

Optimal control over the full counting statistics in a nonadiabatic pump

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We introduce a systematic procedure based on optimal control theory to address the full counting statistics of particle transport in a stochastic system. Our approach enhances the performance of a Thouless pump in the nonadiabatic regime by simultaneously optimizing the average pumping rate while minimizing noise. We demonstrate our optimization procedure on a paradigmatic model for the electronic transport through a quantum dot, both in the limit of vanishing Coulomb interaction and in the interacting regime. Our method enables independent control of the moments associated with charge and spin transfer, allowing for the enhancement of spin current with minimal charge current or the independent tuning of spin and charge fluctuations. These results underscore the versatility of our approach, which can be applied to a broad class of stochastic systems.

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I. INTRODUCTION

Thouless pumping [1,2] captures the topological features of the quantum Hall effect enabling minimal dissipation charge and spin transfer in mesoscopic systems [3–10]. Its significance extends beyond condensed matter physics, sparking intense research interest across diverse scientific domains [11–19]. Notably, robust quantization of pumped charge has driven metrological advancements [20–25]. At its core, pumping is realized by cyclically modulating the system-reservoir couplings, effectively pumping electrons at zero bias. However, the delicate balance between pumping speed and adiabaticity presents a critical challenge. To minimize dissipation, the modulation must be sufficiently slow, ensuring that the system's instantaneous probability vector closely tracks its stationary state. Operating beyond the adiabatic limit inevitably leads to deviations from the stationary state [26], resulting in a sharp decline in the geometric [27] pumping rate.

For a stochastic system coupled to multiple reservoirs, the stochastic dynamics equation bears a strong resemblance to a Schrödinger equation in imaginary time, where the system's instantaneous probability vector, $(p_1(t), \dots, p_N(t))$ associated with the different states $\{1, \dots, N\}$, describes the system state. Within this framework, the breakdown of the unique features of a Thouless pumping at high frequencies can be understood as the emergence of undesired nonadiabatic transitions from the instantaneous adiabatic state. To maintain the efficiency of geometric pumping in the nonadiabatic regime, previous studies [28,29] have applied *shortcut-to-adiabaticity* (STA) techniques [30–32], originally developed in quantum physics.

In practice, such methods often introduce an extra driving term—e.g., the counterdiabatic (CD) correction—to suppress nonadiabatic transitions. However, this approach suffers from several limitations. First, implementing the shortcut strategy is challenging when only a subset of the transition rates can be controlled. Moreover, while STA can extend Thouless pumping beyond the adiabatic regime, the range of accessible frequencies remains limited: at sufficiently high frequencies, the required time-dependent transition rates to implement the counterdiabatic drive would become negative, rendering them unphysical. Most importantly, although STA techniques successfully preserve the average number of particles transferred per cycle, they fail to control the fluctuations in the number of transferred particles. This is a crucial limitation for many applications, particularly in metrology [33–35]. So far, fluctuations in geometric pumping have been mostly analyzed in the adiabatic limit [36]. Full counting statistics (FCS) [37–39] have been investigated in the nonadiabatic regime only for specific examples of Thouless cycles [40], but these studies reported very low pumping efficiency at high frequencies.

Here, we present a systematic procedure for simultaneously controlling both the average flux and the counting statistics of the transfer. To this end, we introduce an effective space of sufficiently large dimension to accommodate the system's state along with additional degrees of freedom governing the FCS dynamics. Applied to charge and spin transport through a quantum dot (QD), we show that our procedure demonstrates unprecedented independent control of charge and spin fluctuations, paving the way for time-dependent applications in spintronics.

II. FULL COUNTING STATISTICS FORMALISM FOR A TWO-STATE SYSTEM

We consider a stochastic system coupled to two reservoirs with time-dependent tunneling rates $\{\Gamma_L^\pm(t), \Gamma_R^\pm(t)\}$. $\Gamma_L^+(t)$ represents particle transfer from the left reservoir to the system, while $\Gamma_L^-(t)$ accounts for the reverse process, as shown

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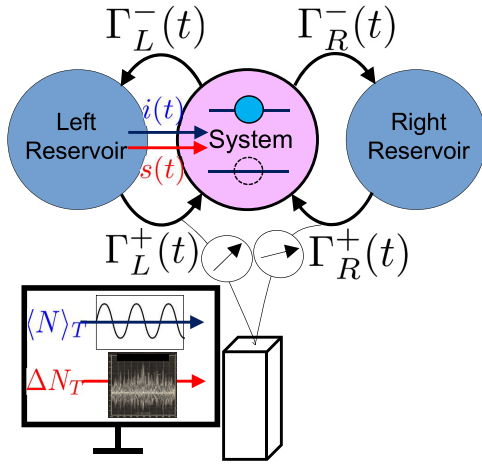


FIG. 1. Schematic model of a quantum dot coupled to two reservoirs. Our procedure optimizes $\Gamma_{L,R}^+(t)$ to maximize the average pumped current while minimizing the associated noise.

in Fig. 1. The system state is described by a probability vector $|\mathbf{p}(t)\rangle = (p_0(t), \dots, p_N(t))^T$, where $p_k(t)$ denotes the probability of having k particles in the system. The coupling to the reservoirs induces a Markovian dynamics governed by the master equation:

$$\partial_t |\mathbf{p}(t)\rangle = \mathcal{L}_0(t) |\mathbf{p}(t)\rangle. \quad (1)$$

This stochastic equation captures the microscopic theory of quantum transport [41]. To illustrate our method, we consider a single-resonance QD coupled to two reservoirs, where the system can accommodate at most one particle (Fig. 1). This simplification is valid when the decay widths $\hbar\Gamma_{L,R}^\pm(t)$ are significantly smaller than both the thermal energy and the chemical potential difference between the reservoirs, specifically $\hbar\Gamma_{L,R}^\pm(t) \ll \max\{k_B T_{\text{res}}, eV\}$ [42]. The operator $\mathcal{L}_0(t)$, defined below [Eq. (2) with $\chi = 0$], depends on the transition rates $\Gamma_{L,R}^\pm(t)$. In accordance with the conservation of the total probability sum, $\langle \mathbf{q}_0 | \mathbf{p}(t) \rangle = 1$, where $\langle \mathbf{q}_0 | = (1, 1)$, it follows that $\langle \mathbf{q}_0 | \mathcal{L}_0(t) = 0$ (each column of $\mathcal{L}_0(t)$ sums to zero). Consequently, $\mathcal{L}_0(t)$ has a zero eigenvalue associated with a stationary solution $|\pi(t)\rangle = (\pi_0(t), \pi_1(t))^T$, along with a negative eigenvalue $-\Gamma(t)$ where $\Gamma(t) = \Gamma_L^+ + \Gamma_L^- + \Gamma_R^+ + \Gamma_R^-$.

A FCS theory of charge transport [37] can be formulated for this system [42]. The characteristic function $\phi(\chi, t) = \langle e^{i\chi N} \rangle$ describes the transfer of N particles from the system to the left reservoir over the time interval $[t_0, t]$. Moments are obtained by differentiation with respect to the counting field χ : $\langle N^m \rangle = \partial_{i\chi}^m \phi(\chi, t)|_{\chi=0}$, where $\langle N^m \rangle \equiv \langle N^m \rangle(t)$. To determine $\phi(\chi, t)$, we solve the master equation $\partial_t |\mathbf{p}(\chi, t)\rangle = \mathcal{L}(\chi, t) |\mathbf{p}(\chi, t)\rangle$ with initial condition $|\mathbf{p}(\chi, t_0)\rangle = |\mathbf{p}(t_0)\rangle$ and a modified Liouvillian [42]:

$$\mathcal{L}(\chi, t) = \begin{pmatrix} -\Gamma_L^+(t) - \Gamma_R^+(t) & \Gamma_L^-(t)e^{-i\chi} + \Gamma_R^-(t) \\ \Gamma_L^+(t)e^{i\chi} + \Gamma_R^+(t) & -\Gamma_L^-(t) - \Gamma_R^-(t) \end{pmatrix}, \quad (2)$$

obtained from $\mathcal{L}_0(t)$ by modifying the left reservoir's tunneling rates as $\Gamma_L^\pm(t) \rightarrow \Gamma_L^\pm(t)e^{\pm i\chi}$, by introducing a counting field factor $e^{\pm i\chi}$ in the off-diagonal elements. This substitution

affects only the left reservoir rates, as we monitor the particle current and the FCS there, and satisfies $\mathcal{L}_0(t) = \mathcal{L}(\chi = 0, t)$. The characteristic function is then $\phi(\chi, t) = \langle \mathbf{q}_0 | \mathbf{p}(\chi, t) \rangle$. To obtain the average particle number and noise transfer in the steady-state regime, we avoid transients by initiating the counting time t_0 when $|\mathbf{p}(t_0)\rangle$ is near the steady-state solution of Eq. (1). Typically, $t_0 = N_0 T$, where T is the cycling period for the transfer rates and $N_0 = 3$ suffices for the numerical examples presented herein (system-reservoir couplings are initiated at $t = 0$).

Let us now evaluate the steady-state moments $\langle N^m \rangle_T = \langle \mathbf{q}_0 | \partial_{i\chi}^m |\mathbf{p}(\chi, t_0 + T) \rangle |_{\chi=0}$ for the pumped particle number per period. The first moment stems from the integration of the particle current $i(t) = \partial_t \langle N \rangle = \langle \mathbf{q}_0 | \hat{J}_1(t) | \mathbf{p}(t) \rangle$ over a period, with $\hat{J}_1(t) = \partial_{i\chi} \mathcal{L}(\chi, t)|_{\chi=0}$ the current operator [40]. The noise current is given by $s(t) = \partial_t [\langle N^2 \rangle - \langle N \rangle^2] = \langle \mathbf{q}_0 | \hat{J}_2(t) | \mathbf{p}(t) \rangle + 2\langle \mathbf{q}_0 | \hat{J}_1(t) | \bar{\mathbf{p}}(t) \rangle - 2i(t)\langle \mathbf{q}_0 | \bar{\mathbf{p}}(t) \rangle$, where we have introduced the operator $\hat{J}_2(t) = \partial_{i\chi}^2 \mathcal{L}(\chi, t)|_{\chi=0}$ and the vector $|\bar{\mathbf{p}}(t)\rangle = |\partial_{i\chi} \mathbf{p}(\chi, t)\rangle|_{\chi=0}$. The dynamical equation for $|\bar{\mathbf{p}}(t)\rangle$ is readily obtained by differentiating the master equation with respect to the counting field χ , namely, $\partial_t |\bar{\mathbf{p}}(t)\rangle = \mathcal{L}_0(t) |\bar{\mathbf{p}}(t)\rangle + \hat{J}_1(t) |\mathbf{p}(t)\rangle$. As $|\mathbf{p}(\chi, t_0)\rangle = |\mathbf{p}(t_0)\rangle$ for any value of χ , the initial condition is $|\bar{\mathbf{p}}(t_0)\rangle = (0, 0)^T$.

III. SHORTCUT-TO-ADIABATICITY DRIVING AND COUNTING STATISTICS

Here, we assess how a standard STA technique [31], recently applied to nonadiabatic Thouless pumping [28,29], affects the FCS. In the STA approach, an additional counterdiabatic driving term forces the instantaneous probability vector $|\mathbf{p}(t)\rangle$ to remain close to the adiabatic trajectory throughout the cycle (see Appendix A). The associated CD driving constrains only the total transition rates, $\Gamma^\pm(t) = \Gamma_L^\pm(t) + \Gamma_R^\pm(t)$, without imposing specific values for the individual left and right rates. This introduces a useful gauge freedom in the shortcut implementation.

As an example, we consider $\Gamma_L^{0(+)}(t) = A + R \cos(\Omega t)$, $\Gamma_R^{0(+)}(t) = A + R \sin(\Omega t)$, and $\Gamma_{L,R}^{0(-)}(t) = \Gamma_0$ with $A = 4\Gamma_0$ and $R = \Gamma_0$ [28]—the results are invariant under a global time rescaling as long as the transport equation (1) holds. From now on, all frequencies are given in units of Γ_0 . By implementing the appropriate CD shortcut on the right side, we get $\Gamma_L^{\text{STA}(\pm)}(t) = \Gamma_L^{0(\pm)}(t)$ and $\Gamma_R^{\text{STA}(\pm)}(t) = \Gamma_R^{0(\pm)}(t) \pm \gamma(t)$. Here, the extra CD term reads $\gamma(t) = \partial_t [\partial_t (\Gamma_0^-(t)/\Gamma_0^0(t))/\Gamma_0^0(t)]$ and $\Gamma_0^0(t) = \Gamma_0^+(t) + \Gamma_0^-(t)$.

We analyze the FCS for the nonadiabatic cycling frequency $\Omega = 10\Gamma_0$. While the STA driving achieves near-perfect average particle transfer, with $\langle N \rangle_{T,\text{STA}}/N_{T,\text{geo}} \simeq 1$, approaching the maximum geometric value $N_{T,\text{geo}} = 2\pi R^2/(4(A+1)^2 - 2R^2)^{3/4} = 6.5 \times 10^{-3}$ obtained in the adiabatic limit, and significantly outperforming of the original protocol ($\langle N \rangle_{T,(0)} \simeq 0.5N_{T,\text{geo}}$), it has minimal impact on the noise. The noise remains much larger than the geometric pumping rate for both protocols: $(\Delta N)_{T,\text{STA}}^2 \simeq (\Delta N)_{T,(0)}^2 \simeq 0.5$, which is detrimental to many applications. To address this, a more sophisticated approach is required, considering the dynamics of both $|\mathbf{p}\rangle$ and $|\bar{\mathbf{p}}\rangle$, as detailed below.

IV. QUANTUMLIKE OPTIMAL CONTROL OVER THE FULL COUNTING STATISTICS OF A TWO-STATE SYSTEM

The dynamics for the global vector $|\mathbf{p}, \bar{\mathbf{p}}\rangle = (|\mathbf{p}\rangle, |\bar{\mathbf{p}}\rangle)^T$ is compactly expressed as $\partial_t |\mathbf{p}, \bar{\mathbf{p}}\rangle = \mathcal{L}_4(\mathbf{u}(t), t) |\mathbf{p}, \bar{\mathbf{p}}\rangle$, where

$$\mathcal{L}_4(\mathbf{u}(t), t) = \begin{pmatrix} \mathcal{L}_0(t) & 0 \\ \hat{f}_1(t) & \mathcal{L}_0(t) \end{pmatrix}, \quad (3)$$

and $\mathbf{u}(t) = (\Gamma_L^+(t), \Gamma_R^+(t), \Gamma_L^-(t), \Gamma_R^-(t))$ is the control vector.

The goal is to maximize particle transfer while minimizing noise in the cyclic evolution of the control parameter. This conditional optimization problem can be readily formulated using the Pontryagin Maximum Principle (PMP) [43,44] (see Appendix B for more details on this procedure). For a system described by the differential equations $\dot{\mathbf{x}} = f(\mathbf{x}(t), \mathbf{u}(t), t)$, the PMP provides a framework to find the control vector $\mathbf{u}(t)$ that minimizes the cost function $C[\mathbf{u}(t)] = \int_0^T dt f_0(\mathbf{x}(t), \mathbf{u}(t), t)$. This is achieved by an effective Hamiltonian formalism, where the Pontryagin Hamiltonian is defined as $H_P(\mathbf{x}(t), \mathbf{v}(t), \mathbf{u}(t), t) = -f_0(\mathbf{x}(t), \mathbf{u}(t), t) + \mathbf{v}(t) \cdot f(\mathbf{x}(t), \mathbf{u}(t), t)$. Here, $\mathbf{v}(t)$ is an adjoint vector (a generalized Lagrange multiplier). The optimal solution $(\mathbf{x}^*(t), \mathbf{v}^*(t), \mathbf{u}^*(t))$ for an unbound control parameter is obtained by solving $\dot{\mathbf{v}} = -\partial_{\mathbf{x}} H_P$, $\dot{\mathbf{x}} = \partial_{\mathbf{v}} H_P$, and $\partial_{\mathbf{u}} H_P|_{\mathbf{u}^*} = 0$. In our case, $\mathbf{x}(t) = |\mathbf{p}(t)\rangle, |\bar{\mathbf{p}}(t)\rangle$, $f(\mathbf{x}(t), \mathbf{u}(t), t) = \mathcal{L}_4(\mathbf{u}(t), t) |\mathbf{p}(t)\rangle, |\bar{\mathbf{p}}(t)\rangle$, and $f_0 = w_1 i + w_2 s$, with $i(t) = \Gamma_L^+ x_1 - \Gamma_L^- x_2$ and $s(t) = \Gamma_L^+ x_1 + \Gamma_L^- x_2 + 2\Gamma_L^+ x_3 - 2\Gamma_L^- x_4 - 2(\Gamma_L^+ x_1 - \Gamma_L^- x_2)(x_3 + x_4)$. To minimize fluctuations, we set $w_2 > 0$. The sign of w_1 determines the direction of the particle current: $w_1 < 0$ for currents flowing from the left reservoir, $i(t) > 0$, and $w_1 > 0$ for currents toward it, $i(t) < 0$. Since the trajectory endpoint $\mathbf{x}(T)$ is kept free, the boundary condition for the adjoint is $\mathbf{v}(T) = \mathbf{0}$. These optimal solutions are obtained numerically using an iterative gradient descent optimization routine on a discrete time grid [45,46].

We present a practical implementation of this method for a two-state stochastic system (representing an empty or occupied single-particle state), with FCS described by the Lagrangian (2). This system corresponds to a QD in the limit of vanishing Coulomb interaction, allowing for independent treatment of spin-up and spin-down electron transport. We begin with the time-dependent pumping rates $\Gamma_{L,R}^{0\pm}(t)$, as previously defined. To maintain the periodicity and positivity of the pumping rates during the iterative process, we define $\Gamma_{L,R}^{\pm}(t) = [\sqrt{\Gamma_{L,R}^{0\pm}(t)} + \sin(\Omega t/2) f_{L,R}(t)]^2$, and allow the functions $f_{L,R}(t)$ to evolve freely according to the optimal control routine, while leaving the rates $\Gamma_{L,R}^{\pm}(t)$ unchanged.

$$\mathcal{L}(\chi, t) = \begin{pmatrix} -\Gamma^+(t) & \Gamma_{L,\uparrow}^-(t)e^{-i\chi_{\uparrow}} + \Gamma_{R,\uparrow}^-(t) & \Gamma_{L,\downarrow}^-(t)e^{-i\chi_{\downarrow}} + \Gamma_{R,\downarrow}^-(t) \\ \Gamma_{L,\uparrow}^+(t)e^{i\chi_{\uparrow}} + \Gamma_{R,\uparrow}^+(t) & -\Gamma_{\uparrow}^-(t) & 0 \\ \Gamma_{L,\downarrow}^+(t)e^{i\chi_{\downarrow}} + \Gamma_{R,\downarrow}^+(t) & 0 & -\Gamma_{\downarrow}^-(t) \end{pmatrix}, \quad (4)$$

where $\Gamma^+(t) = \sum_{k=\{L,R\}} \sum_{\sigma=\{\uparrow,\downarrow\}} \Gamma_{k,\sigma}^+(t)$ and $\Gamma_{\uparrow}^-(t) = \Gamma_{L,\uparrow}^-(t) + \Gamma_{R,\uparrow}^-(t)$. We denote the average number of spin-up

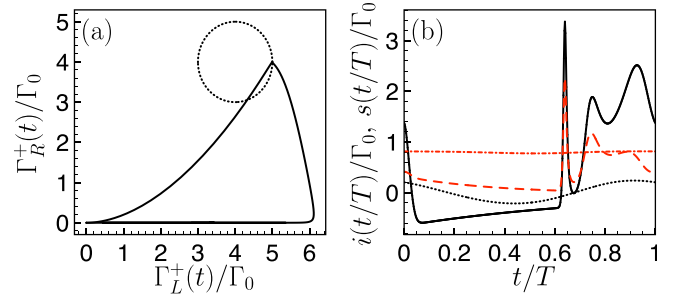


FIG. 2. Optimal transfer: (a) parametric plot of $\Gamma_L^+(t)$ vs $\Gamma_R^+(t)$ showing the initial (dashed line) and optimized (solid line) pumping rate cycles. (b) Final average particle $i(t/T)$ (solid line) and noise $s(t/T)$ (red dashed line) currents, and their initial profiles [dotted line for $i(t/T)$ and red dash-dotted line for $s(t/T)$]. All results scale with Γ_0 and are plotted as a function of the dimensionless time t/T with $T = 2\pi/\Omega$ the cycle period. These results were obtained after 100 iterations for $w_1 = -0.2 = -w_2$, $\epsilon(t) = 0.8\Gamma_0 \sin(\pi t/T)$, and $\Omega = 10\Gamma_0$.

Note that the equation governing the adjoint vector $\mathbf{v}(t)$, $\dot{\mathbf{v}} = -\partial_{\mathbf{x}} f_0 - \mathcal{L}_4^T \cdot \mathbf{v}$, differs from the one for the propagation of the system state $\mathbf{x}$. Since the operator $\mathcal{L}_4$ governing the forward propagation has negative (and null) eigenvalues, the operator $-\mathcal{L}_4^T$ appearing in the adjoint dynamics typically has positive (or null) eigenvalues. This prevents exponential growth during the backward propagation. Figure 2 shows the results of our optimization procedure. The quantities $i(t/T)$ $s(t/T)$ are given in units of Γ_0 . At a frequency $\Omega = 10\Gamma_0$, we achieve $\langle N \rangle_T = 0.22$ and $\Delta N_T^2 = 0.23$. Notably, ΔN_T^2 is reduced by approximately 50% compared to the shortcut trajectory, while $\langle N \rangle_T$ is increased by over an order of magnitude.

V. OPTIMAL CONTROL OVER THE FULL COUNTING STATISTICS IN THE INTERACTING REGIME

To demonstrate the versatility of our method, we now explore the interacting regime, where electrons are pumped into the system at different rates depending on their spin orientation. Let us consider now quantum dot systems at the Coulomb-blockade regime [47–49], for which $\hbar\Gamma \ll k_B T_{\text{res}} \ll \delta \ll U$, where T_{res} stands for the temperature, δ is the QD single-particle mean level spacing, and U is the charging energy. Since δ and U are much larger than the other energy scales, these QD systems are accurately described by a single-resonance QD [42]. In this regime, the system can be either empty or occupied by a single spin-up or spin-down electron. The corresponding FCS Lagrangian, expressed in the $\{|0\rangle, |\uparrow\rangle, |\downarrow\rangle\}$ basis, is given by

(spin-down) particles transferred from the left reservoir to the system per cycle as $\langle N_{\uparrow} \rangle_T$ ($\langle N_{\downarrow} \rangle_T$), with corresponding

current $i_{\uparrow}(t)$ ($i_{\downarrow}(t)$). We associate distinct counting fields, $\chi_{\uparrow}$ and $\chi_{\downarrow}$, with $i_{\uparrow}(t)$ and $i_{\downarrow}(t)$. The probability vector $|\mathbf{p}(t)\rangle$ follows Eq. (1) with $\mathcal{L}_0(t) = \mathcal{L}(\chi = \mathbf{0}, t)$.

We also consider a more realistic model where the transition rates $\Gamma_{k,\sigma}^{\pm}(t)$ are not independent but are determined by experimentally accessible physical parameters, such as applied voltages and magnetic fields, which influence the reservoir filling factors. The sharp dependence of these filling factors on the modulated fields leads to abrupt variations in the transition rates $\Gamma_{k,\sigma}^{\pm}(t)$. Consequently, the CD-STA approach becomes invalid, as the required counterdiabatic driving term involves extremely large or even unphysical negative Γ 's. In contrast, our PMP-based optimization procedure remains applicable to these available physical parameters.

We consider the QD model used in Refs. [40,42], which describes spin transport in the Coulomb-blockade regime. The transition rates are given by $\Gamma_{k\sigma}^{(+)}(t) = f_{k\sigma}(t)\Gamma_k(t)$ and $\Gamma_{k\sigma}^{(-)}(t) = [1 - f_{k\sigma}(t)]\Gamma_k(t)$, where $k \in \{L, R\}$ denotes the reservoir, $\sigma \in \{\uparrow, \downarrow\}$ the electron spin projection, and $f_{k\sigma}(t) = [1 + e^{-\epsilon_{k\sigma}(t)/(k_B T_{\text{res}})}]^{-1}$ the filling factors (for simplicity, we assume both reservoirs have the same temperature T_{res}) [50]. The chemical potential $\epsilon_{k\sigma}(t)$ can be tuned by voltage gates and magnetic fields. Specifically, $\epsilon_{k\sigma}(t) = V_k(t) + (-1)^s \epsilon_{kZ}(t)$ consists of an electric voltage contribution $V_k(t)$ (identical for both spins) and a spin-dependent Zeeman contribution $(-1)^s \epsilon_{kZ}(t)$ induced by a tunable magnetic field (where $\sigma = +1$ corresponds to spin-up).

Next, we investigate the feasibility of generating a net average spin current while minimizing fluctuations and total charge transport, which has potential applications in spintronics [51]. The charge and spin currents are given by $i_q(t) = -e i_{\Sigma}(t)$ with $i_{\Sigma}(t) = i_{\uparrow}(t) + i_{\downarrow}(t)$, and $i_{\sigma}(t) = \frac{\hbar}{2} i_{\Delta}(t)$ where $i_{\Delta}(t) = i_{\uparrow}(t) - i_{\downarrow}(t)$. The applied Zeeman field primarily influences the chemical potentials $\epsilon_{k\sigma}$ and, due to Pauli blocking, generates an unbalance between $i_{\uparrow}$ and $i_{\downarrow}$, without significantly affecting the junctions or the couplings $\Gamma_k(t)$.

The total charge and spin transfer per cycle are proportional to the dimensionless quantities $\langle N \rangle_T = \langle N_{\uparrow} + N_{\downarrow} \rangle_T = \int_{t_0}^{t_0+T} i_{\Sigma}(t) dt$ and $\langle \tilde{S} \rangle_T = \langle N_{\uparrow} - N_{\downarrow} \rangle_T = \int_{t_0}^{t_0+T} i_{\Delta}(t) dt$. Similarly, we quantify the dimensionless spin fluctuations per cycle as $\Delta \tilde{S}_T = \int_{t_0}^{t_0+T} s_{\uparrow\downarrow}(t) dt$ with $s_{\uparrow\downarrow}(t) = \partial_t[(\langle N_{\uparrow} - N_{\downarrow} \rangle^2) - \langle N_{\uparrow} - N_{\downarrow} \rangle^2]$, where higher moments are given by $\langle N_{\uparrow}^{m_1} N_{\downarrow}^{m_2} \rangle = \langle \mathbf{q}_0 | \partial_{i_{\chi_{\uparrow}}}^{m_1} \partial_{i_{\chi_{\downarrow}}}^{m_2} |\mathbf{p}(\chi, t)\rangle |_{\chi=0}$ with $|\mathbf{q}_0\rangle = (1, 1, 1)^T$ ($\langle \mathbf{q}_0 | \mathcal{L}_0(t) = 0$).

The spin currents read $i_{\sigma}(t) = \langle \mathbf{q}_0 | \hat{J}_{1,\sigma} | \mathbf{p}(t) \rangle$ with $\hat{J}_{1,\sigma} = \Gamma_{L,\sigma}^{(+)}(t) |\sigma\rangle \langle 0| - \Gamma_{L,\sigma}^{(-)}(t) |0\rangle \langle \sigma|$. In turn, the spin fluctuations read $s_{\uparrow\downarrow}(t) = \langle \mathbf{q}_0 | \hat{S}_{\uparrow\downarrow}(t) | \mathbf{p}(t) \rangle$ with (see Appendix C)

$$\begin{aligned} \hat{S}_{\uparrow\downarrow}(t) = & [\hat{J}_{2,\uparrow}(t) + \hat{J}_{2,\downarrow}(t)] |\mathbf{p}(t)\rangle + 2 \sum_{\sigma} \hat{J}_{1,\sigma}(t) |\bar{\mathbf{p}}^{\sigma}(t)\rangle \\ & - 2 \sum_{\sigma} \hat{J}_{1,\sigma}(t) |\bar{\mathbf{p}}^{\bar{\sigma}}(t)\rangle - 2 i_{\Delta}(t) [|\bar{\mathbf{p}}^{\uparrow}(t)\rangle - |\bar{\mathbf{p}}^{\downarrow}(t)\rangle], \end{aligned}$$

where $\bar{\sigma} = -\sigma$. Here, $|\bar{\mathbf{p}}^{\sigma}(t)\rangle = \partial_{i_{\chi_{\sigma}}} |\mathbf{p}(\chi, t)\rangle |_{\chi=0}$ evolve dynamically under the Lagrangian $\mathcal{L}_0(t)$, driven by their coupling to the probability vector $|\mathbf{p}(t)\rangle$ through their respective current operators $\hat{J}_{1,\sigma}(t)$ and fulfill the initial condition $|\bar{\mathbf{p}}^{\sigma}(t_0)\rangle = (0, 0, 0)^T$. In summary, the FCS dynamics of this system is governed by a master equation in a nine-dimensional

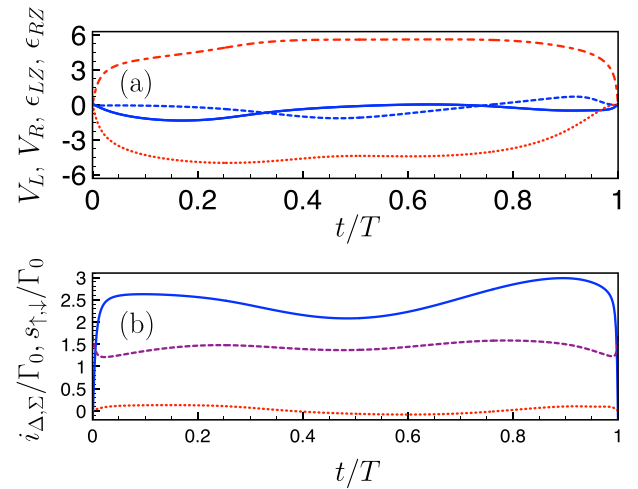


FIG. 3. Optimal transfer in Coulomb-blockade QD system after 100 iterations: (a) Optimized control parameters $V_L(t/T)$ (solid blue line), $V_R(t/T)$ (dashed blue line), $\epsilon_{LZ}(t/T)$ (dash-dotted red line), and $\epsilon_{RZ}(t/T)$ (dotted red line) in units of $V_0 = k_B T_{\text{res}}$ as a function of t/T . (b) Currents $i_{\Delta}(t/T)$ (solid blue line), $i_{\Sigma}(t/T)$ (dotted purple line), and spin fluctuations $s_{\uparrow\downarrow}(t/T)$ (dashed red line) for the optimized cycle as a function of t/T . Parameters: $\Omega = 10\Gamma_0$, $w_1 = -0.5$, $w_2 = 1$, $w_3 = 0.2$, and numerical increment $\epsilon(t) = 2V_0 \sin(\pi t/T)$. Prescribed cycle: $\Gamma_L(t) = A + R \cos(\Omega t)$ and $\Gamma_R(t) = A + R \sin(\Omega t)$ with $A = 4\Gamma_0$ and $R = \Gamma_0$. Initial control parameters: $V_L(t) = (V_0/10) \cos(\Omega t)$, $V_R(t) = (V_0/10) \sin(\Omega t)$, and $\epsilon_{LZ}(t) = \epsilon_{RZ}(t) = V_0/20$.

space, $\partial_t |\mathbf{p}, \bar{\mathbf{p}}^{\uparrow}, \bar{\mathbf{p}}^{\downarrow}\rangle = \mathcal{L}_9(\mathbf{u}(t), t) |\mathbf{p}, \bar{\mathbf{p}}^{\uparrow}, \bar{\mathbf{p}}^{\downarrow}\rangle$, where

$$\mathcal{L}_9(\mathbf{u}(t), t) = \begin{pmatrix} \mathcal{L}_0(t) & 0 & 0 \\ \hat{J}_{1,\uparrow}(t) & \mathcal{L}_0(t) & 0 \\ \hat{J}_{1,\downarrow}(t) & 0 & \mathcal{L}_0(t) \end{pmatrix}. \quad (5)$$

We now apply the PMP to this dynamical system ($\mathbf{x}(t) = |\mathbf{p}, \bar{\mathbf{p}}^{\uparrow}, \bar{\mathbf{p}}^{\downarrow}\rangle$ and $f(\mathbf{x}(t), \mathbf{u}(t), t) = \mathcal{L}_9(\mathbf{u}(t), t) |\mathbf{p}, \bar{\mathbf{p}}^{\uparrow}, \bar{\mathbf{p}}^{\downarrow}\rangle$). We define the cost function as $f_0(t) = w_1 i_{\Delta}(t) + w_2 i_{\Sigma}(t)^2 + w_3 s_{\uparrow\downarrow}(t)$, where the positive weights $w_{2,3} > 0$ are chosen to suppress the average charge current and reduce fluctuations in spin transfer. A negative $w_1 < 0$ ($w_1 > 0$) favors a net rightward transfer of spin-up (spin-down) particles.

We consider the cyclic couplings $\Gamma_L(t) = A + R \cos(\Omega t)$ and $\Gamma_R(t) = A + R \sin(\Omega t)$, and optimize the control parameters $\{V_L(t), V_R(t), \epsilon_{LZ}(t), \epsilon_{RZ}(t)\}$ composing $\mathbf{u}(t)$. To enforce the periodicity of the parameters, we apply a time-dependent increment $\epsilon(t) = \epsilon_0 \sin(\pi t/T)$ at each iteration, ensuring that initial and final values remain unchanged. Figure 3 displays the resulting optimized cycle and the corresponding time-dependent currents $i_{\Delta}(t)$, $i_{\Sigma}(t)$ and spin fluctuations $s_{\uparrow\downarrow}(t)$. The optimization process yields a nearly pure spin current ($\langle \tilde{S} \rangle_{T,\text{opt}} = 1.55$, $\langle N \rangle_{T,\text{opt}} = 0.02$) with the dimensionless spin fluctuations $\Delta \tilde{S}_{T,\text{opt}}^2 = 0.90$, in contrast to a negligible initial spin current ($\langle \tilde{S} \rangle_{T,\text{ini}} = 1.4 \times 10^{-3}$ and $\Delta \tilde{S}_{T,\text{ini}}^2 = 0.83$). The signal-to-noise ratio is improved by several orders of magnitude, while effectively suppressing the charge current. We note that our method also allows for the decoupling of spin and charge transfer statistics. In this case, the cost functional involves simultaneously the spin $s_{\uparrow\downarrow}(t)$ and particle $s_N(t)$ fluctuations (for more details, see Appendix D).

VI. CONCLUSION

We have investigated a nonadiabatic electron pump using full counting statistics with an approach that optimizes both the current and its noise. We demonstrate that our approach matches the STA's efficiency in generating a high-fidelity quantized current, but significantly expands its capabilities through a wider frequency range and the ability to mitigate current fluctuations. We have applied our method to demonstrate optimal control over a Thouless pump in the nonadiabatic regime in which charge and spin fluctuation can be independently tuned. This method is particularly relevant for high-accuracy pumping and can be readily extended to control even higher-order moments $\langle N^m \rangle_T$ with $m \geq 3$ [52]. As such, it can be transposed to a wide range of quantum phenomena in mesoscopic physics [53–57], including charge transport in nanostructures [58], stochastic thermodynamics [59–64], heat transfer [65], and Brownian motors [66]. Finally, counting statistics are also a key element in quantum optics, e.g., in the Hong-Ou-Mandel experiment [67] or to witness nonclassicality [68], so that the reported FCS control procedure may also inspire further developments in this field.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: SHORTCUT TO ADIABATICITY IN A TWO-STATE ADIABATIC PUMP

We briefly recall the main steps for the obtention of STAs in adiabatic pumps provided in Refs. [28,29]. Such STAs have been obtained in the context of two-state systems only. We consider the Liouvillian $\mathcal{L}_0(t) = \mathcal{L}(0, t)$ [$\mathcal{L}(\chi, t)$ given by Eq. (2)] and the associated equation (1) for the probability vector $|\mathbf{p}(t)\rangle$.

For a slow (adiabatic) variation of the Liouvillian with time, one expects the probability vector $|\mathbf{p}(t)\rangle$ to follow closely the instantaneous adiabatic state $|\pi(t)\rangle$, which fulfills by definition $\mathcal{L}_0(t)|\pi(t)\rangle = 0$. One can then recast the dynamical equation (1) as $(\frac{d}{dt} - \mathcal{L}_0(t))|\mathbf{p}(t)\rangle = 0$ and treat the time derivative as a perturbative term. By doing so, one obtains the following expansion for the probability vector $|\mathbf{p}(t)\rangle \simeq |\mathbf{p}_{\text{ad}}(t)\rangle$ [28] with

$$\begin{aligned} |\mathbf{p}_{\text{ad}}(t)\rangle &= |\pi(t)\rangle + |\delta\pi(t)\rangle, \\ |\delta\pi(t)\rangle &= \mathcal{R}(t)|\partial_t\pi(t)\rangle, \end{aligned} \quad (\text{A1})$$

where $\mathcal{R}(t)$ is the generalized inverse of the Liouvillian $\mathcal{L}_0(t)$. The near-adiabatic state $|\mathbf{p}_{\text{ad}}(t)\rangle$ captures first-order

nonadiabatic corrections, which is responsible for the geometric current. The average number of particles transferred per cycle is obtained as $N^{(0)} = \int_{t_0}^{t_0+T} dt \langle \mathbf{q}_0 | \hat{J}(t) | \mathbf{p}_{\text{ad}}(t) \rangle$ [where $\hat{J}(t) = \partial_{i\chi} \mathcal{L}(\chi, t)|_{\chi=0}$] and can be separated into a dynamical contribution arising from the stationary state $|\pi(t)\rangle$ and a geometric contribution (independent of the cycle period T) associated with the near-adiabatic correction $|\delta\pi(t)\rangle$. These read, respectively, $N_{\text{dyn}}^{(0)} = \int_{t_0}^{t_0+T} dt \langle \mathbf{q}_0 | \hat{J}(t) | \pi(t) \rangle$ and $N_{\text{geo}}^{(0)} = \int_{t_0}^{t_0+T} dt \langle \mathbf{q}_0 | \hat{J}(t) \mathcal{R}(t) | \partial_t\pi(t) \rangle$.

As the cycle is accelerated, the probability vector $|\mathbf{p}(t)\rangle$ fails to follow the near-adiabatic state $|\mathbf{p}_{\text{ad}}(t)\rangle$. This departure leads to a drastic diminution of the average number of particles transferred per cycle. To restore the average pumping efficiency, an additional term—referred to as the counterdiabatic term—is incorporated into the Liouvillian, formally expressed as

$$\mathcal{L}_{\text{cd}}(t) = |\partial_t\delta\pi(t)\rangle \langle \mathbf{q}_0| \quad (\text{A2})$$

or in a more explicit form as (there is not a unique choice for this counterdiabatic driving)

$$\mathcal{L}_{\text{cd}}(t) = \gamma(t) \begin{pmatrix} -1 & -1 \\ 1 & 1 \end{pmatrix}, \quad (\text{A3})$$

with $\gamma(t) = \partial_t[\alpha(t)/\Gamma^0(t)]$ and $\alpha(t) = \partial_t[\Gamma^{0(-)}(t)/\Gamma^0(t)]$. The total driving Liouvillian reads $\mathcal{L}_{\text{STA}} = \mathcal{L}_0(t) + \mathcal{L}_{\text{cd}}(t)$. With this extra driving term, the probability vector $|\mathbf{p}(t)\rangle$ follows exactly the near-adiabatic state $|\mathbf{p}_{\text{ad}}(t)\rangle$. As a consequence, the geometric transfer is equal to its adiabatic value $N_{\text{geo}}^{\text{STA}}(\Omega) = N_{\text{geo}}^{(0)}$ independently of the frequency Ω .

The counterdiabatic term $\mathcal{L}_{\text{cd}}(t)$ is implemented through a time-dependent shift in the total positive ($\Gamma^{(+)}(t)$) and negative ($\Gamma^{(-)}(t)$) pumping rates. However, in the high-frequency limit, this procedure yields unphysical negative pumping rates. In practice, the applicability of the counterdiabatic driving method is thus limited to a finite frequency range. Finally, it is not obvious at all to extend the STA approach (explicitly designed for two-state stochastic systems) to systems of higher dimension such as the three-state system considered for the simultaneous spin/charge transport.

APPENDIX B: APPLICATION OF THE PONTRYAGIN MAXIMUM PRINCIPLE IN STOCHASTIC PUMPS

We provide here additional details on the application of the Pontryagin maximum principle in the context of stochastic pumps.

1. General formulation of the Pontryagin Maximum Principle

We first recall the general formulation of the PMP. Let us consider a vector $\mathbf{x}(t)$ (referred to as the system state) following a dynamical equation controlled by a time-dependent vector $\mathbf{u}(t)$:

$$\dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}(t), \mathbf{u}(t), t), \quad (\text{B1})$$

with a given initial condition $\mathbf{x}(t_0) = \mathbf{x}_0$ and a cost functional to be minimized of the form

$$J[\mathbf{x}(t), \mathbf{u}(t)] = \phi(\mathbf{x}(t_f), t_f) + \int_{t_0}^{t_f} f_0(\mathbf{x}(t'), \mathbf{u}(t'), t') dt'. \quad (\text{B2})$$

$f(\mathbf{x}(t), \mathbf{u}(t), t)$ is an arbitrary vectorial function. Solving the control problem boils down to finding the time-dependent profile $\mathbf{u}^*(t)$ (called optimal control parameter) that yields a minimization of the above functional.

One introduces a time-dependent adjoint vector $\mathbf{v}(t)$ [of the same dimension as $\mathbf{x}(t)$] and an effective Hamiltonian (called the Pontryagin Hamiltonian):

$$H_P(\mathbf{x}(t), \mathbf{v}(t), \mathbf{u}(t), t) = -f_0(\mathbf{x}(t), \mathbf{u}(t), t) + \mathbf{v}(t) \cdot f(\mathbf{x}(t), \mathbf{u}(t), t). \quad (\text{B3})$$

The PMP states that if one imposes the following equation of motion for the adjoint vector

$$\dot{\mathbf{v}} = -\frac{\partial H_P}{\partial \mathbf{x}} \quad (\text{B4})$$

together with the boundary condition

$$\mathbf{v}(t_f) = -\left. \frac{\partial \phi(\mathbf{x}(t), t)}{\partial \mathbf{x}} \right|_{t=t_f}, \quad (\text{B5})$$

then the optimal control parameter $\mathbf{u}^*(t)$ maximizes at each instant the Pontryagin Hamiltonian along the trajectory (for $t_0 \leq t \leq t_f$):

$$H_P(\mathbf{x}(t), \mathbf{v}(t), \mathbf{u}^*(t), t) = \max_{\mathbf{u}(t)} [H_P(\mathbf{x}(t), \mathbf{v}(t), \mathbf{u}(t), t)]. \quad (\text{B6})$$

By construction, one also has $\dot{\mathbf{x}} = \partial H / \partial \mathbf{v}$, so the dynamical equations for $\{\mathbf{x}(t), \mathbf{v}(t)\}$ take the form of Hamilton's equations, but we stress that $H_P(\mathbf{x}(t), \mathbf{v}(t), \mathbf{u}^*(t), t)$ is a mere mathematical artifact to conduct the optimization.

The PMP provides us with an iterative procedure to obtain the optimal control $\mathbf{u}^*(t)$. Starting from a guess control parameter $\mathbf{u}^{(0)}(t)$, one obtains the system trajectory $\mathbf{x}^{(0)}(t)$ from Eq. (B1) and then the adjoint trajectory $\mathbf{v}^{(0)}(t)$ by solving the differential system in Eqs. (B4) and (B5). One may then compute the gradient of the Pontryagin Hamiltonian at each instant and assign to the control parameter a value $\mathbf{u}^{(1)}(t) = \mathbf{u}^{(0)}(t) + \epsilon \nabla_{\mathbf{u}} H_P(\mathbf{x}^{(0)}(t), \mathbf{v}^{(0)}(t), \mathbf{u}^{(0)}(t), t)$ with fixed sufficiently small parameter ϵ . One can then iterate the procedure with the control parameter $\mathbf{u}^{(1)}(t)$.

2. Dynamical equations in the context of the stochastic pump

In the context of the two-state system, one has $\mathbf{x}(t) = |\mathbf{p}(t), \bar{\mathbf{p}}(t)\rangle$, $f(\mathbf{x}(t), \mathbf{u}(t), t) = \mathcal{L}_4(\mathbf{u}(t), t)|\mathbf{p}(t), \bar{\mathbf{p}}(t)\rangle$ and the adjoint equation takes the form

$$\dot{\mathbf{v}} = \frac{\partial f_0(\mathbf{x}(t), \mathbf{u}(t), t)}{\partial \mathbf{x}} - \mathcal{L}_4(\mathbf{u}(t), t)^T \cdot \mathbf{v}(t). \quad (\text{B7})$$

In the spin/charge transport problem, one has $\mathbf{x}(t) = |\mathbf{p}, \bar{\mathbf{p}}^\uparrow, \bar{\mathbf{p}}^\downarrow\rangle$ and $f(\mathbf{x}(t), \mathbf{u}(t), t) = \mathcal{L}_9(\mathbf{u}(t), t)|\mathbf{p}, \bar{\mathbf{p}}^\uparrow, \bar{\mathbf{p}}^\downarrow\rangle$ with

an adjoint equation of the similar form:

$$\dot{\mathbf{v}} = \frac{\partial f_0(\mathbf{x}(t), \mathbf{u}(t), t)}{\partial \mathbf{x}} - \mathcal{L}_9(\mathbf{u}(t), t)^T \cdot \mathbf{v}(t). \quad (\text{B8})$$

In either case, the cost functional has no component associated with the final system state, so that $\phi(\mathbf{x}(t_f), t_f) = 0$. The boundary condition fulfilled by the adjoint vector is thus $\mathbf{v}(t_0 + T) = \mathbf{0}$.

The propagation of the adjoint $\mathbf{v}(t)$ follows a different equation than the state vector $\mathbf{x}(t)$. The Liouvillians $\mathcal{L}_{4,9}(\mathbf{u}(t))$ have null or negative eigenvalues, associated with stationary or exponentially decaying solutions. In contrast, the linear terms governing the adjoint equation $[-\mathcal{L}_4(\mathbf{u}(t))$ and $-\mathcal{L}_9(\mathbf{u}(t))$] have typically null or positive eigenvalues. As the adjoint propagation occurs backward, the numerical solutions remain well behaved. Also, the derivative of the cost function provides an additional driving term in the adjoint propagation $\mathbf{v}(t)$. This driving term is obtained by expressing the intensity and fluctuations composing the cost function $f_0(\mathbf{x}(t), \mathbf{u}(t), t)$ in terms of the coordinates of $\mathbf{x}(t)$ [see for instance Eq. (C7)].

APPENDIX C: FCS IN THE SPIN TRANSPORT

Our starting point is the FCS Lagrangian for the transfer of spin-1/2 particles in the strongly interacting regime [Eq. (4)]. The successive moments are given by $\langle N_\uparrow^{m_1} N_\downarrow^{m_2} \rangle = \langle \mathbf{q}_0 | \partial_{i_{\chi_\uparrow}}^{m_1} \partial_{i_{\chi_\downarrow}}^{m_2} | \mathbf{p}(\chi, t) \rangle |_{\chi=0}$ with $|\mathbf{q}_0\rangle = (1, 1, 1)^T$ s.t. $\langle \mathbf{q}_0 | \mathcal{L}(\mathbf{0}, t) = 0$ at all times. Consistently with the Coulomb blockade enabling at most one particle in the system, one retrieves $\langle N_\uparrow^{m_1} N_\downarrow^{m_2} \rangle = 0$ if $m_1 m_2 \neq 0$. The current associated with spin-up particles is obtained as

$$i_\uparrow(t) = \langle \mathbf{q}_0 | \partial_t \partial_{i_{\chi_\uparrow}} \mathbf{p}(\chi, t) |_{\chi=0} = \langle \mathbf{q}_0 | \hat{J}_{1,\uparrow} | \mathbf{p}(t) \rangle. \quad (\text{C1})$$

We introduce the operators

$$\hat{J}_{1,\uparrow} = \partial_{i_{\chi_\uparrow}} \mathcal{L}(\chi, t) |_{\chi=0} = \begin{pmatrix} 0 & -\Gamma_{L,\uparrow}^{(-)}(t) & 0 \\ \Gamma_{L,\uparrow}^{(+)}(t) & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$\hat{J}_{1,\downarrow} = \partial_{i_{\chi_\downarrow}} \mathcal{L}(\chi, t) |_{\chi=0} = \begin{pmatrix} 0 & 0 & -\Gamma_{L,\downarrow}^{(-)}(t) \\ 0 & 0 & 0 \\ \Gamma_{L,\downarrow}^{(+)}(t) & 0 & 0 \end{pmatrix},$$

so that $i_\uparrow(t) = \Gamma_{L,\uparrow}^{(+)} p_0 - \Gamma_{L,\uparrow}^{(-)} p_\uparrow$ and $i_\downarrow(t) = \Gamma_{L,\downarrow}^{(+)} p_0 - \Gamma_{L,\downarrow}^{(-)} p_\downarrow$. The spin transferred per cycle corresponds to $\langle \tilde{S} \rangle_T = \frac{\hbar}{2} \langle N_\uparrow - N_\downarrow \rangle_T$. The charge current and the spin current read, respectively, $i_q(t) = q i_\Sigma(t)$ with $i_\Sigma(t) = i_\uparrow(t) + i_\downarrow(t)$ and $q = -e$, and $i_\sigma(t) = \frac{\hbar}{2} i_\Delta(t)$ with $i_\Delta(t) = i_\uparrow(t) - i_\downarrow(t)$. The spin fluctuations are calculated following the procedure as $\Delta \tilde{S}_T^2 = \frac{\hbar^2}{4} \int_{t_0}^{t_0+T} s_{\uparrow\downarrow}(t) dt$ with

$$s_{\uparrow\downarrow}(t) = \partial_t [\langle (N_\uparrow - N_\downarrow)^2 \rangle - \langle N_\uparrow - N_\downarrow \rangle^2] = \langle \mathbf{q}_0 | (\partial_{i_{\chi_\uparrow}} - \partial_{i_{\chi_\downarrow}})^2 [\mathcal{L}(\chi, t) | \mathbf{p}(\chi, t)] |_{\chi=0} - 2i_\Delta(t) \langle \mathbf{q}_0 | (\overline{|\mathbf{p}(t)\rangle}^\uparrow - \overline{|\mathbf{p}(t)\rangle}^\downarrow) \rangle$$

$$= \langle \mathbf{q}_0 | (\hat{J}_{2,\uparrow}(t) + \hat{J}_{2,\downarrow}(t)) | \mathbf{p}(t) \rangle + 2 \langle \mathbf{q}_0 | \hat{J}_{1,\uparrow}(t) | \overline{|\mathbf{p}(t)\rangle}^\uparrow \rangle + 2 \langle \mathbf{q}_0 | \hat{J}_{1,\downarrow}(t) | \overline{|\mathbf{p}(t)\rangle}^\downarrow \rangle$$

$$- 2 \langle \mathbf{q}_0 | \hat{J}_{1,\uparrow}(t) | \overline{|\mathbf{p}(t)\rangle}^\downarrow \rangle - 2 \langle \mathbf{q}_0 | \hat{J}_{1,\downarrow}(t) | \overline{|\mathbf{p}(t)\rangle}^\uparrow \rangle - 2i_\Delta(t) \langle \mathbf{q}_0 | (\overline{|\mathbf{p}(t)\rangle}^\uparrow - \overline{|\mathbf{p}(t)\rangle}^\downarrow) \rangle, \quad (\text{C2})$$

where we have used that $\partial_{i_{\chi_1}} \partial_{i_{\chi_2}} \mathcal{L}(\chi, t)|_{\chi=0} = 0$ and $\langle \mathbf{q}_0 | \mathcal{L}(\mathbf{0}, t) = 0$ between the second and the third lines. One observes the presence of crossed contributions that simultaneously involve both spin orientations. We have introduced the operator and the three-dimensional vector:

$$\hat{J}_{2,\uparrow}(t) = \partial_{i_{\chi_1}}^2 \mathcal{L}(\chi, t)|_{\chi=0} = \begin{pmatrix} 0 & \Gamma_{L,\uparrow}^{(-)}(t) & 0 \\ \Gamma_{L,\uparrow}^{(+)}(t) & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$|\overline{\mathbf{p}(t)}^\uparrow\rangle = \partial_{i_{\chi_1}} |\mathbf{p}(\chi, t)\rangle|_{\chi=0}. \quad (\text{C3})$$

$\hat{J}_{2,\downarrow}(t)$ and $|\overline{\mathbf{p}(t)}^\downarrow\rangle$ are defined analogously. $|\overline{\mathbf{p}(t)}^{\uparrow(\downarrow)}\rangle$ follows the differential equation $\partial_t |\overline{\mathbf{p}(t)}^{\uparrow(\downarrow)}\rangle = \mathcal{L}(\mathbf{0}, t) |\overline{\mathbf{p}(t)}^{\uparrow(\downarrow)}\rangle + \hat{J}_{1,\uparrow(\downarrow)}(t) |\mathbf{p}(t)\rangle$. Summing up, one has now the coupled dynamics in a 3×3 dimensional space:

$$\frac{\partial |\mathbf{p}, \overline{\mathbf{p}}^\uparrow, \overline{\mathbf{p}}^\downarrow\rangle}{\partial t} = \mathcal{L}_9(\mathbf{u}(t), t) |\mathbf{p}, \overline{\mathbf{p}}^\uparrow, \overline{\mathbf{p}}^\downarrow\rangle,$$

$$\mathcal{L}_9(\mathbf{u}(t), t) = \begin{pmatrix} \mathcal{L}_0(t) & 0 & 0 \\ \hat{J}_{1,\uparrow}(t) & \mathcal{L}_0(t) & 0 \\ \hat{J}_{1,\downarrow}(t) & 0 & \mathcal{L}_0(t) \end{pmatrix},$$

$$|\mathbf{p}, \overline{\mathbf{p}}^\uparrow, \overline{\mathbf{p}}^\downarrow\rangle = \begin{pmatrix} \mathbf{p} \\ \overline{\mathbf{p}}^\uparrow \\ \overline{\mathbf{p}}^\downarrow \end{pmatrix}. \quad (\text{C4})$$

One then finds

$$i_\Delta(t) = (\Gamma_{L,\uparrow}^{(+)} - \Gamma_{L,\downarrow}^{(+)})p_0 - \Gamma_{L,\uparrow}^{(-)}p_\uparrow + \Gamma_{L,\downarrow}^{(-)}p_\downarrow,$$

$$i_\Sigma(t) = (\Gamma_{L,\uparrow}^{(+)} + \Gamma_{L,\downarrow}^{(+)})p_0 - \Gamma_{L,\uparrow}^{(-)}p_\uparrow - \Gamma_{L,\downarrow}^{(-)}p_\downarrow, \quad (\text{C5})$$

and from Eq. (C2)

$$s_{\uparrow\downarrow}(t) = \Gamma_{L,\uparrow}^{(+)}p_0 + \Gamma_{L,\uparrow}^{(-)}p_\uparrow + 2\Gamma_{L,\uparrow}^{(+)}\overline{p}_0^\uparrow - 2\Gamma_{L,\uparrow}^{(-)}\overline{p}_0^\uparrow$$

$$+ \Gamma_{L,\downarrow}^{(+)}p_0 + \Gamma_{L,\downarrow}^{(-)}p_\downarrow + 2\Gamma_{L,\downarrow}^{(+)}\overline{p}_0^\downarrow - 2\Gamma_{L,\downarrow}^{(-)}\overline{p}_0^\downarrow$$

$$- 2(\Gamma_{L,\uparrow}^{(+)}\overline{p}_0^\downarrow - \Gamma_{L,\uparrow}^{(-)}\overline{p}_0^\uparrow) - 2(\Gamma_{L,\downarrow}^{(+)}\overline{p}_0^\uparrow - \Gamma_{L,\downarrow}^{(-)}\overline{p}_0^\downarrow)$$

$$- 2[(\Gamma_{L,\uparrow}^{(+)} - \Gamma_{L,\downarrow}^{(+)})p_0 - \Gamma_{L,\uparrow}^{(-)}p_\uparrow + \Gamma_{L,\downarrow}^{(-)}p_\downarrow]$$

$$\times (\overline{p}_0^\uparrow + \overline{p}_0^\downarrow + \overline{p}_0^\uparrow - \overline{p}_0^\downarrow - \overline{p}_0^\uparrow - \overline{p}_0^\downarrow). \quad (\text{C6})$$

With our conventions, $(p_0, p_\uparrow, p_\downarrow, \overline{p}_0^\uparrow, \overline{p}_0^\downarrow, \overline{p}_0^\uparrow, \overline{p}_0^\downarrow, \overline{p}_0^\uparrow, \overline{p}_0^\downarrow) \equiv (x_1, x_2, x_3, x_4, x_5, x_6, x_7, x_8, x_9)$, corresponding to an alphabetical ordering $(0, \uparrow, \downarrow)$ where the superscript comes first.

For the practical implementation of the optimal control routines, it is useful to write the above quantities in terms of the coordinates of the $\mathbf{x}$ vector representing the instantaneous state in the enlarged vector space:

$$i_\Delta(t) = (\Gamma_{L,\uparrow}^{(+)} - \Gamma_{L,\downarrow}^{(+)})x_1 - \Gamma_{L,\uparrow}^{(-)}x_2 + \Gamma_{L,\downarrow}^{(-)}x_3,$$

$$i_\Sigma(t) = (\Gamma_{L,\uparrow}^{(+)} + \Gamma_{L,\downarrow}^{(+)})x_1 - \Gamma_{L,\uparrow}^{(-)}x_2 - \Gamma_{L,\downarrow}^{(-)}x_3,$$

$$s_{\uparrow\downarrow}(t) = \Gamma_{L,\uparrow}^{(+)}x_1 + \Gamma_{L,\uparrow}^{(-)}x_2 + 2\Gamma_{L,\uparrow}^{(+)}x_4 - 2\Gamma_{L,\uparrow}^{(-)}x_5$$

$$+ \Gamma_{L,\downarrow}^{(+)}x_1 + \Gamma_{L,\downarrow}^{(-)}x_3 + 2\Gamma_{L,\downarrow}^{(+)}x_7 - 2\Gamma_{L,\downarrow}^{(-)}x_8$$

$$- 2(\Gamma_{L,\uparrow}^{(+)}x_7 - \Gamma_{L,\uparrow}^{(-)}x_8) - 2(\Gamma_{L,\downarrow}^{(+)}x_4 - \Gamma_{L,\downarrow}^{(-)}x_6)$$

$$- 2[(\Gamma_{L,\uparrow}^{(+)} - \Gamma_{L,\downarrow}^{(+)})x_1 - \Gamma_{L,\uparrow}^{(-)}x_2 + \Gamma_{L,\downarrow}^{(-)}x_3]$$

$$\times (x_4 + x_5 + x_6 - x_7 - x_8 - x_9). \quad (\text{C7})$$

We can derive a similar expression for the fluctuation current associated with the charge transfer by writing

$$s_N(t) = \partial_t [((N_\uparrow + N_\downarrow)^2) - \langle N_\uparrow + N_\downarrow \rangle^2]$$

$$= \langle \mathbf{q}_0 | (\partial_{i_{\chi_1}} + \partial_{i_{\chi_2}})^2 [\mathcal{L}(\chi, t) |\mathbf{p}(\chi, t)\rangle] |_{\chi=0}$$

$$- 2i_\Sigma(t) \langle \mathbf{q}_0 | (|\overline{\mathbf{p}(t)}^\uparrow\rangle + |\overline{\mathbf{p}(t)}^\downarrow\rangle) \rangle$$

$$= \langle \mathbf{q}_0 | (\hat{J}_{2,\uparrow}(t) + \hat{J}_{2,\downarrow}(t)) |\mathbf{p}(t)\rangle + 2\langle \mathbf{q}_0 | \hat{J}_{1,\uparrow}(t) |\overline{\mathbf{p}(t)}^\uparrow\rangle$$

$$+ 2\langle \mathbf{q}_0 | \hat{J}_{1,\downarrow}(t) |\overline{\mathbf{p}(t)}^\downarrow\rangle + 2\langle \mathbf{q}_0 | \hat{J}_{1,\uparrow}(t) |\overline{\mathbf{p}(t)}^\downarrow\rangle$$

$$+ 2\langle \mathbf{q}_0 | \hat{J}_{1,\downarrow}(t) |\overline{\mathbf{p}(t)}^\uparrow\rangle - 2i_\Sigma(t) \langle \mathbf{q}_0 | (|\overline{\mathbf{p}(t)}^\uparrow\rangle + |\overline{\mathbf{p}(t)}^\downarrow\rangle) \rangle, \quad (\text{C8})$$

so that in terms of the coordinates, one finds

$$s_N(t) = \Gamma_{L,\uparrow}^{(+)}x_1 + \Gamma_{L,\uparrow}^{(-)}x_2 + 2\Gamma_{L,\uparrow}^{(+)}x_4 - 2\Gamma_{L,\uparrow}^{(-)}x_5$$

$$+ \Gamma_{L,\downarrow}^{(+)}x_1 + \Gamma_{L,\downarrow}^{(-)}x_3 + 2\Gamma_{L,\downarrow}^{(+)}x_7 - 2\Gamma_{L,\downarrow}^{(-)}x_8$$

$$+ 2(\Gamma_{L,\uparrow}^{(+)}x_7 - \Gamma_{L,\uparrow}^{(-)}x_8) + 2(\Gamma_{L,\downarrow}^{(+)}x_4 - \Gamma_{L,\downarrow}^{(-)}x_6)$$

$$- 2[(\Gamma_{L,\uparrow}^{(+)} + \Gamma_{L,\downarrow}^{(+)})x_1 - \Gamma_{L,\uparrow}^{(-)}x_2 - \Gamma_{L,\downarrow}^{(-)}x_3]$$

$$\times (x_4 + x_5 + x_6 + x_7 + x_8 + x_9). \quad (\text{C9})$$

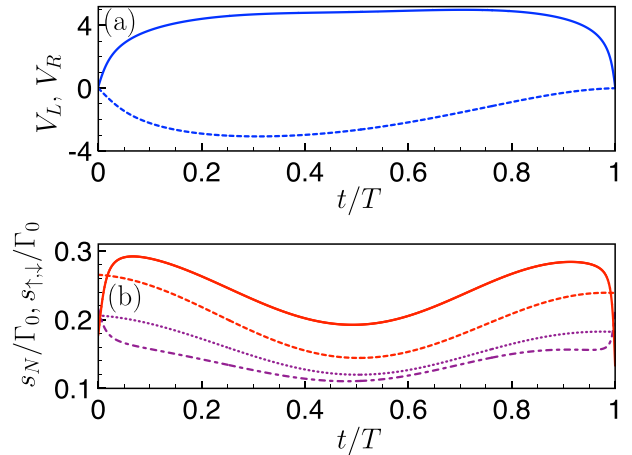


FIG. 4. Separate driving of the spin and charge fluctuations ΔS^2 and ΔN^2 after 100 iterations: (a) Evolution of the control parameters $V_L(t/T)$ (solid blue line) and $V_R(t/T)$ (dashed blue line) as a function of the rescaled time t/T . (b) Dimensionless spin $[s_{\uparrow\downarrow}(t/T)]$ (solid red line) and charge $[s_N(t/T)]$ (dash-dotted purple line) noise currents (in units of Γ_0) as a function of the rescaled time for the optimized cycle. Dashed red and dotted purple lines represent, respectively, the spin and charge fluctuations for the initial cycle. Parameters: $\Omega = 50\Gamma_0$, increment $\epsilon(t) = 4V_0 \sin(\pi t/T)$, $w_1 = 2$, $w_2 = -2$, and $\epsilon = 4$. Prescribed cycle $\{\Gamma_L(t), \Gamma_R(t)\}$, and initial profiles for $\{V_{L,R}(t/T), \epsilon_{L,RZ}(t/T)\}$ identical to Fig. 3.

APPENDIX D: REDUCTION OF THE CORRELATION BETWEEN SPIN AND CHARGE FCS

Here, we apply the method described in the main text to uncorrelate the charge and spin fluctuations. To this end, we consider the cost function $f_0(t) = w_1 s_N(t) + w_2 s_{\uparrow\downarrow}(t)$ with $w_1 > 0$ and $w_2 < 0$ in order to maximize the difference $|\Delta\tilde{S}_T^2 - \Delta N_T^2|$ (here, $\Delta\tilde{S}_{T,\text{ini}} > \Delta N_{T,\text{ini}}$). Figure 4 shows that the spin and charge fluctuations are driven in opposite

directions by the optimization routine. Our optimized cycle generates, respectively, spin and charge fluctuations $\Delta\tilde{S}_{T,\text{opt}}^2 = 0.153$ and $\Delta N_{T,\text{opt}}^2 = 0.087$ against $\Delta\tilde{S}_{T,\text{ini}}^2 = 0.124$ and $\Delta N_{T,\text{ini}}^2 = 0.099$ for the initial cycle. The ratio $\Delta\tilde{S}_T^2/\Delta N_T^2$ is thus increased by 25% through the optimization process. Figure 4 also presents the optimal driving parameters $\{V_L(t/T), V_R(t/T)\}$ (the control procedure yields very small values for the magnetic couplings).

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