Supplemental Material to
'Cross Modulation of Two Laser Beams at the Individual-Photon Level'

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THEORETICAL DESCRIPTION

The system consists of atoms with a four-state N-type level structure $|f\rangle \leftrightarrow |d\rangle \leftrightarrow |c\rangle \leftrightarrow |e\rangle$ as described in the text. Including the decay, the effective Hamiltonian for this system can be written as

$$\begin{aligned} H_{eff}/\hbar = &\sum_k c\,|k|\, a_k^\dagger a_k + (\omega_c - i\kappa/2) b^\dagger b + i\mathcal{E}(b^\dagger - b) \\ &+ (\omega_{fd} - i\Gamma/2)\sum_j |d\rangle_j \langle d| + (\omega_{ce} - i\Gamma/2)\sum_j |e\rangle_j \langle e| + (\omega_{fc} - i\gamma/2)\sum_j |c\rangle_j \langle c| \\ &+ \sum_x \left(\Omega/2\, e^{i\omega_{dc}t} |c\rangle_x \langle d| + g_g a^\dagger(x)\ |f\rangle_x \langle d| + g_s\, b^\dagger\, |c\rangle_x \langle e| + h.c.\right) \end{aligned} \tag{1}$$

Here, c is the speed of light, k is the wavenumber of the signal (free-space) field, ω_c is the cavity frequency and κ is the decay rate of the cavity. The electric field operators for the signal (free-space) and cavity fields can be written as $\hat{\mathcal{E}}_s(x) = \sqrt{\frac{\hbar c k_0}{\epsilon_0 V}} a(x)$ and $\hat{\mathcal{E}}_c = \sqrt{\frac{\hbar\omega_c}{\epsilon_0 V}} b$, where $a(x) = N^{-1/2}\sum_k e^{ikx} a_k$ and b are bosonic annihilation operators, ck_0 is the center frequency of the signal field, and V is the quantization volume. Additionally, $\mathcal{E}$ is the amplitude of the cavity input field, $\omega_{\mu\nu}$ is the atomic transition energy between states μ and ν, Ω is the classical Rabi frequency for the coupling field, Γ is the linewidth of the excited states $|d\rangle$ and $|e\rangle$, γ is decoherence rate of two stable ground states $|f\rangle$ and $|c\rangle$, and g_s, g_c are the bare couplings of the atomic transition to the two fields. We take the gate and signal fields to be resonant with the atoms so that $ck_0 = \omega_{fd}$ and $\omega_c = \omega_{ce}$.

The use of this effective Hamiltonian is sufficient to describe the steady state for the case of weak coherent state input fields $g_s\langle a\rangle \ll \Omega^2/\Gamma$ and $g_c\langle b\rangle \ll \kappa$. In this limit, we can take the approach of [Ref. [1]] to calculate the two-time correlation function between the fields

$$g^{(2)}(x,\tau) = \frac{\langle b^\dagger(t) a^\dagger(x,t+\tau) a(x,t+\tau) b(t)\rangle}{\langle a^\dagger(x,t) a(x,t)\rangle\langle b^\dagger(t) b(t)\rangle} \tag{2}$$

In this limit, we also can write the density matrix as a product state $\rho = |\chi(\tau)\rangle\langle\chi(\tau)|$ and truncate the available states in the system at the level of two excitations from the state with zero photons and all atoms in $|f\rangle$, which we refer to as $|f,0,0\rangle$. The one-excitation states are $|f,1_x,0\rangle = a^\dagger(x)|f,0,0\rangle$, $|f,0,1\rangle = b^\dagger|f,0,0\rangle$, $|c_x,0,0\rangle \equiv \sigma^x_{cf}|f,0,0\rangle$, and $|d_x,0,0\rangle \equiv \sigma^x_{df}|f,0,0\rangle$, where $\sigma^x_{\mu\nu} \equiv |\mu\rangle_x\langle\nu|$. The two-excitation states that are relevant for $g^{(2)}$ are $|f,1_x,1\rangle \equiv a^\dagger(x)|f,0,1\rangle$, $|c_x,0,1\rangle \equiv b^\dagger|c_x,0,0\rangle$, $|d_x,0,1\rangle \equiv b^\dagger|d_x,0,0\rangle$, and $|e_x,0,0\rangle \equiv \sigma^x_{ec}|c_x,0,0\rangle$.

We then expand $|\chi(t)\rangle$ in these states and find the evolution according to $i\frac{d|\chi\rangle}{dt} = H_{eff}|\chi\rangle$ applying the boundary condition that the free space input field is a weak coherent state. The only terms in H_{eff} which create excitations are the driving fields, which are perturbative implying that the amplitude of the one-excitation states are proportional to $\mathcal{E}$ and the two-excitation amplitudes are proportional to $\mathcal{E}^2$.

To calculate $g^{(2)}(\tau)$ we take the picture where the detection corresponds to a quantum jump from the steady state $|\chi_{ss}\rangle$ into the state $a(x,t)|\chi_{ss}\rangle$ for $\tau < 0$ and $b(t)|\chi_{ss}\rangle$ for $\tau > 0$ [Ref. [1]]. To find $g^{(2)}(\tau)$ we can then simply evolve the operator $n_s(t)$ or $n_c(x,t)$ for a time τ under H_{eff} starting from the jump state.

To find the steady state we expand $|\chi(t)\rangle$ in the zero-, one- and two- excitation states

$$\begin{aligned} |\chi(x,t)\rangle = &|f,0,0\rangle + A^1_0(x)|f,1_x,0\rangle + A^1_1(x)|c_x,0,0\rangle + A^1_2(x)|d_x,0,0\rangle + A^1_3|f,0,1\rangle \\ &+ A^2_1(x)|f,1_x,1\rangle + A^2_2(x)|c_x,0,1\rangle + A^2_3(x)|d_x,0,1\rangle + A^2_4(x)|e_x,0,0\rangle \end{aligned} \tag{3}$$

where we neglect the one- and two- excitation states in the normalization because they are perturbative. The equations of motion are for the A_i^j are found from $i\frac{d|\chi\rangle}{dt} = H_{eff}\,|\chi\rangle$.

$$(\partial_t + c\,\partial_x)A_0^1(x) = -ig_s\sqrt{N}\,A_2^1(x), \tag{4}$$

$$\partial_t A_2^1(x) = -\Gamma/2A_2^1(x) - ig_s\sqrt{N}\,A_0^1(x) - i\Omega/2A_1^1(x), \tag{5}$$

$$\partial_t A_1^1(x) = -\gamma/2A_1^1(x) - i\Omega/2A_2^1(x), \tag{6}$$

$$\partial_t A_3^1 = -\kappa/2A_3^1 + \mathcal{E}, \tag{7}$$

$$(\partial_t + c\,\partial_x)A_1^2(x) = -\kappa/2A_1^2(x) + \mathcal{E}A_0^1(x) - ig_s\sqrt{N}A_3^2(x), \tag{8}$$

$$\partial_t A_3^2(x) = -(\Gamma+\kappa)/2A_3^2(x) - ig_s\sqrt{N}A_1^2(x) - i\Omega/2A_2^2(x) + \mathcal{E}A_2^1(x), \tag{9}$$

$$\partial_t A_2^2(x) = -(\kappa+\gamma)/2A_2^2(x) - ig_cA_4^2(x) - i\Omega/2A_3^2(x) + \mathcal{E}A_1^1(x), \tag{10}$$

$$\partial_t A_4^2(x) = -\Gamma/2\,A_4^2(x) - ig_cA_2^2(x) \tag{11}$$

These eight equations are the only ones relevant for $g^{(2)}(t)$, they give the steady state

$$\bar{A}_0^1(x) = \alpha\exp\left(-\frac{2g_s^2N}{\Gamma+\Omega^2/\gamma}\frac{x}{c}\right) = \alpha\exp\left(-\frac{\mathcal{N}}{2(1+\Omega^2/\gamma\Gamma)}\frac{x}{L}\right) \tag{12}$$

$$\bar{A}_1^1(x) = -\frac{2g_s\sqrt{N}\,\Omega}{\Omega^2+\gamma\Gamma}\bar{A}_0^1(x), \tag{13}$$

$$\bar{A}_2^1(x) = -\frac{i2g_s\sqrt{N}}{\Gamma+\Omega^2/\gamma}\bar{A}_0^1(x), \tag{14}$$

$$\bar{A}_3^1 = \frac{\mathcal{E}}{\kappa/2}, \tag{15}$$

$$\frac{\bar{A}_1^2(x)}{\bar{A}_0^1(x)\bar{A}_3^1} = \frac{1}{1+\eta} + \frac{\eta}{1+\eta}\exp\left(-\frac{\mathcal{N}}{2\zeta}\frac{x}{L}\right) + \mathcal{O}(\kappa/\Gamma) \tag{16}$$

where α is the amplitude of the input coherent state, N is the number of atoms, $\mathcal{N} = 4g_s^2NL/c\Gamma$ is the optical depth, L is the length of the medium, $\eta = 4g_c^2/\kappa\Gamma$ is the cooperativity, and we have defined

$$\zeta = \left(1+\frac{\gamma\Gamma}{\Omega^2}\right)\left(1+\frac{\Omega^2/\kappa\Gamma+\gamma/\kappa}{1+\eta}\right) \tag{17}$$

a correction factor that accounts for finite γ and cooperativity η.

When $\tau < 0$ the free space photon is detected first leading to a quantum jump into the state

$$|\chi_J\rangle = \frac{a(L,\tau)\,|\chi_{ss}\rangle}{\sqrt{\langle\chi_{ss}|\,a^\dagger(L,\tau)a(L,\tau)\,|\chi_{ss}\rangle}} = |f,0,0\rangle + \frac{\bar{A}_1^2(L)}{\bar{A}_0^1(L)}\,|f,0,1\rangle \tag{18}$$

Now

$$g^{(2)}(\tau) = \frac{\langle\chi_J(t)|\,b^\dagger(\tau)b(\tau)\,|\chi_J(t)\rangle}{\langle\chi_{ss}|\,b^\dagger(\tau)b(\tau)\,|\chi_{ss}\rangle} = \frac{|\tilde{A}_3^1(t)|^2}{\left|\bar{A}_3^1\right|^2} \tag{19}$$

$$= \left[1-\left(1-e^{-\mathcal{N}/2\zeta}\right)\frac{\eta}{1+\eta}e^{-\kappa_<|\tau|/2}\right]^2 \tag{20}$$

where $\kappa_< = \kappa$ and $\tilde{A}_3^1(t)$ is found from Eq. 7 with the initial condition $\tilde{A}_3^1(0) = \bar{A}_1^2(L)/\bar{A}_0^1(L)$.

For $\tau > 0$ the procedure is the same, except we have to evolve Eqs. 4-6 starting from the initial conditions $A_0^1(x,\tau) = \bar{A}_1^2(x)$, $A_1^1(x,\tau) = \bar{A}_2^2(x)$, and $A_2^1(x,\tau) = \bar{A}_3^2(x)$. This corresponds to the state $|\chi_J\rangle \propto b\,|\chi_{ss}\rangle$. The result can be expressed in the same form as Eq. 19 with $\kappa_<$ replaced by $\kappa_> = \Omega^2/\Gamma + \gamma$.

SCHEME TO PRODUCE POLARIZATION-ENTANGLED STATES

Continuous entanglement of two light beams can be achieved in the following way: prepare the atomic ensemble in the $|F=3, m_F=3\rangle$ and $|F=3, m_F=-3\rangle$ states with equal population in a small magnetic field oriented at small angle to the cavity mode, and apply the signal beam along the magnetic field. The scheme we demonstrate in the manuscript then works independently for both circular polarizations, so that $|\sigma_+\rangle_s |\sigma_+\rangle_c$ (the subscript indicates the signal and cavity mode, respectively, and $\sigma_\pm$ indicates circular photon polarization) becomes $|\sigma_+\rangle_s |0\rangle_c$ or $|0\rangle_s |\sigma_+\rangle_c$, $|\sigma_-\rangle_s |\sigma_-\rangle_c$ becomes $|\sigma_-\rangle_s |0\rangle_c$ or $|0\rangle_s |\sigma_-\rangle_c$, and the states $|\sigma_+\rangle_s |\sigma_-\rangle_c$ and $|\sigma_-\rangle_s |\sigma_+\rangle_c$ are unchanged. When linearly polarized beams are sent into both the signal and cavity paths and we post-select for detecting a photon in each mode, the output is the entangled (Bell) state $\frac{1}{\sqrt{2}}\left(|\sigma_-\rangle_s |\sigma_+\rangle_c + |\sigma_+\rangle_s |\sigma_-\rangle_c\right)$.

[1] H. J. Carmichael, R. J. Brecha, and P. R. Rice, Optics Communications **82**, 73 (1991).