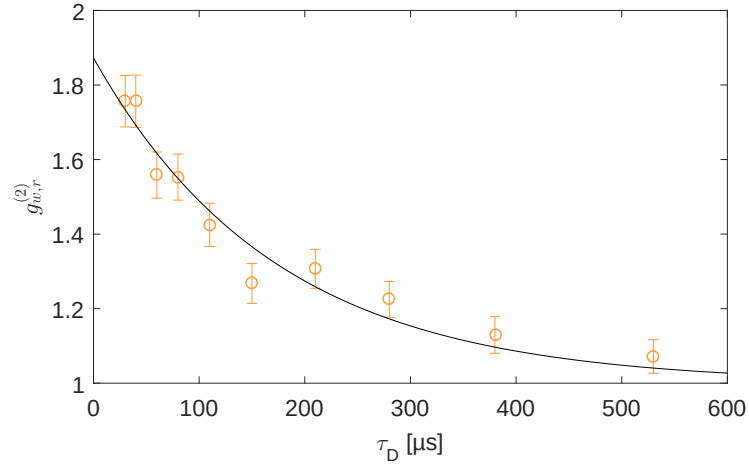


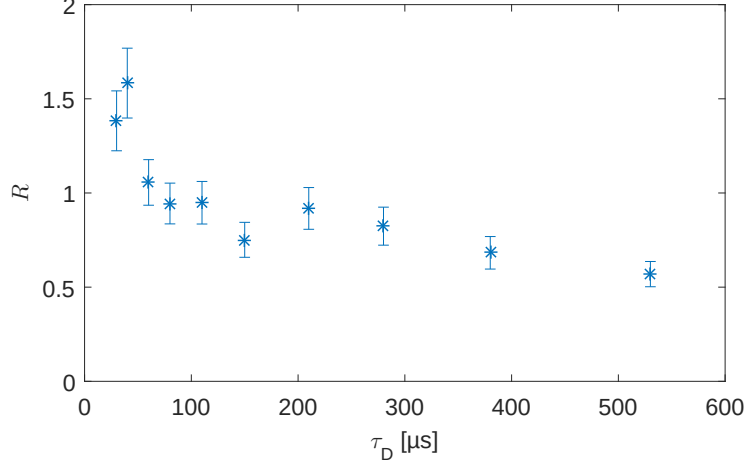
Supplementary Information: Long-lived non-classical correlations towards quantum communication at room temperature

Supplementary Note 1

Correlations decay. Supplementary figures 1 and 2 show the decay of the field correlations. We observe an increase in read leakage noise when the write-read delay increases. This increase is related to the cell cavity birefringence. This results in a faster decay for the observed cross-correlation than for the number of excitations. We can correct for the noise increase by considering the retrieval efficiency as it is done in the main text, thus getting a better estimate for the collective-excitation lifetime.



Supplementary Figure 1: Cross-correlation of write and read detection events versus write-read delay, for $\tau_R = 40 \mu$ s. A fit (line) to the function $g_{w,r}^{(2)}(t) = 1 + C \exp(-t/\tau_g)$ yields a characteristic decay time of $\tau_g = 0.17 \pm 0.02$ ms. Error bars represent statistical uncertainty for the correlations (see Methods in the main text).



Supplementary Figure 2: Cauchy-Schwarz parameter decay versus write-read delay, for $\tau_R = 40 \mu\text{s}$. We observe non-classical correlations ($R > 1$) for $\tau_D < 80 \mu\text{s}$. Error bars represent statistical uncertainty for the correlations (see Methods in the main text).

Supplementary Methods

Read photon shape model. In order to describe the temporal shape of the retrieved photons, we adapt the model from [1]. Here both transitions appearing in fig. 1c in the article are driven simultaneously with the coupling between collective excitations and scattered photons on the heralding (readout) transition described by the coefficient ξ (χ). The simultaneous driving leads to FWM where collective excitations are created and retrieved within the same pulse. The mean number of detected photons as a function of time during the read process is given by:

$$\begin{aligned} \langle \hat{a}_{\text{ro}}^\dagger(t) \hat{a}_{\text{ro}}(t) \rangle = & \chi_r^2 \Omega_R^2(t) \frac{N(t)}{N_0} \cdot \exp \left(t (\xi_r^2 - \chi_r^2) \Omega_R^2(t) \frac{N(t)}{N_0} \right) \cdot n_{\text{ce}} + \quad (1) \\ & \frac{\chi_r^2 \xi_r^2 \Omega_R^2(t)}{\xi_r^2 - \chi_r^2} \frac{N(t)}{N_0} \cdot \left[\exp \left(t (\xi_r^2 - \chi_r^2) \Omega_R^2(t) \frac{N(t)}{N_0} \right) - 1 \right] + \\ & B(\Omega_R^2(t)) + L(t, \Omega_R^2(t)) + C + BG, \end{aligned}$$

where Ω_R is a common Rabi frequency defined for both transitions appearing in fig. 1 (d) in the article, χ_r and ξ_r are reduced coupling coefficients, with the common Rabi frequency factored out. N_0 is the number of atoms initially present in $|g\rangle$, and $N(t) = N_0 e^{-t/T_1}$ accounts for the atomic population decay with a time constant $T_1 \approx 1.1 \text{ ms}$, which we measure independently. n_{ce} is the mean number of collective excitations created by the write pulse. The first term in eq. (1) describes the readout gain for collective excitations present in the ensemble at $t = 0$. When FWM is allowed (non-vanishing ξ_r), excess readout photons are seeded by collective excitations created during the write. The second term describes the FWM noise which is independent of the write process. BG is a fixed offset given by background noise, mostly comprised of

dark counts. $L(t, \Omega_R^2(t))$ is the leakage contribution, which is proportional to the intra-cavity power. The leakage part includes a time-dependency due to birefringence dependent on the atomic population. $B(\Omega_R^2(t))$ represents the spectrally broad contribution that we observed in fig. 2. We expect it to be dependent on the Rabi frequency. The term C is a constant offset that is discussed below. The mean number of photons per pulse after an integration time τ_R is then given by $\langle N_{ro}(\tau_R) \rangle = \int_0^{\tau_R} \langle \hat{a}_{ro}^\dagger(t) \hat{a}_{ro}(t) \rangle dt$.

The coefficients of the model are determined in the following way:

- Background: The background contribution BG is determined from a part of the experimental sequence during which the single-photon detector gate is on and no light is sent to the system.
- Rabi frequency: The common Rabi frequency $\Omega_R(t)$ can be computed from the intra-cavity power of the cell-cavity, obtained via a calibrated photodiode monitoring the cell-cavity transmission and used to lock it. For simplicity, we approximate it to be constant starting from the half-turn on time of the excitation pulse.
- Leakage and broad: In order to estimate the leakage and broad contribution, we realize a fit identical to the one of fig. 2 in the article for consecutive time slices of 32 μs until the entire read detection window is covered - excluding the turn-on and off transients. Each of the fits allows us to obtain a value for the leakage and broad contribution at zero detuning. We observe a clear linear trend for both leakage and broad contribution and thus approximate it with fitting linear functions.
- Number of collective excitations: The number of collective excitations n_{ce} is estimated from the mean number of write detection events. It is corrected for the total detection efficiency (including escape efficiency from the cell cavity), by the write efficiency obtained from the data of fig. 2 in the article, and by the four-wave mixing process during the write, where the write excitation already reads out collective excitations.
- Coupling coefficients: The ratio of the coupling coefficients $\alpha = \xi_r/\chi_r$ is set by the Clebsch-Gordan coefficients at our detuning, and by the cell cavity resonance affecting the coupling strength for the scattering of a photon on each side-band. We neglect time-dependent cell cavity resonance shifts. We determine ξ_r from the write process, where the number of heralding photons for a given Rabi frequency allows us to calculate the coupling ξ_r .

We note that we are required to add an extra constant term C to make the model agree with the data. We emphasize, that this is the only free parameter that we use in the model. We observe a factor $C/(B+C) \approx 0.64 \approx \eta_W$ that is also supported by further data at different powers. This suggests, that the constant term C and broad contribution B have a common origin. For the conditional data we use the same model parameters except fitting the number of collective excitations. This yields $n_{ce} = 0.92$ which agrees with an estimate obtained from using conditional probabilities.

We note, that a few simplifications have been made for the model. Firstly, the cell cavity was not included in the Hamiltonian used to derive the model. Secondly, we neglect decoherence. However, the measured lifetime of 0.27 ms

suggests that decoherence would likely have an effect on the timescale of the read detection window. Thirdly, we do not include any contributions from population residing in other Zeeman states. The latter we believe contributes to the terms B and C , but we lack sufficiently firm evidence to make stringent claims about this.

Supplementary References

- [1] Dąbrowski, M., Chrapkiewicz, R. & Wasilewski, W. Hamiltonian design in readout from room-temperature Raman atomic memory. *Opt. Exp.* **22**, 26076 (2014).