Painting Non-Classical States of Spin or Motion with Shaped Single Photons: Supplemental Material

Department of Physics, Stanford University, Stanford, California 94305, USA

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In this supplement, we elaborate on details of derivations of the results in the main paper. In Sec. I, we provide analytic expressions for the final heralded state, fidelity $F_{\rm min}$, and detection rate at arbitrary drive strength in the spin system. In Sec. II we provide detailed derivations of the limits set by the single-atom cooperativity in painting non-classical states of spin or motion in atomic cavity-QED systems.

I. ANALYTIC RESULTS FOR COLLECTIVE SPIN STATES

The conditional evolution of the spin ensemble or oscillator is governed by the non-Hermitian Hamiltonian

$$H_{\rm eff} = H_{S/M} + \mathcal{E}c^\dagger + \mathcal{E}^* c + \left(\omega_c - i\frac{\kappa}{2}\right) c^\dagger c, \tag{S1}$$

where $H_{S/M}$ are the Hamiltonians for the spin/oscillator defined in Eqs. 1 and 3 of the main text. From $H_{\rm eff}$, we can obtain the composite state of the system and cavity field conditioned on no photon having exited the cavity up to time t,

$$|\Psi(t)\rangle \equiv e^{-iH_{\rm eff}t} |\psi_0\rangle \otimes |0\rangle . \tag{S2}$$

Furthermore, conditioned on one photon exiting the cavity at time t_d, and no photons exiting before t_d, the composite state is given by

$$|\Psi_1\rangle = \sqrt{\kappa} c \hat{\mathcal{T}} e^{-i\int_0^{t_d} H_{\rm eff}(t){\rm d}t} |\psi_0\rangle \otimes |0_c\rangle . \tag{S3}$$

This result is valid for any strength of the drive field.

In the main text, we first proceeded to analyze the conditional dynamics for the limit of a weak drive field, which offers the closest analogy between the spin ensemble and the mechanical oscillator. In the more general case, the spin system is easier to analyze analytically because the atom-light interaction Hamiltonian $\propto c^\dagger c S_z$ commutes with the system Hamiltonian. We here derive the conditional state of the spin system for arbitrary drive strength to arrive at an analytic expression for the fidelity $F_{\rm min}$.

A. Heralded Spin State for Arbitrary Drive Strength

In the spin ensemble, we can get the exact analytic form of the time evolution for arbitrary drive strength, because the cavity response depends on the spin state only through J_z, which is a constant of motion. To solve for the time evolution, we expand the composite state of the spins and field in the Dicke basis,

$$|\Psi(t)\rangle = \sum_m c_m(t) |m\rangle \otimes |\alpha_m(t)\rangle , \tag{S4}$$

where $|\alpha_m(t)\rangle$ denotes a coherent field of the cavity that depends on time and on $J_z = m$, and $c_m(t)$ is a complex amplitude. The Schrödinger equation with effective Hamiltonian $H_{\rm eff}$ then yields

$$i\dot{\alpha}_m = \omega_m \alpha_m + \mathcal{E}(t) \tag{S5a}$$

$$\frac{\dot{c_m}}{c_m} = \frac{1}{2}[\dot{\alpha}_m \alpha_m^* + \alpha_m \dot{\alpha}_m^*] - i\mathcal{E}^*(t)\alpha_m, \tag{S5b}$$

where $\omega_m = \omega_c + m\Omega_S - i\frac{\kappa}{2}$ is the complex frequency of the cavity resonance, accounting the finite cavity linewidth and the atom-induced dispersive shift. To arrive at Eqs. S5, we have used the relation

$$\frac{\partial}{\partial t} |\alpha(t)\rangle = -\frac{1}{2}[\dot{\alpha}(t)\alpha^*(t) + \alpha(t)\dot{\alpha}^*(t)] |\alpha(t)\rangle + \dot{\alpha}(t) a^\dagger |\alpha(t)\rangle . \tag{S6}$$

We solve the differential Equations S5 for initial conditions $\alpha_m(0) = 0$ and $c_m(0) = c_m^0$, corresponding to a product state of the atoms in state $|\psi_0\rangle$ and the vacuum field in the cavity. We thus obtain

$$\alpha_m(t) = -i\int_0^t \mathcal{E}(t')e^{-i\omega_m(t-t')}\mathrm{d}t', \tag{S7a}$$

$$c_m(t) = c_m^0 \exp\left(\frac{1}{2}|\alpha_m(t)|^2 - i\int_0^t \left[\mathcal{E}^*(t')\alpha_m\right]\mathrm{d}t'\right). \tag{S7b}$$

For times $t \geq T$, after the drive pulse has ended, we can conveniently express both $\alpha_m(t)$ and the complex amplitude $c_m(t)$ in terms of the Fourier transform of the drive pulse:

$$\tilde{\mathcal{E}}(\omega) = \frac{1}{\sqrt{2\pi}}\int_0^T \mathcal{E}(t)e^{i\omega t}\mathrm{d}t. \tag{S8}$$

In terms of $\tilde{\mathcal{E}}$, we have

$$\alpha_m(t) = -i\sqrt{2\pi}e^{-i\omega_m t}\tilde{\mathcal{E}}(\omega_m), \tag{S9a}$$

$$c_m(t) = \exp\left[\frac{1}{2}|\alpha_m(t)|^2 - i\int_{-\infty}^{\infty}\frac{|\tilde{\mathcal{E}}(\omega)|^2}{\omega - \omega_m}\right]c_m^0 \tag{S9b}$$

for $t > T$. Thus, for a given Dicke state $|m\rangle$, the coherent field in the cavity is determined by the component of the drive field at the m-dependent complex resonance frequency ω_m. The resulting atom-light entanglement enables the heralded generation of non-classical spin states. In the ideal limit of a weak drive, the complex amplitude $c_m(t)$ is the same as in the initial state, $c_m(t) = c_m^0$, before conditioning on a detected photon. To see the effect of a stronger drive, we now examine the final heralded state for arbitrary drive strength.

Conditioned on detecting a single photon at time t_d, the heralded atomic state is

$$|\psi_1(t_d)\rangle = \sqrt{\kappa}\langle 0|\hat{c}|\Psi(t)\rangle = -i\sqrt{2\pi\kappa}\sum_m \xi_m e^{-i\omega_m t_d}\tilde{\mathcal{E}}(\omega_m)c_m^0|m\rangle, \tag{S10}$$

where

$$\xi_m = \exp\left[-i\int_{-\infty}^{\infty}\frac{|\tilde{\mathcal{E}}(\omega)|^2}{\omega - \omega_m}\right]. \tag{S11}$$

In the limit of a weak drive, $\xi_m = 1$ for all m, and Eq. S10 reduces to the recipe of Eq. 14 of the main text for preparing the target state $|\psi_*\rangle$ with coefficients $c_m^f \propto c_m^0\tilde{\mathcal{E}}(\omega_m)$. At larger drive strength, ξ_m accounts for the effect of light that enters the cavity and leaks back out the input mirror before time t_d. This factor can generically reduce the fidelity $F_\epsilon \equiv |\langle\psi_1|\psi_*\rangle|^2 / \langle\psi_1|\psi_1\rangle$ even for perfect detection. However, for the states and drive strengths considered in this paper, $F_\epsilon \approx 1$ to a very good approximation, with the fidelity instead being limited by the effects of imperfect detection considered in Sec. I B.

B. Fidelity of Heralded Spin States

To determine the fidelity of the final heralded state, we must account for dark counts and finite quantum efficiency in photodetection. Due to these imperfections, we will generically prepare an incoherent mixture of the state produced for a single transmitted photon and states produced when either no photon is transmitted (and we detect a dark count) or multiple photons are transmitted. We calculate the lower bound $F_{\min}$ on the fidelity by assuming that the state conditioned on transmission of a single photon is orthogonal to the states produced when either no photon or more than one photon is transmitted. Then the fidelity is limited by the ratio of successful detector clicks to unsuccessful detector clicks:

$$F_{\min}(t) \equiv \frac{F_\epsilon(t)R_s(t)}{R_t(t) + R_d/Q}, \qquad T < t < t_{\max}, \tag{S12}$$

in terms of the transmission rate $R_t = \kappa\langle c^\dagger c\rangle$, success rate $R_s = \langle\psi_1|\psi_1\rangle$, and dark count rate R_d.

At the modest drive strengths of interest, where $F_\epsilon \approx 1$ (see Sec. I A), the success rate is approximately

$$R_s(t) \approx \kappa \left|\frac{\epsilon}{\Omega}\right|^2 e^{-|\frac{\epsilon}{\Omega}|^2} e^{-\kappa t}, \qquad T < t < t_{\max}, \tag{S13}$$

where ϵ parametrizes the drive strength according to Eq. 14 of the main text. The fidelity (Eq. S12) then reduces to

$$F_{\min}(t) \approx \frac{e^{-|\epsilon/\Omega|^2}}{1+\rho_\epsilon e^{\kappa t}}, \qquad T < t < t_{\max}, \tag{S14}$$

where $\rho_\epsilon = R_d/(Q\kappa\,|\epsilon/\Omega|^2)$. We use Eq. S14 for plotting $F_{\min}$ in Fig. 2a.

It should be emphasized that $F_{\min}$ is a conservative estimate of the fidelity, and we can also determine the actual fidelity at larger drive strengths. We calculate the atomic state for n transmitted photons, then trace over the detection times of $n-1$ photons. This yields a density matrix for the atoms corresponding to the detection at time t_d of a single heralding photon in the window $T < t < t_{\max}$, with $n-1$ additional transmitted but undetected photons. Roughly, the undetected photons smear the state with extra rotations about the z-axis. The smearing angle $\approx \Omega_S/\kappa$ can be small for $\Omega_S/\kappa \lesssim 1$. In Figure 2c ii, we plotted the Wigner function of the density matrix for a larger drive $\epsilon = \Omega_S$. The fidelity is $F = 0.72$, which is significantly higher than $F_{\min} = 0.37$.

II. ROLE OF THE COOPERATIVITY IN ATOMIC ENSEMBLES

Finite atom-light coupling strength limits the spin rotation or phase-space displacement that a single photon can induce in an atomic ensemble within the cavity lifetime. In particular, the dispersive atom-light coupling is accompanied by absorption that broadens the cavity linewidth to a value κ_N at atom number N, decreasing the spin rotation $\Phi_N = \Omega_S/\kappa_N$ or phase-space displacement $D_N \lesssim g_0/\kappa_N$ that the photon imparts within its lifetime $1/\kappa_N$. Fundamental limits on Φ_N and D_N are set by the single-atom cooperativity $\eta = \mathcal{G}^2/(\kappa\Gamma)$, where $\mathcal{G}$ is the vacuum Rabi frequency and Γ is the linewidth of the optical transition to which the cavity mode couples. The average spin rotation produced by a single photon is thus limited to $\Phi_N \leq \sqrt{\eta/(2N)}$, while the average displacement is limited to $D_N \lesssim \sqrt{\eta\omega_r/(2\Omega_{\mathrm{M}})}$, where ω_r is the single-atom recoil frequency. Larger rotations or displacements can be attained only with an exponentially decaying success probability. Below, we elaborate on the derivation of these limits.

A. Atomic Spin Ensembles

To determine the size of the spin rotation that can be induced by a single photon within the cavity lifetime, we consider three-level atoms with ground states $|\downarrow\rangle, |\uparrow\rangle$ and excited state $|e\rangle$. We assume that the cavity mode couples state $|\uparrow\rangle$ to an excited state $|e\rangle$ with vacuum Rabi frequency g, and that the other spin state $|\downarrow\rangle$ is unaffected by the light (e.g., the cavity mode is far detuned from transitions involving state $|\downarrow\rangle$). For a detuning Δ of the resonator mode from the $|\uparrow\rangle \to |e\rangle$ transition, a single photon induces a differential ac Stark shift $\Omega_S = g^2/\Delta$ between the two ground spin states $|\downarrow\rangle$ and $|\uparrow\rangle$.

The interaction time of the photon with the ensemble is limited by the intrinsic loss rate κ_0 of the cavity and an additional loss rate $(N/2)\Gamma_{\mathrm{sc}} = (N/2)(g/\Delta)^2\Gamma$, where Γ is the linewidth of the atomic excited state $|e\rangle$, due to the possibility of the photon being scattered out of the resonator by one of the approximately $N/2$ atoms in state $|\uparrow\rangle$. The atomic scattering results in a broadened cavity linewidth

$$\kappa_N = \kappa_0 + \frac{N}{2}\frac{g^2}{\Delta^2}\Gamma = \kappa_0 + \frac{N\Omega^2\Gamma}{2g^2}. \tag{S15}$$

The phase shift Ω/κ_N imparted in the cavity lifetime is maximized by operating at a detuning $\Delta = \Gamma\sqrt{\eta/(2N)}$, where $\eta = 4g^2/(\kappa_0\Gamma)$ is the single-atom cooperativity. Atomic decay then doubles the cavity linewidth ($\kappa_N = 2\kappa_0$), resulting in a ratio $\Omega_S/\kappa_N = \sqrt{\eta/(8N)}$.

The phase shift $\Phi_N \equiv \Omega/\kappa_N$ imparted in the cavity lifetime is an indication of how large a cat can realistically be prepared. Whereas the success probability is approximately constant for $\Phi < \Phi_N$, for larger angles the success probability decays exponentially. Thus, the threshold cooperativity for preparing an N-atom cat with phase separation Φ is $\eta_{\min} \sim N\Phi^2$. More generally, the fidelity of preparing a cat of a given "size" in units of the coherent state width, $\Phi\sqrt{N}$, is set by the single-atom cooperativity and is independent of atom number.

B. Collective Atomic Motion

The optomechanical interaction Hamiltonian for an ensemble of N atoms linearly coupled to a standing-wave cavity mode is approximately given by [1, 2]

$$H \approx kU_0 c^\dagger c \sum_{i=1}^{N} x_i = NkU_0 c^\dagger c X \equiv g_0 c^\dagger c X/X_0, \tag{S16}$$

where U_0 is the AC Stark shift due to a single atom at an antinode of the standing wave, X represents the center-of-mass coordinate, and the zero-point motion X_0 of the center of mass is related to oscillator length x_0 of a single atom by $X_0 \approx x_0/\sqrt{N}$. Thus, the optomechanical coupling strength is $g_0 = \sqrt{N}U_0\zeta$, where $\zeta = kx_0 = \sqrt{\omega_r/\Omega_\mathrm{M}}$ is the Lamb-Dicke parameter set by the single-atom recoil frequency ω_r at the cavity wavenumber k and by the trap frequency Ω_M.

How large can one make the ratio g_0/κ_N that limits the phase-space displacement D_N? We first note that the ac Stark shift is $U_0 = \mathcal{G}^2/\Delta$ in terms of the detuning Δ of the cavity mode from the atomic excited state, i.e., U_0 is identical to the value which we called Ω_S for the spin system in Sec. II A. Thus, the cooperativity sets a limit $\sqrt{N}U_0/\kappa_N \leq \sqrt{\eta/8}$ at an optimal detuning $\Delta/\Gamma = \sqrt{N\eta}/2$ where $\kappa_N = 2\kappa_0$. Hence, $g_0/\kappa_N \leq \zeta\sqrt{\eta/8}$. The Lamb-Dicke parameter ζ must be small to approximate the sinusoidal potential as a harmonic trap; in seminal experiments by Murch *et al.* [1], $\zeta \approx 0.3$ [1]. Achieving a collective optomechanical coupling $g_0 > \kappa_N$ then requires large *single-atom* cooperativity $\eta \gg 1$. Rewriting the limit on g_0/κ_N in terms of the recoil frequency ω_r, we find that the single-photon displacement within the cavity lifetime is limited to $D_N \lesssim \sqrt{\eta\omega_r/(2\Omega_\mathrm{M})}$. Thus, for preparing motional cat states, it is advantageous to operate in a regime of not only high cooperativity but also relatively low trap frequency, which also facilitates meeting the additional requirement $X_1 = g_0/\Omega_\mathrm{M} > 1$.

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[1] Kater W Murch, Kevin L Moore, Subhadeep Gupta, and Dan M Stamper-Kurn, "Observation of quantum-measurement backaction with an ultracold atomic gas," Nature Physics **4**, 561 (2008).

[2] Monika H Schleier-Smith, Ian D Leroux, Hao Zhang, Mackenzie A Van Camp, and Vladan Vuletić, "Optomechanical cavity cooling of an atomic ensemble," Physical Review Letters **107**, 143005 (2011).