Observation of Quantum Interference Effects in the Frequency Domain

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(Received 24 August 1992)

Interference has been observed in the mixing of signal photons produced by spontaneous down-conversion in two nonlinear crystals, when the optical path difference through the interferometer greatly exceeds the coherence length of the light. The interference shows up as a periodic modulation of the spectrum. We demonstrate that the interference is nonclassical and connected with the entangled state of the signal and idler photons, by inserting the long delay in the path of the idler photons, even though the signal photons are interfering.

PACS numbers: 42.50.Dv, 42.50.Wm

As is well known, interference effects produced by two light beams of similar spectral density are in general observable with one photodetector only when the optical path difference cT between them is shorter than the longitudinal coherence length cT_c. Actually, this "well known" theorem is untrue. Interference occurs no matter how long or short the optical path difference may be, but the effect is manifest as a periodic spatial variation of the light intensity when $T \ll T_c$ and as a spectral variation when $T \gg T_c$. The spectral modulation has been known within the domain of classical optics for many years [1-3] although it is probably less familiar in the quantum domain. Recently there has also been interest in spectral shifts resulting from the interference of light [4-8].

We wish to report an experiment with single photons in which interference shows up as a modulation of the spectrum, even when the long delay T is not inserted directly in one of the two interferometer arms. This is the first reported observation of one-photon interference under the condition $T \gg T_c$. The visibility of the spectral modulation can be controlled by adjustment of a filter of variable transmissivity, as in some previous interference experiments [9,10], and as before the effect is nonclassical. We show that, under certain conditions, the spectral modulation can be carried by each photon rather than by an ensemble of monochromatic photons of different frequencies.

Let us first treat the situation in which the optical fields are classical waves. Let $E_1^{(+)}(t)$ and $E_2^{(+)}(t)$ be the positive frequency parts of two partly correlated electromagnetic waves that come together and are mixed by a beam splitter of reflectivity $\mathcal{R}$ and transmissivity $\mathcal{T}$ from one side and $\mathcal{R}',\mathcal{T}'$ from the other side. If wave 2 is delayed by some time T, then the total complex fields $E_3^{(+)}(t)$ and $E_4^{(+)}(t)$ at the two exit ports of the beam splitter can be written [11]

$$\begin{aligned} E_3^{(+)}(t) &= \mathcal{R}E_1^{(+)}(t) + \mathcal{T}'E_2^{(+)}(t-T)\,, \\ E_4^{(+)}(t) &= \mathcal{T}E_1^{(+)}(t) + \mathcal{R}'E_2^{(+)}(t-T)\,. \end{aligned} \tag{1}$$

If the fields are stationary, the two-time autocorrelation function of $E_3^{(+)}(t)$ is given by

$$\begin{aligned} \Gamma_{33}(\tau) = {} & |\mathcal{R}|^2\Gamma_{11}(\tau) + |\mathcal{T}|^2\Gamma_{22}(\tau) \\ & + \mathcal{R}^*\mathcal{T}'\Gamma_{12}(\tau - T) + \mathcal{T}'^*\mathcal{R}\Gamma_{21}(\tau + T)\,, \end{aligned} \tag{2}$$

where $\Gamma_{ij}(\tau) \equiv \langle E_i^{(-)}(t)E_j^{(+)}(t+\tau)\rangle$ $(i,j=1,2,3)$, and each autocorrelation or cross-correlation function depends only on the difference between the two time arguments. By the Wiener-Khintchine theorem, the Fourier transform with respect to τ of the correlation gives the corresponding spectral density. Hence, if we write $\Phi_{ij}(\omega)$ for the autospectral or cross-spectral density, then

$$\Phi_{ij}(\omega) \equiv \frac{1}{2\pi}\int_{-\infty}^{\infty}\Gamma_{ij}(\tau)e^{i\omega\tau}d\tau \quad (i,j=0,1,2)\,. \tag{3}$$

On making use of the symmetry property $\Phi_{ij}^*(\omega) = \Phi_{ji}(\omega)$, we obtain from Eq. (2) after taking Fourier transforms

$$\begin{aligned} \Phi_{33}(\omega) = {} & |\mathcal{R}|^2\Phi_{11}(\omega) + |\mathcal{T}|^2\Phi_{22}(\omega) \\ & + \mathcal{R}^*\mathcal{T}'\Phi_{12}(\omega)e^{+i\omega T} + \text{c.c.} \end{aligned} \tag{4}$$

Finally, let us assume for simplicity that all Fourier components of $\hat{E}_1^{(+)}(t)$ and $\hat{E}_2^{(+)}(t)$ are correlated to the same degree, which has been called the condition for cross-spectral purity [12,13], so that

$$\Phi_{12}(\omega)/[\Phi_{11}(\omega)\Phi_{22}(\omega)]^{1/2} \equiv \tilde{\gamma}_{12}\,. \tag{5}$$

Let $\Phi_{11}(\omega) = \langle I_1\rangle\phi(\omega)$, $\Phi_{22}(\omega) = \langle I_2\rangle\phi(\omega)$, where $\langle I_j\rangle = \Gamma_{jj}(0)$ is the intensity of field j $(j=1,2,3)$ and $\phi(\omega)$ is the normalized spectral density of either incoming beam, normalized so that it integrates to unity. Then we have from Eqs. (4) and (5) for the spectral density of $\hat{E}_3^{(+)}(t)$,

$$\Phi_{33}(\omega) = \phi(\omega)[|\mathcal{R}|^2\langle I_1\rangle + |\mathcal{T}|^2\langle I_2\rangle + \mathcal{R}^*\mathcal{T}'(\langle I_1\rangle\langle I_2\rangle)^{1/2}\tilde{\gamma}_{12}e^{i\omega T} + \text{c.c.}]\,. \tag{6}$$

The term proportional to the degree of coherence $|\tilde{\gamma}_{12}|$ shows that so long as $\tilde{\gamma}_{12} \neq 0$, a cosine modulation with respect to the optical path difference cT is always present. Suppose that the spectrum of the optical field is centered at frequency ω_0 and has a bandwidth $\Delta\omega$. Then if $T \ll 1/\Delta\omega$, we may replace ωT by $\omega_0 T$ in the exponent to a good approximation, and then integration over all frequencies leads to the familiar interference law [11]

$$\langle I_3\rangle=\Gamma_{33}(0)=\frac{1}{2\pi}\int_0^\infty \Phi_{33}(\omega)d\omega=|\mathcal{R}|^2\langle I_1\rangle+|\mathcal{T}|^2\langle I_2\rangle+2|\mathcal{R}\mathcal{T}|\langle I_1\rangle\langle I_2\rangle|\tilde{\gamma}_{12}|\cos(\omega_0 T+\arg\tilde{\gamma}_{12}+\arg\mathcal{T}'-\arg\mathcal{R}). \qquad (7)$$

But suppose that $T\gg 1/\Delta\omega$. Integration over ω now causes the oscillatory terms in Eq. (6) to integrate to zero. Nevertheless, as is clear from Eq. (6), interference effects are still present, but they show up as a modulation of the original spectral density $\phi(\omega)$ rather than of the total light intensity. Equation (6) is seen to be the more fundamental interference relation, whereas Eq. (7) depends on the special condition $T\ll 1/\Delta\omega$.

If $\langle I_1\rangle=\langle I_2\rangle$ and the relation $|\mathcal{T}|^2+|\mathcal{R}|^2=1$ holds, then Eq. (6) reduces to

$$\Phi_{33}(\omega)=\langle I_1\rangle\phi(\omega)[1+\mathcal{R}^*\mathcal{T}'\tilde{\gamma}_{12}e^{i\omega T}+\text{c.c.}]. \qquad (8)$$

Similarly we may show that the spectral density at the other beam splitter output is given by

$$\Phi_{44}(\omega)=\langle I_1\rangle\phi(\omega)[1+\mathcal{R}'\mathcal{T}^*\tilde{\gamma}_{12}e^{i\omega T}+\text{c.c.}]. \qquad (9)$$

Because $\mathcal{R}^*\mathcal{T}'+\mathcal{R}'\mathcal{T}^*=0$ from the symmetry properties of a beam splitter, the two spectral modulations are in antiphase. Exactly the same conclusions hold for a quantized field when the correlations $\Gamma_{ij}=\langle\hat{E}_i^{(-)}(t)\hat{E}_j^{(+)}(t+\tau)\rangle$ are defined in terms of normally ordered field operators.

Next let us consider the experimental situation illustrated in Fig. 1. Two similar nonlinear crystals NL1 and NL2 are optically pumped by mutually coherent pump beams of midfrequency ω_0 and complex amplitudes $V_1(t),V_2(t)$, respectively, which are derived from one laser. As a result parametric down-conversion can occur at NL1 with the simultaneous emission of a signal s_1 and idler i_1 photon pair, or at NL2 with the simultaneous emission of signal s_2 and idler i_2 photons. The crystals are so arranged that i_1 and i_2 are collinear, and a filter of complex transmissivity $\mathcal{T}$ is inserted between NL1 and NL2. Light fields s_1 and s_2 are mixed at the beam splitter BS_0 with reflectivity $\tilde{\mathcal{R}}$ and transmissivity $\tilde{\mathcal{T}}$ ($|\tilde{\mathcal{R}}|^2+|\tilde{\mathcal{T}}|^2=1$) from one side and $\tilde{\mathcal{R}}',\tilde{\mathcal{T}}'$ from the other, and the combined signals fall on detector D_s. Then the quantized combined field $\hat{E}_3^{(+)}(t)$ at D_s may be given the mode expansion [10]

$$\hat{E}_3^{(+)}(t)=\left(\frac{\delta\omega}{2\pi}\right)^{1/2}\sum_\omega \tilde{\mathcal{R}}\hat{a}_{s1}(\omega)e^{-i\omega(t-\tau_1)}+\tilde{\mathcal{T}}'\hat{a}_{s2}(\omega)e^{i\omega(\boldsymbol{\kappa}_2\cdot\mathbf{r}_2/c-t+\tau_2)}, \qquad (10)$$

where τ_0,τ_1,τ_2 are the propagation times from NL1 to NL2, from NL1 to D_s, and from NL2 to D_s, respectively. We have taken the center of NL1 to be the origin, whereas NL2 is centered at $\mathbf{r}_2$. The quantum state $|\psi(t)\rangle$ of the down-converted light in the interaction picture at a time t after the pump beam is turned on has been calculated before [10], and it is found to be the entangled state given by Eq. (5) of Ref. [10]. From these relations we may readily calculate the autocorrelation function $\langle\hat{E}_3^{(-)}(t)\hat{E}_3^{(+)}(t+\tau)\rangle$, whose Fourier transform with respect to τ gives the spectral density of $\hat{E}_3^{(+)}(t)$, and we obtain

$$\Phi_{33}(\omega)=\langle I_1\rangle|\eta_1|^2(2\pi)|\phi(\omega,\tilde{\omega})|^2\{1+\tilde{\mathcal{R}}^*\tilde{\mathcal{T}}'\mathcal{T}^*\gamma_{12}(\tau_0)e^{i\omega(\tau_2+\tau_0-\tau_1)}+\text{c.c.}\}. \qquad (11)$$

This may be compared with the classical Eq. (8). Here $\gamma_{12}(\tau)$ is the normalized cross-correlation of the pump field at the two crystals with $\langle I_1\rangle=\langle I_2\rangle$, and the constants η_1,η_2 that represent the two down-converter efficiencies are assumed to be equal. The spectral function $|\phi(\omega,\tilde{\omega})|^2$ ($\tilde{\omega}\equiv\omega-\omega_0$) characterizes the spectral density of the down-converted signal and idler light. Once again we note that if the optical propagation time difference $|\tau_2+\tau_0-\tau_1|\gg 1/\Delta\omega$, $\Phi_{33}(\omega)$ carries a spectral modulation as in Eq. (8). The new element is the appearance of the factor $|\mathcal{T}|$ in the visibility v of the spectral interference pattern, which makes $v=|\mathcal{T}|\gamma_{12}(\tau_0)|$ when $|\tilde{\mathcal{R}}|=|\tilde{\mathcal{T}}|=1/\sqrt{2}$ and causes all interference effects to be wiped out when $\mathcal{T}=0$, i.e., when the path from NL1 to NL2 is blocked. A similar conclusion also holds for the spatial interference pattern when $|\tau_2+\tau_0-\tau_1|\ll 1/\Delta\omega$ [9].

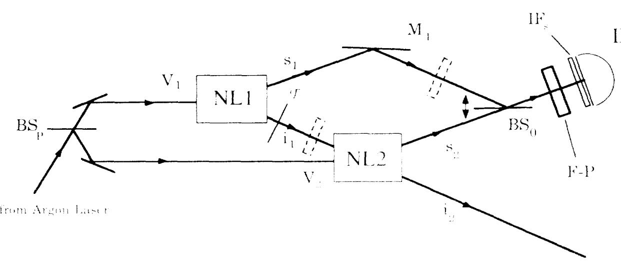

FIG. 1. Outline of the experimental setup.

We have carried out an interference experiment with the apparatus in Fig. 1 in which the spectral modulation in Eq. (11) was observed. The experiment is rather similar, in principle, to that described in Ref. [9]. NL1 and NL2 are two similar nonlinear 25-mm-long crystals of $LiIO_3$, which are optically pumped by mutually coherent pump beams derived from a single laser beam at 351 nm. The interference filter IF_s limits the bandwidth to about 10^{12} Hz and the coherence length to about 0.3 mm. Interference effects show up in the counting rate of D_s when BS_0 is translated through a few microns in the direction indicated by the arrow. It has been observed

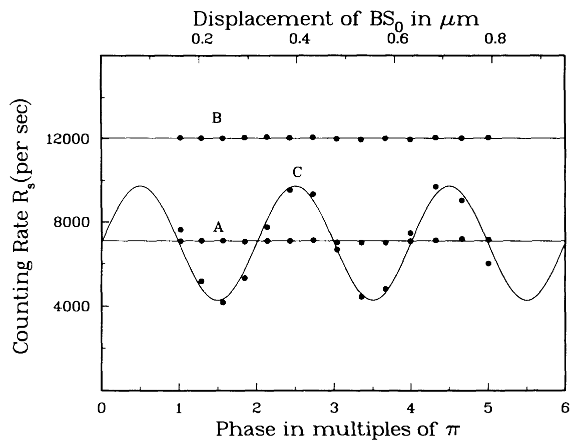

FIG. 2. Results of light intensity measurements A with insertion of the 3-mm delay plate in the s_1 arm; B with 3-mm delay in the i_1 arm; C without any delay inserted. Data B have been displaced upwards by 5000 for clarity.

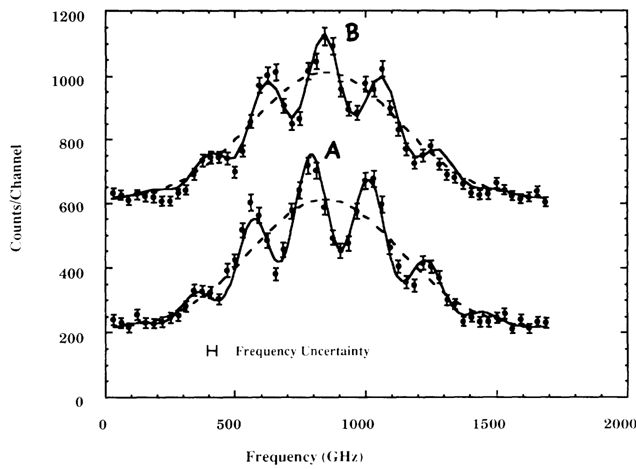

FIG. 3. Results of measurements of the spectral density at detector D_s with the 3-mm delay plate inserted in arm s_1 (A) and in arm i_1 (B). The full curves are based on Eq. (12). The broken curves are the same as in Fig. 4. Data B have been displaced upwards by 400 for clarity.

previously [9,10] that s_1 and s_2 exhibit second-order interference with about 30% visibility when i_1 and i_2 are aligned and the path lengths of s_1,s_2 are balanced so that $\tau_0+\tau_2=\tau_1$, but that the interference disappears when i_1 is blocked and prevented from reaching NL2. More generally, insertion of a filter of amplitude transmissivity $\mathcal{T}$ between NL1 and NL2 results in interference fringes whose visibility is proportional to $|\mathcal{T}|$. The effect can be interpreted in terms of the indistinguishability of the photon paths [9], and has been treated in detail in Ref. [10].

We have modified the original experiment, first by inserting a 3-mm-thick glass plate in the s_1 arm of the interferometer, as shown, that introduces an optical path difference cT of about 1.5 mm between the two arms. As this appreciably exceeds the 0.3-mm coherence length, all interference in the light intensity falling on D_s is lost, as is apparent from the data A in Fig. 2. However, insertion of a scanning Fabry-Pérot interferometer with resolution of about 40 GHz before D_s reveals the spectral modulation expected from Eq. (11). The results of an accumulation of 10000 interferometer scans are shown by data A in Fig. 3, and they clearly exhibit the expected modulation. The continuous curves in Fig. 3 are given by the following equation, which was found by a least-squares procedure:

$$y=212+401e^{-(\nu-847)^2/400^2}\{1+v\cos[2\pi(\nu-\phi)/230]\} \tag{12}$$

with $v=37\%$ for curve A and $v=30\%$ for curve B. The broken curves correspond to $v=0$, and are identical to the curves in Fig. 4.

Up to this point all observations are describable also in classical wave terms, as is apparent from a comparison of Eq. (11) with Eqs. (8) and (9). We now insert a beam stop between NL1 and NL2 so as to block i_1 and make $\mathcal{T}=0$. As shown by the data in Fig. 4, we find that the spectral modulation of the signal beam $\hat{E}_3^{(+)}(t)$ disappears, in agreement with the quantum prediction given by Eq. (11). As was observed in previous experiments [9], s_1 and s_2 then become mutually incoherent for reasons that are not understandable classically, but can be expressed in terms of the distinguishability of the s_1,s_2 photon paths.

Next we remove the delay T from the s_1 interferometer arm and insert it between NL1 and NL2 instead (see Fig. 1). As is shown by data B in Figs. 2 and 3, we observe the same absence of interference in the light intensity $\langle \hat{I}_3\rangle$ and the same spectral modulation of $\hat{E}_3^{(+)}(t)$ as before, even though neither of the two interfering signals s_1,s_2 is delayed or disturbed directly by the insertion. This also is

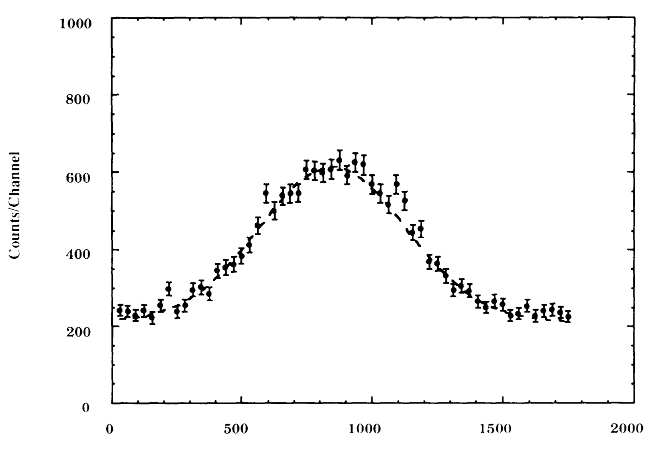

FIG. 4. Results of measurements of the spectral density at detector D_s with the 3-mm delay plate inserted in arm s_1 and beam i_1 blocked ($\mathcal{T}=0$). The broken curve is given by Eq. (12) with $v=0$.

a nonclassical effect that is connected with the entangled state of the signal and idler photons, and is in accord with Eq. (11). The absence of interference in the total light intensity measured by D_s can be understood in terms of the potential distinguishability of the photon paths NL1 to D_s and NL2 to D_s. If an efficient photodetector D_i were inserted in the path of i_2 at distance $c\tau_2$ from NL2, then simultaneous detections by D_s and D_i would be attributable to photons from source NL2, whereas D_i detections that lag behind D_s detections by T would obviously be attributable to photons emitted by NL1. It is this distinguishability, in principle, which wipes out the interference in the total light intensity. It does not, however, wipe out the spectral modulation, because this is measured with an interferometer whose passband is much narrower than $1/T$, which makes the time delay T unresolvable. Once again it is found that the modulation or interference disappears when $\mathcal{T}=0$, for the same reason as before (see Fig. 4).

Finally, we briefly address the question whether the rather complicated spectral distribution in Fig. 3 is to be attributed to an ensemble of monochromatic photons of different frequencies, or whether each photon emerging from one output port of BS_0 should be regarded as carrying the spectrum. The answer depends in a subtle way on the bandwidth $\delta\omega$ of the classical pump field and on whether this field is stationary in time. In practice, with stationary ensembles, the two cases are really indistinguishable. We can make use of Eq. (5) of Ref. [10] to calculate the density operator $\hat{\rho}^{(3)}$ of the light emerging from BS_0 in mode 3. We then find that if the pump field is stationary then $\hat{\rho}^{(3)}$ is strictly diagonal in zero- and one-photon states $|\omega\rangle$, so that the emerging light should be regarded as a mixture of monochromatic photons. On the other hand, if the pump field is not stationary, then we encounter a linear superposition of one-photon $|\omega\rangle$ states of different frequencies within $\delta\omega$, but these superposed states are mixed incoherently over the wider down-converter bandwidth $\Delta\omega$. The condition for one photon to carry a significant portion of the modulated spectrum is evidently that $1/T \ll \delta\omega$. This inequality is not satisfied in our experiments, and therefore the modulated spectrum must be regarded as being carried only by the ensemble, even with a nonstationary pump. We emphasize, however, that the quantum mechanical character of the spectral modulation we have observed does not depend on this issue.

This work was supported by the National Science Foundation and by the U.S. Office of Naval Research.

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