# **Nonclassicality of induced coherence without induced emission**

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(Received 3 July 2019; published 18 November 2019)

Interference of two beams produced at separate biphoton sources was first observed more than two decades ago. The phenomenon, often called "induced coherence without induced emission," has recently gained attention after its applications to imaging, spectroscopy, and measuring biphoton correlations have been discovered. The sources used in the corresponding experiments are nonlinear crystals pumped by laser light. The use of a laser pump makes the occurrence of induced (stimulated) emission unavoidable, and the effect of stimulated emission can be observed in the joint detection rate of the two beams. This fact raises the question whether the stimulated emission also plays a role in inducing the coherence. Here we investigate a case in which the crystals are pumped with a single-photon Fock state. We find that coherence is induced even though the possibility of stimulated emission is now fully ruled out. Furthermore, the joint detection rate of the two beams becomes ideally zero. Our results rule out any classical or semiclassical explanation of the phenomenon and also suggest that it is, in principle, possible to perform similar experiments with fermions, for which stimulated emission is strictly forbidden.

DOI: [10.1103/PhysRevA.100.053839](https://doi.org/10.1103/PhysRevA.100.053839)

# **I. INTRODUCTION**

In 1991 [\[1,2\]](#page-5-0), Zou, Wang, and Mandel (ZWM) induced coherence between two light beams generated by two spatially separated identical biphoton sources. The sources were nonlinear crystals each of which could emit two photons into two separable beams. The crucial technique (originally suggested by Ou) was to send a beam from one of the sources through the other source and to overlap it with the beam of the same photon generated by the latter (Fig. [1\)](#page-1-0). In a recent series of work, this technique has been used for imaging [\[3,4\]](#page-5-0), spectroscopy [\[5\]](#page-5-0), generating a light beam in any state of polarization [\[6\]](#page-5-0), testing the complementarity principle [\[7,8\]](#page-5-0), two-color interferometry [\[9\]](#page-5-0), measuring correlations between two photons [\[10,11\]](#page-5-0), and generating many-particle entangled states [\[12,13\]](#page-5-0).

In all the above-mentioned experiments, laser light has been used to pump the identical nonlinear crystals. Therefore, when a beam from one of the sources is sent through the other source, the occurrence of stimulated (induced) emission becomes, in principle, unavoidable. This fact leads to the question whether stimulated emission is the cause of the induced coherence. This question is important to address mainly because of two reasons: (1) It would otherwise leave the possibility of explaining the generation of coherence semiclassically and (2) more importantly, if stimulated emission plays a key role in inducing the coherence, it will not be

possible to perform similar experiments with fermions for which stimulated emission is strictly forbidden.

ZWM claimed that stimulated emission is not the cause of induced coherence. This point has been reinvestigated by Wiseman and Mølmer [\[14\]](#page-5-0) and, again recently, by Shapiro *et al.* [\[15\]](#page-5-0). However, the conclusion of Refs. [\[1,2,14\]](#page-5-0) is in direct contradiction to the conclusion of Ref. [\[15\]](#page-5-0): while Refs. [\[1,2,14\]](#page-5-0) conclude that stimulated emission does not play any role in inducing coherence, Ref. [\[15\]](#page-5-0) claims exactly the opposite (further details are given in Sec. [VI\)](#page-4-0).

We address this question from a different angle. We show that if the biphoton sources are pumped with a single-photon Fock state, stimulated emission becomes forbidden but the two beams remain mutually coherent. Our results thus establish that stimulated emission is not responsible for the mutual coherence and, therefore, suggest that performing such experiments with fermions is, in principle, possible. We also discuss the similarities and differences between the cases of a single-photon pump and a laser pump.

# **II. DESCRIPTION OF THE PROPOSED EXPERIMENT**

A ZWM interferometer (Fig. [1\)](#page-1-0) uses two spatially separated identical biphoton sources (NL1 and NL2). Each source can emit a photon pair (signal and idler) into a pair of beams. NL1 emits signal and idler photons into beams  $S_1$  and  $I_1$  respectively. Similarly, NL2 emits the photons into  $S_2$  and  $I_2$ . In the experiment, single-photon interference between  $S_1$  and  $S_2$ is observed by erasing which-way information with the help of  $I_1$  and  $I_2$ . The key is to send  $I_1$  through NL2 and to align

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<span id="page-1-0"></span>

FIG. 1. Schematic of the experimental setup. *Q* is a source that generates the pump beam. In our proposed experiment it is a singlephoton source. (In usual experiments it is a laser source.) The pump beam is split into two to illuminate two identical nonlinear crystals, NL1 and NL2. The crystals generate signal  $(S_1, S_2)$  and idler  $(I_1, I_2)$ beams by the process of parametric down-conversion. Signal beams *S*<sup>1</sup> and *S*<sup>2</sup> are combined by a beam splitter *BS*, and the resulting beam is collected by a detector D. The idler beams are aligned through NL2 but never detected.

it with *I*2. In such experiments, weakly pumped nonlinear crystals are used as biphoton sources. Since these crystals are usually pumped by *laser beams*, there is always a nonzero probability of the presence of idler photons generated by the first source at the second source when the down-conversion is taking place at the latter. Therefore, occurrence of stimulated (induced) emission at NL2 becomes, in principle, inevitable.

We propose to replace the pumping laser source with a *single-photon source*. The light generated by the singlephoton source displays antibunching [\[16\]](#page-5-0). Below we discuss in detail the expected outcome of the experiment.

#### **III. THEORY**

In the process of parametric down-conversion, a nonlinear crystal absorbs a pump photon and generates a photon pair (signal and idler); the generation of multiple pairs is also possible depending on the state of the pumping field. For simplicity, we consider single-mode optical fields. We denote the frequencies of pump, signal, and idler fields by  $\omega_P$ ,  $\omega_S$ , and  $\omega_I$ , respectively. The Hamiltonian describing parametric down-conversion at either crystal can be expressed, in the interaction picture, as (see, for example, Ref. [\[17\]](#page-5-0))

$$
\widehat{H}_j(t) = g'e^{i\Delta\omega t}\widehat{a}_{Pj}\widehat{a}_{Sj}^\dagger\widehat{a}_{Ij}^\dagger + \text{H.c.},\tag{1}
$$

where  $j = 1, 2$  labels the two nonlinear crystals,  $g'$  represents the interaction strength; *P*, *S*, and *I* represent pump, signal, and idler photons, respectively;  $\hat{a}$  and  $\hat{a}^{\dagger}$  represent photon annihilation and creation operators respectively;  $\Delta \omega = \omega_s +$  $\omega_I - \omega_P$ ; and H. c. denotes Hermitian conjugation. The quantum state of light generated by each crystal is obtained by the standard perturbative solution (see, for example, Ref. [\[18\]](#page-5-0)) and is given by the well-known expression

$$
|\psi_j\rangle = \left[\mathbb{1} + \frac{1}{i\hbar} \int_0^{\tau} dt_1 \widehat{H}_j(t_1) + \left(\frac{1}{i\hbar}\right)^2 \int_0^{\tau} dt_1 \widehat{H}_j(t_1) \int_0^{t_1} dt_2 \widehat{H}_j(t_2) + \cdots \right] |\psi_{j0}\rangle
$$
  

$$
\equiv \widehat{\mathscr{U}}_j |\psi_{j0}\rangle, \tag{2}
$$

where  $|\psi_{j0}\rangle$  is the state of light before down-conversion,  $\tau$  is the interaction time which is usually the time taken by the pump to travel the crystal's length, and we have dropped the normalization constant. It is important to note that the interaction Hamiltonian is time dependent and one needs to consider the proper ordering of the time integrations while calculating the higher order terms.

For example, by carrying out the integrations in Eq. (2) and dropping the terms with zero contribution, we can express the state generated by NL1 in the following form:

$$
|\psi_1\rangle = \left[1 + \hat{g}\hat{a}_{P1}\hat{a}_{S1}^\dagger\hat{a}_{I1}^\dagger + \frac{g^2}{2}(\hat{a}_{P1}\hat{a}_{S1}^\dagger\hat{a}_{I1}^\dagger)^2 + \tilde{g}^2\hat{a}_{P1}^\dagger\hat{a}_{P1}\hat{a}_{S1}\hat{a}_{S1}^\dagger\hat{a}_{I1}\hat{a}_{I1}^\dagger + \cdots \Big|\,|\psi_{10}\rangle,\right.
$$
 (3)

where  $g$  and  $\tilde{g}$  contain the same order of  $g'$ . Although their explicit forms are not necessary for the purpose of our discussion, they are given in the Appendix.

If the  $I_1$  beam originating from NL1 is sent through NL2 and then perfectly aligned with the  $I_2$  beam (Fig. 1), we have

$$
\widehat{a}_{I_2} = \widehat{a}_{I_1} \exp[i\phi_I], \tag{4}
$$

where  $\phi$ *I* is the phase change due to propagation from crystal NL1 to crystal NL2.

When the two crystals are put into the ZWM setup (Fig. 1), the quantum state of light generated by them is given by (cf. Refs. [\[4,19\]](#page-5-0))

$$
|\Psi\rangle = \widehat{\mathscr{U}_2}\widehat{\mathscr{U}_1}|\psi_0\rangle,\tag{5}
$$

where  $|\psi_0\rangle$  is the initial state of light before any downconversion took place, and Eq. (4) has been substituted into the expression of  $\mathcal{U}_2$ . Equation (5) is applicable to both the cases where laser and single-photon pumps are used. It is the initial state,  $|\psi_0\rangle$ , which makes the difference between the two cases.

We stress that this theoretical treatment is valid only when the interaction is sufficiently weak.

#### **A. Pumping with single photons**

A single-photon source produces light that displays antibunching [\[16\]](#page-5-0); i.e., if the light is sent through a beam splitter, no coincidence counts between the two outputs are registered for an appropriate choice of the delay line. This is because a single photon cannot be broken into further halves and, therefore, cannot be absorbed at more than one place simultaneously. In the experiment (Fig. 1), the two crystals are placed at the two outputs of a beam splitter, and, therefore, they cannot both absorb the same pump photon. Consequently, if the time difference between two consecutive pump photons is sufficiently large, it becomes impossible for an idler photon generated by NL1 to be present at NL2 when down-conversion takes place in the latter. The occurrence of stimulated emission at NL2 thus becomes strictly forbidden. We prove below that even in this case the signal beams,  $S_1$  and *S*2, remain fully coherent.

We assume, for simplicity, that the pump beam has the same intensity at the two crystals. When the pump beam is generated by a single-photon source, the initial state [see

<span id="page-2-0"></span>Eq.  $(5)$ ] is given by

$$
|\psi_0\rangle_{\rm sp} = \frac{1}{\sqrt{2}} (|1_{P_1}\rangle + e^{i\phi_P} |1_{P_2}\rangle) |{\rm vac}_{\{S,I\}}\rangle, \tag{6}
$$

where "vac" implies vacuum (no photon), e.g.,|vac{*S*,*<sup>I</sup>*} signifies no occupation in the signal and the idler modes generated by both crystals; φ*<sup>P</sup>* represents the phase difference between the pump field at the two crystals; and the single-photon Fock state  $|1_{P_i}\rangle$  represents a pump photon for the crystal *j* such that  $\hat{a}_{P_j}|1_{P_j}=\text{[vac}_{P_j})$ . It now follows from Eqs. [\(2\)](#page-1-0)–(6) that the quantum state of light in the system takes the form

$$
|\Psi_{\rm sp}\rangle = \eta |\psi_0\rangle_{\rm sp} + G_{\rm sp} |\text{vac}_{\{P\}}\rangle \big[ \big( \big| 1_{S_1} \big| + e^{i\phi} | 1_{S_2} \big| \big) \big| 1_{I_1} \big| \big], \quad (7)
$$

where  $\eta$  has contributions from all even orders of  $g'$ ,  $G_{sp}$  has contributions from all odd orders of  $g'$ ;  $|\eta| \gg |G_{sp}|$  and  $\phi =$  $\phi_P - \phi_I$ . It is important to note that Eq. (7) provides an exact expression that is obtained without dropping any higher-order term; the terms containing even order of  $g'$  in  $\widehat{\mathscr{U}}_2 \widehat{\mathscr{U}}_1$  yield the initial state  $|\psi_0\rangle_{\text{sp}}$ , and the terms containing odd order of *g*<sup>-</sup> yield the state multiplied with  $G_{\text{sp}}$ .

In order to calculate the photon counting rate, we need to determine the quantized electric field at the detector, *D* (placed after the beam splitter, *BS*, in Fig. [1\)](#page-1-0). The positive frequency part of the field can be represented by

$$
\widehat{E}_S^{(+)} = \widehat{a}_{S_1} + ie^{i\phi_S}\widehat{a}_{S_2},\tag{8}
$$

where  $\phi_s$  is the phase due to the difference between the optical paths from *NL*1 and *NL*2 to *D*. The photon counting rate at *D* is then given by

$$
\mathcal{R}_{sp} = \langle \Psi_{sp} | \widehat{E}_{S}^{(-)} \widehat{E}_{S}^{(+)} | \Psi_{sp} \rangle = 2 |G_{sp}|^{2} (1 + \cos \phi_{in}), \quad (9)
$$

where  $\widehat{E}_S^{(-)} = \{\widehat{E}_S^{(+)}\}^{\dagger}$ ,  $\phi_{\text{in}} = \phi_S + \phi + \pi/2$ , the state  $|\Psi_{\text{sp}}\rangle$  is given by Eq. (7), and we have dropped a constant multiplicative factor that depends on the detection efficiency. We note that Eq. (9) gives an exact expression for the photon counting rate, i.e., *no higher order term has been neglected*.

It is clear from Eq. (9) that the signal beams  $S_1$  and  $S_2$ create a single-photon interference pattern at the detector *D*. This means that the two signal beams are mutually coherent. Since the occurrence of stimulated emission is forbidden (see discussions above), this mutual coherence in the lowest order [\[20\]](#page-6-0) can be explained only from the indistinguishability of the paths for the signal photons arriving at the detector.

The single-photon pump ensures that only one pair of down-converted photons exists in the system. This fact is also supported by the absence of terms with higher photon number in Eq. (7). Since the signal modes generated by both crystals are never simultaneously occupied, there will be no coincidence count if one detects both signal beams (Fig. 2). This point is elaborated in the next section.

#### **B. Comparison with the case of a laser pump**

We represent the laser field by a coherent state. When the two crystals are pumped by a laser, the initial state (before any down-conversion) is given by

$$
|\psi_0\rangle_{lp} = |\alpha_1\rangle_P |\alpha_2\rangle_P |\text{vac}_{\{S,I\}}\rangle, \tag{10}
$$

where the suffix,  $l_p$ , denotes the laser pump,  $|{\rm vac}_{\{S,I\}}\rangle$  signifies zero occupation in all signal and idler modes, and  $|\alpha_j\rangle_p$ 



FIG. 2. Setup for detecting the effect of stimulated emission in coincidence counting. Coincidence detections between beams *S*<sup>1</sup> and  $S_2$  are registered with detectors  $D_1$  and  $D_2$ . The optical path length from  $Q$  to  $D_1$  via  $NL1$  is equal to the optical path length from  $Q$  to *D*<sup>2</sup> via *NL*2.

represents the coherent state of the pump at crystal *j* such that  $\hat{a}_p$ | $\alpha_j$ | $\beta_p = \alpha_j |\alpha_j\rangle_p$ . For simplicity, we again assume that the pump beams have the same intensities at the two crystals, i.e.,  $|\alpha_1| = |\alpha_2| = \alpha$ . It now follows from Eqs. [\(5\)](#page-1-0) and (10) that

$$
|\Psi_{lp}\rangle \approx |\psi_0\rangle_{lp} + [g_{lp}\alpha|\{\alpha_p\}\rangle(|1_{S_1}\rangle + e^{i\phi}|1_{S_2}\rangle)|1_{I_1}\rangle] + [(g_{lp}\alpha)^2|\{\alpha_p\}\rangle(|2_{S_1}\rangle + e^{2i\phi}|2_{S_2}\rangle) + \sqrt{2}e^{i\phi}|1_{S_1}, 1_{S_2}\rangle)|2_{I_1}\rangle + \tilde{g}_{lp}^2(\alpha_1\tilde{a}_{P_1}^{\dagger} + \alpha_2\tilde{a}_{P_2}^{\dagger})|\psi_0\rangle] + \cdots, \qquad (11)
$$

where we have written *g* of Eq. [\(3\)](#page-1-0) as  $g_{lp}$  to distinguish the case of a laser pump and absorbed the phase factor  $\exp[i\arg\{\alpha_1\}]$  into it;  $|\{\alpha_P\}\rangle = |\alpha_1\rangle_P |\alpha_2\rangle_P$ ; the terms containing the same order of  $g'$  are arranged inside the same square brackets;  $\phi = \phi_P - \phi_I$  with  $\phi_P = \arg{\alpha_2} - \arg{\alpha_1}$ ; and we have dropped the normalization constant.

We now determine the photon counting rate at *D* under the assumptions that the crystals are weakly pumped and the rate of down-conversion is low, i.e.,  $|g_{lp}\alpha|^2 \ll 1$ . It follows from Eqs.  $(8)$  and  $(11)$  that the photon counting rate is given by

$$
\mathcal{R}_{lp} = \langle \Psi_{lp} | \widehat{E}_{S}^{(-)} \widehat{E}_{S}^{(+)} | \Psi_{lp} \rangle \approx 2 |g_{lp} \alpha|^{2} (1 + \cos \phi_{\text{in}}), \quad (12)
$$

where  $\phi_{\text{in}} = \phi_s + \phi + \pi/2$  and we have dropped a constant multiplicative factor that depends on the detectors's efficiency. We find that Eq.  $(12)$  and Eq.  $(9)$  have the same form. A comparison between the cases of single-photon pump and laser pump reveals many interesting features, as we discuss below.

We note that for a single-photon pump, the quantum state [Eq. (7)] does not contain any term that involves more than two down-converted photons. In contrast, the quantum state for the case of a laser pump  $[Eq. (11)]$  contains terms that involve more than two down-converted photons. In the single-mode treatment, these terms correspond to stimulated emission [\[21\]](#page-6-0). Furthermore, these terms are also multiplied by the second or higher powers of  $g_{lp} \alpha$ . Neglecting  $|g_{lp} \alpha|^2$  with respect to 1 is, therefore, equivalent to neglecting the effect of stimulated emission. We find from Eq. (12) that this approximation does *not* destroy the interference pattern. Furthermore, the visibility of the patterns for both types of pumps is equal. We thus conclude that *although stimulated emission occurs when a laser pump is used, the mutual coherence between*

*the two signal beams is not due to stimulated emission*; the spontaneous emissions occurring at the two crystals play the dominating role when the crystals are weakly pumped and the rate of down-conversion is low.

A close examination of Eq.  $(11)$  reveals that two kinds of stimulated emission are present for a laser pump: (1) the terms containing  $|2_{S_1}\rangle$  and  $|2_{S_2}\rangle$  correspond to the emission stimulated by photons generated in the same crystal and (2) the term containing  $\sqrt{2}$ |1<sub>*S*1</sub>, 1<sub>*S*<sub>2</sub></sub> refers to the emission at NL2 stimulated by the idler photon generated at NL1. We also note that if the beam  $I_1$  is blocked between NL1 and NL2, the term  $\sqrt{2} |1_{S_1}, 1_{S_2} \rangle |2_{I_1} \rangle$  gets replaced by  $|1_{S_1}, 1_{I_1}, 1_{S_2}, 1_{I_2} \rangle$ . Therefore, the alignment of the idler beams enhances the probability of joint pair emission at NL1 and NL2 by a factor of two. This effect can be observed in the intensity correlation of the signal beams.

Let us, for example, analyze the situation illustrated in Fig. [2.](#page-2-0) Here coincidence detection of  $S_1$  and  $S_2$  is considered when any time delay between them is fully compensated by a coincidence circuit. It follows from Eq. [\(11\)](#page-2-0) that for a laser pump  $(l_p)$ , the coincidence detection rate is given by

$$
\mathscr{C}_{lp}^{S_1,S_2} = \langle \Psi_{lp} | \hat{a}_{S_1}^{\dagger} \hat{a}_{S_2}^{\dagger} \hat{a}_{S_2} \hat{a}_{S_1} | \Psi_{lp} \rangle \approx 2 |g_{lp} \alpha|^4. \qquad (13)
$$

Clearly, the rate of coincidence detection increases quadratically with the pump power  $(|\alpha|^2)$ . This coincidence detection rate is due to the joint pair emission at NL1 and NL2, i.e., due to the term  $\sqrt{2}|1_{S_1}, 1_{S_2}|2_{I_1}\rangle$  in Eq. [\(11\)](#page-2-0). If the idler beams are misaligned, one has  $\sqrt{2}|1_{S_1}, 1_{S_2}\rangle|2_{I_1}\rangle \rightarrow$  $|1_{S_1}, 1_{I_1}, 1_{S_2}, 1_{I_2}\rangle$ , and the corresponding coincidence detection rate becomes

$$
\mathscr{C}_{lp}^{S_1, S_2} \approx |g_{lp}\alpha|^4 \quad \text{(idlers not aligned)}.\tag{14}
$$

Comparing with Eq.  $(13)$  with  $(14)$ , we find that a complete misalignment of the idler beams reduces the  $S_1 - S_2$  coincidence detection rate by a factor of two (Fig. 3). The phenomenon can be physically understood as follows: The presence of a photon in the *I*<sup>1</sup> mode at NL2, makes the emission of another photon in the same mode more probable. Since the emission of an idler photon is always accompanied by the emission of its partner signal photon, the *S*1-*S*<sup>2</sup> coincidence detection rate enhances.

For the case of a single-photon pump (*sp*), the coincidence rate is

$$
\mathscr{C}_{\text{sp}}^{S_1, S_2} = \langle \Psi_{\text{sp}} | \hat{a}_{S_1}^{\dagger} \hat{a}_{S_2}^{\dagger} \hat{a}_{S_2} \hat{a}_{S_1} | \Psi_{\text{sp}} \rangle = 0, \tag{15}
$$

where  $|\Psi_{\rm sp}\rangle$  is given by Eq. [\(7\)](#page-2-0). Clearly, the rate of coincidence detection does not depend on the crystal gain, i.e., the rate of photon production. In the case of a single-photon pump, no stimulated emission is possible even with fully aligned idler beams. Therefore, the  $S_1-S_2$  coincidence detection rate ( $\mathcal{C}_{sp}^{S_1, S_2}$ ) remains unchanged (zero) when the idler beams are misaligned. However, for both types of pumps, the lowest-order correlation between the two signal beams will be completely lost if the idler beams are fully misaligned.

# **IV. CONNECTION WITH STATISTICAL PROPERTIES OF LIGHT**

When a nonlinear crystal is pumped with a single-photon Fock state, the down-converted light is ideally in a two-photon



FIG. 3. Effect of stimulated emission in the case of a laser pump. The coincidence detection rate (dimensionless unit) of two signal photons ( $S_1$  and  $S_2$ ) is plotted against the pump power ( $|\alpha|^2$ ; dimensionless unit) for the two cases: (a) the idler beams are fully aligned and (b) the idler beams are fully misaligned. In both cases, the coincidence detection rate increases quadratically with the pump power. However, if the idlers are fully misaligned, the coincidence rate becomes two times smaller than the value obtained for fully aligned idler beams. The stimulated emission caused by the alignment of idler beams results in the higher coincidence rate on curve (a) compared to curve (b).

Fock state. This down-converted light displays different photon statistics than the down-converted light generated from a laser pump. An undepleted laser pump, which is usually modeled by a coherent state, allows one to treat the pump field classically. In this case, signal and idler beams are individually in thermal (chaotic) states [\[22,23\]](#page-6-0); the quantum state produced by the crystal is a superposition of Fock states, each of which contains equal number of signal and idler photons. However, a single-photon pump cannot be treated classically; in this case, each down-converted beam is individually represented by a single-photon Fock state, which is certainly not a thermal state. This difference between the statistical properties of the down-converted light can be intuitively connected to our results.

It is well-known that the phase space distributions of Fock states are not Gaussian ([\[24\]](#page-6-0), Sec. 11.8.6) and, consequently, the higher-order field correlation functions cannot, in general, be expressed in terms of the lowest-order ones [\[25\]](#page-6-0). The analogous result in our case is the fact that when  $S_1$  and *S*<sup>2</sup> beams contain photons in a Fock state, the lowest-order coherence between the beams is *not* accompanied by any intensity correlation.

The phase space distribution of light in a thermal (chaotic) state is Gaussian (see, for example, Ref. [\[24\]](#page-6-0), Sec. 11.8.6). For such light, all higher-order field correlation functions can be expressed in terms of the lowest-order ones ([\[24\]](#page-6-0), Ch. 13). We have found in our analysis that in the case of thermal *S*<sup>1</sup> and *S*<sup>2</sup> beams (produced by laser pumps), the lowest-order coherence [\[20\]](#page-6-0) is necessarily accompanied by the presence of the intensity correlation between the two beams. When the idler beams are misaligned, the loss of lowest-order coherence between  $S_1$  and  $S_2$  is associated with the reduction of <span id="page-4-0"></span>the coincidence detection rate of the two beams. As discussed above, this reduction is due to the fact that photons generated at NL2 are not stimulated by the emission at NL1.

One can therefore conclude that for a laser pump, the induced lowest-order coherence needs to be accompanied by stimulated emission, even though stimulated emission is not responsible for inducing coherence.

# **V. EXISTING EXPERIMENTAL EVIDENCE**

With the available technology, it is extremely challenging to perform the above mentioned experiments with a singlephoton pump [\[26\]](#page-6-0). There is an alternative way of showing that stimulated emission at NL2 plays no role in inducing lowestorder coherence between the two signal beams. This method, which was introduced in Ref. [\[1\]](#page-5-0), is to insert an attenuator on the path of the idler beam between NL1 and NL2 and then to show that the visibility of the interference pattern is linearly proportional to the amplitude transmission coefficient of the attenuator. It was later shown in Ref. [\[14\]](#page-5-0) that this dependence does not remain linear when the effect of stimulated emission is prominent. A recent paper also analyzes this issue in great detail [\[27\]](#page-6-0).

The coincidence measurement between  $S_1$  and  $S_2$  for a laser pump has been performed by Liu *et al.*, under more general considerations, where they controlled the rate of stimulated emission by placing an attenuator in the idler's path between NL1 and NL2 [\[19\]](#page-5-0). They observed a significant drop in the coincidence detection rate when the idler beam was fully blocked (no stimulated emission), compared to the case when the idler beam was fully transmitted (maximum stimulated emission).

# **VI. DISCUSSION**

In order to put our results into the context of existing work, we now briefly discuss the arguments presented in Refs. [\[1,2,14,15\]](#page-5-0).

Zou, Wang, and Mandel stated that "when the external field is weak, the down-conversion occurs spontaneously and at random" [\[2\]](#page-5-0). Here the external field corresponds to beam *I*<sup>1</sup> and the down-conversion refers to the down-conversion at NL2. A weak field implies that the average photon number generated by down-conversion is very low. Their theoretical analysis, based on this assumption, correctly predicted the experimentally observed [\[1,2\]](#page-5-0) linear dependence of visibility on the amplitude transmission coefficient of the attenuator (see Sec. V).

In Ref. [\[14\]](#page-5-0), Wiseman and Mølmer tested whether the above-mentioned linear dependence can be found when stimulated emission is the cause of induced coherence. They considered a situation in which the emission rates of the crystals can be arbitrarily enhanced. They determined the modulus of the *normalized* lowest-order coherence function between the fields of the two superposed beams  $(S_1 \text{ and } S_2)$  and expressed it as a function of the average photon number generated by down-conversion [[\[14\]](#page-5-0), Eq. [\(11\)](#page-2-0)]. This normalized coherence function gives the highest attainable visibility. They found that if the average photon number is high enough for the stimulated emission to be the cause of induced coherence, the visibility

is *not* proportional to the amplitude transmission coefficient of the attenuator. However, when the mean photon number is very low, i.e., when the photon generation is dominated by spontaneous emission, the visibility becomes proportional to the amplitude transmission coefficient. Based on this observation they confirmed that the linear dependence of the visibility on the amplitude transmission coefficient is the true signature of induced coherence without stimulated emission.

In Ref. [\[15\]](#page-5-0) Shapiro, Venkatraman, and Wong considered a situation that is similar to the situation considered in Ref. [\[14\]](#page-5-0), albeit they used a different terminology. In contrast to Ref. [\[14\]](#page-5-0), the lowest-order coherence function presented in Eq. (22) of Ref. [\[15\]](#page-5-0) is not normalized. This *unnormalized* coherence function decreases as the average photon number decreases and can give the impression that stimulated emission is responsible for the induced coherence. It is important to note that the absolute value of this unnormalized coherence function would always decrease with a decreasing average photon number, even though its normalized version (the highest attainable visibility) could show an entirely different behavior. It can be readily checked that if one normalizes the coherence function obtained by Shapiro *et al.* [[\[15\]](#page-5-0), Eq. (22)], one finds the formula derived by Wiseman and Mølmer [[\[14\]](#page-5-0), Eq.  $(11)$ ]. As mentioned above, this normalized coherence function does not vanish as the average photon number becomes very low [\[28\]](#page-6-0); it rather becomes equal to the amplitude transmission coefficient of the attenuator.

We therefore conclude that in a ZWM-type experiment, the induced lowest-order coherence is not due to stimulated emission. This fact rules out a classical or semiclassical interpretation of the phenomenon like the one suggested in Ref. [\[15\]](#page-5-0).

#### **VII. SUMMARY**

We have proposed to perform a Zou-Wang-Mandel (ZWM) experiment using a *single-photon pump* [\[29\]](#page-6-0). Our theoretical analysis shows that for such a pump no emission stimulated by the light from the first source can occur at the second source. We have explicitly shown that the absence of this stimulated emission does not affect the induced lowestorder coherence [\[20\]](#page-6-0) of the two signal beams, i.e., the beams will produce a single-photon interference pattern if superposed.

A comparison with the case of laser pump and the existing experimental evidences establishes that in any ZWM-type experiment, where the crystals are weakly pumped, the induced lowest-order coherence is not due to stimulated emission. Therefore, our results support the conclusion of Refs. [\[1,2,14\]](#page-5-0) and contradict the conclusion of Ref. [\[15\]](#page-5-0).

#### **VIII. OUTLOOK**

As mentioned in Introduction, the ZWM experiment with photons has found broad applications in quantum optics. Recently, a ZWM-type experiment experiment has been performed with microwave superconducting cavities [\[30\]](#page-6-0). At this point it is important to look beyond the photonic domain and ask whether similar experiments can be performed with other quantum entities. Recent advancements in the fields of trapped

<span id="page-5-0"></span>ions [\[31\]](#page-6-0), atomic systems [\[32,33\]](#page-6-0), and superconducting circuits [\[34\]](#page-6-0) show a very high prospect of research in this direction.

The ZWM-type experiments performed in the photonic domain suggest that there is no fundamental obstacle in performing this type of experiments with other kinds of bosons. However, the case of fermions requires separate attention because stimulated emission is strictly forbidden in this case.

We have shown that stimulated emission plays no role in the quantum interference observed in a ZWM experiment. Therefore, our results suggest that such experiments can, in principle, be performed with fermions and open up the possibility of building an experimental setup for this purpose.

Let us briefly illustrate how the same situation can be conceptually attained with fermions. Consider, for example, electron-electron scattering. Imagine that electrons are produced in a coherent superposition of two paths that are incident onto two scattering samples. One now needs to select only those modes where the scattered electron paths overlap as in the case of the ZWM experiment.

Although building such a setup could be technically challenging at this point, we note that analogs of several optical experiments have already been performed with fermions. For example, electron interference has been observed using a Mach-Zehnder setup [\[35\]](#page-6-0); also, two-particle interference displaying antibunching (Pauli dip) has been realized with electrons [\[36\]](#page-6-0). Furthermore, correlated fermions pairs have also been created in the laboratory [\[37\]](#page-6-0). Based on these

facts, we expect that ZWM experiments with fermions will be performed in the near future.

# **ACKNOWLEDGMENTS**

We thank E. Giese for helpful discussions. This work was supported by the Austrian Academy of Sciences (ÖAW-IQOQI, Vienna) and the Austrian Science Fund (FWF) with SFB F40 (FOQUS) and W1210-2 (CoQus). M.L. also acknowledges support from College of Arts and Sciences and the Office of the Vice President for Research, Oklahoma State University. R.L. was supported by National Science Centre (Poland) grants 2015/17/D/ST2/03471, 2015/16/S/ST2/00424, the Polish Ministry of Science and Higher Education, and the Foundation for Polish Science (FNP) under the FIRST TEAM project, "Spatiotemporal photon correlation measurements for quantum metrology and super-resolution microscopy," cofinanced by the European Union under the European Regional Development Fund.

#### **APPENDIX**

Explicit forms of *g* and  $\tilde{g}$  are obtained by carrying out the integrations in Eq. [\(2\)](#page-1-0). They are given below:

$$
g = \left(\frac{\tau g'}{i\hbar}\right) e^{i\Delta\omega\tau/2} \text{sinc}(\Delta\omega\tau/2),\tag{A1a}
$$

$$
\tilde{g}^2 = \left(\frac{|g'|}{i\hbar}\right)^2 \frac{i\tau}{\Delta\omega} [1 + e^{-i\Delta\omega\tau/2} \text{sinc}(\Delta\omega\tau/2)]. \tag{A1b}
$$

- [1] X. Y. Zou, L. J. Wang, and L. Mandel, Induced Coherence and Indistinguishability in Optical Interference, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.67.318) **[67](https://doi.org/10.1103/PhysRevLett.67.318)**, [318](https://doi.org/10.1103/PhysRevLett.67.318) [\(1991\)](https://doi.org/10.1103/PhysRevLett.67.318).
- [2] L. J. Wang, X. Y. Zou, and L. Mandel, Induced coherence without induced emission, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.44.4614) **[44](https://doi.org/10.1103/PhysRevA.44.4614)**, [4614](https://doi.org/10.1103/PhysRevA.44.4614) [\(1991\)](https://doi.org/10.1103/PhysRevA.44.4614).
- [3] G. B. Lemos, V. Borish, G. D. Cole, S. Ramelow, R. Lapkiewicz, and A. Zeilinger, Quantum imaging with undetected photons, [Nature \(London\)](https://doi.org/10.1038/nature13586) **[512](https://doi.org/10.1038/nature13586)**, [409](https://doi.org/10.1038/nature13586) [\(2014\)](https://doi.org/10.1038/nature13586).
- [4] M. Lahiri, R. Lapkiewicz, G. B. Lemos, and A. Zeilinger, [Theory of quantum imaging with undetected photons,](https://doi.org/10.1103/PhysRevA.92.013832) Phys. Rev. A **[92](https://doi.org/10.1103/PhysRevA.92.013832)**, [013832](https://doi.org/10.1103/PhysRevA.92.013832) [\(2015\)](https://doi.org/10.1103/PhysRevA.92.013832).
- [5] D. A. Kalashnikov, A. V. Paterova, S. P. Kulik, and L. A. Krivitsky, Infrared spectroscopy with visible light, [Nat. Phot.](https://doi.org/10.1038/nphoton.2015.252) **[10](https://doi.org/10.1038/nphoton.2015.252)**, [98](https://doi.org/10.1038/nphoton.2015.252) [\(2016\)](https://doi.org/10.1038/nphoton.2015.252).
- [6] M. Lahiri, A. Hochrainer, R. Lapkiewicz, G. B. Lemos, and A. Zeilinger, Partial polarization by quantum distinguishability, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.95.033816) **[95](https://doi.org/10.1103/PhysRevA.95.033816)**, [033816](https://doi.org/10.1103/PhysRevA.95.033816) [\(2017\)](https://doi.org/10.1103/PhysRevA.95.033816).
- [7] A. Heuer, R. Menzel, and P. W. Milonni, Induced Coherence, Vacuum Fields, and Complementarity in Biphoton Generation, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.114.053601) **[114](https://doi.org/10.1103/PhysRevLett.114.053601)**, [053601](https://doi.org/10.1103/PhysRevLett.114.053601) [\(2015\)](https://doi.org/10.1103/PhysRevLett.114.053601).
- [8] A. Heuer, R. Menzel, and P. W. Milonni, Complementarity in biphoton generation with stimulated or induced coherence, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.92.033834) **[92](https://doi.org/10.1103/PhysRevA.92.033834)**, [033834](https://doi.org/10.1103/PhysRevA.92.033834) [\(2015\)](https://doi.org/10.1103/PhysRevA.92.033834).
- [9] A. Hochrainer, M. Lahiri, G. B. Lemos, R. Lapkiewicz, and A. Zeilinger, Interference fringes controlled by non-interfering photons, [Optica](https://doi.org/10.1364/OPTICA.4.000341) **[4](https://doi.org/10.1364/OPTICA.4.000341)**, [341](https://doi.org/10.1364/OPTICA.4.000341) [\(2017\)](https://doi.org/10.1364/OPTICA.4.000341).
- [10] A. Hochrainer, M. Lahiri, G. B. Lemos, R. Lapkiewicz, and A. Zeilinger, Quantifying the momentum correlation between two light beams by detecting one, [Proc. Natl. Acad. Sci. USA](https://doi.org/10.1073/pnas.1620979114) **[114](https://doi.org/10.1073/pnas.1620979114)**, [1508](https://doi.org/10.1073/pnas.1620979114) [\(2017\)](https://doi.org/10.1073/pnas.1620979114).
- [11] M. Lahiri, A. Hochrainer, R. Lapkiewicz, G. B. Lemos, and A. Zeilinger, Twin photon correlations in single-photon interference, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.96.013822) **[96](https://doi.org/10.1103/PhysRevA.96.013822)**, [013822](https://doi.org/10.1103/PhysRevA.96.013822) [\(2017\)](https://doi.org/10.1103/PhysRevA.96.013822).
- [12] M. Krenn, A. Hochrainer, M. Lahiri, and A. Zeilinger, Entanglement by Path Identity, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.118.080401) **[118](https://doi.org/10.1103/PhysRevLett.118.080401)**, [080401](https://doi.org/10.1103/PhysRevLett.118.080401) [\(2017\)](https://doi.org/10.1103/PhysRevLett.118.080401).
- [13] M. Lahiri, Many-particle interferometry and entanglement by path identity, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.98.033822) **[98](https://doi.org/10.1103/PhysRevA.98.033822)**, [033822](https://doi.org/10.1103/PhysRevA.98.033822) [\(2018\)](https://doi.org/10.1103/PhysRevA.98.033822).
- [14] H. M. Wiseman and K. Mølmer, Induced coherence with and without induced emission, [Phys. Lett. A](https://doi.org/10.1016/S0375-9601(00)00314-5) **[270](https://doi.org/10.1016/S0375-9601(00)00314-5)**, [245](https://doi.org/10.1016/S0375-9601(00)00314-5) [\(2000\)](https://doi.org/10.1016/S0375-9601(00)00314-5).
- [15] J. H. Shapiro, D. Venkatraman, and F. N. C. Wong, Classical imaging with undetected photons, [Sci. Rep.](https://doi.org/10.1038/srep10329) **[5](https://doi.org/10.1038/srep10329)**, [10329](https://doi.org/10.1038/srep10329) [\(2015\)](https://doi.org/10.1038/srep10329).
- [16] H. J. Kimble, M. Dagenais, and L. Mandel, Photon Antibunching in Resonance Fluorescence, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.39.691) **[39](https://doi.org/10.1103/PhysRevLett.39.691)**, [691](https://doi.org/10.1103/PhysRevLett.39.691) [\(1977\)](https://doi.org/10.1103/PhysRevLett.39.691).
- [17] R. Ghosh, C. K. Hong, Z. Y. Ou, and L. Mandel, Interference of two photons in parametric down conversion, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.34.3962) **[34](https://doi.org/10.1103/PhysRevA.34.3962)**, [3962](https://doi.org/10.1103/PhysRevA.34.3962) [\(1986\)](https://doi.org/10.1103/PhysRevA.34.3962).
- [18] Claude Cohen-Tannoudji, B. Diu, and F. Laloe, *Quantum Mechanics*, Volume 1 (John Wiley, Singapore, 2005 and Hermann, Paris, 1977).
- [19] B. H. Liu, F. W. Sun, Y. X. Gong, Y. F. Huang, Z. Y. Ou, and G. C. Guo, Investigation of the role of indistinguishability in photon bunching and stimulated emission, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.79.053846) **[79](https://doi.org/10.1103/PhysRevA.79.053846)**, [053846](https://doi.org/10.1103/PhysRevA.79.053846) [\(2009\)](https://doi.org/10.1103/PhysRevA.79.053846).
- <span id="page-6-0"></span>[20] In coherence theory, single-photon interference effects are explained in terms of second-order field correlations [24]. The same correlation has also been referred to as first-order in literature [38]. We use the term "lowest-order" to avoid confusion.
- [21] In the multimode scenario, two pairs of photon can also be created via spontaneous emission at a nonlinear crystal [39].
- [22] B. R. Mollow and R. J. Glauber, Quantum theory of parametric amplification. II, [Phys. Rev.](https://doi.org/10.1103/PhysRev.160.1097) **[160](https://doi.org/10.1103/PhysRev.160.1097)**, [1097](https://doi.org/10.1103/PhysRev.160.1097) [\(1967\)](https://doi.org/10.1103/PhysRev.160.1097).
- [23] F. Paleari, A. Andreoni, G. Zambra, and M. Bondani, Thermal photon statistics in spontaneous parametric downconversion, [Opt. Express](https://doi.org/10.1364/OPEX.12.002816) **[12](https://doi.org/10.1364/OPEX.12.002816)**, [2816](https://doi.org/10.1364/OPEX.12.002816) [\(2004\)](https://doi.org/10.1364/OPEX.12.002816).
- [24] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, Cambridge, 1995).
- [25] R. J. Glauber, Photon Correlations, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.10.84) **[10](https://doi.org/10.1103/PhysRevLett.10.84)**, [84](https://doi.org/10.1103/PhysRevLett.10.84) [\(1963\)](https://doi.org/10.1103/PhysRevLett.10.84).
- [26] An experiment for demonstrating the effect of single-photon pump is currently being performed in the group of R. W. Boyd at Ottawa (private communication).
- [27] M. I. Kolobov, E. Giese, S. Lemieux, R. Fickler, and R. W. Boyd, Controlling induced coherence for quantum imaging, [J. Opt.](https://doi.org/10.1088/2040-8986/aa64a2) **[19](https://doi.org/10.1088/2040-8986/aa64a2)**, [054003](https://doi.org/10.1088/2040-8986/aa64a2) [\(2017\)](https://doi.org/10.1088/2040-8986/aa64a2).
- [28] Average photon number is equal to zero implies both sources are turned off. In this case, no interference occurs simply because there is no light to detect.
- [29] Single-photon pump was also considered for graphical description of such experiments in the following publication: S. Ataman, The quantum optical description of three experiments [involving non-linear optics using a graphical method,](https://doi.org/10.1140/epjd/e2014-50693-1) Eur. Phys. J. D **[69](https://doi.org/10.1140/epjd/e2014-50693-1)**, [44](https://doi.org/10.1140/epjd/e2014-50693-1) [\(2015\)](https://doi.org/10.1140/epjd/e2014-50693-1).
- [30] P. Lähteenmäki, G. S. Paraoanu, J. Hassel, and P. J. Hakonen, Coherence and multimode correlations from vacuum fluctua-

tions in a microwave superconducting cavity, [Nat. Commun.](https://doi.org/10.1038/ncomms12548) **[7](https://doi.org/10.1038/ncomms12548)**, [12548](https://doi.org/10.1038/ncomms12548) [\(2016\)](https://doi.org/10.1038/ncomms12548).

- [31] R. Blatt and D. Wineland, Entangled states of trapped atomic ions, [Nature \(London\)](https://doi.org/10.1038/nature07125) **[453](https://doi.org/10.1038/nature07125)**, [1008](https://doi.org/10.1038/nature07125) [\(2008\)](https://doi.org/10.1038/nature07125).
- [32] M. Keller, M. Kotyrba, F. Leupold, M. Singh, M. Ebner, and A. Zeilinger, Bose-Einstein condensate of metastable helium for quantum correlation experiments, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.90.063607) **[90](https://doi.org/10.1103/PhysRevA.90.063607)**, [063607](https://doi.org/10.1103/PhysRevA.90.063607) [\(2014\)](https://doi.org/10.1103/PhysRevA.90.063607).
- [33] R. Lopes, A. Imanaliev, A. Aspect, M. Cheneau, D. Boiron, and [C. I. Westbrook, Atomic Hong-Ou-Mandel experiment,](https://doi.org/10.1038/nature14331) Nature (London) **[520](https://doi.org/10.1038/nature14331)**, [66](https://doi.org/10.1038/nature14331) [\(2015\)](https://doi.org/10.1038/nature14331).
- [34] R. Barends *et al.*, Superconducting quantum circuits at the surface code threshold for fault tolerance, [Nature \(London\)](https://doi.org/10.1038/nature13171) **[508](https://doi.org/10.1038/nature13171)**, [415](https://doi.org/10.1038/nature13171) [\(2014\)](https://doi.org/10.1038/nature13171).
- [35] Y. Ji, Y. Chung, D. Sprinzak, M. Heiblum, D. Mahalu, and H. [Shtrikman, An electronic Mach-Zehnder interferometer,](https://doi.org/10.1038/nature01503) Nature (London) **[422](https://doi.org/10.1038/nature01503)**, [1054](https://doi.org/10.1038/nature01503) [\(2003\)](https://doi.org/10.1038/nature01503).
- [36] E. Bocquillon, V. Freulon, J. M. Berroir, P. Degiovanni, B. Plaçais, A. Cavanna, Y. Jin, and G. Fève, Coherence and indistinguishability of single electrons emitted by independent sources, [Science](https://doi.org/10.1126/science.1232572) **[339](https://doi.org/10.1126/science.1232572)**, [1054](https://doi.org/10.1126/science.1232572) [\(2013\)](https://doi.org/10.1126/science.1232572).
- [37] M. Greiner, C. A. Regal, J. T. Stewart, and D. S. Jin, Probing Pair-Correlated Fermionic Atoms Through Correlations in Atom Shot Noise, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.94.110401) **[94](https://doi.org/10.1103/PhysRevLett.94.110401)**, [110401](https://doi.org/10.1103/PhysRevLett.94.110401) [\(2005\)](https://doi.org/10.1103/PhysRevLett.94.110401).
- [38] [R. J. Glauber, The quantum theory of optical coherence,](https://doi.org/10.1103/PhysRev.130.2529) Phys. Rev. **[130](https://doi.org/10.1103/PhysRev.130.2529)**, [2529](https://doi.org/10.1103/PhysRev.130.2529) [\(1963\)](https://doi.org/10.1103/PhysRev.130.2529).
- [39] A. J. H. van der Torren, S. C. Yorulmaz, J. J. Renema, M. P. van Exter, and M. J. A. de Dood, Spatially entangled four-photon states from a periodically poled potassium-titanyl-phosphate crystal, [Phys. Rev. A](https://doi.org/10.1103/PhysRevA.85.043837) **[85](https://doi.org/10.1103/PhysRevA.85.043837)**, [043837](https://doi.org/10.1103/PhysRevA.85.043837) [\(2012\)](https://doi.org/10.1103/PhysRevA.85.043837).