

# Supplemental Information: Violating the Thermodynamic Uncertainty Relation in the Three-Level Maser

Alex Arash Sand Kalaei, Andreas Wacker and Patrick P. Potts  
*Mathematical Physics and NanoLund, Lund University, Sweden*  
 (Dated: July 21, 2021)

The following is a supplementary material detailing and deriving the equations presented in the main text.

## I. STEADY STATE OF THE THREE-LEVEL MASER

The Hamiltonian  $H(t) = H_0 + V(t)$  consists of a bare term

$$H_0 = \omega_l \sigma_{ll} + \omega_u \sigma_{uu} + \omega_x \sigma_{xx}, \quad (\text{S1})$$

and an external classical field

$$V(t) = \epsilon(e^{i\omega_d t} \sigma_{lu} + e^{-i\omega_d t} \sigma_{ul}). \quad (\text{S2})$$

To remove the time dependence of the Hamiltonian we transform to an appropriate rotating frame which simplifies the equation of motion. We define rotated operators by the unitary transformation  $A^{\text{rot}} = U(t)AU^\dagger(t)$  with  $U(t) = e^{iXt}$  and  $X = \omega_l \sigma_{ll} + (\omega_l + \omega_d) \sigma_{uu} + \omega_x \sigma_{xx}$ . In this rotated frame, we obtain

$$\dot{\rho} = -i[\tilde{H}, \rho] + \sum_{\alpha} \{\gamma_{\alpha}(n_{\alpha} + 1) \mathcal{D}_{\sigma_{\alpha x}}[\rho] + \gamma_{\alpha} n_{\alpha} \mathcal{D}_{\sigma_{x\alpha}}[\rho]\}, \quad (\text{S3})$$

with the new Hamiltonian  $\tilde{H} = H^{\text{rot}} - X = -\Delta \sigma_{uu} + \epsilon(\sigma_{ul} + \sigma_{lu})$  where we introduced the detuning parameter  $\Delta = \omega_d - (\omega_u - \omega_l)$ . Note, that the structure of the dissipators is not affected by this transformation. From Eq. (S3), we obtain the equations of motion for  $\rho_{ij} = \langle i | \rho^{\text{rot}} | j \rangle$

$$\dot{\rho}_{xx} = \gamma_l n_l \rho_{ll} + \gamma_u n_u \rho_{uu} - [(n_l + 1)\gamma_l + (n_u + 1)\gamma_u] \rho_{xx}, \quad (\text{S4})$$

$$\dot{\rho}_{uu} = \gamma_u (n_u + 1) \rho_{xx} - \gamma_u n_u \rho_{uu} + i\epsilon(\rho_{ul} - \rho_{ul}^*), \quad (\text{S5})$$

$$\dot{\rho}_{ll} = \gamma_l (n_l + 1) \rho_{xx} - \gamma_l n_l \rho_{ll} - i\epsilon(\rho_{ul} - \rho_{ul}^*), \quad (\text{S6})$$

$$\dot{\rho}_{ul} = i\Delta \rho_{ul} + i\epsilon(\rho_{uu} - \rho_{ll}) - \Gamma \rho_{ul}, \quad (\text{S7})$$

where  $\Gamma = \frac{1}{2}(\gamma_u n_u + \gamma_l n_l)$  is the quantum decoherence rate. In the steady state (superscript <sup>ss</sup>), Eq. (S7) provides

$$\rho_{ul}^{\text{ss}} = \frac{-\epsilon(\rho_{uu}^{\text{ss}} - \rho_{ll}^{\text{ss}})}{\Delta + i\Gamma}. \quad (\text{S8})$$

The steady-state populations are

$$\begin{aligned} \rho_{ll}^{\text{ss}} &= \frac{\gamma_l \gamma_u n_u (n_l + 1) + \gamma_c [\gamma_l (n_l + 1) + \gamma_u (n_u + 1)]}{[\gamma_l (2n_l + 1) + \gamma_c][\gamma_u (2n_u + 1) + \gamma_c] - [\gamma_l (n_l + 1) - \gamma_c][\gamma_u (n_u + 1) - \gamma_c]}, \\ \rho_{uu}^{\text{ss}} &= \frac{\gamma_l \gamma_u n_l (n_u + 1) + \gamma_c [\gamma_l (n_l + 1) + \gamma_u (n_u + 1)]}{[\gamma_l (2n_l + 1) + \gamma_c][\gamma_u (2n_u + 1) + \gamma_c] - [\gamma_l (n_l + 1) - \gamma_c][\gamma_u (n_u + 1) - \gamma_c]}, \\ \rho_{xx}^{\text{ss}} &= 1 - \rho_{uu}^{\text{ss}} - \rho_{ll}^{\text{ss}}, \end{aligned} \quad (\text{S9})$$

where the classical rate reads  $\gamma_c = \frac{2\epsilon^2 \Gamma}{\Delta^2 + \Gamma^2}$ . The lasing populations provide

$$\rho_{uu}^{\text{ss}} - \rho_{ll}^{\text{ss}} = \frac{\gamma_l \gamma_u (n_l - n_u)}{2\gamma_c [3\Gamma + \gamma_u + \gamma_l] + \gamma_l \gamma_u (3n_l n_u + n_l + n_u)},$$

which is proportional to the occupation differences of the baths. (Note that  $n_l > n_u$  yields population inversion). Inserting this into Eq. (S8) we find

$$\rho_{ul}^{\text{ss}} = \frac{(-\Delta + i\Gamma)\epsilon(n_l - n_u)}{(\Delta^2 + \Gamma^2)A + \epsilon^2 B} \quad \text{with } A = 3n_l n_u + n_l + n_u \quad \text{with } B = 2\Gamma[(3n_l + 2)/\gamma_u + (3n_u + 2)/\gamma_l]. \quad (\text{S10})$$

By straightforward algebra, we find that  $|\rho_{ul}^{\text{ss}}|$  has a ridge of maxima as a function of  $\epsilon$  and  $\Delta$  on the curve

$$\epsilon^2 = (\Delta^2 + \Gamma^2) \frac{A}{B}, \quad (\text{S11})$$

with the constant peak value

$$|\rho_{ul}^{\text{ss}}|_{\text{peak}} = \frac{(n_l - n_u)}{2\sqrt{AB}}, \quad (\text{S12})$$

as can be seen in Fig. 4(b) of the main article.

## II. FULL COUNTING STATISTICS

An analytical approach to determine the particle statistics in an open quantum system is provided by Full Counting Statistics (FCS), where counting fields are included in the master equation [1, 2]. Let  $\chi_u$  and  $\chi_l$  be counting fields for the respective reservoirs. The Lindblad master equation becomes

$$\dot{\rho}^{\text{rot}} = -i[\tilde{H}, \rho^{\text{rot}}] + \mathcal{L}_u^{\chi_u}[\rho^{\text{rot}}] + \mathcal{L}_l^{\chi_l}[\rho^{\text{rot}}], \quad (\text{S13})$$

with the modified Lindbladians

$$\mathcal{L}_\alpha^{\chi_\alpha}[\rho] = \gamma_\alpha(n_\alpha + 1)\mathcal{D}_{\sigma_{\alpha x}}^{\chi_\alpha}[\rho] + \gamma_\alpha n_\alpha \mathcal{D}_{\sigma_{\alpha x}}^{-\chi_\alpha}[\rho], \quad (\text{S14})$$

and dissipators

$$\mathcal{D}_\sigma^\chi[\rho] = e^{-i\chi} \sigma \rho \sigma^\dagger - \frac{1}{2} \{ \sigma^\dagger \sigma \rho + \rho \sigma^\dagger \sigma \}. \quad (\text{S15})$$

If we reshape the density matrix into a state vector  $\rho^{\text{rot}} = (\rho_{xx}, \rho_{uu}, \rho_{ll}, \text{Re}[\rho_{ul}], \text{Im}[\rho_{ul}])^T$  we summarize the Lindblad master equation as a matrix equation with the Liouvillian supermatrix  $\mathcal{L}(\chi_u, \chi_l)$  [3]

$$\dot{\rho}^{\text{rot}} = \mathcal{L}(\chi_u, \chi_l) \rho^{\text{rot}}. \quad (\text{S16})$$

The full Liouvillian supermatrix with counting fields is

$$\mathcal{L}(\chi_u, \chi_l) = \begin{bmatrix} -\gamma_u(n_u + 1) - \gamma_l(n_l + 1) & \gamma_u n_u e^{i\chi_u} & \gamma_l n_l e^{i\chi_l} & 0 & 0 \\ \gamma_u(n_u + 1)e^{-i\chi_u} & -\gamma_u n_u & 0 & 0 & -2\epsilon \\ \gamma_l(n_l + 1)e^{-i\chi_l} & 0 & -\gamma_l n_l & 0 & 2\epsilon \\ 0 & 0 & 0 & -\Gamma & \Delta \\ 0 & \epsilon & -\epsilon & -\Delta & -\Gamma \end{bmatrix}. \quad (\text{S17})$$

In the limit  $\chi_u, \chi_l \rightarrow 0$  this reduces to the original Liouvillian supermatrix for proper time evolution. As shown in Sec. III, it is sufficient for our purposes to count the quanta exchanged with bath  $u$ . Therefore, we set  $\chi_l = 0$  and  $\chi_u = \chi$  in the following.

In the large time limit the  $k$ 'th cumulant of the integrated number of quanta emitted into reservoir  $\alpha$  over a time window  $t$  can be determined by [2]

$$C^k(t) = (i\partial_\chi)^k [\zeta(\chi)t + c(\chi)]|_{\chi=0}, \quad (\text{S18})$$

where  $\zeta(\chi)$  is the eigenvalue of  $\mathcal{L}(\chi) \equiv \mathcal{L}(\chi, 0)$  with the largest real part and  $c(\chi)$  is a polynomial depending on the eigenvectors of  $\mathcal{L}(\chi)$ . The first and second cumulants correspond to the mean and variance of the integrated particle current respectively

$$\langle \dot{N} \rangle \simeq i\partial_\chi \zeta(\chi)|_{\chi=0}, \quad \text{var}(\dot{N}) \simeq -\partial_\chi^2 \zeta(\chi)|_{\chi=0}, \quad (\text{S19})$$

where we have dropped the term in Eq. (S18) that does not grow in time.

To determine the mean and variance from the derivatives analytically we follow the method outlined in Ref. [4]. Consider the characteristic polynomial of  $\mathcal{L}(\chi)$

$$\sum_n a_n \zeta^n = 0, \quad (\text{S20})$$

where the terms  $a_n$  are functions of  $\chi$ . Define

$$a'_n = i\partial_\chi a_n|_{\chi=0}, \quad a''_n = (i\partial_\chi)^2 a_n|_{\chi=0} = -\partial_{\chi_u}^2 a_n|_{\chi=0}, \quad (\text{S21})$$

and similarly for  $\zeta$ . We determine the derivative of the polynomial equation in  $\chi$

$$\left[ i\partial_\chi \sum_n a_n \zeta^n \right]_{\chi=0} = \sum_n [a'_n + (n+1)a_{n+1}\zeta'] \zeta^n(0) = 0. \quad (\text{S22})$$

Continuing with the second derivative we find

$$\left[ (i\partial_\chi)^2 \sum_n a_n \zeta^n \right]_{\chi=0} = \sum_n [a''_n + 2(n+1)a'_{n+1}\zeta' + (n+1)a_{n+1}\zeta'' + (n+1)(n+2)a_{n+2}\zeta'^2] \zeta^n(0) = 0. \quad (\text{S23})$$

We assume that the system has a unique steady state for which  $\zeta(0) = 0$ . We know that the zeroth term  $\zeta^0 = 1$  must vanish, hence Eq. (S22) indicates

$$a'_0 + a_1 \zeta' = 0, \quad (\text{S24})$$

which provides the current

$$\langle \dot{N} \rangle = \zeta' = -\frac{a'_0}{a_1}. \quad (\text{S25})$$

We obtain the variance similarly from Eq. (S23)

$$\text{var}(\dot{N}) = \zeta'' = -\frac{a''_0 + 2\langle \dot{N} \rangle(a'_1 + a_2 \langle \dot{N} \rangle)}{a_1} = 2\frac{a'_0 a_1 a'_1 - a_0'^2 a_1'}{a_1^3} - \frac{a_0''}{a_1}. \quad (\text{S26})$$

The expressions (S25) and (S26) hold for all systems with Lindblad dynamics assuming a unique steady state.

The Liouvillian given in Eq. (S17) results in the parameters

$$\begin{aligned} a'_0 &= -2\epsilon^2 \gamma_l \gamma_u (n_l - n_u) \Gamma, \\ a''_0 &= -2\epsilon^2 \gamma_u \gamma_l \Gamma (2n_l n_u + n_l + n_u), \\ a_1 &= \gamma_l \gamma_u [\Delta^2 + \Gamma^2] (3n_l n_u + n_u + n_l) + 4\epsilon^2 \Gamma [3\Gamma + \gamma_l + \gamma_u], \\ a'_1 &= -2\epsilon^2 \gamma_u \gamma_l (n_l - n_u), \\ a_2 &= [\Delta^2 + \Gamma^2 + 4\epsilon^2] (4\Gamma + \gamma_l + \gamma_u) + 2\Gamma \gamma_l \gamma_u (3n_l n_u + n_l + n_u), \end{aligned} \quad (\text{S27})$$

where  $\Gamma = \frac{1}{2}(\gamma_u n_u + \gamma_l n_l)$  is the decoherence rate. This provides the current

$$\langle \dot{N} \rangle = \frac{2\gamma_u \gamma_l \epsilon^2 (n_l - n_u) \Gamma}{[\Delta^2 + \Gamma^2] \gamma_u \gamma_l (3n_l n_u + n_u + n_l) + 4\epsilon^2 \Gamma [3\Gamma + \gamma_u + \gamma_l]}, \quad (\text{S28})$$

and the Fano factor

$$F(\dot{N}) = \frac{\text{var}(\dot{N})}{\langle \dot{N} \rangle} = \frac{a''_0}{a'_0} + 2\langle \dot{N} \rangle \left[ \frac{a'_1}{a'_0} - \frac{a_2}{a_1} \right] = \frac{n_u(n_l + 1) + n_l(n_u + 1)}{n_l - n_u} + 2\langle \dot{N} \rangle \left[ \Gamma^{-1} - \frac{a_2}{a_1} \right]. \quad (\text{S29})$$

We introduce the population and transport terms of the Fano factor, respectively,

$$F_{\text{pop}} = \frac{n_u(n_l + 1) + n_l(n_u + 1)}{n_l - n_u}, \quad F_{\text{tr}} = -2\langle \dot{N} \rangle \left[ \frac{a_2}{a_1} - \Gamma^{-1} \right], \quad (\text{S30})$$

for which  $F = F_{\text{pop}} + F_{\text{tr}}$ . Finally, from the Fano factors we define the thermodynamic uncertainty similarly. Per Eq. (8) in the main text

$$\mathcal{Q} = \ln \left[ \frac{n_l(n_u + 1)}{n_u(n_l + 1)} \right] F_{\text{pop}} + \ln \left[ \frac{n_l(n_u + 1)}{n_u(n_l + 1)} \right] F_{\text{tr}} = \mathcal{Q}_{\text{pop}} + \mathcal{Q}_{\text{tr}}, \quad (\text{S31})$$

where  $\mathcal{Q}_{\text{pop}}$  is the population term and  $\mathcal{Q}_{\text{tr}}$  the transport term. Using the inequalities  $a/(a-b) \ln(a/b) \geq 1$  and  $b/(a-b) \ln(a/b) \geq 1$  we can show that

$$\mathcal{Q}_{\text{pop}} = \ln \left[ \frac{n_l(n_u + 1)}{n_u(n_l + 1)} \right] \frac{n_u(n_l + 1) + n_l(n_u + 1)}{n_l - n_u} \geq 2. \quad (\text{S32})$$

The population term adheres to the TUR limit for all parameters, hence, the transport term is essential for TUR violations. For some parameters  $\mathcal{Q}_{\text{tr}}$  is positive and decreases the engine precision.

### III. CONNECTING FCS TO THE AVERAGE AND VARIANCE OF POWER

In this section, we show that in the long time limit, counting the heat quanta exchanged with the bath  $u$  is sufficient for determining the statistics of all heat and work flows up to the variance. We start by showing that the statistics of the heat quanta exchanged with the two baths are identical. To this end, we consider the unitary superoperator

$$\mathcal{U}\rho = e^{-\frac{i}{2}\chi_l(\sigma_{uu}+\sigma_{ll})}\rho e^{-\frac{i}{2}\chi_l(\sigma_{uu}+\sigma_{ll})}, \quad \mathcal{U}^\dagger\rho = e^{\frac{i}{2}\chi_l(\sigma_{uu}+\sigma_{ll})}\rho e^{\frac{i}{2}\chi_l(\sigma_{uu}+\sigma_{ll})}. \quad (\text{S33})$$

A straightforward calculation shows that

$$\mathcal{U}^\dagger\mathcal{L}(\chi_u, \chi_l)\mathcal{U} = \mathcal{L}(\chi_u + \chi_l, 0). \quad (\text{S34})$$

In the long-time limit, the statistics are fully determined by the eigenvalues of the Liouvillian, which do not change under a unitary transformation. Equation (S34) thus implies that the statistics of the heat quanta exchanged with the two baths are identical and that it is sufficient to consider a single counting field in the long-time limit.

To connect the mean and variance of the exchanged heat quanta to the power operator, we consider the unitary superoperator

$$\mathcal{V}\rho = e^{-\frac{i}{2}\chi\sigma_{uu}}\rho e^{-\frac{i}{2}\chi\sigma_{uu}}. \quad (\text{S35})$$

Transforming the Liouvillian then results in

$$\hat{\mathcal{L}}(\chi)\rho = \mathcal{V}^\dagger\mathcal{L}(\chi)\mathcal{V}\rho = -i\tilde{H}(\chi)\hat{\rho} + i\rho\tilde{H}(-\chi) + \mathcal{L}_u[\rho] + \mathcal{L}_l[\rho], \quad (\text{S36})$$

with

$$\tilde{H}(\chi) = -\Delta\sigma_{uu} + \epsilon\left(\sigma_{ul}e^{\frac{i}{2}\chi} + \sigma_{lu}e^{-\frac{i}{2}\chi}\right). \quad (\text{S37})$$

We may now compute  $\langle\dot{N}\rangle$  and  $\text{var}(\dot{N})$  from  $\hat{\mathcal{L}}$ . Following the methods outlined in Ref. [5], we find for the average

$$\langle\dot{N}\rangle = \text{Tr}\left[\hat{\mathcal{L}}'\rho^{\text{ss}}\right] = \frac{1}{\omega_d}\text{Tr}\left[P^{\text{rot}}\rho^{\text{ss}}\right], \quad (\text{S38})$$

where  $\hat{\mathcal{L}}' = i\partial_\chi\hat{\mathcal{L}}(\chi)|_{\chi=0}$  and the power operator in the rotating frame reads

$$P^{\text{rot}} = U(t)[- \partial_t H]U^\dagger(t). \quad (\text{S39})$$

This confirms that  $\langle\dot{N}\rangle = \langle P \rangle / \omega_d$  denotes the number of photons emitted into the driving field.

The variance can be written as [5] (dropping a term that vanishes for our Liouvillian)

$$\text{var}(\dot{N}) = 2\text{Tr}\left[\hat{\mathcal{L}}'\mathcal{L}^{\text{D}}\hat{\mathcal{L}}'\rho^{\text{ss}}\right], \quad (\text{S40})$$

with the Drazin inverse of  $\mathcal{L} \equiv \mathcal{L}(0)$  given by [6, 7]

$$\mathcal{L}^{\text{D}} = -\int_0^\infty d\tau e^{\mathcal{L}\tau}(\mathcal{I} - \mathcal{P}), \quad (\text{S41})$$

where  $\mathcal{I}$  denotes the identity and  $\mathcal{P}$  projects onto the null space of the Liouvillian, i.e.,  $\mathcal{P}\rho = \rho^{\text{ss}}\text{Tr}[\rho]$ , see also Eq. (S56) below. Equation (S40) can be expressed as

$$\text{var}(\dot{N}) = \frac{1}{\omega_d^2} \int_0^\infty d\tau \text{Tr}\left[\delta P^{\text{rot}} e^{\mathcal{L}\tau} \{\delta P^{\text{rot}}, \rho^{\text{ss}}\}\right] = \frac{1}{\omega_d^2} \int_{-\infty}^\infty \frac{1}{2} \langle \{\delta P(t+\tau), \delta P(t)\} \rangle, \quad (\text{S42})$$

where  $\delta P^{\text{rot}} = P^{\text{rot}} - \text{Tr}[P^{\text{rot}}\rho^{\text{ss}}]$ . To obtain the last equality in Eq. (S42), which connects  $\text{var}(\dot{N})$  to the standard low frequency power fluctuations, we employed standard equalities for two-point correlation functions in open quantum systems [8].

#### IV. A CLASSICAL FORMULATION OF THE THREE-LEVEL MASER

To understand the quantum nature of the SSDB maser we construct a classical equivalent system for reference. We omit the Hamiltonian from the master equation and add the Lindblad jump operators  $\sigma_{ul}$  and  $\sigma_{lu}$  with the coupling rate  $\gamma_c$ . Any coherence vanishes in this formulation and we consider the vectorized density matrix  $\rho^{\text{cl}} = (\rho_{xx}, \rho_{uu}, \rho_{ll})^T$  with the Liouvillian

$$\mathcal{L}^{\text{cl}} = \begin{bmatrix} -\gamma_l(n_l + 1) - \gamma_u(n_u + 1) & \gamma_u n_u e^{i\chi} & \gamma_l n_l \\ \gamma_u(n_u + 1) e^{-i\chi} & -\gamma_c - \gamma_u n_u & \gamma_c \\ \gamma_l(n_l + 1) & \gamma_c & -\gamma_c - \gamma_l n_l \end{bmatrix}. \quad (\text{S43})$$

Similarly to Sec II we determine the polynomial factors with respective derivatives

$$\begin{aligned} a'_0 &= -\gamma_c \gamma_l \gamma_u (n_l - n_u), \\ a''_0 &= -\gamma_c \gamma_l \gamma_u (2n_l n_u + n_l + n_u), \\ a_1 &= 2\gamma_c [3\Gamma + \gamma_l + \gamma_u] + \gamma_l \gamma_u (3n_l n_u + n_l + n_u), \\ a'_1 &= 0, \\ a_2 &= 2\gamma_c + 4\Gamma + \gamma_l + \gamma_u, \end{aligned} \quad (\text{S44})$$

where  $\Gamma = \frac{1}{2}(\gamma_u n_u + \gamma_l n_l)$  is the quantum decoherence rate. The classical steady-state current reads

$$\langle \dot{N}^{\text{cl}} \rangle = -\frac{a'_0}{a_1} = \frac{\gamma_c \gamma_u \gamma_l (n_l - n_u)}{2\gamma_c [3\Gamma + \gamma_u + \gamma_l] + \gamma_l \gamma_u (3n_l n_u + n_l + n_u)}. \quad (\text{S45})$$

Comparing with the quantum current (S28) we note that the two currents coincide  $\langle \dot{N}^{\text{cl}} \rangle = \langle \dot{N} \rangle$  when the coupling rate is defined as

$$\gamma_c = \frac{2\epsilon^2 \Gamma}{\Delta^2 + \Gamma^2}. \quad (\text{S46})$$

Next, we determine the classical Fano factor

$$\begin{aligned} F(\dot{N}^{\text{cl}}) &= \frac{a''_0}{a'_0} - 2\langle \dot{N}^{\text{cl}} \rangle \left[ \frac{a_2}{a_1} - \frac{a'_1}{a'_0} \right] \\ &= \frac{n_l(n_u + 1) + n_u(n_l + 1)}{n_l - n_u} - 2\langle \dot{N}^{\text{cl}} \rangle \frac{2\gamma_c + 4\Gamma + \gamma_l + \gamma_u}{2\gamma_c [3\Gamma + \gamma_l + \gamma_u] + \gamma_l \gamma_u (3n_l n_u + n_l + n_u)}. \end{aligned} \quad (\text{S47})$$

Similarly to Sec II, we can write the Fano factor as the sum  $F^{\text{cl}} = F_{\text{pop}} + F_{\text{tr}}^{\text{cl}}$  where the population term is identical to its quantum counterpart (S30) and

$$F_{\text{tr}}^{\text{cl}} = -2\langle \dot{N}^{\text{cl}} \rangle \frac{2\gamma_c + 4\Gamma + \gamma_l + \gamma_u}{2\gamma_c [3\Gamma + \gamma_l + \gamma_u] + \gamma_l \gamma_u (3n_l n_u + n_l + n_u)}, \quad (\text{S48})$$

defines the classical Fano transport term. Finally, we define the classical thermodynamic uncertainty

$$\mathcal{Q}^{\text{cl}} = \ln \left[ \frac{n_l(n_u + 1)}{n_u(n_l + 1)} \right] F_{\text{pop}} + \ln \left[ \frac{n_l(n_u + 1)}{n_u(n_l + 1)} \right] F_{\text{tr}}^{\text{cl}} = \mathcal{Q}_{\text{pop}} + \mathcal{Q}_{\text{tr}}^{\text{cl}}. \quad (\text{S49})$$

Similarly to the Fano factor,  $\mathcal{Q}^{\text{cl}}$  contains a population term identical to its quantum counterpart (S32). As described in Sec. II this is restricted to values above  $\mathcal{Q}_{\text{pop}} \geq 2$ . Considering the classical transport term  $\mathcal{Q}_{\text{tr}}^{\text{cl}}$  we note that it can never be positive, unlike its quantum counterpart  $\mathcal{Q}_{\text{tr}}$ . Hence,  $\mathcal{Q}_{\text{pop}}$  is an upper bound for the classical thermodynamic uncertainty.

#### V. QUANTUM THERMODYNAMIC UNCERTAINTY RELATION

A quantum formulation of TUR was recently suggested which for Lindblad master equations in our notation reads [9]

$$\frac{\text{var}(\dot{N})}{\langle \dot{N} \rangle^2} \geq \frac{1}{\Upsilon + \Psi}, \quad (\text{S50})$$

where the quantum dynamical activity is the average rate of transitions in the steady-state and reads

$$\Upsilon = \sum_{\alpha \in \{u, l\}} \gamma_{\alpha} (1 + n_{\alpha}) \rho_{xx}^{\text{ss}} + \gamma_{\alpha} n_{\alpha} \rho_{\alpha}^{\text{ss}}, \quad (\text{S51})$$

and the coherent-dynamics contribution reads

$$\Psi = -4\text{Tr}[\mathcal{K}_1 \mathcal{L}^{\text{D}} \mathcal{K}_2 [\rho^{\text{ss}}] + \mathcal{K}_2 \mathcal{L}^{\text{D}} \mathcal{K}_1 [\rho^{\text{ss}}]], \quad (\text{S52})$$

where  $\mathcal{K}_1[\rho] = -i\tilde{H}\rho + \frac{1}{2}\sum_{\alpha}\mathcal{L}_{\alpha}[\rho]$  and  $\mathcal{K}_2[\rho] = i\rho\tilde{H} + \frac{1}{2}\sum_{\alpha}\mathcal{L}_{\alpha}[\rho]$ , and  $\mathcal{L}^{\text{D}}$  is the Drazin inverse of the Liouvillian  $\mathcal{L} = \mathcal{K}_1 + \mathcal{K}_2$ . In Sec II we exploited the fact that the Lindblad master equation preserves hermiticity of the density matrix in order to write the master equation in a reduced basis of the populations and upper-diagonal coherence terms. However, the superoperators  $\mathcal{K}$  do not individually preserve hermiticity of the density matrix, hence, the reduced basis employed in Sec. II becomes inconvenient. For the vectorized density matrix in the basis  $\rho^{\text{rot}} = (\rho_{xx}, \rho_{uu}, \rho_{ll}, \rho_{ul}, \rho_{lu})^T$  the  $\mathcal{K}$ -matrices read

$$\mathcal{K}_1 = \begin{bmatrix} -\frac{\gamma_l(n_l+1)}{2} - \frac{\gamma_u(n_u+1)}{2} & \frac{\gamma_u n_u}{2} & \frac{\gamma_l n_l}{2} & 0 & 0 \\ \frac{\gamma_u(n_u+1)}{2} & i\Delta - \frac{\gamma_u n_u}{2} & 0 & 0 & -i\epsilon \\ \frac{\gamma_l(n_l+1)}{2} & 0 & -\frac{\gamma_l n_l}{2} & -i\epsilon & 0 \\ 0 & 0 & -i\epsilon & i\Delta - \frac{\gamma_u n_u}{2} & 0 \\ 0 & -i\epsilon & 0 & 0 & -\frac{\gamma_l n_l}{2} \end{bmatrix}, \quad (\text{S53})$$

and

$$\mathcal{K}_2 = \begin{bmatrix} -\frac{\gamma_l(n_l+1)}{2} - \frac{\gamma_u(n_u+1)}{2} & \frac{\gamma_u n_u}{2} & \frac{\gamma_l n_l}{2} & 0 & 0 \\ \frac{\gamma_u(n_u+1)}{2} & -i\Delta - \frac{\gamma_u n_u}{2} & 0 & i\epsilon & 0 \\ \frac{\gamma_l(n_l+1)}{2} & 0 & -\frac{\gamma_l n_l}{2} & 0 & i\epsilon \\ 0 & i\epsilon & 0 & -\frac{\gamma_l n_l}{2} & 0 \\ 0 & 0 & i\epsilon & 0 & -i\Delta - \frac{\gamma_u n_u}{2} \end{bmatrix}. \quad (\text{S54})$$

The Drazin inverse can be computed with the help of the Moore-Penrose pseudoinverse  $\mathcal{L}^{\text{MP}} = (\mathcal{L}^{\dagger} \mathcal{L})^{-1} \mathcal{L}^{\dagger}$  as

$$\mathcal{L}^{\text{D}} = (\mathcal{I} - \mathcal{P}) \mathcal{L}^{\text{MP}} (\mathcal{I} - \mathcal{P}), \quad (\text{S55})$$

where  $\mathcal{I}_{ij} = \delta_{ij}$ , with  $\delta_{ij}$  denoting the Kronecker delta, and  $\mathcal{P}$  the projection operator

$$\mathcal{P} = [\rho^{\text{ss}}, \rho^{\text{ss}}, \rho^{\text{ss}}, \vec{0}, \vec{0}], \quad (\text{S56})$$

where the columns are repeated instances of the vectorized density matrix and a vectorized zero state  $\vec{0} = (0, 0, 0, 0, 0)^T$ .

Rearranging (S50) we obtain the time-differentiated quantum TUR

$$\mathcal{Q} = \sigma \frac{\text{var}(\dot{N})}{\langle \dot{N} \rangle^2} \geq \frac{\sigma}{\Upsilon + \Psi} \equiv \mathcal{B}, \quad (\text{S57})$$

as used in Fig. 2 of the main article.

- 
- [1] M. Esposito, U. Harbola, and S. Mukamel, Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems, *Rev. Mod. Phys.* **81**, 1665 (2009).
  - [2] G. Schaller, *Open quantum systems far from equilibrium*, Lecture Notes in Physics, Vol. 881 (Springer, 2014).
  - [3] H.-P. Breuer and F. Petruccione, *The theory of open quantum systems* (Oxford University Press, 2002).
  - [4] M. Bruderer, L. D. Contreras-Pulido, M. Thaller, L. Sironi, D. Obreschkow, and M. B. Plenio, Inverse counting statistics for stochastic and open quantum systems: the characteristic polynomial approach, *New J. Phys.* **16**, 033030 (2014).
  - [5] C. Flindt, T. Novotný, A. Braggio, and A.-P. Jauho, Counting statistics of transport through Coulomb blockade nanostructures: High-order cumulants and non-Markovian effects, *Phys. Rev. B* **82**, 155407 (2010).
  - [6] D. Mandal and C. Jarzynski, Analysis of slow transitions between nonequilibrium steady states, *J. Stat. Mech: Theory Exp.* **2016**, 063204 (2016).
  - [7] M. Scandi and M. Perarnau-Llobet, Thermodynamic length in open quantum systems, *Quantum* **3**, 197 (2019).
  - [8] C. W. Gardiner and P. Zoller, *Quantum Noise*, edited by H. Haken (Springer, 2000).
  - [9] Y. Hasegawa, Quantum thermodynamic uncertainty relation for continuous measurement, *Phys. Rev. Lett.* **125**, 050601 (2020).
  - [10] C. Maes, Frenesy: Time-symmetric dynamical activity in nonequilibria, *Phys. Rep.* **850**, 1 (2020).