Supplementary Material for: Sub-shot-noise Rydberg EIT spectrum

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Atom-field evolution equations

Following the model proposed in Ref. [1–4], we construct a Heisenberg-Langevin model for a ladder-type three-level system to trace the evolution of squeezed probe light during its interaction with cesium vapor. Atoms have two metastable states, $|2\rangle$ and $|3\rangle$, which interact with two optical fields $\hat{a}(z,t)$ and $\Omega_c(t)$, respectively, as shown in Fig. S1. The field $\hat{a}(z,t)$ is a weak quantum field that couples the ground state $|1\rangle$ and the excited state $|2\rangle$. The $|2\rangle \to |3\rangle$ transition is driven resonantly by a classical coherent coupling light with Rabi frequency Ω_c.

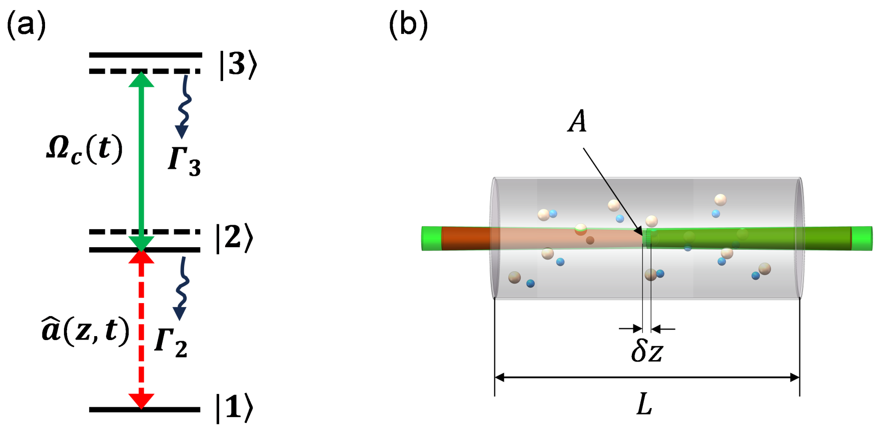

Fig. S1 **(a)** Rydberg EIT level structure. Γ_2 and Γ_3 are the spontaneous emission rate from $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|2\rangle$, respectively. $\hat{a}(z,t)$ is the envelope operator of probe field, and $\Omega_c(t)$ is the coupling light Rabi frequency. **(b)** Schematic diagram of atoms ensemble interacting with laser beams. A is the cross-section area of the beam inside the medium, δz is an infinitesimal slice long the propagation direction. L is the light-atom interaction length, i.e. the effective absorption length.

To perform a quantum analysis of light-atom interactions, the atomic dipole operator $\hat{\sigma}_{\mu\nu}(z,t) = \frac{1}{N_a A\delta z}\sum_{z_k\in\delta z}\hat{\sigma}^j_{\mu\nu}(t)e^{i(\omega_{\mu\nu}/c)(z-ct)}$ at position z in the rotating frame is defined by locally averaging over transverse slices δz containing N atoms. Where $\hat{\sigma}^j_{\mu\nu}(t) = |\mu^j(t)\rangle\langle\nu^j(t)|$ for the jth atom, N_a is the atoms density and A is the cross-section area of the beam. The Hamiltonian of the light-atom interaction after use of the rotating wave approximation can be written as:

$$H = \hbar\begin{bmatrix} 0 & g\hat{a} & 0 \\ g^*\hat{a}^\dagger & \Delta_p & \frac{\Omega_c}{2} \\ 0 & \frac{\Omega_c^*}{2} & \Delta_p+\Delta_c \end{bmatrix} \tag{1}$$

where probe light is described by quantum mechanical operators, the operator $\hat{a}$ is the photon annihilation operator. $g = \wp_{12}\sqrt{\omega_p/2\varepsilon_0 V\hbar}$ is the atom-field coupling constant, $\wp_{12}$ is the atomic dipole moment for the $|1\rangle \to |2\rangle$, ω_p is the angular frequency of the probe light, V is the interaction volume and ε_0 is the permittivity in a vacuum. Δ_p and Δ_c are single-photon detunings of the probe light and the coupling light. Considering the decay term of the atom, etc., the evolution of the system is described using the Heisenberg-Langevin equation:

$$\frac{\partial}{\partial t}\hat{\sigma}_{\mu\nu} = -\frac{i}{\hbar}[H,\hat{\sigma}_{\mu\nu}] + L(\hat{\sigma}_{\mu\nu}) + \hat{F}_{\mu\nu} \tag{2}$$

The $L(\hat{\sigma}_{\mu\nu})$ are the terms produced by decay, dephasing and population exchange:

$$L(\hat{\sigma}_{\mu\nu}) = \begin{bmatrix} -\gamma_c\hat{\sigma}_{11} + (\Gamma_2+\gamma_c)\hat{\sigma}_{33} & -\frac{(\gamma_0+\gamma_c+\Gamma_2)}{2}\hat{\sigma}_{12} & -\frac{(\Gamma_3+2\gamma_0+2\gamma_c)}{2}\hat{\sigma}_{13} \\ -\frac{(\gamma_0+\gamma_c+\Gamma_2)}{2}\hat{\sigma}_{21} & \Gamma_3\hat{\sigma}_{33} - \Gamma_2\hat{\sigma}_{22} & -\frac{(\Gamma_2+\Gamma_3+\gamma_0+\gamma_c)}{2}\hat{\sigma}_{23} \\ -\frac{(\Gamma_3+2\gamma_0+2\gamma_c)}{2}\hat{\sigma}_{31} & -\frac{(\Gamma_2+\Gamma_3+\gamma_0+\gamma_c)}{2}\hat{\sigma}_{32} & \gamma_c\hat{\sigma}_{11} - (\Gamma_3+\gamma_c)\hat{\sigma}_{33} \end{bmatrix} \tag{3}$$

where Γ_2 is the spontaneous emission rate from the intermediate state $|2\rangle$ to the ground state $|1\rangle$, Γ_3 is the spontaneous emission rate from the Rydberg state $|3\rangle$ to the intermediate state $|2\rangle$, and γ_0 is off-diagonal dephasing rate caused by atoms entering and leaving the interaction region. γ_c is population exchange between Rydberg state and ground state, which needs to be considered when atomic pumping is not optimal [3]. We assume that the population exchange from $|1\rangle$ to $|3\rangle$ is same as the population exchange from $|3\rangle$ to $|1\rangle$. The $\hat{F}_{\mu\nu}$ is δ-correlated Langevin noise operators, which represents the noise introduced by radiative decay when atoms are coupled to vacuum modes. The correlation functions can be calculated via the quantum regression theorem [5]:

$$\begin{aligned} \left\langle \hat{F}_{\mu\nu}(z_1,t_1)\hat{F}_{\alpha\beta}(z_2,t_2)\right\rangle = \frac{L}{N}\langle D(\hat{\sigma}_{\mu\nu}\hat{\sigma}_{\alpha\beta}) - D(\hat{\sigma}_{\mu\nu})\hat{\sigma}_{\alpha\beta} \\ -\hat{\sigma}_{\mu\nu}D(\hat{\sigma}_{\alpha\beta})\rangle\,\delta(z_2-z_1)\delta(t_2-t_1) \end{aligned} \tag{4}$$

The Dirac δ function represents the shot memory of the reservoir of vacuum modes responsible for the Langevin forces. L is the light-atom interaction length. N is the number of atoms per unit volume. The equations of the evolution of atomic operators over time obtained by substituting (1), (3) in (2) together with Maxwell's equations of the evolution of the light field constitute the equations of the system evolution, as follows:

$$\begin{aligned}
\dot{\hat{\sigma}}_{11} &= -\gamma_c\hat{\sigma}_{11} - ig\hat{a}\hat{\sigma}_{21} + ig^*\hat{a}^\dagger\hat{\sigma}_{12} + \Gamma_2\hat{\sigma}_{22} + \gamma_c\hat{\sigma}_{33} + \hat{F}_{11} \\
\dot{\hat{\sigma}}_{12} &= ig\hat{a}(\hat{\sigma}_{11} - \hat{\sigma}_{22}) + i\Delta_p\hat{\sigma}_{12} + i\frac{\Omega_c^*}{2}\hat{\sigma}_{13} - \frac{\gamma_c+\gamma_0+\Gamma_2}{2}\hat{\sigma}_{12} + \hat{F}_{12} \\
\dot{\hat{\sigma}}_{13} &= -ig\hat{a}\hat{\sigma}_{23} + i\frac{\Omega_c}{2}\hat{\sigma}_{12} + i(\Delta_p+\Delta_c)\hat{\sigma}_{13} - (\frac{\Gamma_3}{2} + \gamma_0 + \gamma_c)\hat{\sigma}_{13} + \hat{F}_{13} \\
\dot{\hat{\sigma}}_{21} &= -ig^*\hat{a}^\dagger(\hat{\sigma}_{11} - \hat{\sigma}_{22}) - i\Delta_p\hat{\sigma}_{21} - i\frac{\Omega_c}{2}\hat{\sigma}_{31} - \frac{\Gamma_2+\gamma_c+\gamma_0}{2}\hat{\sigma}_{21} + \hat{F}_{21} \\
\dot{\hat{\sigma}}_{22} &= -ig^*\hat{a}^\dagger\hat{\sigma}_{12} + ig\hat{a}\hat{\sigma}_{21} + i\frac{\Omega_c^*}{2}\hat{\sigma}_{23} - i\frac{\Omega_c}{2}\hat{\sigma}_{32} - \Gamma_2\hat{\sigma}_{22} + \Gamma_3\hat{\sigma}_{33} + \hat{F}_{22}
\end{aligned}$$

$$
\begin{aligned}
\dot{\hat{\sigma}}_{23} &= -ig^*\hat{a}^\dagger\hat{\sigma}_{13} + i\frac{\Omega_c}{2}(\hat{\sigma}_{22}-\hat{\sigma}_{33}) + i\Delta_c\hat{\sigma}_{23} - \frac{\Gamma_2+\Gamma_3+\gamma_0+\gamma_c}{2}\hat{\sigma}_{23} + \hat{F}_{23}\\
\dot{\hat{\sigma}}_{31} &= ig^*\hat{a}^\dagger\hat{\sigma}_{32} - i\frac{\Omega_c^*}{2}\hat{\sigma}_{21} - i(\Delta_p+\Delta_c)\hat{\sigma}_{31} - (\frac{\Gamma_3}{2}+\gamma_c+\gamma_0)\hat{\sigma}_{31} + \hat{F}_{31}\\
\dot{\hat{\sigma}}_{32} &= ig\hat{a}\hat{\sigma}_{31} + i\frac{\Omega_c^*}{2}(\hat{\sigma}_{33}-\hat{\sigma}_{22}) - i\Delta_c\hat{\sigma}_{32} - \frac{\Gamma_2+\Gamma_3+\gamma_c+\gamma_0}{2}\hat{\sigma}_{32} + \hat{F}_{32}\\
\dot{\hat{\sigma}}_{33} &= \gamma_c\hat{\sigma}_{11} - i\frac{\Omega_c^*}{2}\hat{\sigma}_{23} + i\frac{\Omega_c}{2}\hat{\sigma}_{32} - (\Gamma_3+\gamma_c)\hat{\sigma}_{33} + \hat{F}_{33}
\end{aligned}
$$

$$
c\frac{\partial}{\partial z}\hat{a} = igN\hat{\sigma}_{12} \tag{5}
$$

In order to solve the propagation equations, we usually assume that the Rabi frequency of the quantized probe field is much lower than that of the classical coupling field, and that the photon number density in the quantized field is much lower than the atomic number density. In this case, the probe field can be regarded as the disturbance field in the EIT line. To explore the propagation behavior of probe photons in a medium, we use the Heisenberg-Langevin equation and the Maxwell-Schrödinger equation to solve the following first-order perturbation equation:

$$
\begin{aligned}
\dot{\hat{\sigma}}_{12} &= ig\hat{a}(\hat{\sigma}_{11}-\hat{\sigma}_{22}) + i\Delta_p\hat{\sigma}_{12} + i\frac{\Omega_c^*}{2}\hat{\sigma}_{13} - \frac{\gamma_c+\gamma_0+\Gamma_2}{2}\hat{\sigma}_{12} + \hat{F}_{12}\\
\dot{\hat{\sigma}}_{13} &= -ig\hat{a}\hat{\sigma}_{23} + i\frac{\Omega_c}{2}\hat{\sigma}_{12} + i(\Delta_p+\Delta_c)\hat{\sigma}_{13} - (\frac{\Gamma_3}{2}+\gamma_0+\gamma_c)\hat{\sigma}_{13} + \hat{F}_{13}
\end{aligned}
$$

$$
c\frac{\partial}{\partial z}\hat{a} = igN\hat{\sigma}_{12} \tag{6}
$$

Fourier transform the above three sets of equations into the frequency domain via

$$
\hat{F}(z,\omega) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{+\infty}\hat{F}(z,t)e^{i\omega t}dt \tag{7}
$$

The average field expansion method is used, that is, the operator is expressed as the steady-state average of the operator plus the fluctuation term. Solving 6 can obtain the Fourier form of the detector light annihilation operator.

$$
\frac{d}{dz}\hat{a}(z,\omega) = -\chi(\omega)\hat{a}(z,\omega) + \frac{gN}{c}\hat{F}(z,\omega) \tag{8}
$$

where

$$
\begin{aligned}
\chi(\omega) &= \frac{|g|^2N}{c}\frac{2(-((\Gamma_3+2\gamma_0+2\gamma_c-2i(\Delta_p+\Delta_c+\omega)))\langle\hat{\sigma}_{11}-\hat{\sigma}_{22}\rangle + i\Omega_c^*\langle\hat{\sigma}_{23}\rangle)}{(\gamma_0+\gamma_c+\Gamma_2-2i(\Delta_p+\omega))(\Gamma_3+2\gamma_0+2\gamma_c-2i(\Delta_p+\Delta_c+\omega))+|\Omega_c|^2}\\
\hat{F}(z,\omega) &= \frac{(2i\Gamma_3+4i\gamma_0+4i(\Delta_p+\Delta_c+\omega+i\gamma_c))\hat{F}_{12}-2\Omega_c^*\hat{F}_{13}}{(\gamma_0+\gamma_c+\Gamma_2-2i(\Delta_p+\omega))(\Gamma_3+2\gamma_0+2\gamma_c-2i(\Delta_p+\Delta_c+\omega))+|\Omega_c|^2}
\end{aligned} \tag{9}
$$

Integrating z can obtain the output field after interacting with the atom medium.

$$
\hat{a}(L,\omega) = e^{-\chi(\omega)L}\hat{a}(0,\omega) + \frac{gN}{c}\int_0^L e^{-\chi(\omega)(L-s)}\hat{F}(s,\omega)ds \tag{10}
$$

Solving the steady-state average value of the atomic operator can obtain the $\langle\hat{\sigma}_{11}\rangle$, $\langle\hat{\sigma}_{22}\rangle$ and $\langle\hat{\sigma}_{33}\rangle$ and the off-diagonal term $\langle\hat{\sigma}_{23}\rangle$, and then obtain the analytical solution of $\hat{a}(L,\omega)$. For the convenience of calculation, in the process of solving the above system of equations, we make the following approximation, solve the first order of $\hat{a}$, Γ_2, Γ_3, γ_c and γ_0, and assume that $\Omega_c^2 \gg (\Gamma_3^2, \Gamma_3\gamma_c, \Gamma_3\gamma_0)$, $\Gamma_2^2 \gg (\Gamma_3^2, \Gamma_3\gamma_c, \Gamma_3\gamma_0)$ and $\Delta_p = \Delta_c = 0$. Thus,

$$
\langle\hat{\sigma}_{11}\rangle = 1 - 2\frac{\gamma_c}{\Gamma_2}, \langle\hat{\sigma}_{22}\rangle = \frac{\gamma_c}{\Gamma_2}, \langle\hat{\sigma}_{33}\rangle = \frac{\gamma_c}{\Gamma_2}, \langle\hat{\sigma}_{23}\rangle = i\frac{\gamma_c}{\Omega_c}. \tag{11}
$$

Introduce the quadrature amplitude and phase operators of the probe light, defined as $\hat{X}_1(L,\omega) = \hat{a}(L,\omega)+\hat{a}^+(L,-\omega)$ and $\hat{X}_2(L,\omega) = -i[\hat{a}(L,\omega)-\hat{a}^+(L,-\omega)]$. The power spectrum $S_p(\omega)$ is expressed as:

$$S_p(\omega)\delta(\omega+\omega') = \frac{c}{L}\langle \hat{X}(L,\omega)\hat{X}(L,\omega')\rangle \tag{12}$$

The non-zero terms of the Langevin noise operator correlation equation are as follows:

$$\begin{aligned}
\langle \hat{F}_{13}^{\dagger}(z1,\omega1)\hat{F}_{13}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}[\gamma_c\langle\hat{\sigma_{11}}\rangle + (2\gamma_0+\gamma_c)\langle\hat{\sigma}_{33}\rangle] \\
\langle \hat{F}_{13}(z1,\omega1)\hat{F}_{13}^{\dagger}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}[(\Gamma_3+2\gamma_0+\gamma_c)\langle\hat{\sigma}_{11}\rangle + \Gamma_2\langle\hat{\sigma}_{22}\rangle + \gamma_c\langle\hat{\sigma}_{33}\rangle] \\
\langle \hat{F}_{12}^{\dagger}(z1,\omega1)\hat{F}_{12}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}[(\gamma_0+\gamma_c)\langle\hat{\sigma}_{22}\rangle + \Gamma_3\langle\hat{\sigma}_{33}\rangle] \\
\langle \hat{F}_{12}(z1,\omega1)\hat{F}_{12}^{\dagger}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}[(\gamma_0+\Gamma_2)\langle\hat{\sigma}_{11}\rangle + \Gamma_2\langle\hat{\sigma}_{22}\rangle + \gamma_c\langle\hat{\sigma}_{33}\rangle] \\
\langle \hat{F}_{12}^{\dagger}(z1,\omega1)\hat{F}_{13}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}(\gamma_0+\gamma_c)\langle\hat{\sigma}_{23}\rangle] \\
\langle \hat{F}_{13}^{\dagger}(z1,\omega1)\hat{F}_{12}(z2,\omega2)\rangle &= \frac{\delta(z_1-z_2)\delta(\omega_1+\omega_2)}{N_a A}(\gamma_0+\gamma_c)\langle\hat{\sigma_{32}}\rangle
\end{aligned} \tag{13}$$

The Fourier spectrum of the output probe light considering the popultion exchange and off- diagonal dephasing can be obtained through (12) and (13). Assuming that the incident probe light spectrum is constant and equal to S_{in} at the analysis frequency, we can obtian:

$$S_p(\omega) = S_{in}\eta(L,\omega) + [1-\eta(L,\omega)](1+N_f) \tag{14}$$

Here $\eta(L,\omega) = e^{-[\chi(\omega)+\chi^*(\omega)]L}$ is the absorption and phase shift of the medium, $\chi(\omega)$ is the susceptibility of the medium

$$\chi(\omega) = \frac{|g|^2 N}{c}\frac{2(2\gamma_0(\Gamma_2-3\gamma_c)+\Gamma_2(\Gamma_A-\gamma_c)-3\gamma_c\Gamma_A)}{\Gamma_2(2\gamma_0^2+\Gamma_3\gamma_c+2\gamma_c^2+\Gamma_2\Gamma_A+\gamma_0\Gamma_B-2i\Gamma_3\omega-6i\gamma_c\omega-4\omega^2+|\Omega_c|^2)} \tag{15}$$

$$N_f = \frac{\gamma_c(-\Gamma_3(\Gamma_3(\Gamma_2+\Gamma_3)+\gamma_0(7\Gamma_2+6\Gamma_3))-4(2\gamma_0+\Gamma_3)\omega^2-(4\gamma_0+2\Gamma_2+\Gamma_3)|\Omega_c|^2)}{2\Gamma_2\gamma_c(\Gamma_3^2+2\omega^2)+3\Gamma_3\gamma_c|\Omega_c|^2-\Gamma_2(\Gamma_3+\gamma_c)|\Omega_c|^2+3\gamma_0\gamma_c\Gamma_C-\gamma_0\Gamma_2(\Gamma_C-5\Gamma_3\gamma_c)} \tag{16}$$

where $\Gamma_A = \Gamma_3+2\gamma_c-2i\omega$, $\Gamma_B = 2\Gamma_2+\Gamma_3+4\gamma_c-6i\omega$, $\Gamma_C = \Gamma_3^2+4\omega^2+2|\Omega_c|^2$. ω is the Fourier frequency. Normalizing Eq. 14 to shot noise

$$S_p(\omega) = S_{PN} + S_{AN} \tag{17}$$

The terms in Eq.17 are the contribution from shot noise (PN), atomic noise (AN). S_1+S_2 stems from two decoherence channels, one is absorption-driven noise $S_1 = (S_{in}-1)\eta(L,\omega)$ arising from vacuum noise coupling and dephasing dissipation, the other is excess noise $S_2 = [1-\eta(L,\omega)]N_f$, generated by atomic population exchange.

For $L = 0.075$ m, $S_{in} = 0.676$ (corresponding to -1.7 dB squeezing), and $\omega = 3$ MHz, we systematically investigate the response characteristics of the output power spectrum S_p to variations in N under different combinations of dephasing γ_0 and population exchange γ_c, as detailed in Fig. S2. When $\gamma_c = 0$, the robustness of S_p against variations in N rapidly deteriorates with increasing γ_0. At $\gamma_0 = 40\Gamma_3$ and $N = 1.2\times10^7$, the noise squeezing advantage in the output field vanishes ($S_p = 0$). Crucially, for $\gamma_c = 0$, no amount of increasing γ_0 can yield $S_p > 0$, i.e., γ_0 only degrades squeezing without introducing excess noise above the shot noise level, as shown in Fig. S2 (a). When $\gamma_0 = 0$, increasing γ_c drives noise level of the output field above the shot noise limit, with larger γ_c values yielding higher S_p (indicating increased noise). This demonstrates that γ_c introduces excess noise into the atomic system. During light-atom interactions, this noise transfers to the optical field, thereby compromising the input field noise squeezing

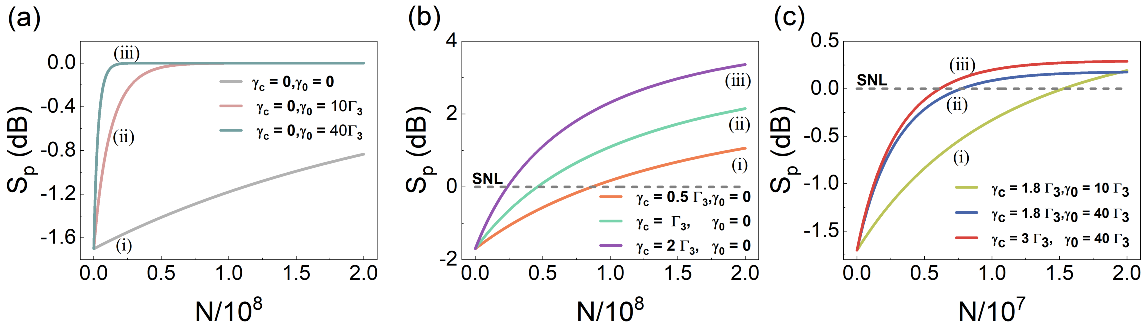

Fig. S2 Power spectrum S_p as a function of number of atoms N in the interaction volume. (a) (i) $\gamma_c = 0$ and $\gamma_0 = 0$. (ii) $\gamma_c = 0$ and $\gamma_0 = 10\Gamma_3$. (iii) $\gamma_c = 0$ and $\gamma_0 = 40\Gamma_3$. **(b)** (i) $\gamma_c = 0.5\Gamma_3$ and $\gamma_0 = 0$. (ii) $\gamma_c = \Gamma_3$ and $\gamma_0 = 0$. (iii) $\gamma_c = 2\Gamma_3$ and $\gamma_0 = 0$. **(c)** (i) $\gamma_c = 1.8\Gamma_3$ and $\gamma_0 = 10\Gamma_3$. (ii) $\gamma_c = 1.8\Gamma_3$ and $\gamma_0 = 40\Gamma_3$. (iii) $\gamma_c = 3\Gamma_3$ and $\gamma_0 = 40\Gamma_3$. Here, SNL represent the shot noise limit, indicated by the dotted line. $\wp_{12} = -5.477a_0e$ and $L = 0.075$ m. The beam radius of probe light is 134 μm and the quantized volume $V = 4.23 \times 10^{-3}\ mm^3$. Coupling light Rabi frequency $\Omega_c/(2\pi)$ is 13.30 MHz. The spontaneous emission rate $\Gamma_2/2\pi = 5.2 \times 10^6$ MHz from intermediate state to the ground state. The spontaneous emission rate $\Gamma_3/2\pi = 3.9 \times 10^3$ kHz from Rydberg state to intermediate state.

advantage while introducing excess noise, as shown in Fig. S2 (b). When both γ_c and γ_0 are non-zero, achieving $S_p < 0$ imposes stricter requirements on N. For $\gamma_c \geq 1.8\Gamma_3$ and $\gamma_0 \geq 10\Gamma_3$, S_p approaches a saturation value as N increases. This indicates that the presence of γ_0 counteracts the unilateral influence of γ_c, as shown in Fig. S2 (c). In summary, there always exists a viable parameter space in which the noise squeezing advantage in the output field can be preserved.

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