



Entangling Different-Color Photons via Time-Resolved Measurement and Active Feed Forward

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Entangling independent photons is not only of fundamental interest but also of crucial importance for quantum-information science. Two-photon interference is a major method for entangling independent identical photons. If two photons are different in color, perfect two-photon coalescence can no longer happen, which makes the entangling of different-color photons difficult to realize. In this Letter, by exploring and developing time-resolved measurement and active feed forward, we have entangled two independent photons of different colors for the first time. We find that entanglement with a varying form can be identified for different two-photon temporal modes through time-resolved measurement. By using active feed forward, we are able to convert the varying entanglement into uniform entanglement. Adopting these measures, we have successfully entangled two photons with a frequency separation 16 times larger than their linewidths. In addition to its fundamental interest, our work also provides an approach for solving the frequency-mismatch problem for future quantum networks.

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Entangling independent photons through two-photon interference [1,2] is ubiquitous in photonic quantum-information experiments [3–5]. When two identical photons are superimposed on a beam splitter, the probabilities that both photons are transmitted or both are reflected interfere with each other and result in two-photon coalescence. Such a two-photon interference effect was first observed by Hong, Ou, and Mandel [6]. From a more fundamental point of view, this interference is due to the bosonic nature of photons [7]. Two identical photons have a symmetric wave function, thus, their spacial wave function has to be symmetric, which leads to photon coalescence after passing through a beam splitter. Therefore, only an antisymmetric two-photon state will lead to a coincidence between different output ports of a beam splitter, which constitutes the physical basis of measuring Bell states and entangling independent photons. What if the input photons are different color? Can we still make Bell-state measurements and entangle independent photons as usual, for instance, in the degrees of polarization, time bin, and momentum?

Entangling different-color photons also has strong practical applications. In quantum networking [8], photons from separate quantum systems are often different in color for various reasons. For instance, in the condensed matter systems, such as quantum dots and nitrogen-vacancy centers, photons from two separate emitters are usually different in frequency due to their different local environments [9,10]. For all quantum systems, when they are moving with a high speed (e.g., in a satellite or an airplane), the Doppler effect will give rise to significant frequency

shifts for the emitting photons. Additionally, the strong interest in hybrid quantum networking by combining the advantages of each physical system also necessitates the entangling operation between different-color photons. Preliminary studies have been carried out on the quantum beat of two different-color photons both theoretically [11,12] and experimentally [7,13]. Without making use of time-resolved measurement [7], rather poor interference visibility has been observed. By making use of time-resolved measurement [13], high-visibility interference shows up in a time-dependent fast-oscillating manner; however, perfect two-photon coalescence could only happen through narrow temporal filtering. In addition, interfering different-color photons through a dual Mach-Zehnder interferometer have also been studied previously and interesting interference patterns were reported [14].

In this Letter, we study the entangling of independent different-color photons. In order to erase the frequency-distinguishable information during two-photon interference, we make use of time-resolved measurements. Entanglement states can be selected from an otherwise mixed state for each combination of temporal modes. In our experimental demonstration, we start from two pairs of entangled photons which are different color and make use of the entanglement-swapping process. In contrast to prior experiments, a time-resolved Bell-state analyzer is utilized and reveals a random phase shift for the two photons after entanglement swapping. The random phase shift is later compensated through active feed forward with a Pockels cell. By taking these measures, we have successfully

entangled two independent photons with a frequency separation of 80 MHz, which is 16 times larger than their frequency linewidths. With photon detectors of much better time resolution, independent photons with much larger frequency separation can become entangled with our method.

We consider two single photons, both of which are initialized in the state of $(1/\sqrt{2})(|H\rangle + |V\rangle)$, where H refers to horizontal polarization and V refers to vertical polarization. Ideally, when these two photons are indistinguishable from each other, after passing through a polarizing beam splitter (PBS) that transmits horizontal polarization and reflects vertical polarization, they will become entangled in a state of $(1/\sqrt{2})(|H\rangle_1|H\rangle_2 - |V\rangle_1|V\rangle_2)$, with the subscripts 1 and 2 denoting the two output ports of the PBS shown in Fig. 1, if we consider only the case that two photons exit from different ports. However, if the input two photons are different colors, say, photon a has a frequency of ω_a and photon b has a frequency of ω_b , the output two-photon state will change to $|\psi\rangle_{12} = (1/\sqrt{2})(|H, \omega_b\rangle_1|H, \omega_a\rangle_2 - |V, \omega_a\rangle_1|V, \omega_b\rangle_2)$, where the polarization degree is coupled with the external degree of frequency. If we consider the polarization degree only, the reduced state is thus a maximally mixed state which has little application for quantum information. Generally, this type of coupling with external degrees or environments can be eliminated using the quantum erasing technique. In order to erase the frequency-distinguishable

information, we can make use of fast detections. The output state $|\psi\rangle_{12}$ can be decomposed in the temporal degree with the form

$$|\psi\rangle_{12} = \frac{1}{\sqrt{2}} \iint dt_1 dt_2 [g(t_1)f(t_2)|H, t_1\rangle_1|H, t_2\rangle_2 - e^{i(\omega_a - \omega_b)(t_1 - t_2)} f(t_1)g(t_2)|V, t_1\rangle_1|V, t_2\rangle_2], \quad (1)$$

where $f(t)$ and $g(t)$ denote the temporal shape for the ω_a photon and the ω_b photon, respectively. If the frequency linewidths of the two photons are similar, thus $f(t) \approx g(t)$, and the temporal information is determined much better than $2\pi/|\omega_a - \omega_b|$, the output state will be in an entangled state of $|\phi\rangle_{12} = (1/\sqrt{2})(|H\rangle_1|H\rangle_2 - e^{i\Delta\omega\Delta t}|V\rangle_1|V\rangle_2)$, with $\Delta\omega = \omega_a - \omega_b$ and $\Delta t = t_1 - t_2$, conditioned on a temporal mode combination of $|t_1\rangle_1|t_2\rangle_2$. Therefore, a time-resolved measurement enables us to erase the different-color information and select entangled states out of otherwise mixed states.

While the determination of the temporal information without affecting the polarization of single photons requires the highly demanding technique of nondemolition measurement [17,18], in our experiment we instead make use of the entanglement swapping process [19–21]. As shown in Fig. 1, two pairs of entangled photons are generated from separate sources through cavity-enhanced spontaneous

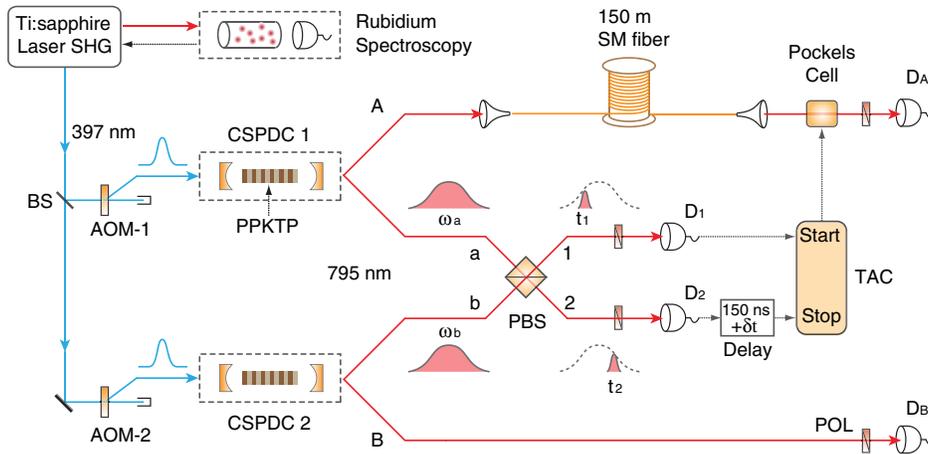


FIG. 1 (color online). Experimental setup. Two cavity-enhanced spontaneous parametric down-conversion (CSPDC) sources are being used to create two pairs of entangled photons. The pumping beam is generated through second-harmonic generation (SHG) of a Ti:sapphire laser working at 795 nm and stabilized through rubidium spectroscopy. Two acousto-optic modulators (AOM) are used to chop the pumping beam into short pulses with a repetition rate of 2 MHz and to tune the frequencies of the narrow band entangled photons. Each entanglement source is basically made up of a linear cavity with a 25 mm long nonlinear crystal (PPKTP) and a 5 mm long KTP crystal inside. The KTP is utilized to achieve double resonance for both CSPDC photons through temperature tuning [15]. The measured linewidths for the two cavities are $\gamma_1/2\pi = 4.2$ MHz and $\gamma_2/2\pi = 5.6$ MHz, respectively. Within each source, by controlling the double-resonance condition and making use of additional filtering etalons [16], the two down-converted photons are configured to have the same frequency, which is exactly one-half of the pumping beam. Photons A and a are polarization entangled, and so are B and b . A polarizing beam splitter (PBS) along with two polarizers (POL) are utilized for the Bell-state measurement. Detection-time differences between D_1 and D_2 are fed forward to a Pockels cell to cancel the random phase shifts in order to recover the entanglement between photons A and B . To compensate the feedback delay due to the single-photon detectors, the time-to-amplitude converter (TAC), and the high-voltage driver of the Pockels cell, a single-mode (SM) fiber loop with a length of 150 m is inserted for photon A .

parametric down-conversion (CSPDC) [22]. The first pair has a frequency of ω_a and a state of $|\Phi^+\rangle_a = (1/\sqrt{2})(|H\rangle_A|H\rangle_a + |V\rangle_A|V\rangle_a)$. The second pair has a frequency of ω_b and a similar state of $|\Phi^+\rangle_b = (1/\sqrt{2})(|H\rangle_B|H\rangle_b + |V\rangle_B|V\rangle_b)$. In order to entangle photons A and B , we need to make a joint Bell-state measurement for photons a and b . As shown in Fig. 1, Photon a from the first pair and photon b from the second pair are superimposed on a PBS, and we only consider the case that two photons leave from different output ports, which gives a final state of

$$\begin{aligned} |\Psi\rangle_f &= (1/\sqrt{2})(|H\rangle_A|H, \omega_b\rangle_1|H, \omega_a\rangle_2|H\rangle_B \\ &\quad - |V\rangle_A|V, \omega_a\rangle_1|V, \omega_b\rangle_2|V\rangle_B) \\ &= \frac{1}{2} \iint dt_1 dt_2 f(t_1) f(t_2) [(|H\rangle_A|H\rangle_B \\ &\quad - e^{i\Delta\omega\Delta t}|V\rangle_A|V\rangle_B) \otimes |\Phi^+\rangle_{12} \\ &\quad + (|H\rangle_A|H\rangle_B + e^{i\Delta\omega\Delta t}|V\rangle_A|V\rangle_B) \otimes |\Phi^-\rangle_{12}], \quad (2) \end{aligned}$$

where $|\Phi^\pm\rangle_{12} = (1/\sqrt{2})(|H, t_1\rangle_1|H, t_2\rangle_2 + |V, t_1\rangle_1|V, t_2\rangle_2)$ can be distinguished unambiguously from a coincidence event of either $|+\rangle_1|+\rangle_2$ or $|-\rangle_1|-\rangle_2$ and $|\Phi^-\rangle_{12} = (1/\sqrt{2})(|H, t_1\rangle_1|H, t_2\rangle_2 - |V, t_1\rangle_1|V, t_2\rangle_2)$ can be distinguished from a coincidence event of either $|+\rangle_1|-\rangle_2$ or $|-\rangle_1|+\rangle_2$, with $|\pm\rangle = (1/\sqrt{2})(|H\rangle \pm |V\rangle)$. Thus a Bell-state measurement result of $|\Phi^\pm\rangle_{12}$ projects the remaining two photons into an entangled state of $|H\rangle_A|H\rangle_B \mp e^{i\Delta\omega\Delta t}|V\rangle_A|V\rangle_B$. In contrast to the traditional frequency-degenerate case ($\Delta\omega = 0$), conditioned on a Bell-state measurement result, the quantum state of photons A and B after entanglement swapping is no longer in a definite entangled state, but in an entangled state with its internal phase depending on the detection-time difference and the frequency separation. Since the detection-time difference Δt varies from event to event, on average, the final state after entanglement swapping is thus in a mixed state. This problem can be solved by using tight temporal filtering by only selecting the events with $\Delta t \approx 0$, which, however, will lead to significant reduction of photon flux. A much better method is to make time-resolved measurements and actively compensate the Δt -dependent phase shift.

Prior to studying entanglement swapping with different-color photon pairs, we verify that our experimental setup works well for the same frequency case ($\Delta\omega = 0$). Photons from CSPDC are usually correlated in frequency [16], which severely limits the fidelity of entanglement swapping with these sources [23]. One solution is to use short pumping pulses with its width smaller than the coherence time for the down-converted photons [24,25]. To verify the elimination of frequency correlation, we measure the polarization-correlation visibilities of photons A and B on the basis of $|H\rangle/|V\rangle$ and $|+\rangle/|-\rangle$ conditioned on

two-photon coincidence events of $|+\rangle_1|+\rangle_2$. Fourfold coincidence counts are analyzed with a commercial multi-channel time analyzer (Agilent U1051A). By setting the pulse width for the pumping beam to 50 or 300 ns, we measure the visibilities for different coincidence time windows. For frequency-correlated photon pairs, the polarization visibility after entanglement swapping drops as the coincidence time window gets larger [23]. For photon pairs without frequency correlation, the visibility will stay constant. The experimental result is shown in Fig. 2, which shows clearly that, under the pulse width of 50 ns, frequency correlation is eliminated very well. Thus, for all remaining measurements, the pulse width for the pump is set to this value. Under the coincidence time window of 300 ns, the two-photon polarization visibilities in the $|H\rangle/|V\rangle$ and $|+\rangle/|-\rangle$ bases are 0.89(3) and 0.80(4), respectively, which are much higher than the threshold of 0.71 to violate the Bell-state Clauser-Horne-Shimony-Holt inequality [26]. There are several origins for the imperfect visibilities, including a slight difference of $f(t)$ and $g(t)$ due to linewidth mismatch between the two CSPDC sources, contribution of multipair events from each CSPDC, imperfect spacial overlapping on the PBS, etc.

Next, we set ω_a and ω_b to be different by changing the acousto-optic modulator working frequencies for the pumping beams as shown in Fig. 1. To compensate the

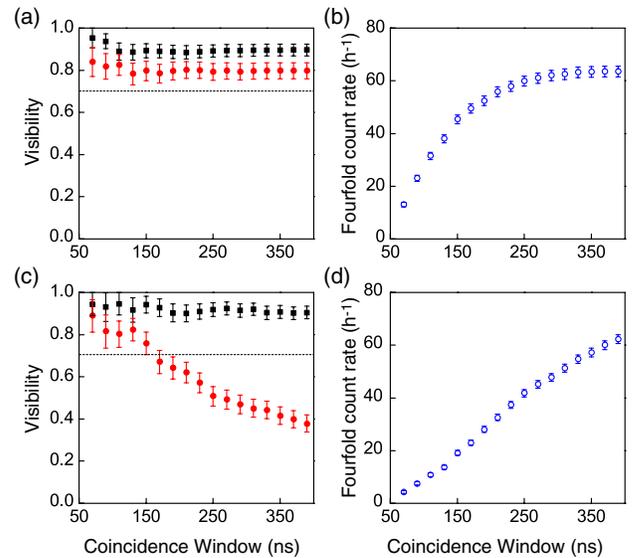


FIG. 2 (color online). Measured correlation visibilities and fourfold count rates as a function of the coincidence time window. (a),(b) Pumping pulse width is set to 50 ns. (c),(d) Pumping pulse width is set to 300 ns. Data points in black filled squares are measured in the basis of $|H\rangle/|V\rangle$, and data points in red filled circles are measured in the basis of $|+\rangle/|-\rangle$. In this measurement, twofold coincidence count rates are 310 and 190 s^{-1} , respectively, for the two CSPDC sources. Error bars represent statistical errors.

random phase shift $\Delta\omega\Delta t$ in Eq. (2), we make use of a home-built time-to-amplitude converter (TAC) and a fast Pockels cell. The home-built TAC converts the detection-time difference into a voltage amplitude, which is fed into the high-voltage driver for the Pockels cell. The systematic time delay of this feedback system is about 360 ns and we use a fiber loop of 150 m to compensate it. The output state becomes the desired state of $|H\rangle_A|H\rangle_B \mp |V\rangle_A|V\rangle_B$ when the phase modulated for photon A by the Pockels cell is equal to $-\Delta\omega\Delta t$. Since the TAC is unipolar and Δt fluctuates in the range of ± 150 ns, we add a fixed delay of 150 ns plus an adjustable time delay δt for the electronic output signal of D_2 . Thus, the actual phase modulated by the Pockels cell is $\Delta\omega(150 \text{ ns} + \delta t - \Delta t)$. In order to measure the entanglement quality after entanglement swapping and to test the validity of this feed-forward system, we measure the A - B correlation visibilities in the basis of $|+\rangle/|-\rangle$ for a series of δt points conditioned on the two-photon coincidence events of $|+\rangle_1|+\rangle_2$. As a comparison, we also make the same measurement for the case without active phase feed forwarding. Both experimental results are shown in Fig. 3. It clearly shows that the $|+\rangle/|-\rangle$ visibilities are recovered by using active phase feed forward. Fitted visibilities for $\Delta\omega = 2\pi \times 40$ MHz and $\Delta\omega = 2\pi \times 80$ MHz are 0.77(4) and 0.80(5), respectively. These results are similar to the $|+\rangle/|-\rangle$ visibilities observed for the same frequency case ($\Delta\omega = 0$), which implies that the main limitations for the visibilities are still due to our imperfect CSPDC

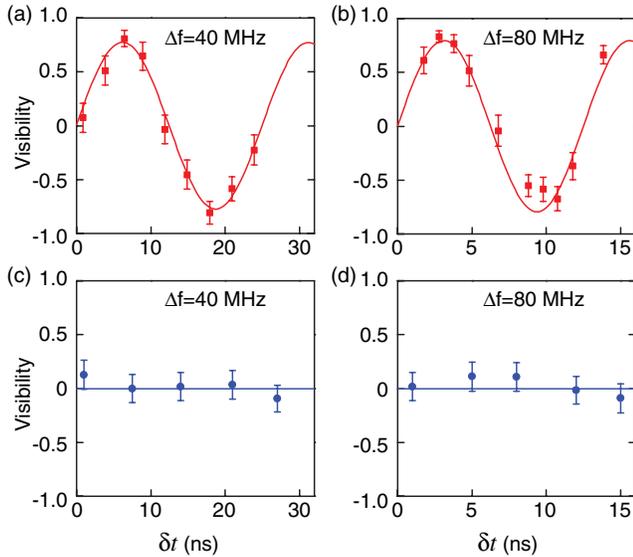


FIG. 3 (color online). Visibilities in the basis of $|+\rangle/|-\rangle$ for photons A and B as a function of detection time delay δt . (a),(b) Active phase feed forwarding is applied. (c),(d) Active phase feed forwarding is not applied. Error bars represent statistical errors. For a given value of $\Delta\omega$, all data points are measured with the same setting for the feed-forward circuitry. In these measurements, the fourfold count rate is 24 h^{-1} and the twofold count rates are 200 and 140 s^{-1} , respectively.

sources. By using sources with better linewidth matching, reducing the excitation probabilities, and optimizing the spacial overlapping, these visibilities could increase significantly.

In order to further and directly verify the entanglement after entanglement swapping, we adopt the method of entanglement witness [27]. As the desired state of photons A and B is $(1/\sqrt{2})(|H\rangle_A|H\rangle_B + |V\rangle_A|V\rangle_B)$, we select an entanglement witness with the form of $W = (1/4)(\hat{I} - \sigma_x \otimes \sigma_x + \sigma_y \otimes \sigma_y - \sigma_z \otimes \sigma_z)$ [28]. If $W < 0$, it implies that photons A and B are in a genuine entangled state [29]. For a maximally entangled state, W gets its minimum value of -0.5 . Experimental results are shown in Fig. 4. When applying the active phase feed forward, the measured results are $W(40 \text{ MHz}) = -0.37(2)$ and $W(80 \text{ MHz}) = -0.35(3)$, respectively, both of which clearly prove that photons A and B are genuinely entangled. In comparison, we also measure the W values for the cases without active phase feed forward, and the results are $W(40 \text{ MHz}) = 0.00(4)$ and $W(80 \text{ MHz}) = 0.02(5)$, respectively, which imply that no entanglement can be detected.

Since commercial single-photon detectors with moderate time resolution (~ 350 ps) have been used in our experiment, the maximal frequency separation allowed is calculated to be ~ 630 MHz, which will allow compensation of the Doppler effect for an airplane. If state-of-the-art fast electronic single-photon detectors (~ 30 ps) [30] are used, entanglement swapping with a frequency separation of ~ 7.3 GHz can be achieved, which will allow entangling dissimilar nitrogen-vacancy centers [10] through two-photon interference or compensating the Doppler effect for a satellite. While sum frequency generation with ultrafast laser pulses enables photon detection much faster than electronics, time

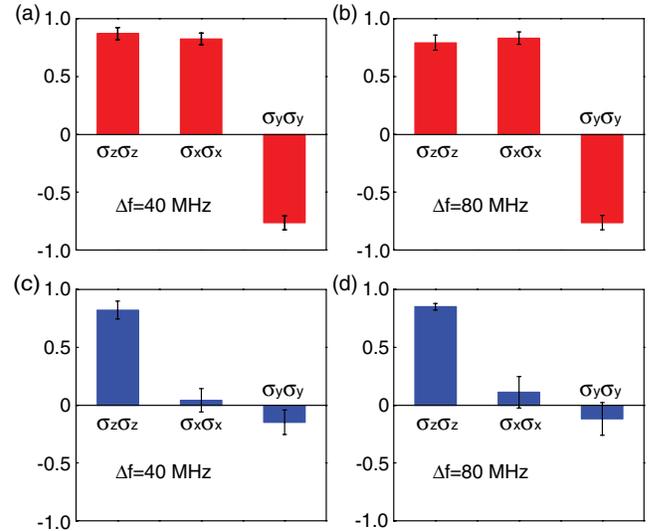


FIG. 4 (color online). Measurement results of $\sigma_i \otimes \sigma_i$ for photons A and B with $i = x, y, \text{ and } z$. (a),(b) Active phase feed forward is applied. (c),(d) Active phase feed forward is not applied. Error bars represent statistical errors.

resolution of ~ 150 fs has been demonstrated by Kuzucu *et al.* [31]. If such a detection technique is used, it will enable entanglement swapping with a frequency separation as large as ~ 1.5 THz, which will allow entangling dissimilar quantum dots [9] with our method.

In summary, we have experimentally entangled two different-color photons in polarization by time-resolved measurement and active feed forward. The time-resolved measurement enables us to select entangled states out of otherwise totally mixed states. Active feed forward enables us to compensate a random phase given from the time-resolved measurement. From a fundamental point of view, our experiment shows that two-photon interference entangles not only identical photons but also different-color photons. From a practical point of view, our experiment provides a ubiquitous approach to solving the frequency-mismatch problem for the interconnection of dissimilar quantum systems, and thus may become an essential tool for future quantum networks.

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