Observation of the Preservation of the Two-Photon Coherence in Plasmon-assisted Transmission

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We experimentally study two-photon coherence in plasmon-assisted transmission with a two-photon Mach-Zehnder (MZ) interferometer. Two collinear photons of identical or orthogonal polarization are simultaneously incident on one optically thick metal film, perforated with a periodic array of subwavelength holes. The de Broglie wavelength of plasmon-assisted transmitted photons is measured, which shows that two photons are re-eradiated by the plasmons and the quantum coherence of biphoton is preserved in the conversion process of transforming biphoton to plasmons and then back to biphoton.

PACS numbers: 03.67.Mn, 42.50Dv, 71.36.+c, 73.20.Mf

It has long been observed that there is an unusually high optical transmission efficiency in metal films perforated with a periodic array of subwavelength apertures[1]. Generally, it is believed that metal surface plays a crucial role and the phenomenon is mediated by surface plasmons (SPs) and there is a process of transform photon to surface plasmon and back to photon[2, 3, 4, 5]. In 2002, Ebbesen et al.[6] first addressed the question of whether the entanglement survives in this extraordinary enhancement light transmission. They showed that quantum entanglement of transmitted photon pair can be preserved when they respectively travel through a hole array. Therefore, the macroscopic surface plasmon polarizations, a collective excitation wave involving typically $10^{10}$ free electrons propagating at the surface of conducting matter, have a true quantum nature. Although several theory models[6, 7, 8] have been proposed to explain this quantum coherence preservation, and extraordinary enhancement light transmission through subwavelength apertures in metal plates, up to our knowledge, a prevalent theory has not yet been developed. Many more experiments were preformed for a rounded understanding of this problem[9, 10, 11, 12, 13, 14]. For example, Young’s double-slit experiment is revisited by an experimental and theoretical study of the optical transmission of a thin metal screen perforated by two subwavelength slits, separated by many optical wavelengths[15]. And there is also a demonstration showing the preservation of the energy-time entanglement of a pair of photons after a photon-plasmon-photon conversion[16]. These works combines of two fields of research, namely quantum information and nano-structured metal optics. Apart from its fundamental interest, this offers a possibility to transfer entanglement between photons and condensed-matter, for storing, modulation and even engineering of quantum information with subwavelength metal optics.

Since there is a process of transforming photon to surface plasmon and then back to photon[2, 3, 4, 5], do two simultaneously input collinear photons excite two plasmon modes or only one? Whether or how those plasmon modes interact with each other? How many photons will be emitted and what is the relationship between those photons? With these problems, we simultaneously input two collinear photons of identical or orthogonal polarization into one optically thick metal film, perforated with a periodic array of subwavelength holes. Previous

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FIG. 1: Hole array transmittance as a function of wavelength. The dashed vertical line indicates the wavelength of 702nm used in the experiment. Inset, scanning electron microscope picture of a typical hole array. After subsequently evaporating a 3-nm titanium bonding layer and a 135-nm gold layer onto a 0.5-mm-thick silica glass substrate, a Focused Ion Etching system (FIB, DB235 of FEB Co.) is used to produce cylindrical holes (200 nm diameter) arranged as a square lattice (period 600 nm). The total area of the hole array is $30 \mu m \times 30 \mu m$ and it is actually made up with four hole arrays of $15 \mu m \times 15 \mu m$ area for the technical reason.
studies on the revealed quantum coherence preservation for single photon, whereas our study emphasizes on two-photon quantum coherence. It is observed that two photons are still re-eradiated by the surface-plasmon waves tunnelling through the holes after excited by two incident photons. We employ a Mach-Zehnder (MZ) type two-photon polarization interferometer and measure the de Broglie wavelength of plasmon-assisted transmitted photons. The measured de Broglie wavelength of re-eradiated photons equals that of Fock state [2], which shows that there are two photons re-eradiated and the quantum coherence of biphoton is preserved in the conversion process of transforming biphoton to plasmon and then back to biphoton.

Fig. 1 shows the hole array transmittance as a function of wavelength. The dashed vertical line indicates the wavelength of 702 nm used in our interference experiment. The transmission of the array at 702 nm is about 3.2%, which is much larger than the value of 0.55% obtained from classical theory [17]. After subsequently evaporating a 3-nm titanium bonding layer and a 135-nm gold layer onto a 0.5-mm-thick silica glass substrate, a Focused Ion Beam Etching system (FIB, DB235 of FEB Co.) is used to produce cylindrical holes (200 nm diameter) arranged as a square lattice (period 600 nm). The total area of the hole array is 30 \( \mu \text{m} \times 30 \mu \text{m} \) and is actually made up of four hole arrays of 15 \( \mu \text{m} \times 15 \mu \text{m} \) area for the technical reason. From a calculation based on the geometry of the array and the optical constants of gold and glass, 702 nm wavelength light is associated with the surface plasmon modes (0,±1) or (±1,0) on the glass-metal interface. The metal plate is set between two lenses of 35 cm focal length, so that the light is normally incident on the hole array, where it has a cross section diameter of about 20\( \mu \text{m} \) and covers hundreds of holes.

The standard method of type-II collinear spontaneous parametric down-conversion is employed to generate a pair of degenerate photons in 702 nm wavelength, as shown in Fig. 2. A 400 mW continuous-wave Ar\(^{+}\)-Laser at the wavelength of 351 nm is directed onto a type-II 2.0mm thick BBO crystal. The pump laser is vertically polarized and the down-converted photon pair of 702 nm wavelength are in |HV\rangle mode propagating in the same direction. A reflector (351 nm) and an interference filter IF (4-nm bandwidth centered at 702nm) are employed to reflect and filter the pump light (not shown in the figure). A birefringent crystal (BC) is used to make the two collinear down-converted photon pair in the |HV\rangle mode simultaneously arrive at the half wavelength plate (HWP) or the hole array. Silicon avalanche photodiode (APD) photon counters are used to record counts.

Firstly, we consider the case when the metal plate is removed from the twin-lenses. When the first HWP is placed either before or after the twin-lenses, the two collinear down-converted photons in the state |HV\rangle will be changed into the state (|HH\rangle − |VV\rangle)/\( \sqrt{2} \). Then these two photons travel two birefringent crystals, where they get a phase difference \( \Delta \phi \) between horizontal and vertical polarization modes (In the figures below, we use \( \Delta L = \Delta \phi/2\pi \times 702 \text{nm} \) as axis label). Two-photon state is thus in the form of (|HH\rangle − e\(^{−\Delta \phi}\) |VV\rangle)/\( \sqrt{2} \).

FIG. 2: Experimental set-up. We pump a type-II 2.0mm thick BBO crystal with an Ar\(^{+}\)-Laser at 351nm. The optical pump power is approximately 400 mW and the pump laser is vertically polarized. Via spontaneous parametric down-conversion (SPDC) pairs of photons(702nm) in HV polarization are produced. Reflector (351 nm) and Interference filter IF (4-nm bandwidth centred at 702nm) are used to reflect the pump light (not shown in the figure). The effect of the pump photon on the background can be neglected. Birefringent crystals(BC) are used to make the photon pairs arrive at the hole array(HA) in the same time. A HWP(at 702nm) is placed first before and then after the twin-lenses. We control the phase difference in the interferometer by rotating a twin-birefringent crystal(T-BC). Coincidences in the HV basis and HH basis (separately shown) are recorded separately.

FIG. 3: (a) and (b) show the coincidences in the HV basis and HH basis after the PBS respectively. The metal plate is removed from the twin-lenses. Case 1(square dots), the first HWP is placed before the twin-lenses. The solid line comes from a fit of the experimental data with \( R_{|HV|,|VV|} = a/\sqrt{4(1 + \cos(2\Delta \phi))} \) using least square method. Case 2(round dots), the first HWP is placed after the twin-lenses. The solid line comes from a fit of the experimental data with \( R_{|HV|,|VV|} = a/\sqrt{8(1 - \cos(2\Delta \phi))} \) using least square method. It can be seen that twin lenses without metal plate has little effect to the interference patterns.
After passing the second HWP (at 702nm), their state are transformed into 
\[
\frac{1}{\sqrt{2}}\{(|H\rangle - e^{i2\Delta \phi}|V\rangle)\}
\]
Then the two photons are separated by a polarization beam splitter (PBS). The coincidence click of the two detectors directly behind PBS projects these photons in the basis of different polarization (|HV\rangle or |VH\rangle due to the indistinguishability of the two photons). The experiment result is shown in Fig. 3a, which fits nicely with the theoretical interference pattern of the coincidence rates in the |HV\rangle and |VH\rangle basis.

\[
R_{HV(VH)} = \frac{1}{4}(1 + \cos(2\Delta \phi)).
\]  
(1)

In addition, we can place a lossless 50%-50% beam splitter with two detectors behind one output port of the PBS and then project photons in the |HH\rangle basis. The experiment results is shown in Fig. 3b, which also fits well the theoretical interference patterns of the coincidence rates.

\[
R_{HH} = \frac{1}{8}(1 - \cos(2\Delta \phi)).
\]  
(2)

The visibility of these interference patterns is about 93%. These results show that this Mach-Zehnder (MZ) type interferometer can directly measure the de Broglie wavelength of collinear polarization photons. The de Broglie wavelength of the down-converted collinear biphoton is \(\lambda/2\), agreeing with the Fock state \(|2\rangle\) result. It can also be seen that two lenses without metal plate has little effect to the interference patterns.

Now we put the metal plate with hole array between the twin lenses. First of all, we study the case when the two collinear photons of identical polarization simultaneously incident on the hole array. In this case, the first HWP is placed before the two lenses containing the metal plate and changes the two photons from the state |HV\rangle to the state \(|HH\rangle - |VV\rangle)/\sqrt{2}\). Then these two collinear photons in the state \(|HH\rangle - |VV\rangle)/\sqrt{2}\) simultaneously arrive the hole array and excite surface plasmon. Similar to the case without metal plate, the zero order reradiated photons, namely, the surface plasmon assisted transmission photons, pass through a twin birefringent crystal assembly, and get a phase difference between horizontal and vertical polarization modes, before travelling to the second HWP and being detected. Fig. 4a shows the interference pattern of the coincidence rate between the two detectors directly behind PBS projects. This corresponds to project them in the |HV\rangle or |VH\rangle basis. The resultant interference pattern is very similar to the case that metal plate is removed, and can be nicely fitted to

\[
R_{HV(VH)} = \frac{1}{4}(1 + \cos(2\Delta \phi + 0.98\pi))
\]  
(3)

Similarly, we also place a lossless 50%-50% beam splitter behind one output port of the PBS and project these photons in |HH\rangle basis. The interference pattern is shown in Fig. 4b, which corresponds to

\[
R_{HH} = \frac{1}{8}(1 - \cos(2\Delta \phi + 0.93\pi))
\]  
(4)

The visibility of these two interference patterns is about 92%. We can conclude the de Broglie wavelength of reradiated photons in this case is \(\lambda/2\). Two photons are reradiated by the surface plasmon, excited simultaneously by two collinear photons of identical polarization. The quantum coherence of the biphoton is preserved in this biphoton \(\rightarrow\) surface plasmon \(\rightarrow\) biphoton conversion.

Secondly, we study the case when the two photons simultaneously exciting surface plasmons have different polarization, by placing the first HWP behind the metal plate. The down-converted collinear photon pair in |HV\rangle

\[\text{FIG. 4: (a) and (b) show the coincidences in the HV basis and HH basis after the PBS respectively. The metal plate is placed between the twin lenses, and the first HWP is placed before the twin-lenses, so the photon pair acts on the hole array is in HHI-VV polarization.}\]

\[\text{FIG. 5: (a) and (b) show the coincidences in the HV basis and HH basis after the PBS respectively. The metal plate is placed between the twin lenses, and the first HWP is placed behind the twin-lenses, so the photon pair acts on the hole array is in HV polarization.}\]
modes are directly incident on the hole array. And the re-eradiated photons by the surface plasmons, simultaneously excited by two photons of orthogonal polarization, travel through the Mach-Zehnder (MZ) type interferometer of collinear polarization photons so that their de Broglie wavelength is measured. The resultant interference patterns of projection in $|HV\rangle$ and $|HH\rangle$ basis are respectively shown in Fig. 5a and Fig. 5b, which can also be nicely fitted respectively with

$$R_{HV(VH)} = \frac{1}{4} \left(1 + \cos(2\Delta \phi - 0.12\pi)\right)$$

$$R_{HH} = \frac{1}{8} \left(1 - \cos(2\Delta \phi - 0.13\pi)\right).$$

The visibility is also about 92%. The de Broglie wavelength of re-eradiated photons is still equal to that of Fock state $|1\rangle$. There are still two photons re-eradiated by the surface plasmon, excited simultaneously by two collinear photons of orthogonal polarization. The wave packets of the two photons are kept superposed coherently in the transmission of biphoton in $|HV\rangle$ mode.

If there is any decoherence in the plasmon-assisted transmission, it corresponds to adding a random phase to the transmitted photons. In the Ref.1, two photons in $(|HV\rangle - |VH\rangle)/\sqrt{2}$ mode are non-collinear. When only one photon travelling through the hole array, it may add a phase $\Delta \phi_{12}(t)$ (or $\Delta \phi_{21}(t)$) to the single photon in $|H\rangle$ (or $|V\rangle$) mode, the experiment result of entanglement measurement shows that $\Delta \phi_{H}(t) - \Delta \phi_{V}(t) = \text{constant}$. In the present experiment, two collinear photons simultaneously incident on the hole array and excite surface plasmon. When the incident two photons are in $(|HH\rangle - |VV\rangle)/\sqrt{2}$ mode, the decoherence corresponds to adding a phase of $\Delta \phi_{HH}(t)$ (or $\Delta \phi_{VV}(t)$) to the biphoton in $|H\rangle$ (or $|V\rangle$) mode. Due to the possible interaction between the surface plasmons excited two photons, there is no clear relationship between $\Delta \phi_{HH}(t)$ (or $\Delta \phi_{VV}(t)$) and $\Delta \phi_{H}(t)$ (or $\Delta \phi_{V}(t)$). The present de Broglie wavelength measurement shows that $\Delta \phi_{HH}(t) - \Delta \phi_{VV}(t)$ is a constant value. To determine the exact value of $\Delta \phi_{HH}(t)$ and $\Delta \phi_{VV}(t)$ (or $\Delta \phi_{H}(t)$ and $\Delta \phi_{V}(t)$), a Mach-Zehnder (MZ) interference measurement of biphoton (or single photon) with separate paths is needed. However, it is still impossible to determine the random phase $\Delta \phi_{HV}(t)$ in the decoherent transmission of biphoton in $|HV\rangle$ even the MZ interferometer with separate paths.

We note that there is a $\pi$-phase ($1.10\pi$ and $1.06\pi$ for HV coincidence and HH coincidence respectively) between the case when the two photons is first transformed into the state $(|HH\rangle - |VV\rangle)/\sqrt{2}$ by the first HWP before incident on the metal plate, and the case when the two photons in the state $|HV\rangle$ first excite the hole array before travelling to the first HWP. Although the detailed mechanism for it is not definitely identified, we speculate that it is not the decoherence of hole array, but may be accounted by the fact that our metal plate is actually made up of four hole arrays of $15\mu m \times 15\mu m$ area and there is a mismatch and in-homogeneity among different hole arrays. Thus this metal plate has an effect similar to $\lambda/4$ birefringent plate and can cause $\pi/2$ phase difference between horizontal and vertical polarization mode. When the two photons in the state $(|HH\rangle - |VV\rangle)/\sqrt{2}$ travel the metal plate, this birefringent effect add a phase $\pi$ between $|HH\rangle$ and $|VV\rangle$ modes so that the re-eradiated two photons will be in the state $(|HH\rangle - e^{i\pi}|VV\rangle)/\sqrt{2}$. For the case that two photons in $|HV\rangle$ mode pass the metal plate, the re-eradiated photons will be in $e^{i\pi/2}|HV\rangle$. This global phase has no effect to the de Broglie wavelength measurement. Actually, we have checked this conjecture by the single photon polarization state tomography measurement, which shows the $|H\rangle + |V\rangle$ (or $|H\rangle - |V\rangle$) mode photon excited plasmon will re-eradiate photon in $|H\rangle - i|V\rangle$ (or $|H\rangle + i|V\rangle$) mode. Surprisingly, such birefringent effect of hole array is not mentioned in the previous experiments. In another metal plate with hole array fabricated in the same FIB procedure, phase birefringent effect of about $\pi$ is observed. Much more work is still needed to fully understand the birefringent effect of the metal plate with medley hole array.

In conclusion, we have measured the de Broglie wavelength of re-eradiated photons of the surface plasmons simultaneously excited by two collinear photons of either identical or orthogonal polarization, which shows that two photons are re-eradiated and the quantum coherence of the biphoton is preserved in the conversion process of transforming biphoton to plasmon and then back to biphoton. The decoherence in two $|HH\rangle$ and $|VV\rangle$ mode photons transmission, if there is any, cause similar random phases ($\Delta \phi_{HH}(t) - \Delta \phi_{VV}(t) = \text{constant}$). These results may give us more hints to the understanding of the plasmon-assisted transmission of photons.

In the preparation of the present manuscript, we note that there is an experiment showing quantum superposition and entanglement of mesoscopic plasmons excited by time-bin entangled photons.

Acknowledgments

This work was funded by the National Fundamental Research Program (2001CB309300), National Nature Science Foundation of China (10304017), the Innovation Funds from Chinese Academy of Sciences. ZYO is also supported by the US National Science Foundation under Grant No.0245021.

[1] T. W. Ebbesen, et. al. Nature 391, 667 (1998).
[2] H. Raether, Surface Plasmons on Smooth and Rough Surfaces and on Gratings, Springer Tracts in Modern Physics, Springer, Berlin, 1988 Vol. 111.
[3] R. H. Ritchie, Phys. Rev. 106, 874 (1957).
[4] D. E. Grupp, et. al. Appl. Phys. Lett. 77 1569 (2000).
[5] M. Moreno, et. al. Phys. Rev. Lett. 86, 1114 (2001).
[6] E. Altwischer, et. al. Nature 418 304 (2002).
[7] E. Moreno, et. al. Phys. Rev. Lett. 92 236801 (2004).
[8] J. L. van Velsen, et. al. Phys. Rev. A 68, 043807 (2003).
[9] H. F. Ghaemi, et. al. Phys. Rev. B 58 6779 (1998).
[10] K. G. Lee and Q-Han Park, Phys. Rev. Lett. 95 103902 (2005).
[11] W. L. Barnes, et. al. Phys. Rev. Lett. 92 107401 (2004).
[12] F. Intravaia and A. Lambrecht, Phys. Rev. Lett. 94 110405 (2005).
[13] K. J. K. Koerkamp, et. al. Phys. Rev. Lett. 92 183901 (2004).
[14] T. A. Kelf, et. al. Phys. Rev. Lett. 95 116802 (2005).
[15] H. F. Schouten, et. al. Phys. Rev. Lett. 94 053901 (2005).
[16] S. Fasel, et. al. Phys. Rev. Lett. 94 110501 (2005).
[17] H. A. Bethe, Phys. Rev. 66, 163”C182 (1944).
[18] G. P. Guo, et. al. in preparation.
[19] S. Fasel, quant-ph/0512022