Experimental detection of quantum coherent evolution through the violation of Leggett-Garg inequalities

Zong-Quan Zhou,1,2 Susana F. Huelga*,3 Chuan-Feng Li1,2 and Guang-Can Guo1,2
1Key Laboratory of Quantum Information, University of Science and Technology of China, CAS, Hefei, 230026, China
2Synergetic Innovation Center of Quantum Information and Quantum Physics, University of Science and Technology of China, Hefei, 230026, P.R. China
3Institut für Theoretische Physik, Albert-Einstein-Allee 11, Universität Ulm, D-89069 Ulm, Germany
(Dated: July 8, 2014)

We present an experimental violation of Leggett-Garg-type inequalities (LGI) with delocalized single-photon excitations sustained in millimeter-sized crystals. Through the reversible transfer of photonic polarization states and the collective atomic excitation of two quantum memories, we prepared a superposition state of a delocalized excitation in two crystals separated by a distance of 4 mm. The phase relation between the states in the superposition is governed by the dynamics of a large number of individual atoms and can be controlled with a frequency detuned and polarization dependent atomic frequency comb. By separately benchmarking the Markovian character of the evolution and the translation invariance of the conditional probabilities, the observed violation of a LGI is unambiguously attributed to the quantum coherent character of the process. These results confirm the persistence of quantum features within systems of increasing complexity.

Ever since the birth of quantum mechanics (QM) it has been stressed the difficulty to reconcile the behavior of quantum particles with the intuition stemming from dealing with macroscopic objects, which should occupy definite states at all times and independently of the observers [1]. Different types of inequalities that set lower bounds on the statistical predictions of QM have been proposed in an attempt to test the validity of the quantum mechanical description under conditions where classicality is expected to emerge. Those include Bell inequalities [2], Kochen-Specker’s [3] and the so-called Leggett-Garg inequalities (LGI) [4–6]. In each case, a set of testable inequalities is obtained which are obeyed by classical stochastic theories but violated by QM. Unlike the Bell inequality, which involves statistical correlations between states of two spatially separated systems, LGI are not related to locality properties but concerned with the correlations of the state of a single system at different times (autocorrelations) [4–6]. Introduced as the Bell inequalities in time, the violation of LGI excludes a hidden-variable description based upon the assumptions of macroscopic realism per se (a macroscopic system must at any time be in a definite one of its macroscopically distinct states) and the possibility to perform noninvasive measurements (measurements do not influence the actual state or the subsequent system dynamics of the system). LGI therefore provide a criterion to characterize the boundary between the quantum and the classical domains.

Until now, the experimental violation of LGI has been performed with microscopic objects, such as photons [7, 8], electron or nuclear spins [9–11] and the micrometer-sized superconducting ‘transmon’ system [12]. In practice, the main experimental challenge comes from the implementation of truly non-invasive measurements. However, the use of weak measurements [12] and schemes involving ancillary systems [11] have recently allowed for the first experimental results in the microscopic domain. Alternatively, to avoid the need of performing measurements at intermediate times, as required in the original formulation of the inequalities, the additional assumption of stationarity has been considered [13, 14]. According to this assumption, the conditional probability \( Q_{ij}(t_1, t_2) \) to find a system in state \( j \) at time \( t_2 \), if it is in state \( i \) at time \( t_1 \) only depends on the time difference \( t_2 - t_1 \). When stationarity holds, LG-like inequalities can then be formulated as follows [13],

\[
K_\mp = K(0, 2t) \mp 2K(0, t) \geq -1, \tag{1}
\]

where \( K(t_1, t_2) \) denotes the autocorrelation function defined as \( K(t_1, t_2) = \langle M(t_1)M(t_2) \rangle \) for certain dichotomic observable \( M(t) \). Note that within a realist framework, a macroscopic object which has two available macroscopically distinct states is assumed to be at any time \( t \) in one of these states, and therefore the dichotomic observable \( M(t) \) always produces the value +1 or -1. Inequalities Eqs. (1) are violated by quantum mechanical unitary dynamics and are easily testable by using projective measurements noting that, given the initial condition \( p_i(0) = 1 \), so that the system is prepared initially in state \( i \), we can write [14]

\[
K(0, t) = 2Q_{i,i}(0, t) - 1 \tag{2}
\]

and an analogous expression for the autocorrelation \( K(0, 2t) \). While the assumption of stationarity immediately leads to testable inequalities, it was known to narrow down the class of macrorealist theories which are put to the test [13] and it is only recently that its implications have become fully understood [15]. Provided
that the system can be initialized in a well defined state, stationarity implies the time-translational invariance of the probabilities as well as the Markovianity of the evolution \cite{21,23}. Remarkably, these are assumptions that could be tested independently, as opposed for instance to the situation encountered with standard Bell inequalities, where the so called fair sampling assumption is not directly testable \cite{10}. As a result, if the stationarity condition does hold for the considered experimental set up, we argue that the inequalities of the form Eqs. (1) provide a quantitative way to witness the persistence of coherent effects. Testing the validity of standard quantum theory over larger and larger scales and increasing levels of complexity is important for assessing the relevance of genuinely quantum traits beyond purely microscopic scales \cite{17,19}.

This is the viewpoint we adopt here and present the experimental violation of this form of a LGI by generating and probing a delocalized atomic excitation over macroscopically separated crystals. Our approaches relies on the quantum interface between two solid-sate quantum memories and a single-photon source. A coherent superposition of delocalized states over the macroscopic crystals is produced by heralded absorption of a single photon. The dynamical evolution of the atomic excitation is controlled with polarization dependent atomic frequency comb (AFC) and will be monitored through the violation of LGI of the form Eq. (3). The validity of the stationary assumption in this system is experimentally verified with independent tests to assess both the stationarity of the conditional probabilities and the Markovianity of the evolution. The former can be performed directly by using projective measurement after selected time intervals while the Markovianity will be assessed in terms of the monotonic decrease of the trace distance between quantum states \cite{20}.

The physical system undergoing evolution is composed of about 10^{10} ions, which are spatially distributed over two crystal separated by 4 mm. The experimental sample, which was similar to our memory hardware presented in \cite{21}, is composed of two pieces of Nd^{3+}:YVO_{4} crystals (doping level: 5 ppm, thickness: 3 mm) sandwiching a 45° half-wave plate (HWP). Horizontal (H)- and vertically (V)-polarized photons with wavelength of approximately 880 nm can be independently processed by the first and second crystal, respectively \cite{21,22}. Here, the polarization direction is defined by the crystal’s c-axis.

Polarization dependent AFC. We utilize the atomic frequency comb technique to realize the reversible transfer between photonic polarization states and atomic excitations \cite{21,23,27}. By independently preparing two frequency detuned AFC in the two crystals, we obtained a polarization dependent AFC, which enables us to initialize, control and analyze the collective excitation states in the crystals.

The AFC protocol requires a tailored absorption profile with a series of periodic and narrow absorbing peaks separated by \Delta (see Fig. 1). The single photon input is then collectively absorbed and diffracted by the atomic frequency grating. The atomic state with and without the photon excitation can be represented by \(|e \rangle_{N} = \sum_{j}^{N} C_{j} e^{-i k z_{j}} e^{2 \pi i \delta_{j} t} |g_{1} \cdots e_{j} \cdots g_{N} \rangle \) and \(|g \rangle_{N} = |g_{1} \cdots g_{j} \cdots g_{N} \rangle\), respectively. Here \(N\) is the total number of atoms in the comb; \(|g_{j}\rangle\) and \(|e_{j}\rangle\) represent the ground and excited states, respectively, of atom \(j\); \(z_{j}\) is the position of atom \(j\); \(k\) is the wavenumber of the input field; \(\delta_{j}\) is the detuning of the atom with respect to the laser frequency and the amplitudes \(C_{j}\) depend on the frequency and on the position of atom \(j\). Due to the periodic structure of AFC, \(\delta_{j} \approx m_{j} \Delta\) with \(m_{j}\) integer. With the two AFC of the two crystals prepared with the same periodicity \(\Delta\), when the input photon’s polarization is chosen as \(H + e^{i \varphi} V\), the AFC of the two crystals will be excited simultaneously. The collective excitation in the two crystals can be represented by a coherent superposition of the form \(|\Psi(t)\rangle = 1/\sqrt{2} (|e\rangle_{N} |g\rangle_{N} + |g\rangle_{N} |e\rangle_{N}) e^{i (2 \pi \delta t + \varphi_{0})}\) where \(N(1)\) is the number of involved atoms in the first (second) crystal. \(\delta\) is the frequency detuning between the two AFC, as shown in Fig. 1(a). The two crystals are actually in an entangled state, as already demonstrated in \cite{28}.

To characterize the coherent nature of the delocalized collective atomic state, we define the dichotomic observable \(M(t) = |D\rangle \langle D| - |A\rangle \langle A|\), where the linear state \(|D\rangle = 1/\sqrt{2} (|e\rangle_{N} |g\rangle_{N} + |g\rangle_{N} |e\rangle_{N})\) defines the basis state \(|0\rangle\) with eigenvalue equal to +1 and the orthogonal state \(|A\rangle = 1/\sqrt{2} (|e\rangle_{N} |g\rangle_{N} - |g\rangle_{N} |e\rangle_{N})\) corresponds to basis state \(|1\rangle\) with eigenvalue equal to -1. The atomic state in two crystals can now be written in a compact form as,

\[
|\Psi(t)\rangle = 1/2 (1 + e^{i \varphi}) |D\rangle + 1/2 (1 - e^{i \varphi}) |A\rangle = \cos(\varphi/2) |D\rangle - i \sin(\varphi/2) |A\rangle,
\]

where \(\varphi = 2 \pi \delta t + \varphi_{0}\). To probe the atomic state, we need to collectively transfer the delocalized atomic excitation back into a single photon state. Thanks to the periodic structure of AFC, a strong rephasing echo occurs after a time \(\tau_{r} = 1/\Delta\). The photon is re-emitted in the forward direction as a result of a collective interference between all of the atoms that are in phase. The atomic state \(|D\rangle\) and \(|A\rangle\) corresponds to readout photon polarization of \(H + V\) and \(H - V\), respectively. Since the fidelity of quantum state transfer between single photons and atomic excitation is well above 0.99 as demonstrated in \cite{21}, it is reliable to probe the atomic state by measuring the polarization of the retrieved single photons.

Heralded storage of single photons in polarization dependent AFC. Fig. 2 shows the experimental setup for investigating the evolution of collective atomic excitations in macroscopically separated crystals. It has three different parts: The source of narrow band photon pairs, the laser system for polarization-dependent AFC preparation and the sample with Nd^{3+}:YVO_{4} crystals for the dynamical evolution of the delocalized excitation.

Photon pairs are produced with type-II spontaneous parametric down conversion (SPDC) process in a 20-mm
PPKTP crystal. The non-linear crystal is pumped by pulsed light at 440 nm, yielding degenerate photon pairs with the H-polarized idler photons and the V-polarized signal photons. The initial spectral width of the photons is approximately 150 GHz, which is a factor of 10³ larger than the 100 MHz bandwidth of the AFC quantum memories. To improve the signal-to-noise (SNR) ratio, the photons are filtered by the gratings and the etalon to achieve a bandwidth of approximately 700 MHz. The pulsed operation of the photon source further improves the SNR as a result of the temporal separation of the retrieved photons from the transmitted noise.

The polarization dependent AFC are prepared in two pieces of Nd³⁺:YVO₄ crystals with working wavelength of approximately 880 nm. The pulse sequence for AFC preparation is shown in Fig. 1(b), in which the pump light’s frequency is swept over 100 MHz in 500 µs cycle and its amplitude has been modulated periodically to give an comb structure. The H- and V-polarized light are independently programmed to give two AFC structures with the same periodicity ∆ and the frequency detuning of δ. The signal photons are coherently absorbed by the AFC in the two crystals. The dynamical evolution of the atomic excitation is controlled by the detuning of the two AFC. To analyze the quantum states of the atomic ensembles, the atomic excitation is converted back to photons and measured with polarization dependent single photon detection systems. The readout efficiency of the quantum memories for bandwidth matched laser pulses at different evolution time is shown in Fig. 1(c). The low doped crystals show a better performance of storage efficiency than that of 10-ppm doped crystals (see Methods).

We first verify that the non-classical correlations between signal (s) and idler (i) photons are preserved during the storage and retrieval process. Fig. 3(a) shows an example of storage the $H+V$-polarized single photons and readout with $H−V$ polarizations after 125 ns delay. The frequency detuning of two AFC is set as 5 MHz. Due to mismatched bandwidth of the signal photons and the quantum memories, most of the input photons are transmitted directly through the sample. The extinction ratio for orthogonal polarizations is approximately 1000:1 in our setup, so a small transmission peak still can be seen at $t=0$. A strong echo is emitted after 125 ns. Due to the 100-MHz bandwidth of the AFC, the retrieved photons has a temporal width of approximately 10 ns. The small peak at 250 ns is from the second order echo emission. The cross-correlation function $g_{si}^{(2)}$ (see Methods) is measured in the the H or V polarization basis to avoid polarization rotation caused by the AFC. The measured results are shown in Fig. 3(b). Since the values of $g_{si}^{(2)}$ at all times are significantly larger than the classical bound of 2, any classical description of light is eliminated. The lowest measured $g_{si}^{(2)}$ value is 14.3 after storage time of 250 ns. This allows to upper bound the autocorrelation of the heralded signal photon to $g_{1ss}^{(2)} ≤ 0.27$, which confirms that the single-photon character is well preserved during the storage and retrieval process. We note that all the results presented here are from raw data, without any background subtraction.

Test on the validity of stationary assumption. The derivation of Eqs 14 makes use of the so-called-stationarity assumptions which relies on the condition of time invariance of the conditional probabilities $Q_{ij}$ and the Markovianity of the evolution 15. We now present experimental verification on the validity of stationary assumption in our system.

1. Time translational invariance. The input photon’s polarization is rotated to $H+V$ ($H−V$) by the HWP. To detect the atomic state in $D(A)$ at different readout time $t$, the probe photon state should be set as $H+e^{i\varphi_0}V$ ($H−e^{i\varphi_0}V$) with $\varphi_0 = -2\pi\delta t$ by carefully adjusting the phase plate $\theta$. This photon state is directly mapped to the initial state of atomic excitation. The subsequent system evolution is recorded as the function of the time difference $\tau$. Fig. 4(a) shows the conditional probability $Q_{ij}(t,t+\tau)$ to find a system in state $j$ at time $t+\tau$, if it is in state $i$ at time $t$. It can be seen that $Q_{ij}(t,t+\tau)$ only depends on time difference $\tau$ and show little dependence on the evolution time $t$ within experimental errors, thus the conditional probabilities $Q_{ij}$ are time-translational invariant.

2. Markovianity. The stationarity test is completed by assessing the absence of memory effects in our sys-
FIG. 2: Experimental setup for violation of LGI with delocalized single-photon excitation. The photon pairs are generated in the periodically poled potassium titanyl phosphate (PPKTP) crystal via SPDC. The pump source is obtained by sending the master laser to a frequency doubler. The blue pump light is modulated into pulses by the acousto-optic modulators (AOM1). The pump light is then removed by the dichroic mirror (DM). The photon pairs are spectrally filtered by the gratings and the etalon. The two photons in pair are separated by the polarization beam splitter (PBS). The H-polarized photons are directed to a single mode fiber (SMF)-coupled single photon detector (SPD). The AOM3 and AOM4 generate the H- and V-polarized AFC preparation light for H- and V-polarized AFC preparation, respectively. The AOM2 is used to shift the AFC frequency to match with the center frequency of photon pairs. The signal photons’ polarization is controlled by the polarization beam splitters (PBS), the half-wave plate (HWP) and the phase plate (θ). The AFC preparation light and single photon pulses are combined with beam splitter and collected into a SMF to ensure perfect mode matching. After a programmable delay, the polarization of the retrieved signal photons is analyzed with a quarter-wave plate (QWP), a HWP and a PBS. The single photon signals are analyzed with time interval analyzers (TIA). The phase locked mechanical choppers are used to protect the SPD during the preparation procedure. Further details are provided in the section on Methods.

FIG. 3: (a). Example of storage of the $H + V$ polarized photons in the polarization dependent AFC with readout polarization of $H - V$. Most parts of the transmitted parts are blocked by the PBS. A strong echo is emitted at 125 ns with rotated polarization states. The time-bin size is 2 ns and the integration time is 4 hours. (b). Cross-correlations $g_{\alpha\beta}^{(2)}$ as a function of storage time. The blue dashed lines corresponds to the classical limit $g_{\alpha\beta}^{(2)} = 2$ for two-mode squeezed states.

tem’s evolution. To do so, we evaluate a measure able to detect deviations from Markovian behaviour [29], as provided by the the trace distance $D(\rho_1, \rho_2) = 1/2 tr |\rho_1 - \rho_2|$ between any two states $\rho_1$ and $\rho_2$ [30]. Markovian processes, characterized by the absence of information backflow, tend to continuously reduce the distinguishability of physical states and the trace distance should monotonically decrease during the time evolution. To determine the behaviour of the trace distance in our system, the two initial states are chosen as $H + V$ and $H - V$, which guarantees the optimality of the measure [30], and makes $D(\rho_1(t), \rho_2(t)) = 1$ at $t = 0$. The atomic states are read out after different evolution time $t$. Quantum state tomography [31] is carried out to determine the polarization states of the retrieved signal photons. The measurement basis are chosen as $H, V, H + iV$, and $H + V$ by adjusting the waveplates. In the experiment, the density matrix is estimated by the maximum-likelihood procedure. The trace distance $D(\rho_1(t), \rho_2(t))$ is calculated from the reconstructed density matrix of the two states [30]. The results are shown in Fig. 4.b., illustrating that the trace distance monotonically decreases in our experiment. This completes the test to verify the stationarity assumption.

Violation of LGI in macroscopic quantum memories. We proceed now to demonstrate the violation of the LGI Eqs. (1). Using Eq.(5) as the specification of our quantum state, we can easily obtain the correlation function $K(t) = \cos(2\pi \delta t)$. With the same analysis, $K(2t) = \cos(4\pi \delta t)$. The functions $K_\pm$ in Eqs.(3) can be calculated as $K_\mp = \cos(2\pi \delta t) \mp 2 \cos(2\pi \delta t)$. It can be seen that $K_-$ reaches its minimum value of -1.5 when $2\pi \delta t = \pi/3$ and $K_+$ reaches its minimum value of -1.5 when $2\pi \delta t = 2\pi/3$, which yields to maximal violation of the corresponding inequalities in Eq.(1).

Fig. 5(a) shows the time evolution of the $|\Psi(t)|^2$ with an initial state specified by $|D\rangle$. With the AFC prepared with $\delta$ of 5 MHz, the possibility to find the state $|D\rangle$ or $|A\rangle$ oscillates with a periodicity of 200 ns. In agree-
ment with the results in Fig. 4(b), the selected time scale corresponds to a low noise domain with almost complete population inversion. Based on the measurement results in Fig. 3(b), the evolution of \( K_- \) and \( K_+ \) is shown in Fig. 5(b). The solid lines are theoretical predictions based on Eq. (5). \( K_+ \) reaches \(-1.48 \pm 0.07\) at 62.5 ns, which violates the classical limit of -1 by about 6.9 standard deviations. The user-programmable AFC polarization detuning \( \delta \) determines the phase evolution speed of the atomic state. The violation of LGI should hold for a different \( \delta \), i.e., a different evolution speed. We further measured the envelope evolution of \( K_- \) with a \( \delta \) of 2 MHz. The corresponding atomic state oscillates with a periodicity of 500 ns. \( K_- \) reaches \(-1.41 \pm 0.08\) at 83.3 ns.

As a result, our experiment shows clear deviations from the classical bound \( K_-^{\text{min}} = -1 \). Given that we have independently verified the stationarity of the evolution, the violation of the LGI Eq. (11) is attributed to the coherent nature of the process. In this sense, the presented results address the reverse situation to the one analyzed in [32], where LGI are used as non-Markovianity witnesses for the evolution of microscopic systems.

One could argue that despite the prepared quantum states involve delocalized excitation over a mm scale, attending to the original formulation of Leggett [4–6], the considered states do not have a sufficiently high degree of disconnectivity and therefore their macroscopicity would be arguable. However, the two states \( |D\rangle \) and \( |A\rangle \) differ by a collective phase shared by \( \sim 10^{10} \) ions and it is this phase which dictates whether or not the system behaves as a quantum coherent superposition or as a (classical) statistical mixture. Considering the AFC bandwidth \( \Gamma_{\text{AFC}} = 100 \) MHz and the homogeneous linewidth of Nd\(^{3+} \) ions \( \Gamma_h = 11 \) kHz, we can estimate that at least \( \Gamma_{\text{AFC}} / \Gamma_h \sim 10^4 \) different ions have to contribute to the AFC absorption and the envelope evolutions. The elementary AFC composed of about \( 10^4 \) ions just acts like a diffraction grating in the frequency domain, which governs the evolution of the atomic coherence and leads to a temporal delayed echo emission. To substantially improve macroscopicity, it will be a good choice to combine the atomic memory with the multi-photon entangled source [33–36]. With macroscopic number of excitations...
prepared in crystals, the current scheme should provide a powerful platform for exploring the validity of QM in a macroscopic regime and finding the answer to the question of whether superpositions of macroscopically distinct states in the sense originally envisaged by Leggett are indeed possible.

The present results demonstrate that the dynamics of a collective excitation which is distributed in two macroscopically separated crystals is well described by quantum mechanics. The phase relation between the considered states is governed by a macroscopic number of atoms and therefore the observed interference effects are macroscopic in origin although the interfering states themselves are of low Leggett-disconnectivity. These results confirm the persistence of quantum coherence effects in systems of increasing complexity which can be seen with a naked eye. The polarization dependent AFC technique, which is able to drive the dynamical evolution of atomic excitations, may find further applications in both quantum and classical information processing.

**Methods**

**Setup.** The master laser is a frequency stabilized Ti:Sapphire laser (MBR-110, Coherent) with frequency ν₀ of 340.69656 THz. The frequency of the master laser is stabilized to a low-drift Fabry-Pérot Interferometer (FPI) with PDH technique. The FPI is placed in vacuum housing with temperature controlled to a stability of 5 mK. The linewidth of the master laser is below 50 kHz.

Part of the laser output is fed into a cavity enhanced frequency doubler to generate blue light. The blue light is controlled with an -200-MHz AOM1. The photon pairs are filtered with the optical gratings and the etalon, combined with SMF collection. The signal photons are collected with SMF and ready for use. Another part of the master laser output is used for AFC preparation. The AOM2 with center frequency of -250 MHz is used in double-pass configuration to down shift the laser frequency. The AOM3 and AOM4 have a center frequency of 200 MHz and are also used in double-pass configuration. Thus the center frequency of the AFC preparation matches with the photon source at 340.69646 THz.

The AFC preparation light and single-photon pulses are combined with 10:90 BS and collected with SMF. The light focus to a diameter of 100 μm with a lens (f=250 mm). The sample is placed in a cryostat (Oxford Instruments, SpectromagPT) at a temperature of 1.5 K and with a superconducting magnetic field of 0.3 T in the horizontal direction. The two parallel Nd³⁺:YVO₄ crystals’ c-axes are placed in the horizontal direction. Because each crystal only strongly absorbs H-polarized light, the H-polarized components of input photons are stored in the first crystal, and the V-polarized components are stored in the second crystal after polarization rotation by the HWP. The detection efficiencies for both photons are approximately 35% with 50-Hz dark counts.

The initial absorption depth of the sample is d ≈ 8.0. With optimized spectral tailoring, we can prepare AFC with extremely small background absorption d₀ ≤ 0.05. The Zeeman population relaxation lifetime is measured by spectral-hole burning, T₂ = 42.9 ms in the current crystals, which is 7 times longer than that of 10 ppm doped crystals [22]. With two-pulse photon echoes, we measured an optical coherence time T₂ = 28.4 μs (homogeneous linewidth Γₜ = 11 kHz). Because of the better coherence properties and the more efficient optical pumping achieved in the current sample, the AFC efficiency is significantly higher than that obtained in 10 ppm doped crystals [21].

The preparation and measurement timing is controlled by three arbitrary function generators (AFG, Tektronix, AFG3252). It takes 12.5 ms for AFC preparation. To avoid the fluorescence noise caused by the classical pump light, the measurement cycle begins after waiting for time Tₘ = 1.5 ms after the preparation cycle is completed. N trials of single photon pulses are stored in the sample in the 10-ms measurement phase. The complete preparation and measurement cycles are repeated at a frequency of 40 Hz. A schematic drawing of the timing sequence of the experiment is given in the Supplemental Information.

**Details of the photon source.** The phase matching temperature to generate degenerate photon pairs is 44.50 °C. The power of the 440 nm pump light is 20 mW for all the measurements. With continuous pump, the coincidence rate of the unfiltered photon pairs is 1.4 MHz. The heralding efficiency is 0.20. The diffraction efficiency of the gratings (1200 lines per millimeter) is 0.88. The transmission efficiency of the etalon is 0.95. The etalon has a bandwidth of 700 MHz and a free spectral range of 50 GHz. The temperature drift of the etalon is smaller than 10 mK. After spectral filtering, the coincidence rate of the photon pairs is 6.1 kHz and the heralding efficiency is 0.10. The photon source is switched into pulsed operations for interface with the AFC. The temporal width of blue pump light is 32 ns. The transmission efficiency from the photon source to the final SPD for the signal photons is approximately 0.23.

A figure of merit for the nonclassical nature of the photon correlations is the normalized cross-correlation function g(2)ₓᵧ = pₓᵧ/pₓpᵧ, where pₓ (pᵧ) is the probability to detect a signal (idler) photon and pₓᵧ is the probability to detect a coincidence in a specified time window. In practice, pₓ and pᵧ are determined by the number of coincidences in the time window centered on and away from the coincidence peak, respectively. Assuming that the second-order auto-correlations of signal and idler g(2)ₓ (where x=’s’ for signal or ‘i’ for idler) satisfying 1 ≤ g(2)ₓ ≤ 2, then the non-classicality is proved by measuring g(2)ₓᵧ greater than 2 [22, 28, 23]. We note that, compared with the unfiltered source, g(2)ₓ rises significantly after spectral filtering. We measured g(2)ₓᵧ ≈ 452 between the transmitted signal photon and the idler photon. The temporal width of the coincidence window is 2 ns and the
detection window for noise is 400 ns away from the transmitted signal peak. The $g_{si}^{(2)}$ drops after storage because of low recall efficiency, the temporal broadening and the rising contributions of dark counts. The temporal width of the coincidence window for retrieved signal is 10 ns. The detection window for related noise is placed at 400 ns away from the retrieved signal peak. An example of the measurement on the $g_{si}^{(2)}$ is given in the Supplemental Information.

We are most grateful to A. J. Leggett for his comments on the preliminary version of this manuscript and to C. Brukner and M. Paternostro for their feedback on the revised version. This work was supported by the National Basic Research Program (2011CB921200), the CAS, National Natural Science Foundation of China (Grant Nos. 11274289, 11325419, 61327901.), the Fundamental Research Funds for the Central Universities (wk2030380004 and wk2470000011), and the EU STREP project PARAPET.

[1] Schrödinger, E. Die gegenwärtige Situation in der Quantenmechanik. Naturwissenschaften 23, 807, 823, 844 (1935).
[2] Bell, J. S. On the Einstein Podolsky Rosen Paradox. Physics 1, 195 (1964).
[3] Kochen, S. and Specker, E. The problem of hidden variables in quantum mechanics. J. Math. Mech. 17, 59 - 87 (1967).
[4] Leggett, A. J. Macroscopic quantum systems and the quantum theory of measurement. Prog. Theor. Phys. Supplement 69, 80 (1980).
[5] Leggett, A. J. and Garg, A. Quantum mechanics versus macroscopic realism: Is the flux there when nobody looks? Phys. Rev. Lett. 54, 857 (1985).
[6] Leggett, A. J. Realism and the physical world. Rep. Prog. Phys. 71, 022001 (2008).
[7] Xu, J. S., Li, C. F., Zou, X. B. and Guo, G. C. Experimental violation of the Leggett-Garg inequality under decoherence. Scientific Reports 1, 101 (2011).
[8] Goggin, M. E. et al. Violation of the Leggett-Garg inequality with weak measurements of photons. Proc. Natl Acad. Sci. 108, 1256 (2011).
[9] Waldherr, G., Neumann, P., Huelga, S. F., Jelezko, F. and Wrachtrup, J. Violation of a temporal Bell inequality for single spins in a diamond defect center. Phys. Rev. Lett. 107, 090401 (2011).
[10] Dressel, J., Broadbent, C. J., Howell, J. C. and Jordan, A. N. Experimental Violation of Two-Party Leggett-Garg Inequalities with Semiweak Measurements. Phys. Rev. Lett. 106, 040402 (2011).
[11] Knee, G. C. et al. Violation of a Leggett-Garg inequality with ideal non-invasive measurements. Nature Communications 3, 606 (2012).
[12] Palacios-Laloy, A. et al. Experimental violation of a Bell’s inequality in time with weak measurement. Nature Physics 6, 4-42 (2010).
[13] Huelga, S. F., Marshall, T. W. and Santos, E. Proposed test for realist theories using Rydberg atoms coupled to a high-Q resonator. Phys. Rev. A. 52, R2497 (1995).
[14] Huelga, S. F., Marshall, T. W. and Santos, E. Temporal Bell-type inequalities for two-level Rydberg atoms coupled to a high-Q resonator. Phys. Rev. A. 54, 1798 (1996).
[15] C. Emary, N. Lambet, and F. Nori, Rep. Prog. Phys. 77, 016001 (2014). See section 2.4.
[16] Clauser, J. F. and Shimony, A. Bell’s theorem. Experimental tests and implications. Rep. Prog. Phys. 41, 1881 (1978).
[17] Wilde M. M, McCracken J. M and Mizel A. Could light harvesting complexes exhibit non-classical effects at room temperature? Proceedings of the Royal Society A 466 1347 (2010).
[18] Fleming G. R., Huelga, S. F. and Plenio, M. B. Focus on quantum effects and noise in biomolecules. New J. Phys. 13, 115002 (2011).
[19] Li, C-M., Lambert, N., Chen, Y-N., Chen, G-Y. and Nori, F. Witnessing Quantum Coherence: from solid-state to biological systems. Scientific Reports 2, 885 (2012).
[20] Breuer, H.-P., Laine, E.-M. and Piilo, J. Measure for the Degree of Non-Markovian Behavior of Quantum Processes in Open Systems. Phys. Rev. Lett. 103, 210401 (2009).
[21] Zhou, Z. Q., Lin, W. B., Yang, M., Li, C. F. and Guo, G. C. Realization of reliable solid-state quantum memory for photonic polarization-qubit. Phys. Rev. Lett. 108, 190505 (2012).
[22] Hastings-Simon, S. R., Afzelius, M., Minář, J., Staudt, M. U., Lauritzen, B., de Riedmatten, H. and Gisin, N. Spectral hole-burning spectroscopy in Nd$^{3+}$:YVO$_4$. Phys. Rev. B. 77, 125111 (2008).
[23] Afzelius, M., Simon, C., de Riedmatten, H. and Gisin, N. Multimode quantum memory based on atomic frequency combs. Phys. Rev. A. 79, 052329 (2009).
[24] de Riedmatten, H., Afzelius, M., Staudt, M. U., Simon, C. and Gisin, N. A solid-state light-matter interface at the single-photon level. Nature 456, 773 (2008).
[25] Clausen, C., Bussières, F., Afzelius, M., Gisin, N. Quantum storage of polarization qubits in birefringent and anisotropically absorbing materials. Phys. Rev. Lett. 108, 190503 (2012).
[26] Gündoğan, M., Ledingham, P. M., Almasi, A., Cris-tiani, M., de Riedmatten, H. Quantum Storage of a Photonic Polarization Qubit in a Solid. Phys. Rev. Lett. 108, 190504 (2012).
[27] Clausen, C., Usmani, I., Bussières, F., Sanguoard, N., Afzelius, M., de Riedmatten, H., and Gisin, N. Quantum storage of photonic entanglement in a crystal. Nature 469, 508 (2011).
[28] Usmani, I. et al. Heralded quantum entanglement between two crystals. Nature Photonics 6, 234 (2012).
[29] Rivas, A., Huelga, S. F. and Plenio, M. B. Quantum Non-Markovianity: Characterization, Quantification and Detection. arXiv:1405.0303 (Rep. Prog. Phys. to appear 2014).
[30] Liu, B. H., Huang, Y. F., Li, C. F., Guo, G. C., Laine,
E. M., Breuer, H. P. and Pillo, J. Experimental control of the transition from Markovian to non-Markovian dynamics of open quantum systems. *Nature Physics* **7**, 931 (2011).

[31] James, D. F. V., Kwiat, P. G., Munro, W. J. and White, A. G. Measurement of qubits. *Phys. Rev. A* **64**, 052312 (2001).

[32] Souza, A. M., Li, J., Soares-Pinto, D. O., Sarthour, R. S., Oliveira, S., Huelga, S. F., Paternostro, M. and Serniao, F. L. Experimental Demonstration of non-Markovian Dynamics via a Temporal Bell-like Inequality. arXiv:1308.5761.

[33] Huang, Y. F., Liu, B. H., Peng, L., Li, Y. H., Li, L., Li, C. F. and Guo, G. C. Experimental generation of an eight-photon Greenberge-Horne-Zeilinger state. *Nature Communications* **2**, 546 (2011).

[34] Iskhakov, T. Sh., Agafonov, I. N., Chekhova, M. V. and Leuchs, G. Polarization-Entangled Light Pulses of $10^5$ Photons. *Phys. Rev. Lett.* **109**, 150502 (2012).

[35] Bruno, N., Martin, A., Sekatski, P., Sangouard, N., Thew, R. T. and Gisin, N. Displacement of entanglement back and forth between the micro and macro domains. *Nature Physics* **9**, 545 (2013).

[36] Lvovsky, A. I., Ghobadi, R., Chandra, A., Prasad, A. S. and Simon, C. Observation of micro-macro entanglement of light. *Nature Physics* **9**, 541 (2013).

**Author Contributions**

C.-F.L. and Z.-Q.Z. planned and designed the experiments. Z.-Q.Z. implemented the experiments. S.F.H. proposed the tests the validity of stationary assumption. S.F.H. and Z.-Q.Z. carried out the theoretical analysis and developed the interpretation. C.-F.L. and G.-C.G. supervised the project. All authors contributed to writing the manuscript and discussed the experimental procedures and results.

**Additional Information**

**Competing Financial Interests**

The authors declare no competing financial interests.

---

**Supplemental Information**

**Timing sequence.** As shown in Fig. S1, the AFC preparation takes 12.5 ms. The measurement phase starts 1.5 ms after the preparation completes. The chopper before SPD is opened in the measurement phases to enable detection of single photons. During the measurement cycle, $N = 25000$ trials of 440 nm laser pulses are sent into the PPKTP crystal with a periodicity of 400 ns. The generated single photon pulses are stored in the sample and retrieved after a programmable time $\tau_s$. Only for the measurements presented in Fig. 4(b), to probe the quantum states for longer storage time, $N = 10000$ trials of single-photon pulses are sent into the sample with a periodicity of 1000 ns.

**Measurement on $g_{si}^{(2)}$.** An example of measurement on $g_{si}^{(2)}$ is shown in Fig. S2. V-polarized single-photon pulses are mapped into the sample and retrieved 50 ns later. The noise at 400 ns is from the next pump pulse of SPDC. To determine the $g_{si}^{(2)}$ of the transmitted pulses, the 2-ns coincidence window is placed at $t = 0$ to measure the signal and the detection window for related noise is placed at $t = 400$ ns. Because of the temporal broadening of the retrieved signal pulses, the coincidence window for retrieved signal has the width of 10 ns. The detection window for related noise is also placed at 400 ns away from the retrieved signal. $g_{si}^{(2)} = 54.7$ after a storage time of 50 ns.
Fig. S1: The timing sequence used for the experiment. The complete preparation and measurement cycles are repeated at a frequency of 40 Hz.

Fig. S2: Measurement on $g_\text{sr}^{(2)}$ after a storage time of 50 ns. The blue parts shows the transmitted components which have been intentionally decreased by 50 times for visual effect. The red dashed lines and shaded area define the 10-ns detection windows for the coincidences and noise used to compute cross-correlations of the retrieved signal, respectively. The time-bin size is 2 ns and the integration time is 10 minutes.