Monotonic quantum-to-classical transition enabled by positively-correlated biphotons

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(Dated: July 3, 2017)

Multiparticle interference is a fundamental phenomenon in the study of quantum mechanics. It was discovered in a recent experiment [Ra, Y.-S. et al, Proc. Natl Acad. Sci. USA 110, 1227(2013)] that spectrally uncorrelated biphotons exhibited a nonmonotonic quantum-to-classical transition in a four-photon Hong-Ou-Mandel (HOM) interference. In this work, we consider the same scheme with spectrally correlated photons. By theoretical calculation and numerical simulation, we found the transition not only can be nonmonotonic with negative-correlated or uncorrelated biphotons, but also can be monotonic with positive-correlated biphotons. The fundamental reason for this difference is that the HOM-type multi-photon interference is a differential-frequency interference. Our study may shed new light on understanding the role of frequency entanglement in multi-photon behavior.

PACS numbers: 42.50.St, 03.65.Ud, 42.65.Lm, 42.50.Dv

I. INTRODUCTION

Indistinguishability plays an important role in multi-photon interference, which is a fundamental phenomenon in the study of quantum mechanics [1-6]. It was believed that, with the increase of indistinguishability, the multi-photon interference pattern changes monotonically [1]. For example, in the case of Hong-Ou-Mandel (HOM) interference demonstrated in 1987 [7], the two-fold coincidence counts show a monotonic increase when the time delay scanned from zero to infinite. This HOM interference can be interpreted from the viewpoint of indistinguishability: with the increasing of the time delay, the temporal distinguishability (or the decoherence) of the biphoton was also increasing and leading to a quantum-to-classical transition [1-4]. Such a monotonic indistinguishability dependence was also observed in the case of four-photon [10] and six-photon [11] HOM-type interference, where all photons are detected in one output port of the beamsplitter.

However, recent works [1,12,13] both that such monotonic quantum-to-classical transition was only an exception, i.e., only valid for two-photon cases and for bunching detection in multi-photon cases. For example, in the four-photon HOM-type experiment [1], where two pair of biphotons were sent to two input ports of a 50:50 beamsplitter and four detectors were prepared at the two output ports (see Fig.1), by changing the detection schemes, different interference patterns can be obtained: in a 2/2 detection (with two detectors at one output port and two detectors at the other port, shown in Fig. 1(a)), the four-fold coincidence counts showed a nonmonotonic indistinguishability dependence; in contrast, the 4/0 detection scheme as shown in Fig. 1(c), achieved a monotonic dependence. This study on the transition between quantum and classical in Ref. [1] is important for deeper understanding of the multi-particle behavior in quantum mechanics.

The interesting phenomenon in Ref. [1] was realized by spectrally uncorrelated biphotons. Now a question comes naturally: what phenomenon will be if the biphotons are spectrally correlated? In other words, with the introduction of frequency entanglement, will the interference patterns, especially the monotonicity dependence, be changed? To answer this question, in this paper, we consider the same scheme with spectrally correlated (frequency entangled) biphotons. It will be seen that spectrally correlated biphotons show different interference patterns from the patterns by uncorrelated biphotons. For example, under the 2/2 detection scheme, the spectrally negative- and non-correlated biphotons shows a nonmonotonic dependence, while the spectrally positively-correlated biphotons shows a monotonic dependence. In contrast, the monotonicity is not affected by the spectral correlation in the 4/0 and 3/1 detection schemes.

This paper is organized as follow: in the Introduction section, we provide the background and motivation of this research. Then, in the Theory section we develop a multi-mode theory for four-photon HOM-type interference, where the spectral correlation between the signal and idler photons are concerned. Next, in the Analysis

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section, we first simulate the HOM-type interference patterns using biphotons with three different spectral correlations: no-correlation, positive-correlation, negative-correlation. Then, we provide comprehensive discussions on the simulation results. Finally, we summarize the paper in the Conclusion section. More details for the derivation of the relative equations are given in the Appendix.

II. THEORY

In this paper, we consider a four-photon HOM-type interference with the experimental model shown in Fig. 1. The four photon state $|\psi\rangle$ is generated from the two-pair components in a spontaneous parametric downconversion (SPDC) process.

$$|\psi\rangle = \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \times \hat{a}^\dagger_1(\omega_1) \hat{a}^\dagger_2(\omega_2) \hat{a}^\dagger_3(\omega_3) \hat{a}^\dagger_4(\omega_4) \{0000\},$$  \hspace{1cm} (1)

where $\hat{a}^\dagger(\omega)$ is the creation operator at angular frequency $\omega$, the subscripts $s$ and $i$ denote the signal and idler photons from the first pair, while $s'$ and $i'$ denote the signal and idler photons from the second pair. $f(\omega_1, \omega_2)$ and $f(\omega_3, \omega_4)$ are their joint spectral amplitude (JSA).

As calculated in detail in the Appendix, the fourth-fold coincidence probability $P_{22}(\tau)$ in the 2/2 detection scheme is

$$P_{22}(\tau) = \frac{1}{12} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{22}(\tau)|^2,$$  \hspace{1cm} (2)

with

$$|I_{22}(\tau)|^2 = |(f_{13} f_{42} + f_{14} f_{23}) e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{31} f_{42} + f_{32} f_{41}) e^{-i\omega_3 \tau} e^{-i\omega_4 \tau} - (f_{12} f_{34} + f_{14} f_{32}) e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{12} f_{43} + f_{13} f_{42}) e^{-i\omega_1 \tau} e^{-i\omega_4 \tau} - (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_4 \tau} - (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} |^2,$$  \hspace{1cm} (3)

where $f_{mn} = f(\omega_m, \omega_n)$ and $\omega_{mn}(m,n)=1, 2, 3, 4$ is the frequency of the detection field for the detectors $D_n$.

The coincidence probability $P_{31}(\tau)$ in the 3/1 detection scheme is

$$P_{31}(\tau) = \frac{1}{128} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{31}(\tau)|^2,$$  \hspace{1cm} (4)

with

$$|I_{31}(\tau)|^2 = | - (f_{13} f_{42} + f_{14} f_{23}) e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{31} f_{42} + f_{32} f_{41}) e^{-i\omega_3 \tau} e^{-i\omega_4 \tau} - (f_{12} f_{34} + f_{14} f_{32}) e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} + (f_{12} f_{43} + f_{13} f_{42}) e^{-i\omega_1 \tau} e^{-i\omega_4 \tau} - (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} + (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_4 \tau} |^2.$$

The coincidence probability $P_{40}(\tau)$ in the 4/0 detection scheme is

$$P_{40}(\tau) = \frac{1}{1024} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{40}(\tau)|^2,$$  \hspace{1cm} (6)

with

$$|I_{40}(\tau)|^2 = |(f_{13} f_{42} + f_{14} f_{23}) e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{31} f_{42} + f_{32} f_{41}) e^{-i\omega_3 \tau} e^{-i\omega_4 \tau} + (f_{12} f_{34} + f_{14} f_{32}) e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} + (f_{12} f_{43} + f_{13} f_{42}) e^{-i\omega_1 \tau} e^{-i\omega_4 \tau} + (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} + (f_{23} f_{41} + f_{24} f_{31}) e^{-i\omega_2 \tau} e^{-i\omega_4 \tau} |^2.$$

It is interesting to compare the six items in $|I_{22}(\tau)|^2$, $|I_{31}(\tau)|^2$ and $|I_{40}(\tau)|^2$: the first and second terms in $|I_{22}(\tau)|^2$ are positive; the second, fourth and sixth items in $|I_{31}(\tau)|^2$ are positive; all the six items in $|I_{40}(\tau)|^2$ are positive. As calculated in the Appendix, the sign of these terms results from the sign of the transmission and reflection terms after the beam splitter (BS) in Fig. 1. These equations can be further simplified by assuming the exchanging symmetry of $f(\omega_s, \omega_i) = f(\omega_i, \omega_s)$.

III. ANALYSIS

For a given JSA of $f(\omega_s, \omega_i)$, using the equations of $P_{22}(\tau)$, $P_{31}(\tau)$ and $P_{40}(\tau)$, it is possible to simulate the HOM-type interference patterns. Three kinds of JSAs are shown in Fig. 2(a1-c1), with (a1) spectrally uncorrelated, (b1) positively-correlated and (c1) negatively-correlated. Without the loss of generality, we set the center wavelength of the JSAs at 1584 nm, and set the bandwidth (full width at half maximum) of the signal and idler photons at 2 nm. Although the shape of the three JSAs is different, the marginal distributions for the signal and idler photons are the same. In other words, from the viewpoint of single photons, all the signal and idler...
FIG. 2: (color online) Three different JSA $f(\omega_s, \omega_i)$: (a1) uncorrelated, (b1) negatively-correlated and (c1) positively-correlated. The corresponding HOM-type interference patterns are shown in (a2-c4): (a2-c2) are for 2/2 detection scheme; (a3-c3) are for 3/1 detection scheme; (a4-c4) are for 4/0 detection scheme. All the y axes in (a2-c4) are normalized.

photon have the same spectral distribution in Fig 2a1-c1).

Figure 2a2-c2 show the HOM-type interference patterns for 2/2 detection schemes. It is noteworthy that, for the uncorrelated state (a1) and negatively correlated state (b1), the coincidence probability changes in a non-monotonic manner, when the time delay changes from 0 to 10 ps. In contrast, the positively correlated state (c1) shows a monotonic interference pattern. Figure 2a3-c3 show the HOM-type interference patterns for 3/1 detection schemes, with all the figures in dips, i.e., the interference patterns are monotonic when the time delay changes from 0 to infinite. The patterns for 4/0 detection are shown in Fig. 2a4-c4, with all the figures in bumps, i.e., the interference patterns show monotonic dependence.

In Fig 2, biphotons with different correlations show different interference patterns, but what is the underlying physics for such phenomena? To answer this question, we need to further simplify the Eqs. 3 4 7. As an example, by assuming $f_{mn} = f_{nm}$, Eq. 3 can be simplified as

\[ |I_{22}(\tau)|^2 = (f_{12}f_{34})^2 + (f_{13}f_{24})^2 + (f_{14}f_{23})^2 + f_{12}f_{34} \rho_{14}\rho_{23} f_{13}f_{24} f_{14}f_{23} \]

Obviously, Eq. 3 is a function of $\omega_m - \omega_n$. Similar results can also be derived for Eq. 5 and Eq. 7. So, it can be concluded that HOM-type multi-photon interference is a differential-frequency interference. This is true not only for the two-photon HOM interference 14-16, but also for the four-photon HOM interference. Therefore, positively-correlated biphotons, i.e., around $\omega_s - \omega_i = 0$, exhibit different patterns from the one by the uncorrelated biphotons ($\omega_s$ and $\omega_i$ are arbitrary) or negatively-correlated biphotons ($\omega_s + \omega_i = \omega_p$, with $\omega_p$ as the angular frequency of the pump).

The interference patterns in Fig 2c2-c4 are “fatter” (the coherence time is longer) than the patterns in (a2-a4) or (b2-b4). It can also be explained from the above conclusion that HOM type interference is a differential-frequency interference. In fact, Eq. 3 can be viewed as a Fourier transform from frequency-domain to time domain. Consequently, the width of the time-domain-interference-pattern is determined by the spectral-domain distribution along the direction of ($\omega_s - \omega_i$). The value of ($\omega_s - \omega_i$) in Fig 2c1 is the smallest among (a1-c1) in frequency domain, so the corresponding width in the interference patterns are the largest in time domain, thanks to the spectral positive-correlation in (c1).

It should be emphasized that the theoretical model of our scheme is different from the model in Refs. 11 12, where the spectral correlations are not included. The photons in the model of Refs. 11 12 17 is spectrally uncorrelated, therefore, the experiment results in Refs. 11 12 only correspond to Fig 2a2, a3, a4 in our simulation.

Many literatures have been dedicated to theoretically analyze the multi-photon interference using multi-mode theory. Ou et al analyzed the multi-photon interference using multi-mode theory from spectral modes 2 10 11; Chen et al modeled the photons as wave packets in time domain 13: Ra et al considered Schmidt decomposition on the temporal modes of the photons in their theoretical model 11 12 13 However, in all these theoretical model, the role of spectral correlation is not deeply investigated. To the best of our knowledge, our model is the first theoretical model for multi-photon interference with spectral correlation included.

It is interesting to compare the four-photon HOM in-
terference with the case of the traditional two-photon HOM interference \cite{7, 20}. The two-fold coincidence probability between two output ports of a beamsplitter (anti-bunching test) can be written as

$$P_{11}(\tau) = \frac{1}{4} \int_{0}^{\infty} d\omega_1 d\omega_2 |I_{11}(\tau)|^2,$$  \hspace{1cm} (9)

with

$$|I_{11}(\tau)|^2 = |f(\omega_2, \omega_1)e^{-i\omega_1 \tau} - f(\omega_1, \omega_2)e^{-i\omega_2 \tau}|^2 \hspace{1cm} (10)$$

In contrast, the two-fold coincidence probability of one output port of the beamsplitter (bunching test) can be written as

$$P_{20}(\tau) = \frac{1}{16} \int_{0}^{\infty} d\omega_1 d\omega_2 |I_{20}(\tau)|^2,$$  \hspace{1cm} (11)

with

$$|I_{20}(\tau)|^2 = |f(\omega_2, \omega_1)e^{-i\omega_1 \tau} + f(\omega_1, \omega_2)e^{-i\omega_2 \tau}|^2 \hspace{1cm} (12)$$

We also simulated $P_{11}(\tau)$ and $P_{20}(\tau)$ using the three JSA s shown in Fig. 2(a1-c1). It was found that the monotonicity was not affected by the spectral correlations, i.e., all the three $P_{11}(\tau)$ patterns show dips, while all the three $P_{20}(\tau)$ patterns show bumps for the JSA s in Fig. 2(a1-c1).

It is also important to rethink the prerequisite condition for 100% visibility in the two-photon and four-photon HOM interference. In the two-photon case, exchanging symmetry of $f(\omega_1, \omega_2) = f(\omega_2, \omega_1)$ is required to achieve 100% visibility, i.e., $P_{11}(0) = 0$ \cite{21, 21}. In contrast, the prerequisite condition is complex for the four-photon HOM interference to achieve 100% visibility. For example, in the case of 3/1 detection, $P_{31}(0) = 0$ implies $-(f_{13}f_{23} + f_{14}f_{24}) + (f_{31}f_{42} + f_{32}f_{43}) - (f_{12}f_{31} + f_{14}f_{34} + f_{13}f_{42}) - (f_{23}f_{43} + f_{24}f_{34}) + (f_{21}f_{34} + f_{23}f_{41}) = 0$, which is an upgraded version of the exchanging symmetry for the four-photon case.

In the theoretical model in Eq. (1), the four-photon state is generated from a double pair emission, which has a spectral distribution of $f(\omega_s, \omega_i)f(\omega_s', \omega_i')$. In the future, it is possible to directly generate a four-photon state with a spectral distribution of $f(\omega_s, \omega_i)f(\omega_s', \omega_i')$. This state may be generated from, say, a fourth-order spontaneous parametric down conversion process, where a higher-energy photon “splits” into four lower-energy photons. For example, a 1600 nm photon may be downconverted to four 400 nm photons. This is the inverse process of a fourth harmonic generation. The direct generation of three-photon state has been chased by several groups for a long time \cite{22, 25}. It is also interesting to study the case of four-photon state \cite{26, 26}. In this case, the spectral correlations and the HOM interference might be different from the case discussed in this paper. It will be an interesting topic to investigate in the future. Another future work is to expand the theoretical model to the case of six-photon and more photons. Although the equations might be complex, the expansion method is direct, i.e., similar as what we did in the work.

For the future experimental demonstration, our scheme has been ready to be realized with the state-of-art technologies. The spectrally uncorrelated JSA in Fig. 2(a1) can be generated by filtering a PPKTP downconversion source at 1584 nm \cite{23, 31}. The spectrally negatively correlated JSA in Fig. 2(b1) has been generated in a ps-pulse-pumped PPSLT crystal \cite{21}, while the spectrally positively correlated JSA in Fig. 2(c1) has been prepared in a fs-pulse-pumped PPKTP crystal \cite{21}. For detection, we can use the similar setup demonstrated recently \cite{52}.

Our work have several applications in the future. Higher-order correlations in many-body system are very important for characterizing a quantum system and became to be a hot topic in study of quantum optics \cite{17, 33, 34}. In this work, we studied the role of spectral correlation in a four-photon quantum interference, which actually corresponds to a fourth-order temporal correlation in a four-body system. Therefore, this work may make contribution to the deep understanding of higher-order correlations of a quantum system. Another possible application of our work is for quantum sensing based on Hong-Ou-Mandel interference \cite{33, 37} . Thirdly, the spectral correlation may be applied to the reduction of detection noise in a dispersive medium, which has been recently demonstrated in Ref. \cite{38} with only two photons. In the case of four photons, the noise-reduction effect might be enhanced.

IV. CONCLUSION

In conclusion, we have investigated the role of spectral correlation (frequency entanglement) in quantum-to-classical transition in a four-photon Hong-Ou-Mandel interference. By theoretical calculation and numerical simulation based on a multi-mode theory for spectrally correlated photons, it was found that the transition can be monotonic for positively-correlated biphotons, and can be nonmonotonic negative-, or non-correlated biphotons in the 2/2 detection scheme. In contrast, the mononicity was not changed in the 3/1 and 4/0 detection schemes. The fundamental reason for these difference is: the HOM-type interference is a differential-frequency interference. Our theoretical scheme can be easily demonstrated in experiment using the state-of-art technologies. This study may shed new light on understanding the role of entanglement in multi-photon behavior.

Acknowledgements

The authors are grateful to M. Takeoka for helpful discussions. R.-B. J. is supported by Fund from the Educational Department of Hubei Province, China (Grant No. D20161504). C. L. R. is supported by Youth Innovation Promotion Association (CAS) No.2015317,
National Natural Science Foundations of China (Grant No.11605205), Natural Science Foundations of Chong Qing (No.cstc2015jcyjA00021). H. J. is supported by National Natural Science Foundations of China (Grant No. 11474087).

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Here we deduce the equations for the four photon Hong-Ou-Mandel (HOM) type interference in detail. The setup of HOM interference with 2/2 detection scheme is shown in Fig 4(a). The two-pair component from a spontaneous parametric down conversion (SPDC) process is expressed as (Eq. (1) in the main text).

\[ |\psi\rangle = \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \psi(\omega_1, \omega_2) \psi(\omega_3, \omega_4) \times \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0000\rangle, \quad (13) \]

The meaning of each parameter are explained in the main text. The detection field operator of detector \(D_n\) \((n = 1, 2, 3, 4)\) is

\[ \hat{E}_n^{(+)}(t_n) = \frac{1}{\sqrt{2\pi}} \int_0^\infty d\omega_n \hat{a}_n(\omega_n) e^{-i\omega_n t_n}, \quad (14) \]

where \(\omega_n\) is the frequency of the detection field. \(\hat{a}_n\) is the annihilation operator of the detection field. The transformation rule of a 50/50 beamsplitter is

\[ \frac{1}{\sqrt{2}} (\hat{a}_{n1} + \hat{a}_{n2}) \] and \(\frac{1}{\sqrt{2}} (\hat{a}_{n1} - \hat{a}_{n2})\), where the subscripts \(n1\) and \(n2\) denote the two output ports of the beamsplitter, while the \(in_1\) and \(in_2\) denote the two input ports.

So, we can write the detection fields as

\[ \hat{E}_1^{(+)}(t_1) = \int_0^\infty d\omega_1 [\hat{a}_s(\omega_1) e^{-i\omega_1 \tau} + \hat{a}_i(\omega_1)] e^{-i\omega_1 t_1}, \]
\[ \hat{E}_2^{(+)}(t_2) = \int_0^\infty d\omega_2 [\hat{a}_s(\omega_2) e^{-i\omega_2 \tau} + \hat{a}_i(\omega_2)] e^{-i\omega_2 t_2}, \]
\[ \hat{E}_3^{(+)}(t_3) = \int_0^\infty d\omega_3 [\hat{a}_s(\omega_3) e^{-i\omega_3 \tau} - \hat{a}_i(\omega_3)] e^{-i\omega_3 t_3}, \]
\[ \hat{E}_4^{(+)}(t_4) = \int_0^\infty d\omega_4 [\hat{a}_s(\omega_4) e^{-i\omega_4 \tau} - \hat{a}_i(\omega_4)] e^{-i\omega_4 t_4}, \]

where the phase term \(e^{-i\omega_n \tau}\) is introduced by the time delay \(\tau\). The coincidence probability \(P_{22}\) as a function of delay time \(\tau\) can be expressed as

\[ P_{22}(\tau) = \int dt_1 dt_2 dt_3 dt_4 \times \langle \psi | \hat{E}_1^{(-)}(t_1) \hat{E}_2^{(-)}(t_2) \hat{E}_3^{(+)}(t_3) \hat{E}_4^{(+)}(t_4) | \psi \rangle. \quad (16) \]

First, let us consider the \(\hat{E}_1^{(+)} \hat{E}_2^{(+)} \hat{E}_3^{(+)} \hat{E}_4^{(+)} | \psi \rangle\). For simplicity, the key components can be written as:

\[ [\hat{a}_s(\omega_1) + \hat{a}_i(\omega_1)] [\hat{a}_s(\omega_2) + \hat{a}_i(\omega_2)] [\hat{a}_s(\omega_3) - \hat{a}_i(\omega_3)] [\hat{a}_s(\omega_4) - \hat{a}_i(\omega_4)]. \]

Only 6 out of 16 terms exist: \(\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s, \hat{a}_i \hat{a}_i \hat{a}_i \hat{a}_i, -\hat{a}_s \hat{a}_i \hat{a}_s \hat{a}_s, -\hat{a}_i \hat{a}_s \hat{a}_i \hat{a}_i, -\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_i\), and \(-\hat{a}_i \hat{a}_i \hat{a}_i \hat{a}_s\). The first term \((\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s)\) is

\[ \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \psi(\omega_1) \psi(\omega_2) \psi(\omega_3) \psi(\omega_4) e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} e^{-i\omega_4 \tau} \times \] \[\int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle = \frac{1}{16} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 f(\omega_1, \omega_2) f(\omega_3, \omega_4) \hat{a}_1^\dagger(\omega_1) \hat{a}_2^\dagger(\omega_2) \hat{a}_3^\dagger(\omega_3) \hat{a}_4^\dagger(\omega_4) |0\rangle

\[ f f_1 = [f(\omega_1, \omega_3) f(\omega_2, \omega_4) + f(\omega_1, \omega_4) f(\omega_2, \omega_3)] e^{-i\omega_1 \tau} e^{-i\omega_2 \tau}. \quad (18) \]

In the above calculation, the following relationship is used.

\[ \hat{a}_s(\omega_1) \hat{a}_s(\omega_2) \hat{a}_s(\omega_3) \hat{a}_s(\omega_4) |0\rangle = [\delta(\omega_1 - \omega_2) \delta(\omega_3 - \omega_4) + \delta(\omega_1 - \omega_3) \delta(\omega_2 - \omega_4)] |0\rangle \quad (19) \]

Similarly, the second term \((\hat{a}_i \hat{a}_i \hat{a}_i \hat{a}_i)\) is
\[
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_2 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle,
\]

where

\[
ff_2 = [f(\omega_3, \omega_1) f(\omega_4, \omega_2) + f(\omega_3, \omega_2) f(\omega_4, \omega_1)] e^{-i\omega_3 \tau} e^{-i\omega_4 \tau}.
\]

The third term \((-\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s)\) is

\[
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_3 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle,
\]

where

\[
ff_3 = [f(\omega_1, \omega_2) f(\omega_3, \omega_4) - f(\omega_1, \omega_4) f(\omega_3, \omega_2)] e^{-i\omega_1 \tau} e^{-i\omega_3 \tau}.
\]

The fourth term \((-\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s)\) is

\[
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_4 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle,
\]

where

\[
ff_4 = [-f(\omega_1, \omega_2) f(\omega_4, \omega_3) - f(\omega_1, \omega_3) f(\omega_4, \omega_2)] e^{-i\omega_1 \tau} e^{-i\omega_3 \tau}.
\]

The fifth term \((-\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s)\) is:

\[
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_5 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle,
\]

where

\[
ff_5 = [-f(\omega_2, \omega_1) f(\omega_3, \omega_4) - f(\omega_2, \omega_4) f(\omega_3, \omega_1)] e^{-i\omega_2 \tau} e^{-i\omega_3 \tau}.
\]

The sixth term \((-\hat{a}_s \hat{a}_s \hat{a}_s \hat{a}_s)\) is:

\[
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_6 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle,
\]

where

\[
ff_6 = [-f(\omega_2, \omega_1) f(\omega_4, \omega_3) - f(\omega_2, \omega_3) f(\omega_4, \omega_1)] e^{-i\omega_2 \tau} e^{-i\omega_4 \tau}.
\]

Combine these six terms:

\[
\hat{E}_1^{(+)} \hat{E}_2^{(+)} \hat{E}_3^{(+)} \hat{E}_4^{(+)} |\psi\rangle =
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_1 + ff_2 + ff_3 + ff_4 + ff_5 + ff_6 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} |0\rangle.
\]

Then

\[
\left\langle \psi \left| \hat{E}_4^{(-)} \hat{E}_3^{(-)} \hat{E}_2^{(-)} \hat{E}_1^{(-)} \hat{E}_1^{(+)} \hat{E}_2^{(+)} \hat{E}_3^{(+)} \hat{E}_4^{(+)} \right| \psi \right\rangle =
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_1 + ff_2 + ff_3 + ff_4 + ff_5 + ff_6 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4} \times
\frac{1}{8} \left( \frac{1}{2\pi} \right)^2 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \times ff_1 + ff_2 + ff_3 + ff_4 + ff_5 + ff_6 e^{-i\omega_1 t_1} e^{-i\omega_2 t_2} e^{-i\omega_3 t_3} e^{-i\omega_4 t_4}.
\]

where, \(ff^*\) is the complex conjugate of \(ff\).

Finally,
\[ P_{22}(\tau) = \int dt_1 dt_2 dt_3 dt_4 \left\langle \psi \left| \tilde{E}^{(-)}_1 \tilde{E}^{(-)}_2 \tilde{E}^{(+)}_1 \tilde{E}^{(+)}_2 \right| \psi \right\rangle \]

\[ = \frac{1}{64} \int dt_1 dt_2 dt_3 dt_4 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \int_0^\infty d\omega'_1 d\omega'_2 d\omega'_3 d\omega'_4 (f f_1 + f f_2 + f f_3 + f f_4 + f f_5 + f f_6) \]

\[ = \frac{1}{64} \int dt_1 dt_2 dt_3 dt_4 \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 \int_0^\infty d\omega'_1 d\omega'_2 d\omega'_3 d\omega'_4 (f f_1 + f f_2 + f f_3 + f f_4 + f f_5 + f f_6) \]

\[ (f f_1 + f f_2 + f f_3 + f f_4 + f f_5 + f f_6) = \frac{1}{64} \left( \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{22}(\tau)|^2 \right)^2 \] \hspace{1cm} (32)

In the above calculation, the relationship of \( \delta(\omega - \omega') = \frac{1}{2\pi} \int_0^\infty e^{i(\omega - \omega')t} dt \) is used.

In conclusion, the four-fold coincidence probability in the 2/2 detection scheme is

\[ P_{22}(\tau) = \frac{1}{64} \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{22}(\tau)|^2, \] \hspace{1cm} (33)

with

\[ |I_{22}(\tau)|^2 = |(f_{13}f_{24} + f_{14}f_{23})e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{11}f_{24} + f_{12}f_{21})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{12}f_{33} + f_{14}f_{22})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{13}f_{43} + f_{13}f_{44})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{21}f_{32} + f_{21}f_{33}e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} - (f_{21}f_{43} + f_{23}f_{41})e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} |^2, \] \hspace{1cm} (34)

where \( f_{ij} = f(\omega_i, \omega_j) \).

In the 3/1 detection, the key components can be written as \([\tilde{a}_s(\omega_1) + \tilde{a}_s(\omega_1)][\tilde{a}_s(\omega_2) + \tilde{a}_s(\omega_2)][\tilde{a}_s(\omega_3) + \tilde{a}_s(\omega_3)]/\tilde{a}_s(\omega_1) - \tilde{a}_s(\omega_1) \]. Only 6 out of 16 terms exist:

- \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \), \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \), \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \)

Following the similar method as in the case of 2/2 detection, the coincidence probability \( P_{31}(\tau) \) in the 3/1 detection scheme can be calculated as

\[ P_{31}(\tau) = \frac{1}{728} \int_0^\infty \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{31}(\tau)|^2, \] \hspace{1cm} (35)

with

\[ |I_{31}(\tau)|^2 = \left| (f_{13}f_{24} + f_{14}f_{23})e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{11}f_{24} + f_{12}f_{21})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{12}f_{33} + f_{14}f_{22})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} + (f_{13}f_{43} + f_{13}f_{44})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{21}f_{32} + f_{21}f_{33})e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} + (f_{21}f_{43} + f_{23}f_{41})e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} |^2. \] \hspace{1cm} (36)

In the 4/0 detection, the key components can be written as \([\tilde{a}_s(\omega_1) + \tilde{a}_s(\omega_1)][\tilde{a}_s(\omega_2) + \tilde{a}_s(\omega_2)][\tilde{a}_s(\omega_3) + \tilde{a}_s(\omega_3)] \tilde{a}_s(\omega_1)\tilde{a}_s(\omega_1)\tilde{a}_s(\omega_1)\tilde{a}_s(\omega_1)\]. Only 6 out of 16 terms exist:

- \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \), \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \), \( -\tilde{a}_s \tilde{a}_s \tilde{a}_s \tilde{a}_s \)

Following the similar method as in the case of 2/2 detection, the coincidence probability \( P_{40}(\tau) \) in the 4/0 detection scheme is

\[ P_{40}(\tau) = \frac{1}{1024} \int_0^\infty \int_0^\infty d\omega_1 d\omega_2 d\omega_3 d\omega_4 |I_{40}(\tau)|^2, \] \hspace{1cm} (37)

with

\[ |I_{40}(\tau)|^2 = \left| (f_{13}f_{24} + f_{14}f_{23})e^{-i\omega_1 \tau} e^{-i\omega_2 \tau} + (f_{11}f_{24} + f_{12}f_{21})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{12}f_{33} + f_{14}f_{22})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} + (f_{13}f_{43} + f_{13}f_{44})e^{-i\omega_1 \tau} e^{-i\omega_3 \tau} - (f_{21}f_{32} + f_{21}f_{33})e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} + (f_{21}f_{43} + f_{23}f_{41})e^{-i\omega_2 \tau} e^{-i\omega_3 \tau} \right|^2. \] \hspace{1cm} (38)