sum_{j=1}^N N_j^{(\text{ini})} \hbar \Omega_j, \quad (8)$$

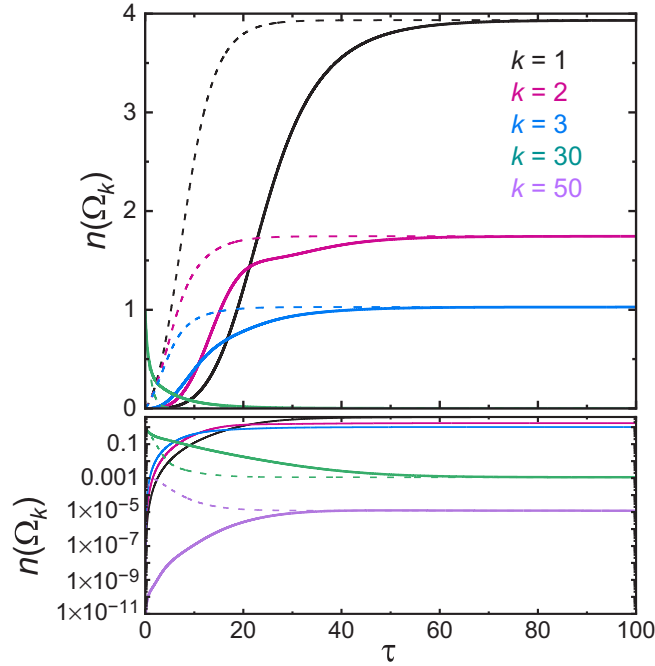


FIG. 2. Time-dependent populations on various photon modes k of the multimode resonator when initialized on the $k_{\text{ini}} = 30$ level with $N_{30} = 1$ at $\tau \equiv \Delta\Omega t = 0$. The solid lines correspond to the coupling of form $|A_{pq}|^2 = g^2 e^{-\gamma|p-q|}$, $|B_r|^2 = r$ [16], and $\Gamma_0 \equiv \frac{4\pi}{\hbar^2} v_0 g^2 = 0.01 \Delta\Omega$, with $\gamma = 1$. The dashed lines on the other hand correspond to the coupling $\frac{4\pi}{\hbar^2} |A_{pq}|^2 |B_r|^2 = 0.03$ for all p, q, r .

where $N_j^{(\text{ini})}$ represents the initial population in state j . If one eventually at $t \rightarrow \infty$ reaches the state with populations $n_B(\Omega_j)$, the final internal energy is

$$U_{\text{fin}} = \sum_{j=1}^N \frac{\hbar\Omega_j}{e^{\beta\hbar\Omega_j} - 1}. \quad (9)$$

Equating $U_{\text{fin}} = U_{\text{ini}}$, we then find β . For a sufficiently large initial energy the final state spreads over many modes, and we can approximate the sum in Eq. (9) by an integral, with the result

$$U_{\text{fin}} \approx \frac{\pi^2}{6\hbar\Delta\Omega\beta^2}. \quad (10)$$

Then, for instance, if we initialize the system with an excitation on the level k_{ini} , the temperature is given approximately by

$$\beta\hbar\Delta\Omega \approx \pi/\sqrt{6k_{\text{ini}}}. \quad (11)$$

Next we present numerical results of the dynamics of the system. First we analyze the time dependence of the populations in different modes after the system is initialized in a given state. Figure 2 is an example of such dynamics, presenting the case of a system initially occupying the mode $k_{\text{ini}} = 30$. Populations at different time instants $\tau = \Delta\Omega t$ of modes $k = 1, 2, 3, 30$, and 50 are shown. We assume that

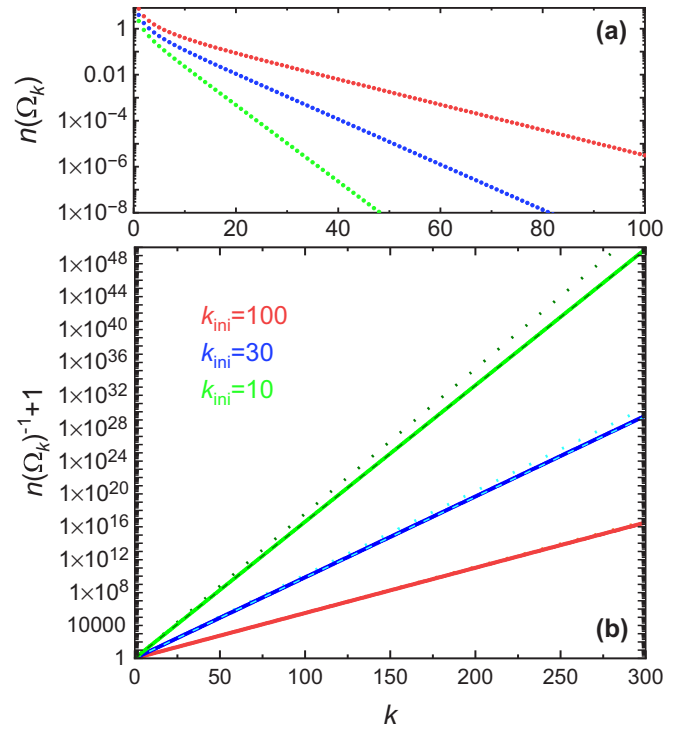


FIG. 3. Long-term asymptotic distribution of oscillator mode populations in the form $n(\Omega_k)^{-1} + 1$ for three different initial states, $k_{\text{ini}} = 10, 30$, and 100 . The symbols are from the numerics, the dashed lines are the exact Bose-Einstein distributions assuming energy conservation, and the dotted line is the approximate result based on Eq. (11).

there are $N = 800$ lowest modes available. In the calculation we assume that the three-wave coupling is parametrized as $|A_{pq}|^2 = g^2 e^{-\gamma|p-q|}$, $|B_r|^2 = r$ [16], and the overall relaxation rate $\Gamma_0 \equiv \frac{4\pi}{\hbar^2} v_0 g^2 = 0.01 \Delta\Omega$, with $\gamma = 1$. We see that the system finds its asymptotic state at times $\tau \gg 100$ in this case. Yet this time is simply scaling as Γ_0^{-1} , thus demonstrating that there is no lower or upper bound of it within the validity of our weak-coupling model. Very similar dynamics can be found with other choices of the coupling parameters, for instance assuming constant $|A_{pq}|^2$ and $|B_r|^2$ as demonstrated by the second set of curves (dashed lines) in Fig. 2. To highlight the rarely populated states, we plot $n(\Omega_k)$ for the same states on a logarithmic scale in the lower panel of Fig. 2. Based on the numerical evolution in Fig. 2 we expect the system to be stable and find a state which is potentially the given steady-state solution $n_B(\Omega)$. We may test the analytical predictions by plotting the distribution at a time when populations are stable in time, here in Fig. 3 at $\tau = 150$ for the couplings with the same set of parameters as above. In Fig. 3(a) we present these asymptotic populations $n(\Omega_k)$ versus k . To test the results against the predicted ones, we plot in Fig. 3(b) $n(\Omega_k)^{-1} + 1$ versus k for three different initial values of $k_{\text{ini}} = 10, 30$, and 100 , demonstrating exponential dependence over nearly 50 orders of magnitude. For comparison, we show $n_B(\Omega_k)^{-1} + 1 = e^{\beta\hbar\Omega_k}$ where β is obtained by equating U_{ini} from Eq. (8) with U_{fin} either from Eq. (9) (accurate, dashed line) or from Eq. (11) (integral approximation, dotted line). The first one coincides perfectly with the numerical $n(\Omega_k)^{-1} + 1$ over all

k , whereas the approximation becomes less accurate for lower k_{ini} as expected.

Discussion. Our nonlinear system loses information of its initial state, apart from its total energy, thus exhibiting full thermalization. This is contrary to several other model systems, like a one-dimensional chain of hard-core bosons [24] or quadratically coupled two-level systems [14]. They relax to their maximum entropy state [24] according to the principle of the generalized Gibbs ensemble [24,25], still remembering the initial state even at long times due to, e.g., the excitation number (and energy) conservation in the latter model. There are a few more subtleties to keep in mind when assessing the results of our model. First, since the evolution of the isolated system is unitary, one does not obtain a purely diagonal density matrix even in the long-time limit. Yet the thermal populations given by the diagonal elements are the key essence of our result; i.e., as soon as the system populations are measured, one cannot distinguish whether the density matrix was diagonal or not. Second, in our analysis we employed the perturbative approach [Kubo formula, Eq. (2)] and the continuum approximation of states when securing the energy conservation in the step between Eqs. (12) and (13) in the Supplemental Material [23]. The implication of the latter approximation is still not quite clear to us, and it remains to be seen in further work whether the true thermalization without Poincaré recoveries can be achieved only in multimode resonators with very small free spectral range $\Delta\Omega$ [26,27], what determines this condition, and what the

influence of nonuniformity of it is. We find an analogous situation in the problem of electron-electron relaxation [28], where electrons in metals at low temperatures form a Fermi-Dirac distribution within a relaxation time of about 1 ns [28,29] after a “quench,” whereas these electrons relax to the external bath formed by lattice phonons over timescales of about 100 μs [30]. Such a huge separation of timescales allows one to assume electrons to form an isolated interacting quantum system that reaches a quasiequilibrium thermal state. In metallic samples the density of states is large, of the order of inverse 1 mK, due to the large number of electrons, securing internal thermalization. A long (1–100 m) superconducting waveguide yields a free spectral range of the same order. Moreover, superconducting waveguides can have quality factors in excess of 10^5 , lending a regime of quasi-isolation over times of order 1 s. Therefore, long nonlinear superconducting waveguides [27] could serve as a test bed for our model. A concrete Josephson junction circuit would be an array formed of asymmetric multijunction loops (SNAILs), which is linear in zero magnetic field, but can provide nonlinearities otherwise. In summary, solid-state systems, in particular superconducting quantum circuits, provide a new, highly controllable pathway to the studies of thermalization in the quantum regime.

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- [1] D. A. Abanin, E. Altman, I. Bloch, and M. Serbyn, Many-body localization, thermalization, and entanglement, *Rev. Mod. Phys.* **91**, 021001 (2019).
 - [2] T. Mori, T. N. Ikeda, E. Kaminishi, and M. Ueda, Thermalization and prethermalization in isolated quantum systems: A theoretical overview, *J. Phys. B: At. Mol. Opt. Phys.* **51**, 112001 (2018).
 - [3] L. D’Alessio, Y. Kafri, A. Polkovnikov, and M. Rigol, From quantum chaos and eigenstate thermalization to statistical mechanics and thermodynamics, *Adv. Phys.* **65**, 239 (2016).
 - [4] R. Nandkishore and D. A. Huse, Many-body localization and thermalization in quantum statistical mechanics, *Annu. Rev. Condens. Matter Phys.* **6**, 15 (2015).
 - [5] T. Kinoshita, T. Wenger, and D. S. Weiss, A quantum Newton’s cradle, *Nature (London)* **440**, 900 (2006).
 - [6] M. Gring, M. Kuhnert, T. Langen, T. Kitagawa, B. Rauer, M. Schreitl, I. Mazets, D. Adu Smith, E. Demler, and J. Schmiedmayer, Relaxation and prethermalization in an isolated quantum system, *Science* **337**, 1318 (2012).
 - [7] A. M. Kaufman *et al.*, Quantum thermalization through entanglement in an isolated many-body system, *Science* **353**, 794 (2016).
 - [8] M. Kjaergaard *et al.*, Superconducting qubits: Current state of play, *Annu. Rev. Condens. Matter Phys.* **11**, 369 (2020).
 - [9] C. Neill *et al.*, Ergodic dynamics and thermalization in an isolated quantum system, *Nat. Phys.* **12**, 1037 (2016).
 - [10] F. Chen *et al.*, Observation of strong and weak thermalization in a superconducting quantum processor, *Phys. Rev. Lett.* **127**, 020602 (2021).
 - [11] S. Sundelin, M. A. Aamir, V. M. Kulkarni, C. Castillo-Moreno, and S. Gasparinetti, Quantum refrigeration powered by noise in a superconducting circuit, *arXiv:2403.03373*.
 - [12] J. M. Magán, Random free fermions: An analytical example of eigenstate thermalization, *Phys. Rev. Lett.* **116**, 030401 (2016).
 - [13] P. Schlagheck and D. L. Shepelyansky, Dynamical thermalization in Bose-Hubbard systems, *Phys. Rev. E* **93**, 012126 (2016).
 - [14] J. P. Pekola, B. Karimi, M. Cattaneo, and S. Maniscalco, Long-time relaxation of a finite spin bath linearly coupled to a qubit, *Open Syst. Inf. Dyn.* **30**, 2350009 (2023).
 - [15] N. E. Frattini *et al.*, 3-wave mixing Josephson dipole element, *Appl. Phys. Lett.* **110**, 222603 (2017).
 - [16] N. Mehta, R. Kuzmin, C. Ciuti, and V. E. Manucharyan, Down-conversion of a single photon as a probe of many-body localization, *Nature (London)* **613**, 650 (2023).
 - [17] A. B. Zorin, Josephson traveling-wave parametric amplifier with three-wave mixing, *Phys. Rev. Appl.* **6**, 034006 (2016).
 - [18] N. Roch, E. Flurin, F. Nguyen, P. Morfin, P. Campagne-Ibarcq, M. H. Devoret, and B. Huard, Widely tunable, nondegenerate three-wave mixing microwave device operating near the quantum limit, *Phys. Rev. Lett.* **108**, 147701 (2012).
 - [19] A. Ganesan, C. Do, and A. Seshia, Phononic frequency comb via intrinsic three-wave mixing, *Phys. Rev. Lett.* **118**, 033903 (2017).

- [20] S. Simbierowicz *et al.*, A flux-driven Josephson parametric amplifier for sub-GHz frequencies fabricated with side-wall passivated spacer junction technology, *Supercond. Sci. Technol.* **31**, 105001 (2018).
- [21] A. Ranadive *et al.*, Kerr reversal in Josephson meta-material and traveling wave parametric amplification, *Nat. Commun.* **13**, 1737 (2022).
- [22] D. J. Parker, M. Savytskyi, W. Vine, A. Laucht, T. Duty, A. Morello, A. L. Grimsmo, and J. J. Pla, Degenerate parametric amplification via three-wave mixing using kinetic inductance, *Phys. Rev. Appl.* **17**, 034064 (2022).
- [23] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevResearch.6.L042023> for details about derivation of stationary population for quadratic coupling and Eq. (5) in the text.
- [24] M. Rigol, V. Dunjko, V. Yurovsky, and M. Olshanii, Relaxation in a completely integrable many-body quantum system: An *ab initio* study of the dynamics of the highly excited states of 1D lattice hard-core bosons, *Phys. Rev. Lett.* **98**, 050405 (2007).
- [25] E. T. Jaynes, Information theory and statistical mechanics, *Phys. Rev.* **106**, 620 (1957); Information theory and statistical mechanics. II, **108**, 171 (1957).
- [26] Y. P. Zhong *et al.*, Violating Bell's inequality with remotely connected superconducting qubits, *Nat. Phys.* **15**, 741 (2019).
- [27] J. Qiu, Y. Liu, J. Niu, L. Hu, Y. Wu, L. Zhang, W. Huang, Y. Chen, J. Li, S. Liu *et al.*, Deterministic quantum teleportation between distant superconducting chips, [arXiv:2302.08756](https://arxiv.org/abs/2302.08756).
- [28] H. Pothier, S. Guéron, Norman O. Birge, D. Esteve, and M. H. Devoret, Energy distribution function of quasiparticles in mesoscopic wires, *Phys. Rev. Lett.* **79**, 3490 (1997).
- [29] T. T. Heikkilä, *The Physics of Nanoelectronics: Transport and Fluctuation Phenomena at Low Temperatures* (Oxford University Press, Oxford, 2013).
- [30] F. Giazotto, T. T. Heikkilä, A. Luukanen, A. M. Savin, and J. P. Pekola, Opportunities for mesoscopes in thermometry and refrigeration: Physics and applications, *Rev. Mod. Phys.* **78**, 217 (2006).