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Testing the Bell inequality on frequency-bin entangled photon pairs using time-resolved detection

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Entanglement, describing the inseparability of a quantum multiparty system, is one of the most intriguing features of quantum mechanics. Violation of Bell inequality, for ruling out the possibility of local hidden-variable theories, is commonly used as a strong witness of quantum entanglement. In previous Bell test experiments with photonic entanglement based on two-photon coincidence measurement, the photon temporal wave packets were absorbed completely by the detectors. That is, the photon coherence time is much shorter than the detection time window. Here we demonstrate the generation of frequency-bin entangled narrowband biphotons, and for the first time, to the best of our knowledge, test the Clauser–Horne–Shimony–Holt (CHSH) Bell inequality $|S| \leq 2$ for their nonlocal temporal correlations with time-resolved detection. We obtain a maximum $|S|$ value of 2.52 ± 0.48, which violates the CHSH inequality. Our result will have applications in quantum information processing involving time-frequency entanglement.

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1. INTRODUCTION

As one of the most important features of quantum mechanics, entanglement is essential in quantum information processing, quantum computation, and quantum communication [1]. Photonic entanglement has been realized in diverse degrees of freedom, including polarization [2–4], position momentum [5,6], orbital angular momentum [7], and time frequency [8–14]. The nonlocal correlations between distant entangled photons provide a standard platform for the Bell test [15–17] and confirm that quantum mechanics is incompatible with the local hidden-variable theories, which is the essence of the Einstein–Podolsky–Rosen (EPR) paradox [18]. Recently, a loophole-free test of Bell's theorem has been demonstrated [19,20]. On the other side, violation of Bell inequality is often used as an entanglement witness.

In most previous experimental tests of Bell's theorem with entangled photons, including these loophole-free tests [19,20], the photon wave packets were absorbed completely by the coincidence detectors. That is, the photon coherence time is much shorter than the detection time window. In these works, the photon coincidence detection is modelled as integral over the entire wave packets. Surprisingly, although time-frequency entanglement has been studied intensively [8–10,12–14], the nonlocal correlation between the arrival times of frequency-bin entangled photons on the detectors (or the biphoton temporal correlation) has never been used for testing Bell's theorem. In all previous experiments of the Bell test, the Bell parameter S is a function of the detectors' local settings (such as polarizations in polarization entanglement [19,20], phases in a Franson interferometer [8]), which do not include the arrival times of the photons on the distant detectors.

The recent development of narrowband biphoton generation makes it possible to reveal the rich temporal quantum state information directly using time-resolved single-photon counters. Biphotons have been produced with a bandwidth narrower than 50 MHz with spontaneous parametric down-conversion inside a cavity [21,22], spontaneous four-wave mixing (SFWM) in a hot atomic vapor cell [23], or laser-cooled atoms [24–30]. On the other hand, time-resolved measurement has been demonstrated as a powerful tool in generating pure heralded single photons [31,32] and biphoton entanglement [33].

In this article, we demonstrate the generation of frequency-bin entangled narrowband biphotons using SFWM in cold atoms for testing Bell's theorem. For the first time, we test the Clauser–Horne–Shimony–Holt (CHSH) Bell inequality $|S| \leq 2$ for their nonlocal temporal correlations with time-resolved detection. We obtain a maximum $|S|$ value of 2.52 ± 0.48, which violates the CHSH inequality. Our result also reveals the connection between the visibility of the two-photon quantum temporal beating resulting from the frequency entanglement and the violation of the Bell inequality. Our work not only provides a new case of Bell test, but also opens a new way to study the time-frequency entanglement with time-resolved detection.

2. GENERATION OF FREQUENCY-BIN ENTANGLED NARROWBAND BIPHOTONS

Our experimental configuration of double-path SFWM and relevant atomic energy level diagram are illustrated in Fig. 1(a) and 1(b). We work with laser-cooled ^{85}Rb atoms trapped in a two-dimensional (2D) magneto-optical trap (MOT) [34]. In the presence of counter-propagating pump (ω_p) and coupling (ω_c) laser beams along the longitudinal z axis of the 2D MOT, correlated Stokes (ω_s) and anti-Stokes (ω_{as}) photons are spontaneously generated in opposite directions and collected into two spatial symmetric single-mode paths (paths 1 and 2, respectively). We send the Stokes photons on path 1 through a frequency-phase shifter (FPS) ($\delta = 2\pi \times 10$ MHz, $\Delta\phi_s$) so that the frequencies of these Stokes photons become $\omega_s + \delta$ and obtain a relative phase $\Delta\phi_s$ to those on path 2. Symmetrically, we add a second FPS (δ, $\Delta\phi_{as}$) to the anti-Stokes photons on path 2. We then combine the two paths with two beam splitters (BS_1 and BS_2, 50%:50%). The single-mode outputs of the beam splitters are detected by single-photon counting modules ($D_{s\pm}$ and $D_{as\pm}$) as shown in Fig. 1(a). A detailed description of our experimental setup is presented in Supplement 1 [35]. The photon pairs from the beam-splitter outputs are frequency-bin entangled and their biphoton states can be described as

$$|\Psi_{XY}\rangle = \frac{1}{\sqrt{2}}[|\omega_s + \delta\rangle|\omega_{as}\rangle + XY e^{i(\Delta\phi_{as}-\Delta\phi_s)}|\omega_s\rangle|\omega_{as} + \delta\rangle], \quad (1)$$

where XY, as a product of the signs (+, −), represents the Stoke-to-anti-Stokes combinations from the beam-splitter outputs: $|\Psi_{XY}\rangle$ is detected by (D_{sX}, D_{asY}).

The Glauber correlation function of the biphoton state in Eq. (1) exhibits a quantum beating [35]:

$$G^{(2)}_{XY}(\tau; \Delta\phi_s, \Delta\phi_{as}) = \frac{1}{2}[G^{(2)}_0(\tau) - N_0] \times [1 + XY \cos(\delta\tau + \Delta\phi_s - \Delta\phi_{as})] + N_0, \quad (2)$$

where $\tau = t_{as} - t_s$. $G^{(2)}_0(\tau)$ is the biphoton Glauber correlation function before the beam splitters, which is the same for both paths 1 and 2. N_0 is the uncorrelated accidental coincidence rate. We then have the normalized biphoton correlation function,

$$g^{(2)}_{XY}(\tau; \Delta\phi_s, \Delta\phi_{as}) = G^{(2)}_{XY}(\tau; \Delta\phi_s, \Delta\phi_{as})/N_0 = \frac{1}{2}[g^{(2)}_0(\tau) - 1] \times [1 + XY \cos(\delta\tau + \Delta\phi_s - \Delta\phi_{as})] + 1, \quad (3)$$

where $g^{(2)}_0(\tau) = G^{(2)}_0(\tau)/N_0$. When N_0 is nonzero, the beating visibility slowly varies as a function of τ:

$$V(\tau) = \frac{G^{(2)}_0(\tau) - N_0}{G^{(2)}_0(\tau) + N_0} = \frac{g^{(2)}_0(\tau) - 1}{g^{(2)}_0(\tau) + 1}. \quad (4)$$

In the ideal case under the limit of zero accidental coincidence counts, the visibility of cosine modulation in the quantum beat approaches 100%. Experimentally, the two-photon temporal correlation is measured as coincidence counts between the detectors:

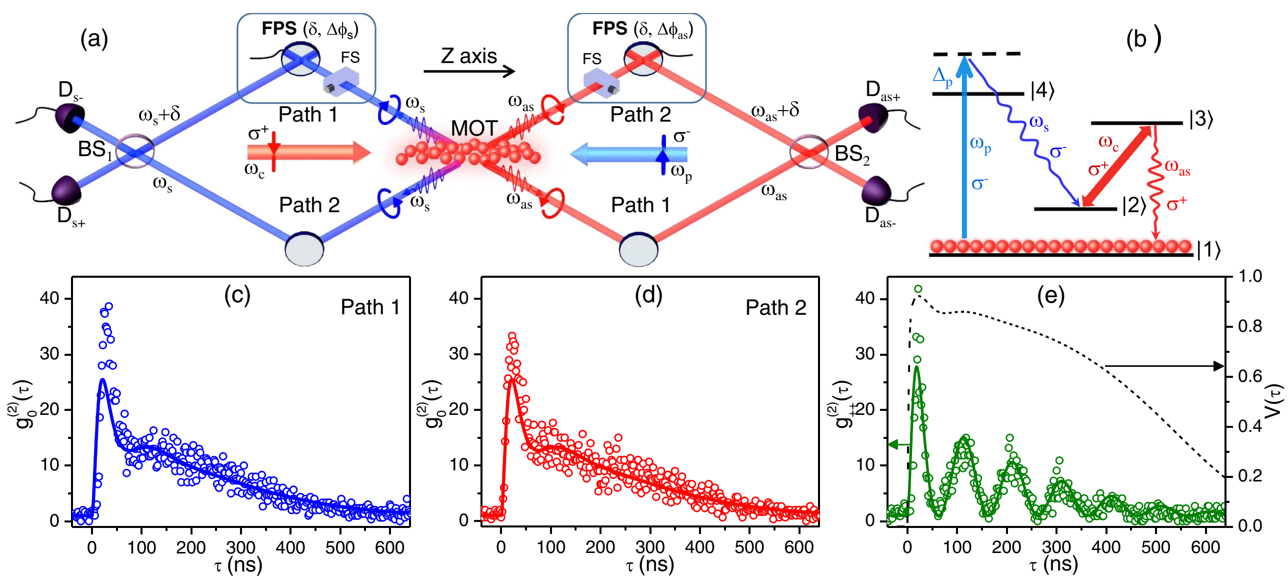

Fig. 1. Generation of frequency-bin entangled narrowband biphotons. (a) Experimental schematics of generating biphotons with double-path SFWM in cold ^{85}Rb atoms. Backward and paired Stokes (ω_s) and anti-Stokes (ω_{as}) photons are spontaneously produced into paths 1 and 2, respectively, which are symmetric with angles of ±3° to the longitudinal axis. The Stokes photons on path 1 go through FPS (δ, $\Delta\phi_s$), while the anti-Stokes photons on path 2 go through FPS (δ, $\Delta\phi_{as}$). BS_1 and BS_2 are two beam splitters. $D_{s\pm}$ and $D_{as\pm}$ are single-photon counting modules. (b) ^{85}Rb atomic energy level diagram for SFWM. The atomic hyperfine levels are chosen as $|1\rangle = |5S_{1/2}, F = 2\rangle$, $|2\rangle = |5S_{1/2}, F = 3\rangle$, $|3\rangle = |5P_{1/2}, F = 3\rangle$, and $|4\rangle = |5P_{3/2}, F = 3\rangle$. The circularly polarized (σ^-) pump laser (780 nm) is blue-detuned by 60 MHz from the transition $|1\rangle \to |4\rangle$, whereas the coupling laser (σ^+, 795 nm) is on resonance to the transition $|2\rangle \leftrightarrow |3\rangle$. Panels (c) and (d) show the biphoton temporal correlations on paths 1 and 2, respectively. (e) Biphoton quantum beating measured between detectors D_{s+} and D_{as+} and its visibility with the phase setting $\Delta\phi_s = 3\pi/2$ and $\Delta\phi_{as} = -\pi/4$. The solid curves in panels (c)–(e) are obtained from the SFWM biphoton theory [35].

$$C_{XY}(\tau;\Delta\phi_s,\Delta\phi_{as}) = G^{(2)}_{XY}(\tau;\Delta\phi_s,\Delta\phi_{as})\eta\xi\Delta t_{\text{bin}}T, \quad (5)$$

where η is the joint detection efficiency, ξ is the duty cycle, Δt_{bin} is the detector time bin width, and T is the data collection time.

Figure 1(c) and 1(d) shows the biphoton correlations for paths 1 and 2, respectively, measured without the presence of the two beam splitters. They are nearly identical to each other with coherence time of about 300 ns, corresponding to the bandwidth of 1.28 MHz. With the two beam splitters presented, the correlation $g^{(2)}_{++}(\tau;3\pi/2,-\pi/4)$ displays a quantum beating [shown in Fig. 1(e)], as predicted in Eq. (3).

3. BELL INEQUALITY OF FREQUENCY-BIN ENTANGLEMENT

As shown in Eqs. (2)–(5) and confirmed in our experiment, the two-photon coincidence counts are functions of the relative arrival time delay $\tau = t_{as} - t_s$ between the Stokes and anti-Stokes photons at the two distant detectors and the relative phase difference $\Delta\phi_s - \Delta\phi_{as}$. We set the photon counters $D_{s\pm}$ distant enough from $D_{as\pm}$, as well as the two FPSs, to make our Bell test locality-loophole-free. Meanwhile, we take the fair-sampling assumption. To test the CHSH-type Bell inequality [17], we define the measurement output as +1 for coincidence between D_{s+} and D_{as+} (or D_{s-} and D_{as-}), and −1 for coincidence between D_{s+} and D_{as-} (or D_{s-} and D_{as+}). Then the Bell correlation coefficient can be obtained from

$$E(\tau;\Delta\phi_s,\Delta\phi_{as}) = \frac{C_{++}+C_{--}-C_{+-}-C_{-+}}{C_{++}+C_{--}+C_{+-}+C_{-+}}. \quad (6)$$

As shown in Fig. 1(c) and 1(d), the biphoton waveforms for paths 1 and 2 are nearly identical with a likeness of 98% [35]. Therefore, the two SFWM paths are nearly symmetrical. Considering the symmetries of the two SFWM paths and the beam splitters, Eq. (6) can be reduced to

$$E(\tau;\Delta\phi_s,\Delta\phi_{as}) = \frac{C_{++}(\tau;\Delta\phi_s,\Delta\phi_{as}) - C_{++}(\tau;\Delta\phi_s^{\perp},\Delta\phi_{as})}{C_{++}(\tau;\Delta\phi_s,\Delta\phi_{as}) + C_{++}(\tau;\Delta\phi_s^{\perp},\Delta\phi_{as})}, \quad (7)$$

which requires only two photon detectors with $\Delta\phi_s^{\perp} = \Delta\phi_s + \pi$. Then the CHSH Bell parameter S can be estimated as

$$S(\tau) = E(\tau;\Delta\phi_s,\Delta\phi_{as}) - E(\tau;\Delta\phi'_s,\Delta\phi_{as}) + E(\tau;\Delta\phi_s,\Delta\phi'_{as}) + E(\tau;\Delta\phi'_s,\Delta\phi'_{as}). \quad (8)$$

Under local realism, the Bell inequality holds $|S(\tau)| \le 2$.

For the biphoton source described by Eqs. (1)–(5), setting $\Delta\phi'_s = \Delta\phi_s - \pi/2$ and $\Delta\phi'_{as} = \Delta\phi_{as} - \pi/2$, we derive [35]

$$S(\tau) = 2\sqrt{2}V(\tau)\cos\left(\delta\tau + \Delta\phi_s - \Delta\phi_{as} + \frac{\pi}{4}\right). \quad (9)$$

For frequency-bin entanglement approaching the ideal case without accidental coincidence counts, i.e., $V(\tau) \to 1$, our theory predicts $|S|_{\text{max}} \to 2\sqrt{2}$, which violates the CHSH Bell inequality. It is clear that $S(\tau)$ exhibits a sinusoidal oscillation pattern with $2\sqrt{2}V(\tau)$ as the slowly varying envelope.

The measured $S(\tau)$ and Bell correlations $E(\tau)$ are shown in Fig. 2, with the phase setting $\Delta\phi_s = 0$, $\Delta\phi_{as} = \pi/4$, $\Delta\phi'_s = -\pi/2$, and $\Delta\phi'_{as} = -\pi/4$. At $\tau = 52$ ns, we have $S = -2.52 \pm 0.48$, which violates the classical limit. The solid theoretical curve in Fig. 2(a) is predicted from Eq. (9) and agrees well with the experiment. The oscillation amplitude follows the dashed envelope plotted from $\pm 2\sqrt{2}V(\tau)$ well. By adjusting the phase setting, we can violate the Bell inequality $|S| \le 2$ for $0 < \tau \le 350$ ns, where the visibility satisfies $V > 1/\sqrt{2}$.

4. NONLOCAL PHASE CORRELATION

At a fixed relative time delay of τ, the two-photon correlation in Eq. (2) is identical to that obtained in polarization entanglement, where $\Delta\phi_s$ and $\Delta\phi_{as}$ correspond to the orientations of distant polarizers. To confirm this, in Fig. 3(a) we plot the measured quantum beating temporal correlations with different phase settings. Figure 3(b) shows the nonlocal phase correlation as a function of $\Delta\phi_s$ at $\tau = 252$ ns under two different $\Delta\phi_{as}$. Similarly to the polarization entanglement, the system has a rotation symmetry and the correlation depends only on the relative phase difference $\Delta\phi_s - \Delta\phi_{as}$. Therefore, the visibility of the nonlocal phase

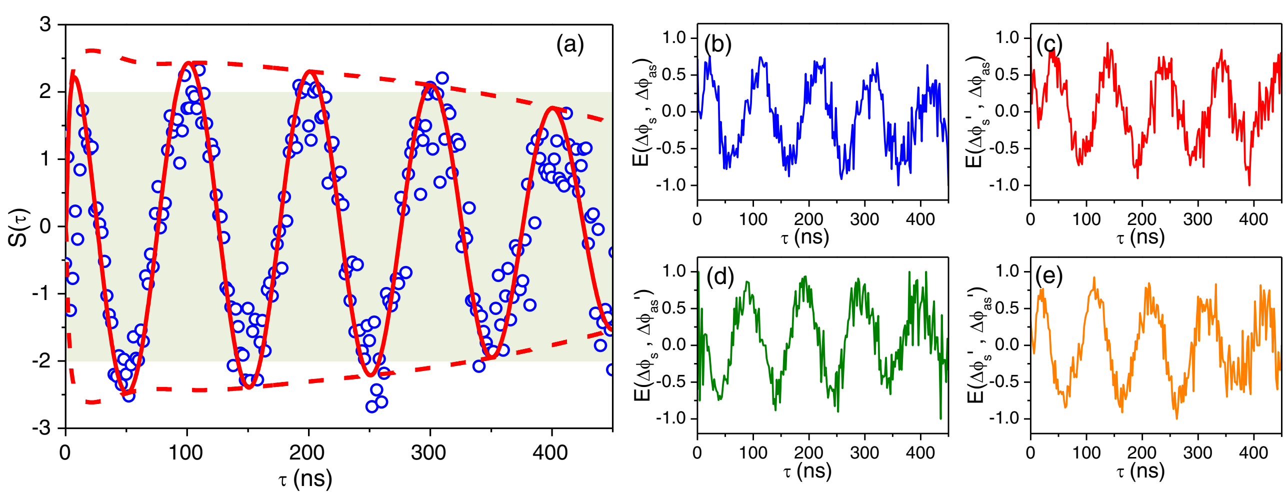

Fig. 2. CHSH Bell inequality of frequency-bin entanglement. (a) S as a function of a two-photon relative time delay of τ. The circles are experimental data. The solid curve is predicted by the theory. The dashed envelopes are plotted from $\pm 2\sqrt{2}V(\tau)$, where the visibility $V(\tau)$ is determined by Eq. (4). The shadow area is the classical regime where $|S| \le 2$. Panels (b)–(e) show the measured Bell correlations. The phase settings are $\Delta\phi_s = 0$, $\Delta\phi_{as} = \pi/4$, $\Delta\phi'_s = -\pi/2$, and $\Delta\phi'_{as} = -\pi/4$.

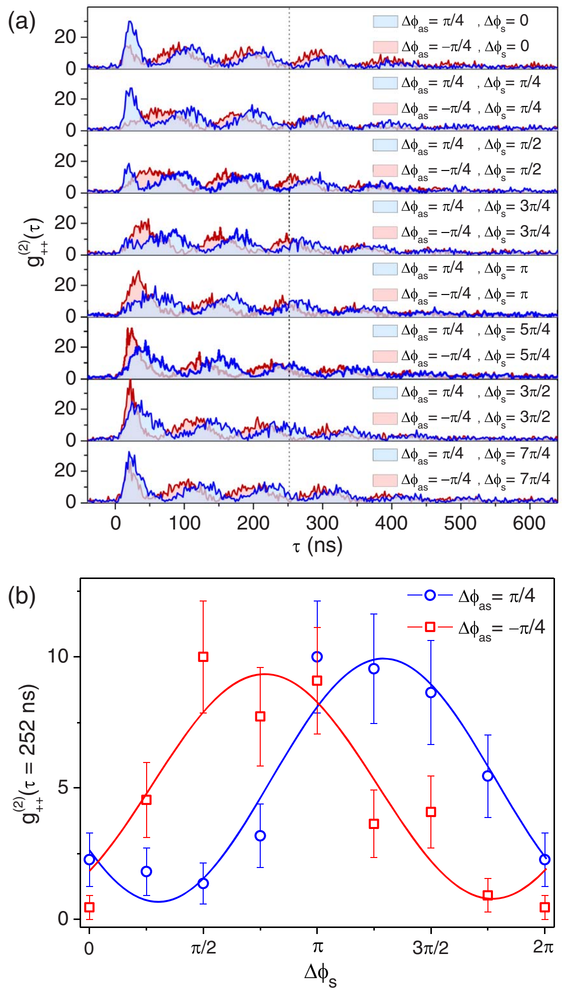

Fig. 3. Two-photon nonlocal phase correlation. (a) Measured biphoton temporal correlations with different phase settings. (b) Phase correlations at $\tau = 252$ ns. The error bars are standard deviations resulting from the statistical uncertainties of coincidence counts. The solid lines are the best fitting curves, with visibilities of $V_1 = 0.86 \pm 0.09$ for $\Delta\phi_{as} = \pi/4$ and $V_2 = 0.85 \pm 0.13$ for $\Delta\phi_{as} = -\pi/4$.

correlation $V > 1/\sqrt{2}$ is an indication of violation of the Bell inequality. The solid lines in Fig. 3(b) are the best fitting sinusoidal curves with visibilities of $V_1 = 0.86 \pm 0.09$ for $\Delta\phi_{as} = \pi/4$ and $V_2 = 0.85 \pm 0.13$ for $\Delta\phi_{as} = -\pi/4$. These visibilities are consistent with the visibility envelope of the quantum beating $V(\tau = 252 \text{ ns}) = 0.78$ in Fig. 1(e). We estimate the S value from $S = \sqrt{2}(V_1 + V_2) = 2.42 \pm 0.31$, which violates the Bell inequality.

5. BIPHOTON TEMPORAL BEATING

The Bell inequality was derived for general local experimental apparatus settings. For the polarization entanglement, these settings are the orientations of the polarizers. For the time-frequency entanglement, one can choose the detection time as the local detector setting parameter. In this experiment, we take t_s and t_{as} as the two distant local parameters. For the frequency-bin entangled state we prepared here, quantum mechanics predicts that the two-photon temporal correlation exhibits a temporal quantum beating [Eq. (2)], which in mathematical form is similar to the polarization correlation from polarization entanglement.

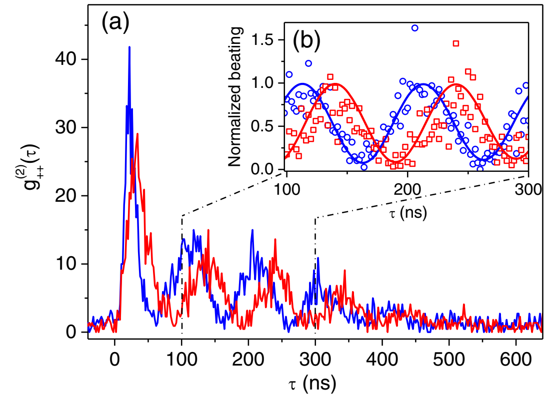

Fig. 4. Biphoton temporal beating. (a) Measured two-photon temporal correlations with different phase settings. The blue data are measured with $\Delta\phi_s = 3\pi/2$, $\Delta\phi_{as} = -\pi/4$ and the red data with $\Delta\phi_s = \pi$, $\Delta\phi_{as} = -\pi/4$. (b) Normalized beating signals. The solid curves are the best fittings.

Figure 4(a) shows the temporal beatings of two-photon correlation at two different phase settings. To compare with that in a conventional polarization entanglement measurement, we normalize the quantum beating $g^{(2)}_{++}(\tau)$ to the correlation envelope $g^{(2)}_0(\tau)$ without subtracting the contribution from accidental photon coincidence counts. The normalized beating signals are plotted in Fig. 4(b). With sinusoidal curve fitting for the normalized beating over 200 ns, we obtain the visibilities to be $\bar{V} = 0.77 \pm 0.04$ and $\bar{V} = 0.76 \pm 0.06$, which clearly surpasses the 0.5 limit of a classical probability theory [2,37]. The corresponding $|S|$ values are 2.17 ± 0.11 and 2.16 ± 0.17.

6. SUMMARY AND DISCUSSION

In summary, we generate frequency-bin entangled narrowband biphotons from SFWM in cold atoms with a double-path configuration, where the phase difference between the two spatial paths can be controlled independently and nonlocally. The two-photon correlation exhibits a temporal quantum beating between the entangled frequency modes whose phase is determined by the relative phase difference between the two paths. We have successfully tested the CHSH Bell inequality and our best result is $|S| = 2.52 \pm 0.48$, which violates the Bell inequality $|S| \leq 2$. With V as the visibility of the two-photon temporal beating, the quantum theory predicts $|S|_{max} = 2\sqrt{2}V$, which is confirmed by the experiment. Therefore, the visibility $V > 1/\sqrt{2} = 71\%$ of the two-photon temporal beating is sufficient to violate the Bell inequality. The experimental value of $|S|$ is below $2\sqrt{2}$ because the uncorrelated accidental coincidence counts from stray light and dark counts, and the randomness of photon pair generation in the spontaneous process reduce the beating visibility. We can reduce the pump laser power for lower accidental coincidence counts and thus for a higher visibility, but it will take a longer time for collecting data. The error bar of $|S|$ is the standard deviation resulting from the statistical uncertainties of coincidence counts, which can also be reduced with a longer data collection time. The time-resolved single-photon detection is a powerful tool for quantum-state control, such as entanglement swapping and teleportation [14]. Our result, for the first time, tests the Bell inequality in a nonlocal temporal correlation of frequency-bin entangled narrowband biphotons with time-resolved detection, and will have applications in quantum information processing involving time-frequency entanglement [38]. Moreover, through the Bell test, we have unambiguously demonstrated the generation of frequency-bin entangled narrowband (about 1 MHz) biphotons, which can efficiently interact with stationary atomic quantum nodes in an atom–photon quantum network [39]. Because of their narrow bandwidth, these biphotons can be stored and retrieved from a quantum memory with high efficiency.

After submission of this article, we became aware that a work of creating and manipulating single photons in superposition states of two discrete frequencies was published recently [38]. Our narrowband frequency-bin entangled biphoton source in this work can be ideally implemented to produce pure heralded single photons in a two-color qubit state with a tunable phase, making use of entanglement, linear optics, and time-resolved detection [31,32,35].

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See Supplement 1 for supporting content.

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