1 Theory of homodyne intensity autocorrelation measurement

1.1 Second-order correlation function

In our experiment, we follow the proposals of Refs. [1, 2] and perform a homodyne intensity autocorrelation measurement. For this, we collect part of the resonance fluorescence (RF) field, combine it with a local oscillator at a beam-splitter and perform an autocorrelation measurement on one of its outputs. Using equations 1 and 2 of the main text, we obtain the following term which appears second order in E_{LO} :

$$G_{2}^{(2)}(t,\tau,\phi) = E_{\rm LO}^{2} \left(V^{2} e^{2i\phi} \left\langle \hat{E}_{\rm RF}^{(-)}(t) \hat{E}_{\rm RF}^{(-)}(t+\tau) \right\rangle \right) + V^{2} e^{-2i\phi} \left\langle \hat{E}_{\rm RF}^{(+)}(t+\tau) \hat{E}_{\rm RF}^{(+)}(t) \right\rangle + V^{2} \left\langle \hat{E}_{\rm RF}^{(-)}(t+\tau) \hat{E}_{\rm RF}^{(+)}(t) \right\rangle + V^{2} \left\langle \hat{E}_{\rm RF}^{(-)}(t) \hat{E}_{\rm RF}^{(+)}(t+\tau) \right\rangle + \left\langle \hat{E}_{\rm RF}^{(-)}(t+\tau) \hat{E}_{\rm RF}^{(+)}(t+\tau) \right\rangle + \left\langle \hat{E}_{\rm RF}^{(-)}(t) \hat{E}_{\rm RF}^{(+)}(t) \right\rangle \right)$$
(1)

where V is the interferometer visibility which includes spatial mode overlap and the effect of low count rates. From then on, we will consider resonance fluorescence in the stationary regime (i.e. continuous laser excitation), where $G_2^{(2)}$ only has a (τ, ϕ) -dependence, and show how it can be related to the normally ordered auto-correlation of the quadrature standard deviation, $\langle : \Delta \hat{X}_{\phi}(0) \Delta \hat{X}_{\phi}(\tau) : \rangle$.

First, we define:

$$\Delta G_j^{(2)}(\tau,\phi) = G_j^{(2)}(\tau,\phi) - \lim_{\tau \to \infty} G_j^{(2)}(\tau,\phi)$$
(2)

With:

$$\lim_{\tau \to \infty} G_2^{(2)}(\tau, \phi) = E_{\rm LO}^2 \left(V^2 e^{2i\phi} \left\langle \hat{E}_{\rm RF}^{(-)} \right\rangle^2 + V^2 e^{-2i\phi} \left\langle \hat{E}_{\rm RF}^{(+)} \right\rangle^2 + 2V^2 \left\langle \hat{E}_{\rm RF}^{(-)} \right\rangle \left\langle \hat{E}_{\rm RF}^{(+)} \right\rangle + 2V^2 \left\langle \hat{E}_{\rm RF}^{(-)} \hat{E}_{\rm RF}^{(+)} \right\rangle \right)$$

$$(3)$$

Subtracting the long-delay auto-correlations yields:

$$\Delta G_{2}^{(2)}(\tau,\phi) = V^{2} E_{\rm LO}^{2} \left[\left(e^{2i\phi} \left\langle \hat{E}_{\rm RF}^{(-)}(0) \hat{E}_{\rm RF}^{(-)}(\tau) \right\rangle + e^{-2i\phi} \left\langle \hat{E}_{\rm RF}^{(+)}(\tau) \hat{E}_{\rm RF}^{(+)}(0) \right\rangle \right. \\ \left. + \left\langle \hat{E}_{\rm RF}^{(-)}(\tau) \hat{E}_{\rm RF}^{(+)}(0) \right\rangle + \left\langle \hat{E}_{\rm RF}^{(-)}(0) \hat{E}_{\rm RF}^{(+)}(\tau) \right\rangle \right) \\ \left. - \left(e^{2i\phi} \left\langle \hat{E}_{\rm RF}^{(-)} \right\rangle^{2} + e^{-2i\phi} \left\langle \hat{E}_{\rm RF}^{(+)} \right\rangle^{2} + 2 \left\langle \hat{E}_{\rm RF}^{(-)} \right\rangle \left\langle \hat{E}_{\rm RF}^{(+)} \right\rangle \right) \right]$$

$$(4)$$

Defining $\hat{X}_{\phi} = \frac{1}{2|\epsilon|} (e^{i\phi} \hat{E}_{RF}^{(-)} + e^{-i\phi} \hat{E}_{RF}^{(+)})$, we get: $\Delta G_2^{(2)}(\tau, \phi) = 4V^2 E_{\text{LO}}^2 |\epsilon|^2 \left(\left\langle : \hat{X}_{\phi}(0) \hat{X}_{\phi}(\tau) : \right\rangle - \left\langle \hat{X}_{\phi} \right\rangle^2 \right)$ $= 4V^2 E_{\text{LO}}^2 |\epsilon|^2 \left\langle : \Delta \hat{X}_{\phi}(0) \Delta \hat{X}_{\phi}(\tau) : \right\rangle$ (5)

The colons $\langle : ...: \rangle$ denote normal ordering of field operators. The Fourier transform of $\langle : \Delta \hat{X}_{\phi}(0)\Delta \hat{X}_{\phi}(\tau) : \rangle$ gives the squeezing spectrum of resonance fluorescence, while the value at $\tau = 0$ gives the stationary squeezing of the resonance fluorescence field. We remark that, in our notation, $|\epsilon|^2$ corresponds to the product of the radiative decay rate and the total collection and detection efficiency. For the normalization of the measured correlations, we use the detected RF count rate divided by ρ_{22} to obtain $|\epsilon|^2$. From an

experimental point of view, Eqn. 5 shows that for a given integration time, the measurement of one quadrature $(\Delta G_2^{(2)}(\tau, \phi))$ will show a signal-to-noise ratio (SNR) comparable to the SNR obtained in a measurement of photon antibunching.

1.2 Extraction of second-order term from the raw data

The total correlation function $\Delta G_{\text{total}}^{(2)}$ contains terms from zeroth- to fourth-order in E_{LO} . In this work, we are solely interested in the second-order term which, as shown in the previous section, directly gives the normally ordered quadrature variances. $G_3^{(2)}(\phi)$ and $G_4^{(2)}(\phi)$ are time independent and are removed by taking $\Delta G_{\text{total}}^{(2)}$. The zeroth-order contribution is independent of LO power, and we can straightforwardly remove its contribution by taking a reference correlation measurement with the LO path blocked and subtracting it from the measured $\Delta G_{\text{total}}^{(2)}$. The first-order term can be shown to be purely 2π periodic:

$$\Delta G_1^{(2)}(\phi) = 2V E_{LO} \left\langle : \Delta \hat{E}_{RF} \Delta \hat{I}_{RF} : \right\rangle \Big|_{\phi=0} \cos(\phi) \tag{6}$$

We use this periodicity, and the fact that all other terms are either phase-independent or π periodic to extract the first order term from the measured correlation function as follows:

$$\Delta G_1^{(2)}(t,\phi) = \frac{\Delta G_{\text{total}}^{(2)}(t,\phi=0) - \Delta G_{\text{total}}^{(2)}(t,\phi=\pi)}{2} \cos(\phi)$$
(7)

The data plotted in Figure 2 of the main text is then:

$$\Delta g_2^{(2)}(t,\phi) = \frac{\Delta G_{\text{total}}^{(2)}(t,\phi) - \Delta G_0^{(2)}(t,\phi) - \Delta G_1^{(2)}(t,\phi)}{4V^2 E_{LO}^2 |\epsilon|^2} = \left\langle : \Delta \hat{X}_{\phi}(0) \Delta \hat{X}_{\phi}(t) : \right\rangle$$
(8)

2 Wigner Functions

We consider the steady state atomic density matrix under exactly resonant excitation[3]:

$$\rho_{QD} = \begin{pmatrix} \frac{\Gamma^2 + |\Omega|^2}{\Gamma^2 + 2|\Omega|^2} & \frac{i\Omega^*\Gamma}{\Gamma^2 + 2|\Omega|^2} \\ \frac{-i\Omega\Gamma}{\Gamma^2 + 2|\Omega|^2} & \frac{|\Omega|^2}{\Gamma^2 + 2|\Omega|^2} \end{pmatrix}$$
(9)

where Γ is the radiative decay rate of the excited state population and Ω is the Rabi frequency which is given by $|\Omega| = \Gamma \sqrt{s/2}$, where $s = P/P_{\text{sat}}$ is the saturation parameter used in the main text. In the far field limit, we have a simple relation between the source-field and the atomic operator [3], giving: $\hat{a}(t) \propto \hat{\sigma}_{-}(t - \frac{r}{c})$. From this, we can write down a heuristic expression for the density matrix of the photon by mapping the steady state atomic density operator onto the $\{ |0\rangle, |1\rangle \}$ subspace of the outgoing field. For illustrative purposes, we furthermore rewrite the density matrix as a mixture of vacuum and a pure superposition state:

$$\rho_{\rm RF} = A \left| \Psi \right\rangle \left\langle \Psi \right| + B \left| 0 \right\rangle \left\langle 0 \right| \tag{10}$$

where:

$$\begin{split} \Psi \rangle &= \alpha \left| 0 \right\rangle + \beta \left| 1 \right\rangle \\ A &= \frac{\Gamma^2 + \left| \Omega \right|^2}{\Gamma^2 + 2 \left| \Omega \right|^2} \\ B &= \frac{\left| \Omega \right|^2}{\Gamma^2 + 2 \left| \Omega \right|^2} \\ \alpha &= \frac{\Gamma}{\Gamma + i \left| \Omega \right|} \\ \beta &= \frac{-i\Omega}{\Gamma + i \left| \Omega \right|} \end{split}$$
(11)

The Wigner function of the full state is then:

$$W_{\rho_{\rm RF}}(x,p) = AW_{|\Psi\rangle\langle\Psi|}(x,p) + BW_{|0\rangle\langle0|}(x,p)$$
(12)

where x and p are dimensionless conjugate variables corresponding in our case to electric field quadratures, and the Wigner function is defined as follows [4]:

$$W_{|\Psi\rangle\langle\Psi|}(x,p) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dy e^{-ipy} \Psi^*(x-\frac{y}{2})\Psi(x+\frac{y}{2})$$
(13)

For a n-photon Fock state, the position representation is:

$$\Psi_n(x) = \frac{1}{\pi^{\frac{1}{4}}} e^{-\frac{x^2}{2}} H_n(x) \tag{14}$$

Where $H_n(x)$ is the *n*th Hermite polynomial. Combining equations 11,13 and 14, we obtain:

$$W_{|\Psi\rangle\langle\Psi|}(x,p) = |\alpha|^2 W_{|0\rangle\langle0|} + |\beta|^2 W_{|1\rangle\langle1|} + \frac{4}{\pi} \Re(\alpha^*\beta(x-ip))e^{-x^2-p^2}$$
(15)

where $W_{|n\rangle\langle n|}$ is the *n*-photon Fock state Wigner function which is in general phase-invariant:

$$W_{|n\rangle\langle n|} = \frac{(-1)^n}{\pi} e^{-p^2 - x^2} L_n \left[2(p^2 + x^2) \right]$$
(16)

Here, $L_n[x]$ is the *n*-th order Laguerre polynomial. From equation 15 it is clear that all phase dependence in the steady state of resonance fluorescence arises from the third term, proportional to the generated steady-state atomic coherence. From inspection of equations 9, 11 and 12, it follows that the mixedness of the steady state increases and the phase dependence is consequently lost as the Rabi frequency increases. This explains qualitatively the power dependence shown in the inset of Fig. 2 of the main text.

3 Theoretical power dependence for Figure 3

We can use the correspondence between atomic and field operators in resonance fluorescence to calculate the maximum degree of squeezing as the excitation power is varied.

$$\left\langle : \Delta \hat{E}^{2} : \right\rangle = \left\langle : \hat{E}^{2} : \right\rangle - \left\langle \hat{E} \right\rangle^{2}$$
$$= \left\langle \hat{E}^{+2} \right\rangle e^{+2i\phi} + \left\langle \hat{E}^{-2} \right\rangle e^{-2i\phi}$$
$$+ 2 \left\langle \hat{E}^{-} \hat{E}^{+} \right\rangle - \left\langle \hat{E}^{+} e^{+i\phi} + \hat{E}^{-} e^{-i\phi} \right\rangle^{2}$$
(17)

Using the relation between atomic and field operators:

$$\hat{E}^{-} = |\epsilon|\,\hat{\sigma}_{+} \tag{18}$$

Where $|\epsilon|^2$ is the product of the radiative decay rate with the collection and detection efficiency. We then obtain:

$$\left\langle :\Delta \hat{E}^{2}:\right\rangle = |\epsilon|^{2} \left(\left\langle \hat{\sigma}_{-}^{2} \right\rangle e^{+2i\phi} + \left\langle \hat{\sigma}_{+}^{2} \right\rangle e^{-2i\phi} + 2\left\langle \hat{\sigma}_{-} \hat{\sigma}_{+} \right\rangle - \left\langle \hat{\sigma}_{-} \right\rangle^{2} e^{+2i\phi} - \left\langle \hat{\sigma}_{+} \right\rangle^{2} e^{-2i\phi} - 2\left\langle \hat{\sigma}_{-} \right\rangle \left\langle \hat{\sigma}_{+} \right\rangle \right) = |\epsilon|^{2} \left(2 \left(\rho_{22} - |\rho_{12}|^{2} \right) - 2 \left| \rho_{12} \right|^{2} \cos(2\phi) \right)$$
(19)

where we have used $\langle \hat{\sigma}_{-}^2 \rangle = \langle \hat{\sigma}_{+}^2 \rangle = 0$. The minimum and maximum quadrature variances with respect to phase are then:

$$\left\langle : \Delta \hat{X}_{\phi}^{2} : \right\rangle \Big|_{\min} = \frac{1}{2} \left(\rho_{22} - 2 \left| \rho_{12} \right|^{2} \right)$$
$$\left\langle : \Delta \hat{X}_{\phi}^{2} : \right\rangle \Big|_{\max} = \frac{\rho_{22}}{2}$$
(20)

The solid lines in Figure 3 a of the main text are obtained by inserting the steady-state density matrix elements of equation (9) into equation (20). The dashed line is a numerical simulation obtained by solving the Bloch equations and includes phase noise in the interferometer and finite timing resolution of the correlation setup.

References

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